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TABLE OF CONTENTS

Page

APPENDICES

Page

List of tables

List of figures

Page

Acronyms and abbreviations

ACCENT	Atmospheric Composition Change the European Network of Excellence
ACE-Asia	Asian Pacific Regional Aerosol Characterization Experiment
ACPD	Atmospheric Chemistry and Physics Discussions
AEROCE	Atmosphere/Ocean Chemistry Experiment
AeroCOM	Aerosol Comparisons between Observations and Models (a global aerosol model intercomparison project)
AIRS	Atmospheric Infrared Sounder
AMMA	African Monsoon Multidisciplinary Analysis
AO	Arctic/Atlantic Oscillation
AOD	Aerosol Optical Properties
Aqua	A NASA polar-orbiting satellite
AR	Arctic
Aura	A NASA polar-orbiting satellite
BB-NO_x	global NO_x emissions from Biomass Burning
BC	Black Carbon
C	carbon
CAFE	Clean Air for Europe
CAI-Asia	Clean Air Initiative-Asia
CAIR	Clean Air Interstate Rule
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations
CAPTEX	Cross-Appalachian Tracer Experiment
CCB	Cold Conveyor Belt
CEPMEIP	Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance
CH_4	methane
CIAM	Centre for Integrated Assessment Modelling (part of EMEP)
CIFEX	Cloud Indirect Forcing Experiment
CLRTAP	Convention on Long-range Transboundary Air Pollution
CMAQ	Community Multiscale Air Quality Model
CO	carbon monoxide
CO_2	carbon dioxide
CORINAIR	Coordination of Information on the Environment – Air
CTM	Chemical Transport Model
DA	Dry Airstream
DAYS60	Number of days with 8 hours running average ozone concentrations in excess of 60 ppbv
DG Environment	Directorate-General for Environment (European Commission)
DMS	dimethylsulfide
EA	East Asia
EANET	Acid Deposition Monitoring Network in East Asia
EARLINET	European Aerosol Research Lidar Network
EC	elemental carbon
EDGAR	Emissions Database for Global Atmospheric Research
EMEP	Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
ENSO	El Niño-Southern Oscillation
ENVISAT	European Space Agency Environmental Satellite
EPER	European Pollutant Emission Register
ETEX	European Tracer Experiment
EU	European Union; in chapter 5 designates the source and receptor region mainly covering Europe
FAO	Food and Agricultural Organization of the United Nations
GAINS	Greenhouse gas and Air pollution Interactions and Synergies

GAP	Global Atmospheric Pollution Forum
GAW	Global Atmosphere Watch Programme (within WMO)
GCM	General Circulation Model
GEIA	Global Emissions Inventory Activity
GEO	Group on Earth Observations
GEOS-CHEM	A global 3-D atmospheric composition model driven by data from the Goddard Earth Observing System
GEOSS	Global Earth Observing System of Systems
GFED	Global Fire Emissions Database
GHG	Greenhouse Gases
GISS	Goddard Institute for Space Studies (part of NASA)
GOCART	Goddard Global Ozone Chemistry Aerosol Radiation Transport
GOME	Global Ozone Monitoring Experiment
H_2SO_4	sulphuric acid
HCHO	formaldehyde
Hg	mercury
HIRDLS	High Resolution Dynamics Limb Sounder
hPa	hectopascal
HTAP	Hemispheric Transport of Air Pollution
IAGOS	Integration of routine Aircraft measurements into a Global Observing System
ICAO	International Civil Aviation Organization
ICARTT	International Consortium for Atmospheric Research on Transport and Transformation
ICT	Intercontinental Transport
IEA	International Energy Agency
IFB-NO_X	NO_X from industrial activities (I), fossil fuel combustion (F) and biofuel combustion (B)
IGAC	International Global Atmospheric Chemistry
IGACO	Integrated Global Atmospheric Chemistry Observations
IGBP	International Geosphere-Biosphere Programme (part of the International Council of Scientific Unions)
IIASA	International Institute for Applied Systems Analysis
IMO	International Maritime Organization
IMPROVE	Interagency Monitoring of Protected Visual Environments
INTEX	Intercontinental Chemical Transport Experiment
IPCC	Intergovernmental Panel on Climate Change
ITCT	Intercontinental Transport and Chemical Transformation
ITCZ	Intertropical Convergence Zone
JJA	June, July, August
LRTAP	Long-range Transboundary Air Pollution
MBE	Mean Bias Error
MICS-Asia	Model Inter-Comparison Study for Asia
MILAGRO	Megacity Initiative: Local and Global Research Observations
MISR	Multiangle Imaging Spectro-Radiometer
MLS	Microwave Limb Sounder
MODIS	Moderate Resolution Imaging Spectroradiometer
MOPITT	Measurements Of Pollution In the Troposphere
MOZAIC	Measurement of Ozone on Airbus In-service Aircraft
MOZART	Model of Ozone and Related Tracers
MSA	methanesulfonic acid
MSC-E	Meterological Synthesizing Centre-East
MSC-W	Meterological Synthesizing Centre-West
NA	North America
NAAQS	National Ambient Air Quality Standards, United States
NAM	Northern Annular Mode
NAO	North Atlantic Oscillation

NARSTO	North American Research Strategy for Tropospheric Ozone
NASA	National Aeronautics and Space Administration, United States
NATAIR	Natural and biogenic emissions and assessment of impacts on air quality
NCEP	National Center for Environmental Prediction, United States
NEI	National Emission Inventory, United States
NH	Northern Hemisphere
NH_3	ammonia
NILU	Norweigan Institute for Air Research
NMHCs	Non-Methane Hydrocarbons
NMVOCs	Non-Methane Volatile Organic Compounds
NO_2	nitrogen dioxide
NO_3	nitrate
NOAA	National Oceanic and Atmospheric Administration, United States
NO_X	nitrogen oxide
NO_y	total reactive nitrogen
Nr	reactive nitrogen (combination of NO_Y and NH_X)
nss-$SO_4^=$	non-seasalt sulphate
O_3	ozone
OC	organic carbon
OECD	Organisation for Economic Co-operation and Development
OMI	Ozone Monitoring Instrument
PAN	peroxyacetyl nitrate
PBL	Planetary Boundary Layer
PCFA	Post Cold Front Airstream
PEM-West B	Pacific Exploratory Mission, Session B
PHOENICS	Particles of Human Origin Extinguishing Natural solar radiation In Climate Systems
PM	Particulate Matter
POET	Precursors of Ozone and their Effects in the Troposphere (an EU project)
POLARCAT	Polar study using Aircraft, Remote sensing, surface measurements and models, of Climate chemistry, Aerosols and Transport
POM	Polycyclic/Particulate Organic Matter
POP	Persistent Organic Pollutant
ppbv	parts per billion by volume
QUANTIFY	Quantifying the climate impact of global and European transport systems (an EU project)
RAINS	Regional Air Pollution Information and Simulation model
RAPID-C	Regional Air Pollution In Developing Countries
REAS	Regional Emissions Inventory in Asia
RETRO	Re-analysis of the tropospheric chemical composition
S/R	source-receptor
SA	South Asia
SBUV	Solar Backscatter Ultaviolet Radiance
SCIAMACHY	Scanning Imaging Absorption Spectrometer for Atmospheric Chartography/Chemistry
SH	Southern Hemisphere
SO_2	sulphur dioxide
SO_4	sulphate
SON	September, October, November
SO_X	anthropogenic sulphur (combination of SO_2 and SO_4)
SPARC	Stratospheric Processes and their Role in Climate (part of WMO)
SRES	Special Report on Emissions Scenarios
SST	Sea Surface Temperatures
STE	Stratosphere-Troposphere Exchange
TEAM model	TNO Emission Assessment Model
TERRA	A NASA polar-orbiting satellite

TES	Tropospheric Emission Spectrometer
TF HTAP	Task Force on Hemispheric Transport of Air Pollutants
TFEIP	Task Force on Emission Inventories and Projections
TOMS	Total Ozone Mapping Spectrometer
TRACE-P	Transport and Chemical Evolution over the Pacific
TRANSCOM	Atmospheric Tracer Transport Model Intercomparison Project
U.S. EPA	United States Environmental Protection Agency
UK DEFRA	United Kingdom Department of Environment, Food, and Rural Affairs
UNECE	United Nations Economic Commission for Europe
UNFCCC	United Nations Framework Convention on Climate Change
VOCs	Volatile Organic Compounds
WCB	Warm Conveyor Belt
WCRP	World Climate Research Programme (part of WMO)
WMO	World Meteorological Organization

Executive Summary

The Task Force on Hemispheric Transport of Air Pollution was established by the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution in December 2004. The Task Force was mandated to obtain a better understanding of intercontinental transport of air pollution and to provide estimates of source-receptor relationships for intercontinental air pollution. The 2007 interim assessment report provides a first report from the Task Force, which aims to provide the Convention with necessary information for its first review of the 1999 Gothenburg Protocol. The Gothenburg Protocol addresses emission sources that contribute to tropospheric ozone, acidifying and eutrophying deposition, and fine particles. The interim assessment report is based on written contributions from some 50 international experts and the deliberations of the Task Force at its third meeting, held from 30 May to 1 June 2007 in Reading, United Kingdom. The Task Force will produce a more comprehensive assessment report in 2009, which will also address mercury and persistent organic pollutants, substances also of concern to the Convention.

Major findings

Key processes driving intercontinental transport

- Observations from the ground, aircraft and satellites provide a wealth of evidence that ozone (O_3) and fine particle concentrations in the UNECE region and throughout the Northern Hemisphere are influenced by intercontinental and hemispheric transport of pollutants.

- The processes that determine the overall patterns of transport at this scale are relatively well understood and our ability to quantify the magnitude of transport is improving. Figure E.1 (below) illustrates the primary intercontinental transport pathways in the Northern Hemisphere. Our improved understanding comes from an increasing body of observational evidence, including new information from intensive field campaigns and satellite-borne instruments, improved emissions inventories and global and regional chemical transport models. The better models can reproduce much of the observed spatial and seasonal patterns of intercontinental transport, as well as describe individual transport events. However, there are differences between models with respect to quantitative estimates of source-receptor (S-R) relationships. The Task Force's hemispheric transport of air pollution (HTAP) model intercomparison has provided the first set of comparable estimates of intercontinental S/R relationships from multiple models. Continuing efforts will enable us to assess, and ultimately reduce, the variability and uncertainty in model estimates.

Estimates of source-receptor relationships

- For ground-level ozone, there is a hemispheric background concentration of 20-40 ppbv (parts per billion by volume) that includes a large anthropogenic and intercontinental component. As part of the HTAP model intercomparison, a set of emission perturbation experiments were conducted to compare model estimates of how emission changes in one region of the world impact air quality in other regions. The preliminary results of these experiments suggest that, under current conditions, local and regional emission changes have the greatest impact on surface air quality, but that changes in intercontinental transport can have small yet significant effects on surface concentrations. The benefits of measures to decrease intercontinental transport would be distributed across the Northern Hemisphere.

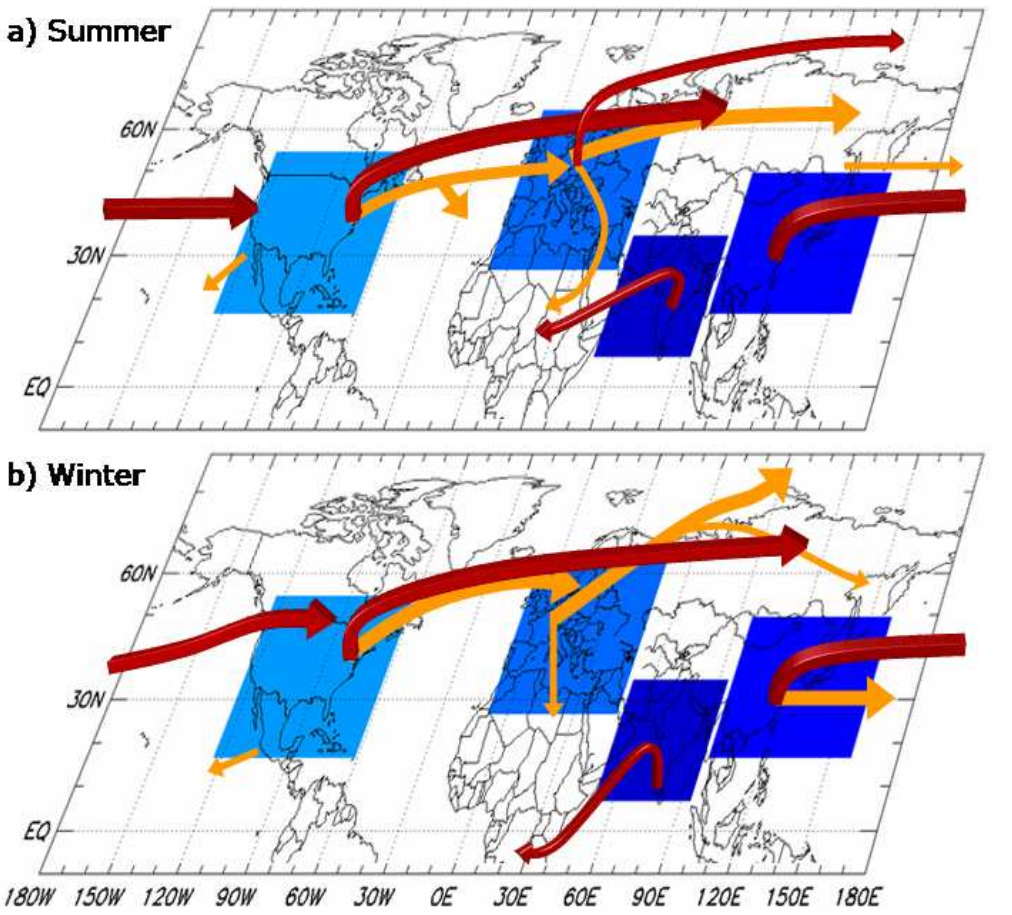

Figure E.1 Intercontinental transport pathways in the Northern Hemisphere. The coloured boxes indicate the four source and receptor regions used in HTAP intercomparison Experiment Set 1. The arrows approximate the magnitude of main transport pathways in summer (June, July, August) and winter (December, January, February), based on modelled average carbon monoxide transport over 8–10 day periods. Light arrows indicate transport generally near ground level (less than 3 km above the surface) and dark arrows indicate transport higher in the atmosphere (more than 3 km above the surface). Adapted from figure 2 of Stohl and Eckhardt (2004), with kind permission of Springer Science and Business Media.

- For fine particles, the impact of intercontinental transport on surface air quality is primarily episodic, especially associated with major emission events such as fires or dust storms. The intercontinental transport of both ozone and fine particles has large impacts on total atmospheric column loadings, which have significant implications for climate change.

- The first set of coordinated experiments under the HTAP model intercomparison examined the global impacts of 20 per cent emission reductions of relevant anthropogenic pollutants in four model regions, approximating North America, Europe, South Asia, and East Asia (see figure E-1). The model experiment results suggest that a 20 per cent decrease in anthropogenic emissions of nitrogen oxides in any three of these regions together would achieve a 30 to 70 per cent reduction in annual mean O_3 concentrations in the fourth region as compared with a 20 per cent decrease of emissions in that region itself. For the mean concentration in the peak O_3 season, this import-to-domestic response ratio is 10 to 30 per cent. The perturbation experiments also suggest that changes in anthropogenic emissions of carbon monoxide and non-methane volatile organic compounds also have significant impacts on hemispheric O_3 levels. Perturbation experiments focused on methane suggest that a 20 per cent decrease in global methane concentrations may have as large, or larger, impacts on

surface O_3 concentrations as decreases in the intercontinental transport of other O_3 precursors, as well as decreasing climate forcing of both methane and O_3.

- For fine particles, the perturbation experiments suggest that a 20 per cent change in anthropogenic emissions in any three world regions combined would achieve between 4 and 18 per cent of the change in annual mean anthropogenic fine particle concentrations (sulphate plus carbonaceous aerosols) that would result from a 20 per cent decrease in these emissions within the fourth region. The import-to-domestic response ratios for surface deposition of sulphate, reactive nitrogen, and carbonaceous aerosols are similar to those for surface concentrations. The import-to-domestic response ratio of the annual mean aerosol column loading is significantly larger than that for the surface concentrations: 30 to 59 per cent for sulphate and 13 to 30 per cent for carbonaceous aerosols.
- These results represent the average of the ensemble of participating models. There are differences between the models, as well as potential biases introduced by the experiments' design, that have yet to be investigated.

Future changes

- The significance of intercontinental transport for the achievement of environmental policy objectives may change in the future due to variation in the magnitude and spatial distribution of emissions. Such changes could be caused by the continuing implementation of pollution control measures, regional differences in the pace of economic development, the growth in shipping and aviation emissions, and the implementation of climate change mitigation measures. In addition, shifts in transport patterns and emissions sources due to climate change, as well as changes in health and environmental objectives based on new knowledge about the impacts of air pollutants, may affect the significance of intercontinental transport.

Necessity of further research

- The variability in current model estimates of transport magnitudes and the inability to explain some of the observed trends suggests that more research is needed to satisfactorily assess the significance of intercontinental transport. In particular, further efforts need to improve the accuracy and spatial and temporal resolution of emissions estimates; the spatial, temporal, vertical and chemical resolution of the current observational system; and the description of some chemical and physical processes in current models.

Recommendations

Improving our assessment of intercontinental and hemispheric transport will require an integrated approach in which the best available knowledge from observations, emissions and models is combined. A robust observational system using multiple observational platforms and methods is needed to provide data for the evaluation and improvement of chemical transport models and emissions inventories. Further analytical efforts planned by the Task Force over the next few years are expected to decrease the range of current model estimates for S-R relationships and improve our confidence in the assessment of intercontinental S-R relationships.

Key Challenges

Some of the key challenges that we face are outlined below. Addressing each of these challenges requires the linking of information across the areas of observation, emissions and modelling:

- To improve the modelling of transport processes using existing and new field campaign data. Focused evaluations of models using field campaign data are needed to improve descriptions of small-scale boundary layer venting, atmospheric subsidence, wet scavenging and transport processes in the tropics.
- To improve global emissions inventories using existing information at the national and subnational scale and using inverse modelling and other methods to compare emissions estimates to ground-based, aircraft and satellite observations.

- To identify and explain observed long-term trends by filling gaps in the observing system, developing reliable emission trends and improving model descriptions. The current observational system has limited coverage and resolution in most regions of the world and provides limited information about the vertical distribution of pollutants. Better observational information is essential for improving the ability to detect and explain long-term changes.
- To develop a robust understanding of current source-receptor relationships using multiple modelling techniques and analyses of observations. The initial results of the HTAP intercomparison provide some useful information about the significance of intercontinental transport, but further detailed analyses are needed.
- To estimate future S-R relationships under changing emissions and climate. Such scenarios should consider future years from 2020 to 2050 and 2100 and be coordinated with efforts under the Intergovernmental Panel on Climate Change.
- To improve organizational relationships and information management infrastructures to facilitate necessary research and analysis. Efforts should further the implementation of the strategy for Integrated Global Atmospheric Chemistry Observations, building upon the World Meteorological Organization's Global Atmospheric Watch program and contributing to the Global Earth Observation System of Systems.

Role of the Task Force

Addressing the challenges outlined above will require the combined effort of many individual scientists, national research organizations, international research programmes, (e.g. the International Geosphere-Biosphere Programme and the World Climate Research Programme) as well as governmental authorities. In this community effort, the Task Force can continue to play an important role as a forum:

- For identifying scientific consensus concerning the current understanding of intercontinental and hemispheric transport and the priorities for future research and development as well as a forum for fostering information exchange and collaboration.
- For raising awareness of transboundary and intercontinental air pollution in regions where the concept is less well known and for facilitating crucial links among institutions both within countries and across regional and hemispheric scales.

References

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1. INTRODUCTION

Current emissions create pollution levels that exceed air quality standards and other environmental objectives at many locations throughout the Northern Hemisphere. While local or regional emissions and environmental conditions are responsible for most of these problems, air quality and pollutant deposition are also influenced by emissions, transport and transformation processes at the intercontinental and global scales. There is well-documented evidence for the intercontinental transport of ozone, fine particles, and their precursors, as well as mercury and some persistent organic pollutants (POPs). The significance of this intercontinental influence for the design of air pollution control policies, however, has not been well understood.

This interim report summarizes the current state of understanding of the significance of intercontinental transport and hemispheric pollution on ozone and fine particle concentrations and sulphur and nitrogen deposition in the Northern Hemisphere. This report was produced by the Task Force on Hemispheric Transport of Air Pollution of the 1979 Convention on Long-range Transboundary Air Pollution, primarily for informing the first review of the Convention's 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone; it may also be informative for other international organizations and national administrations. This is an interim report, prepared in anticipation of a more comprehensive report planned for 2009 aimed at addressing in depth a number of policy-relevant science questions adopted by the Task Force at its first meeting (see box 1). The 2009 report aims to include intercontinental transport of mercury and POPs, which are also of concern for the Convention.

This report builds on written contributions from about 50 experts and addresses important aspects of assessment of intercontinental transport of air pollution. The authors of the report were drawn from a roster of experts set up following a general invitation to nominate authors. Draft chapters of the report were posted on the website of the Task Force (http://www.htap.org) with a general invitation to provide comments. The chapters were updated in the light of comments and the revised drafts were presented to the Task Force at its third meeting held from 30 May to 1 June 2007 in Reading, United Kingdom. At that meeting, all main elements of the report were agreed. The Co-Chairs of the Task Force finalized the executive summary for submission to the Convention's Steering Body of EMEP[1] in September 2007. Amendment and alignment of the chapters took place during the summer of 2007.

The report begins, in chapter 2, with a conceptual overview of hemispheric and intercontinental transport of ozone and fine particles in the Northern Hemisphere. Chapter 3 summarizes the observational evidence from surface sites and networks, aircraft and field campaigns, and satellite instruments for hemispheric and intercontinental transport; it discusses the current state of observational systems for characterizing intercontinental transport. The status and implications of available inventories and future projections for anthropogenic and natural emissions are discussed in chapter 4. Chapter 5 describes different

[1] The Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.

approaches using models for characterizing hemispheric pollution and intercontinental transport and summarizes available modelling results for ozone, fine particles and deposition, including the initial results of the ongoing model intercomparison and evaluation effort organized under the Task Force. Each chapter includes a discussion of recommendations for future research and analysis. Chapter 6 summarizes the main conclusions of this interim assessment and key recommendations for future work as related to the seven policy-relevant science questions in box 1. An overview of the background and activities of the Task Force between 2004 and summer 2007 is given in appendix A. Appendix B provides definitions for some of the transport-related terms used in the report.

Box 1: The Task Force on Hemispheric Transport of Air Pollution's Policy-relevant Science Questions

1. How does the intercontinental or hemispheric transport of air pollutants affect air pollution concentration or deposition levels in the Northern Hemisphere for ozone and its precursors, fine particles and their precursors, compounds that contribute to acidification and eutrophication, mercury, and POPs?
   - What evidence do we have of transport pathways and mechanisms from intensive field studies? From observations? From model predictions?
   - How do the transport pathways differ by pollutant? By source region? By season?
   - What processes need to be better understood to describe the relative significance of intercontinental transport?
   - How do processes at the intercontinental or hemispheric scale affect processes at the local or global scales? (Synoptic scale meteorological events/cycles, Hadley circulation, etc.)

2. More specifically, for each region in the Northern Hemisphere, can we define source-receptor relationships and the influence of intercontinental transport on the exceedance of established standards or policy objectives for the pollutants of interest?
   - What observational evidence exists for attributing pollutant concentrations or deposition levels to source regions or countries?
   - Using predictive chemical transport models, what are possible methods for calculating S/R relationships? At what spatial resolution (geographic region, individual countries) can such methods be reasonably applied?
   - How can models with different spatial resolutions be nested within one another to provide an appropriate level of spatial resolution for the entire hemisphere or globe?
   - What improvements are needed for global and regional transport models to better simulate atmospheric processes to enhance S/R predictions?

3. How confident are we of our ability to predict these S/R relationships? What is our best estimate of the quantitative uncertainty in our estimates of current source contributions or our predictions of the impacts of future emissions changes?
   - What metrics and techniques are most appropriate for evaluating global and regional model simulations with observations and for quantifying uncertainties?
   - Do we have a sufficient database of observed concentrations and deposition levels to evaluate the predictions of current models? How can this observational database be improved for the purposes of evaluating models? Should we develop a set of standard observational platforms and measurements to enhance data consistency globally?
   - Do we have sufficient observational databases to track long term progress and change in transport and deposition patterns?

- Do we have sufficient data on emissions and the trends in driving forces needed for making reasonable future projections? How can this data be improved?
- What physical or chemical processes must be better understood to improve our confidence in our estimates of S/R relationships? What is the minimum level of certainty in our understanding of these processes that must be attained before reasonable/useful estimates can be made?

4. For each country in the Northern Hemisphere, how will changes in emissions in each of the other countries in the Hemisphere change pollutant concentrations or deposition levels and the exceedance of established standards or policy objectives for the pollutants of interest?
   - Is there a simple relationship between changes in emissions and changes in pollutant concentrations and deposition levels?
   - How is the predicted relationship affected by the spatial resolution of the model?

5. How will these source-receptor relationships change due to expected changes in emissions over the next 20 to 50 years?
   - How might emission quantities and spatial distributions change over the next 20 to 50 years?
   - How should future emission scenarios be constructed?

6. How will these source-receptor relationships be affected by changes in climate or climate variability?
   - How will meteorological changes predicted by climate modelling studies affect major transport or chemical processes?
   - Are there significant feedbacks between the transported air pollutants and regional climate and meteorology?
   - Are there significant feedbacks between transported air pollution and potential changes in land use, vegetation or ecosystems, especially with respect to natural emission sources?
   - Are there predictive relationships between climate system indices that can be used to estimate the impact of changing climates on hemispheric transport of air pollutants?

7. What efforts need to be undertaken to develop an integrated system of observational data sources and predictive models that address the questions above and leverage the best attributes of all components?

2. CONCEPTUAL OVERVIEW OF HEMISPHERIC OR INTERCONTINENETAL TRANSPORT PROCESSES

This chapter describes the most important aspects of hemispheric transport processes, focusing on the rapid intercontinental transport of large pollution plumes by mid-latitude cyclones, the venting of the atmospheric boundary layer by deep convection, and small-scale processes that contribute to increasing the background concentrations of various pollutants. The impacts of these transport processes are discussed for a variety of pollutants. To be brief, only one or two pollutant examples are provided for each process and these were chosen according to the greatest amount of information available. Issues related to the influence of climate change on transport processes are also addressed. The chapter concludes with a discussion of outstanding issues. Definitions for some of the transport-related terms are provided in appendix B.

2.1 Major emissions regions

The magnitude and impact of hemispheric and intercontinental scale transport of pollutants is largely controlled by the global distribution of the human population and anthropogenic emissions, as well as their spatial relation to the major meteorological transport pathways. To place the emissions in context, focus is placed primarily on nitrogen oxide (NO_x) emissions, which are important for ozone formation and acid deposition. Global emissions of NO_x from industrial activities (I), fossil fuel combustion (F) and biofuel combustion (B), collectively referred to as IBF-NO_x, are estimated to be 30.3 Tg N yr^{-1} (year 2000 EDGAR1[1] estimate) (Olivier and Berdowski, 2001). Global NO_x emissions from biomass burning (referred to as BB-NO_x) are estimated to be 7.5 Tg N yr^{-1}. Estimates of natural sources of NO_x from lightning and soil vary widely but are considered to be about one third of the anthropogenic emissions (IPCC, 2001). The Northern Hemisphere contains the great majority of humans (88%) and 91 per cent of IBF-NO_x emissions, and 41 per cent of BB-NO_x emissions (figure 2.1). Per capita emissions vary widely across nations, but broadly speaking, North America has the highest per capita IBF-NO_x emissions, while Asia and the northern Africa/Middle East region have the lowest (figure 2.2). The major IBF-NO_x emission regions of the Northern Hemisphere are located in the eastern United States and southeastern Canada, western and central Europe, and southern and eastern Asia (figure 2.1). Emissions of other trace gases related to industrial activity and biofuel and fossil fuel combustion and usage such as sulphur dioxide (SO_2), carbon monoxide (CO), and volatile organic compounds (VOCs) have similar distributions.

Less than 10 per cent of global biomass burning is due to wildfires which occur primarily in temperate and boreal forests (Crutzen and Andreae, 1990). By far the majority of global biomass burning occurs in the tropics and is mainly caused by human activity. The primary biomass burning regions of the Northern Hemisphere are sub-Saharan northern Africa, Southeast Asia, Central America and northern South America (figure 2.1). On an inter-seasonal basis, the IBF-NO_x emissions are relatively constant, while BB-NO_x emissions have a strong seasonal variation as well as interannual variability. Biomass burning peaks in December-February for sub-Saharan northern Africa, in February-April for Southeast Asia, in April-May for Central America, and in March for northern South America (Duncan et al., 2003). Biomass burning is also a major source of global CO emissions.

VOCs include a wide variety of non-methane hydrocarbons (NMHCs) and oxygenated NMHCs. VOCs sources fall into three main categories: (a) emissions from fossil fuel production; distribution and combustion; (b) biomass burning; and (c) vegetation (IPCC, 2001). The largest global source is from vegetation, about two-thirds of all emissions, emitted primarily in the tropics. VOCs emissions from fossil fuel usage (approximately 25% of total emissions) and biomass burning (about 5% of total emissions) have distributions similar to NO_x (figure 2.1).

The distribution of methane (CH_4) emissions resulting from anthropogenic activity is slightly different from trace gases strongly associated with fuel combustion and biomass burning. Year 2000

[1] Emissions Database for Global Atmospheric Research.

estimates of global anthropogenic methane emissions indicate that livestock and rice cultivation accounted for 37 per cent of the total, with 29 per cent coming from fossil fuel production (Olivier and Berdowski, 2001). In general, the distribution of the methane emission regions resembles the global human population distribution, with India and China being the two regions with the greatest emissions. Anthropogenic methane emissions are 50–70 per cent of global emissions, with the primary natural source being wetlands (IPCC, 2001). Similar to methane, anthropogenic ammonia (NH_3) emissions are closely tied to agriculture and domestic animals, with these two sources accounting for two thirds of global emissions from anthropogenic and natural sources (IPCC, 2001).

Airborne particulate matter has a broad range of primary and secondary sources. Sea salt and mineral dust dominate global primary particle emissions, with anthropogenic sources making only a small contribution, albeit in regions that are generally more heavily populated than regions where sea salt (oceans) and mineral dust (deserts) are emitted. Particulate matter (PM) formed as secondary aerosols is formed from precursor emissions of SO_2, NO_x, NH_3, VOCs and dimethylsulfide (DMS). The major mineral dust emission regions are associated with topographical lows in the desert regions of North Africa, the Middle East, China, and Central and South Asia, all in the Northern Hemisphere (Prospero et al., 2002). The largest dust storms associated with intercontinental transport originate in the Gobi Desert, which can reach western North America in spring (Liu et al., 2003), and in North Africa, which routinely impact the Caribbean and southern United States during summer (Prospero, 1999).

The location of the emissions regions with respect to the dominant atmospheric transport patterns has a strong influence on the frequency and strength of intercontinental pollution transport events. For example, the emissions regions along the east coasts of Asia and North America are at the origins of the North Atlantic and North Pacific mid-latitude cyclone storm tracks, which can loft the emissions and transport them to the free troposphere above downwind continents in a matter of days. With western Europe located at the end of the North Atlantic storm track, its emissions are not lofted to the same extent as those on the east coasts of Asia and North America. Instead, European emissions are exported at relatively low altitudes and have a strong impact on the Arctic (Duncan and Bey, 2004; Stohl et al., 2002). These relationships will be described in more detail below.

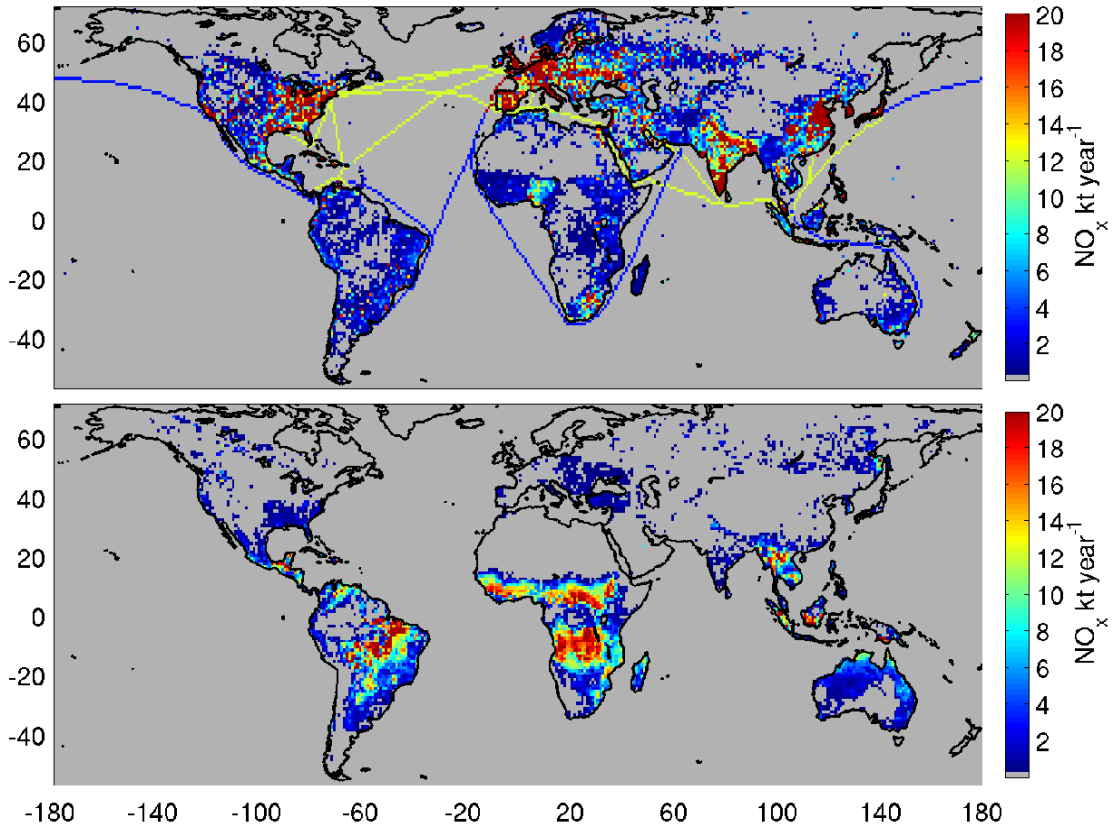

Figure 2.1 Distribution of global IBF-NO_x emissions (top), and BB-NO_x from both natural and anthropogenic fires (bottom), according to the EDGAR 3.2 Fast Track 2000 data set, which estimates year 2000 emissions using the EDGAR 3.2 estimates for 1995 and trend analyses for the individual countries. EDGAR uncertainty estimates are roughly 50 per cent or greater (Olivier and Berdowski, 2001).

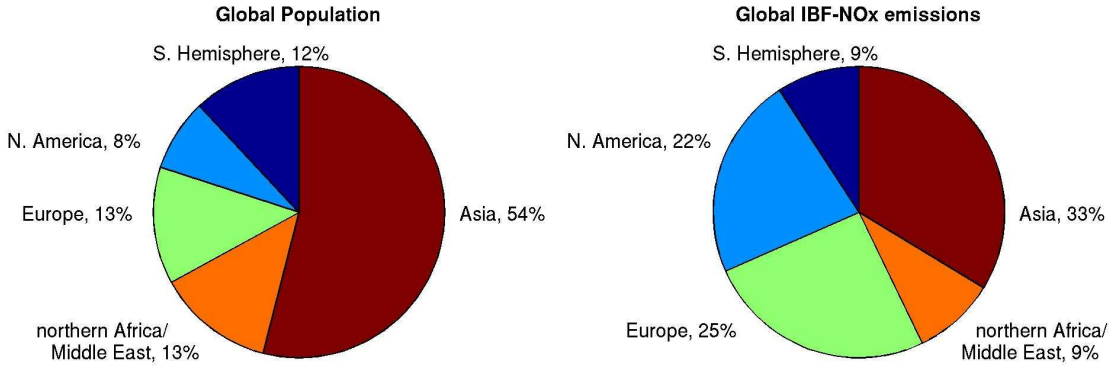

Figure 2.2 Global distribution of human population and year 2000 IBF-NOx emissions. Population data from CIA (2007), emissions data from EDGAR 3.2 (Olivier and Berdowski, 2001).

2.2 Source-receptor relationships

The term "source-receptor relationship" describes the sensitivity of concentrations or deposition at a "receptor" location to a changes in emissions from a "source" location (Seibert and Frank, 2004); it is a key concept used in this document to assess the impact of emissions from an upwind continent or region to a downwind receptor location (Derwent et al., 2004; Fiore et al., 2002; Wild et al., 2001). In general, source-receptor relationships are dependent on the emissions strength of the source; the transport pathway from the source to receptor; and the pollutant transformation, production, and loss processes that occur along the pathway. The transport component of a source-receptor relationship can be expressed independently of the strength of emissions from the source region and is a measure of (a) the quantity of air from the source that reaches the receptor over a given time period; and (b) the amount of time the air spends over the source region. Take, for example, Yosemite National Park in eastern California, located at times downwind of San Francisco. When the prevailing winds transport air directly from San Francisco to Yosemite, the transport source-receptor relationship would be stronger than on a day when the wind did not produce such a direct transport path. Or given two separate days when the wind produces a similar transport pathway from San Francisco to Yosemite, the day with the stronger transport source-receptor (S/R) relationship would be the day when the air spends more time above San Francisco.

Another important term is the source contribution, which is a measure of the quantity of a particular pollutant at a receptor that was emitted from a particular source. The source contribution depends upon (a) the transport S/R relationship; (b) the emission flux of the pollutant from the source region; and (c) any loss (or production) of the pollutant as it is transported from the source to the receptor. For example, the source contribution made by San Francisco to NO_x concentrations at Yosemite is determined by multiplying the transport source-receptor relationship by the NO_x emission flux from San Francisco and subtracting any NO_x molecules that are removed from the pollution plume by physical or chemical processes during transit.

In terms of transport pathway, the source contribution is influenced by (a) the direct transport of plumes from the source to the receptor site; and (b) the contribution of the source to the overall background concentrations at a receptor site (i.e. the fraction of the observed concentration that cannot be directly attributed to local emission sources or discrete transport plumes). While the direct transport of a plume involves a distinct episodic transport event occurring on a relatively short time and spatial scale, the background concentration is the result of the cumulative effect of episodic transport events on longer time and spatial scales.

In terms of constituent loss of the pollutant, the source contribution is affected by chemical transformations, scavenging, deposition and sedimentation processes. The relative impact of these processes varies both spatially and temporally. Within the atmospheric boundary layer, transport speeds are often slow, chemical loss is frequently high, and surface deposition is important. For pathways where air parcels are lofted from the atmospheric boundary layer (e.g. by convection or mid-latitude cyclones), the resultant cooling is usually sufficient to cause precipitation and, therefore, wet scavenging of aerosols and soluble species. The source contribution for species with rapid losses (e.g. lifetimes of a few days) will be determined by the fastest and most direct transport pathways between the source and the receptor, with the background concentration essentially zero far from the source. In contrast, the source contribution of long-lived species (e.g. methane, with an atmospheric lifetime of 8–9 years) is primarily determined by the contribution of the source to the background concentration (see table 2.1 for a list of approximate pollutant lifetimes).

As will be described in further detail below, the most spectacular pollution transport events are related to export from the east coasts of North America or Asia with subsequent transport to the west coasts of Europe and North America, respectively. While the concentrations of pollutants in these plumes are quite high in the free troposphere above the downwind continents, the receptor locations of most interest are the surfaces of the downwind continents where humans live. To impact the surface, the plumes must subside several kilometers from the middle or upper troposphere, which can take several days over which time the pollutant concentrations are diluted. While these subsiding plumes can have strong impacts on remote locations such as the Azores in the North Atlantic or mountaintop sites such as the Alps, their impact in highly populated low-lying areas is masked by the

local pollution. Thus the question of the impact of pollution from an upwind source on a receptor located in a locally polluted atmospheric boundary layer appears to be an issue of the degree to which a plume enhances the local pollution rather than that of the plume replacing local pollution with concentrated pollution from an upwind continent.

Table 2.1 Approximate lifetimes of trace gases and particulate matter in the atmospheric boundary layer and the free troposphere. Lifetimes are highly variable depending on altitude, time of day and year, and proximity to polluted regions.

Trace gas or PM	Approximate lifetime in the atmospheric boundary layer	Approximate lifetime in the free troposphere
NO_2	hours	days
SO_2	days	days to weeks
CO	Weeks to months	weeks to months
VOCs	hours to months	hours to months
CH_4	8-9 years	8-9 years
NH_3	days	days
PM	hours to days	days to weeks
O_3	hours to days	weeks to months

2.3 Major types of intercontinental transport processes

2.3.1 General circulation regimes

The great majority of intercontinental pollutant transport occurs within the troposphere which extends from the surface to about 8 km in cold polar regions, and up to about 18 km in hot tropical regions (figure 2.3). Tropospheric circulation can be divided into three regimes: the subtropics and tropics, the mid-latitudes (30^o-60^o) and the polar regions. The tropics and subtropics are dominated by the Hadley cell, with rising motion within deep convective cumulus towers in the deep tropics (often within the Intertropical Convergence Zone), which is balanced by subsident motion in the subtropics. The meridional winds associated with the Hadley cell are poleward throughout much of the free troposphere, but with a return equatorward flow confined near the surface (figure 2.3). In the tropics and subtropics, intercontinental transport is generally from east to west, with the trade winds throughout the lower and middle troposphere (figure 2.4). For much of the year, the mid-latitudes are dominated by mid-latitude cyclones with strong meridional stirring along tilted surfaces of constant potential temperature (see discussion below and figure 2.4). During the summer months, however, deep convective processes play an important role over land. The following considerations apply to mid-latitude transport (see figures 2.3 and 2.4):

(a) Mid-latitude zonal mean winds are generally westerly throughout the troposphere, causing intercontinental transport to primarily occur from west to east.

(b) Wind speeds generally increase with height, causing pollutants at higher altitudes to be transported rapidly. This is especially true in the vicinity of the polar and subtropical jet streams, which vary in location and intensity from day to day. Thus, processes that loft pollutants into the middle to upper troposphere are most conducive to long-range intercontinental transport.

(c) Winds are generally stronger in winter than in summer, causing intercontinental transport to be more rapid during winter months.

(d) Meridional winds are generally weaker than zonal winds, thus pollutants tend to be transported zonally.

The lower Arctic troposphere tends to be isolated to some extent from low latitude emissions, particularly in summer. Pollutant transport into the lower Arctic troposphere occurs preferentially from Europe. Of the three regions (tropical, mid-latitude and polar), transport processes are best understood in the mid-latitudes.

A number of studies have shown impediments to transport (i.e. transport barriers) between the extra-tropics and the tropics (Bowman and Carrie, 2002; Bowman and Erukhimova, 2004; Hess,

2005; Pierrehumbert and Yang, 1993). These barriers imply that air within the mid-latitudes tends to recirculate within the mid-latitude region, and air within the tropics and subtropics tends to recirculate within the tropics and subtropics. Thus, the background concentration at mid-latitude sites is most immediately affected by mid-latitude emissions, with a much smaller contribution from tropical sources. The mass exchange between the extratropical and tropical regions is estimated to be 1–2 per cent per day of the extratropical mass, with lag times between the Northern and Southern Hemisphere tracer concentrations approaching 2 years (Bowman and Erukhimova, 2004). Equatorward transport between the extra-tropics and tropics occurs primarily in shallow flow near the Earth's surface associated with the equatorward branch of the Hadley circulation, and often occurs behind cold fronts (Bowman and Carrie, 2002; Bowman and Erukhimova, 2004). The return poleward flow occurs in a more diffuse circulation in the upper troposphere associated with the upper branch of the Hadley cell.

Air masses tend to follow transport paths that lie along surfaces of constant potential temperature. The potential temperature is a measure of an air mass's temperature, adjusted for the temperature changes that are caused by variations in atmospheric pressure as the air mass is transported vertically. In general, the potential temperature of the atmosphere increases with altitude and an air mass with a typical (for the middle to upper troposphere) potential temperature of 310 ^{o} K that lies above an air mass with a potential temperature of 305 ^{o} K is in a thermally stable position with respect to the air mass below. These two air masses will not mix unless energy is added to the lower layer or removed from the upper layer so that the two air masses achieve equal potential temperature. As a result, surfaces of constant potential temperature strongly constrain atmospheric transport (figure 2.4), which follows surfaces of constant potential temperature in the absence of heating. Sensible heating at the Earth's surface and the latent heat of condensation are the primary processes by which the atmosphere is warmed (increasing a parcel's potential temperature); these processes are balanced in the mean by radiative cooling (which decreases a parcel's potential temperature by 1–2 degrees per day, on average). In the free troposphere, heating generally occurs on small spatial and temporal scales, due in part to the condensation of water vapor within convective updrafts or within the warm sector of mid-latitude cyclones, particularly the warm conveyor belt. On the other hand, subsidence and tropospheric cooling occur on large spatial scales and long timescales.

At the large scale, transport tends to occur primarily along potential temperature surfaces. These surfaces are generally oriented upward and poleward (figure 2.4); therefore, poleward moving air tends to ascend, while equatorward moving air tends to sink. Mid-latitude meridional stirring occurs along constant potential temperature surfaces on timescales up to 10 days (Bowman and Carrie, 2002; Bowman and Erukhimova, 2004). Transport along potential temperature surfaces that intersect the surface of the Earth occurs primarily through the action of mid-latitude cyclones (Hess, 2005). However, above the 310^{o} K potential temperature surface,

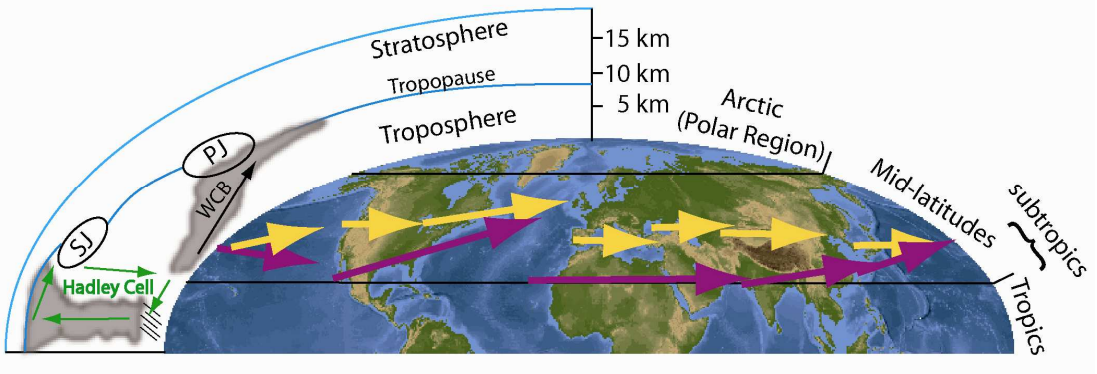

Figure 2.3 Schematic diagram showing some of the main features of the atmosphere related to the transport of air pollutants. The vertical cross-section shows the average location of the tropopause (the boundary between the troposphere and stratosphere) and the polar (PJ) and subtropical (SJ) jet streams during winter. Vertical transport during winter is dominated by deep convective clouds in the tropics (the upward branch of the Hadley cell) and warm conveyor belts (WCB) in the mid-latitudes, as described in Section 2.3.2. The average location of the jet stream is shown across the entire Northern Hemisphere for winter (magenta arrows) and summer (yellow arrows).

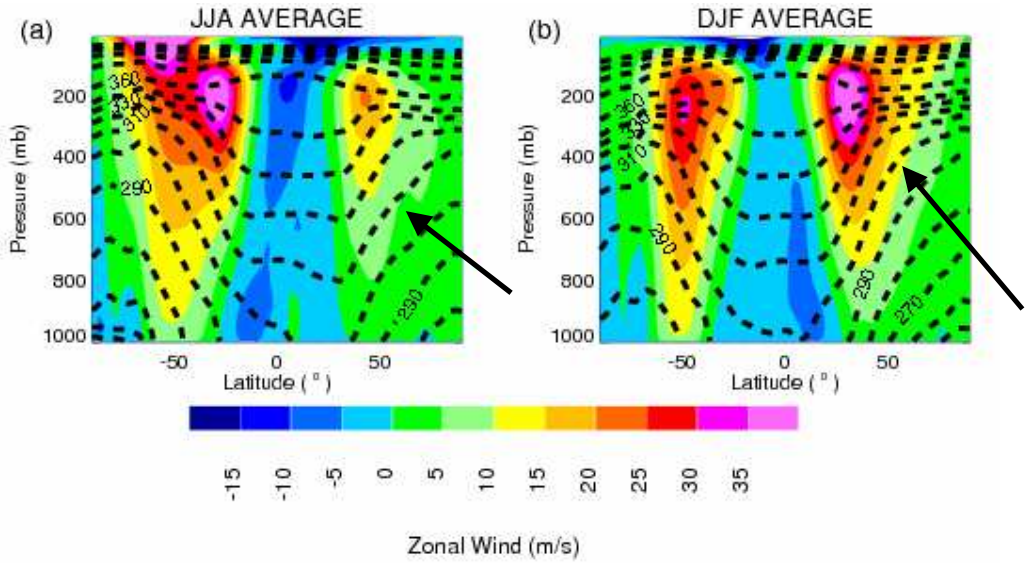

Figure 2.4 Zonal winds and potential temperatures. Zonal winds (colour) and potential temperature (dashed lines) for (a) June, July and August (JJA) and (b) December, January and February (DJF). Fields are from 5 years of the National Centers for Environmental Prediction (NCEP) reanalysis. Potential temperature surfaces are contoured every 10^{o} between 250^{o} K and 340^{o} K and every 40^{o} between 360^{o} K and 520^{o} K. Arrows indicate the 310^{o} K potential temperature surface above which air parcels have been transported vertically through convective clouds. Positive zonal winds indicate winds from the west, while negative zonal winds indicate winds from the east.

most of the air parcels have been vertically transported through a deep convective cloud on a timescale of 40 days (Hess, 2005). Therefore, most of the tropics and subtropics and upper and middle troposphere during the Northern Hemisphere summer are primarily influenced by the transport through deep convection (see figure 2.4). Due to their elevated potential temperatures, transport pathways through deep convective clouds will likely influence surface concentrations only on the long timescales required for subsequent radiative cooling. As a result, radiative cooling impedes rapid long-range transport from warm to cold regions.

2.3.2 The mid-latitude cyclone airstreams

Mid-latitude cyclones tracking from west to east are important mechanisms for the export of trace gases and PM from the east coasts of Asia and North America throughout the year, even in summer when these weather systems are weaker (Cooper et al., 2002a; Cooper et al., 2002b; Merrill and Moody, 1996; Stohl et al., 2002). The cyclones are typically composed of four airstreams (figure 2.5) that influence trace gas mixing ratios and relationships in the troposphere (Bethan et al., 1998; Cooper et al., 2001; Stohl and Trickl, 1999). Three of these airstreams, the warm conveyor belt, cold conveyor belt, and dry airstream produce the distinctive comma cloud of a mature mid-latitude cyclone (Bader et al., 1995; Browning and Monk, 1982; Browning and Roberts, 1994; Carlson, 1998). The warm conveyor belt is located on the eastern side of the cyclone, ahead of the surface cold front. The air originates at low altitudes in the warm sector of the cyclone and travels poleward, ascending into the middle and upper troposphere, above the cold conveyor belt. The dry airstream, which is associated with stratospheric intrusions, originates at high altitudes in the upper troposphere and lowermost stratosphere on the poleward side of the cyclone and descends into the middle and lower troposphere on the polar side of the cold front. The post cold front airstream is the cold, dry, and stable air mass in the lower and middle troposphere that flows behind the cyclone cold front and beneath the dry airstream (Cooper et al., 2001).

The warm conveyor belt is the most important airstream for rapid intercontinental pollutant transport because of its ability to loft polluted atmospheric boundary layer air from the cyclone warm sector to the upper troposphere in the vicinity of the jet stream. Following the mid-latitude cyclone

storm tracks, the jet stream then rapidly transports the pollutants downwind. Transport times from the moment North American atmospheric boundary layer air is lifted within the warm conveyor belt until it reaches the European free troposphere are typically three to four days (Eckhardt et al., 2004; Stohl et al., 2002), and in some cases less than 2 days (Stohl et al., 2003b). Several additional days are required for the North American emissions to reach the European surface with the greatest influence over the Mediterranean due to trapping and anticyclonic descent of the emissions through the Azores' high (Stohl et al., 2002). The greater distance associated with trans-Pacific transport results in slightly longer transport times, and in some instances requires two warm conveyor belts to transport emissions from Asia to North America (Cooper et al., 2004).

Mid-latitude cyclones can also export pollutants from Asia and North America at low altitudes as discussed in section 2.2.5, either when the warm sector of the storm pushes offshore and the warm conveyor belt is too weak to loft the pollutants, or when the cold stable air in the post cold front airstream quickly advects fresher emissions offshore.

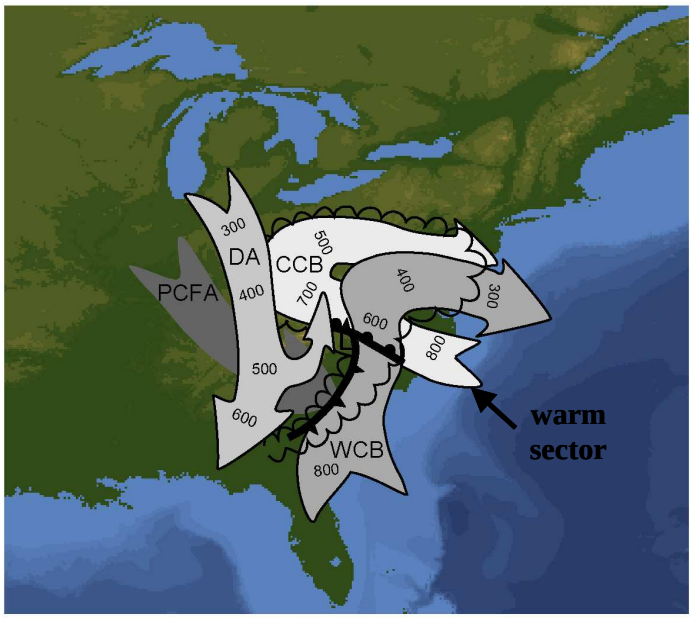

Figure 2.5 Conceptual model of the airstreams within a mid-latitude cyclone: warm conveyor belt (WCB), cold conveyor belt (CCB), dry airstream (DA), and the post-cold front airstream (PCFA). The relationships of the airstreams to the centre of the surface low (L) and the warm and cold fronts are shown, as is the edge of the cloud shield formed by the WCB and CCB (scalloped lines). The numbers on the WCB and CCB indicate the pressure (hPa) at the top of these airstreams, while numbers on the DA indicate the pressure at the bottom of the airstream. After Cooper et al. (2002b).

2.3.3 Deep convection

Deep convection is an important mechanism for vertically transporting air pollutants out of the atmospheric boundary layer into the middle and upper troposphere (Dickerson et al., 1987; Lelieveld and Crutzen, 1994), where the stronger winds can rapidly transport them across intercontinental distances. Convection is triggered when the Earth's surface is warmer than the overlying air, creating unstable conditions such as during daytime over land or when cooler air masses are advected over a warm ocean surface. Condensation of water vapour in convective cells releases latent heat and leads to cloud formation.

Convective systems encompass small-scale fair weather cumuli, active thunderstorms (cumulonimbus clouds), and mesoscale convective systems (Cotton et al., 1995). The corresponding lifetime of these systems increases with their size from minutes to about half a day. Another weather system shaped by organized deep convection is the tropical cyclone (including tropical depressions, tropical storms, hurricanes and typhoons), whose lifetime is in the order of a week. Furthermore, in

summertime over land, even the warm conveyor belts of mid-latitude cyclones are characterized by embedded deep convection, and the distinction between deep convection and warm conveyor belt ascent becomes somewhat arbitrary (Kiley and Fuelberg, 2006). An extreme form of convection is the so-called pyro-convection occurring over large forest fires, which can inject large quantities of aerosols and trace gases into the upper troposphere and also deep into the stratosphere (Fromm et al., 2000; Fromm et al., 2005; Jost et al., 2004).

Globally, the mass flux out of the atmospheric boundary layer due to deep convection is comparable to the mass flux caused by the large-scale ascent in mid-latitude cyclones (Cotton et al., 1995). In addition, the ascent in deep convective cells takes only minutes, whereas the ascent in cyclones takes from hours up to two days. For trace gases with a rather short lifetime in the lower troposphere, this has the consequence that they can reach the upper troposphere in convective cells but not within warm conveyor belts. For instance, Speidel et al. (2007) observed large enhancements of SO_2 in the upper troposphere over Europe, after lifting by deep convection over North America followed by intercontinental transport.

2.3.4 Diffuse or small-scale atmospheric boundary layer venting

Export of pollution from the atmospheric boundary layer to the free troposphere can occur whenever an air parcel is transferred above the boundary-layer height. Since over land the atmospheric boundary layer has a distinct diurnal cycle with a maximum during the day and a minimum during the night, a residual layer is formed upon the transition from day to night (Stull, 1988). This residual layer is decoupled from the surface and experiences higher wind speeds than the air in the atmospheric boundary layer, particularly when a nocturnal low-level jet is present (Angevine et al., 1996). The residual-layer air can (partly) remain in the free troposphere the next day if the atmospheric boundary layer is less deep than on the previous day, or if other vertical transport processes lift it to higher altitudes.

Topography has a large influence on vertical pollution transport, as it generates variability in the atmospheric boundary layer height, and the formation and breaking of gravity waves above the atmospheric boundary layer, which can cause mixing between different air masses. In particular, the mountain-valley wind systems encountered in mountainous regions can trigger vertical lofting. For instance, Henne et al. (2004) estimated that under fair weather conditions in summer, three times the valley volume can be lofted into the free troposphere per day. Once pollutants are vented from the atmospheric boundary layer they are subject to the long-range transport processes discussed above. On the other hand, during stable conditions (particularly in winter), pollutants can also accumulate in the valley atmosphere, preventing them from being transported over long distances. Mountain ranges also influence the large-scale flow, sometimes preventing transport over and across the mountain range.

2.3.5 Slow, low altitude flow

Air masses can also be transported over long distances without being lifted. Often, this involves the formation of a residual layer following the collapse of a daytime atmospheric boundary layer. It can also happen in a strongly stable atmosphere, where hardly any convective atmospheric boundary layer forms, for instance at high latitudes in winter. Because dry deposition and the potential for cloud formation are limited under such conditions, aerosols and trace gases can be transported over long distances, even though transport speeds are lower than in the upper troposphere. Arctic haze (Barrie, 1986), which can cover large parts of the Arctic in winter and spring, is often the result of such low-level long-range transport (Klonecki et al., 2003; Stohl, 2006). The phenomenon has also been observed downwind of North America, where layers with extremely high concentrations of oxidized nitrogen were found far downwind over the North Atlantic Ocean (Neuman et al., 2006). These layers can even reach the Azores (Owen et al., 2006) and probably Europe (Guerova et al., 2006; Li et al., 2002). Similar transport pathways have been identified across the Pacific (Holzer et al., 2005; Liang et al., 2004). The large-scale haze layers over the Indian Ocean generated by the winter monsoon outflow from southern Asia also have limited vertical extent until the flow reaches the intertropical convergence zone (Ramanathan et al., 2001b).

2.4 Impact of intercontinental transport pathways on global and local pollution distributions

2.4.1 Impact of large-scale export events

The major pathways of intercontinental transport are summarized in figure 2.6, while the global distribution of tracers from the world's major population centers are shown in figure 5.1 in chapter 5. Large-scale export events from Asia and North America have strong episodic impact on the chemical composition of the free troposphere above downwind continents, while their impact on surface sites of downwind continents is less frequent and more dilute. Focusing first on transport from Asia to North America, modelling studies indicate that episodic long-range transport of CO from Asia to the northeastern North Pacific region occurs year-round every 10, 15 and 30 days in the upper, middle and lower troposphere, respectively (Liang et al., 2004). In springtime when transport is strongest, 3–5 Asian plumes impact the west coast of the United States' atmospheric boundary layer between February and May (Yienger et al., 2000). Aircraft studies have detected strong Asian plumes in the lower and middle troposphere above the eastern North Pacific (Heald et al., 2003; Nowak et al., 2004) and the U.S. west coast (Cooper et al., 2004; Jaeglé et al., 2003; Jaffe et al., 1999; Jaffe et al., 2003a) with CO on occasion reaching 300 ppbv. Most of these free tropospheric observations, all with CO in excess of 200 ppbv, were associated with warm conveyor belt export from Asia; however, some low altitude events were associated with export and transport in the lower troposphere. When these plumes have impacted the surface of the coastline of Washington State, CO has reached as high as 180 ppbv. These events are not always associated with elevated ozone mixing ratios, and the enhancements are difficult to detect at the surface (Hudman et al., 2004). As Asian pollution plumes continue travelling east, clear enhancements of ozone and aerosol concentrations can also be found over Europe (Stohl et al., 2007b). Asian pollution can also reach Europe via westward transport with the monsoon circulation from India to Africa and the Mediterranean (Lawrence et al., 2003). The overall impact of the summertime Asian monsoon is to advect relatively clean air from the Indian Ocean into southern Asia in the lower troposphere (Stohl et al., 2002), while the associated convection lofts anthropogenic emissions from northeast India and southwest China into the upper troposphere, where they can recirculate above southern Asia within the semi-permanent summertime upper tropospheric anticyclone (Li et al., 2005). During winter, the Asian monsoon advects pollution from the continent southward over the Indian Ocean (Lelieveld et al., 2001).

In terms of large-scale North American export events influencing Europe, confirmation of their strong impact has been made in the free troposphere above Europe (Stohl and Trickl, 1999; Stohl et al., 2003a; Trickl et al., 2003), and in limited cases at high-altitude sites in the Alps (Huntrieser et al., 2005), all involving warm conveyor belts. Concerning low altitude surface sites, Derwent et al. (1997) found five probable cases of North American emissions influencing Mace Head on the west coast of Ireland, but the pollutant concentrations were quite low. The only major North American export event to show a strong impact on low-altitude European surface sites involved smoke plumes from the widespread forest fires in Canada that caused CO mixing ratios to reach 175 ppbv at Mace Head during August 1998 (Forster et al., 2001).

High-latitude Europe and Siberia are cold enough to allow direct transport of air pollution from these regions into the Arctic lower troposphere (Barrie, 1986; Stohl, 2006), contributing to Arctic haze, a mixture of sulfate and particulate organic matter and, to a lesser extent, ammonium, nitrate, black carbon, and dust aerosols (Quinn et al., 2007). Typically, this pollution from Europe and Asia traverses the polar region of the Arctic and reaches high-latitude North America (Sharma et al., 2006).

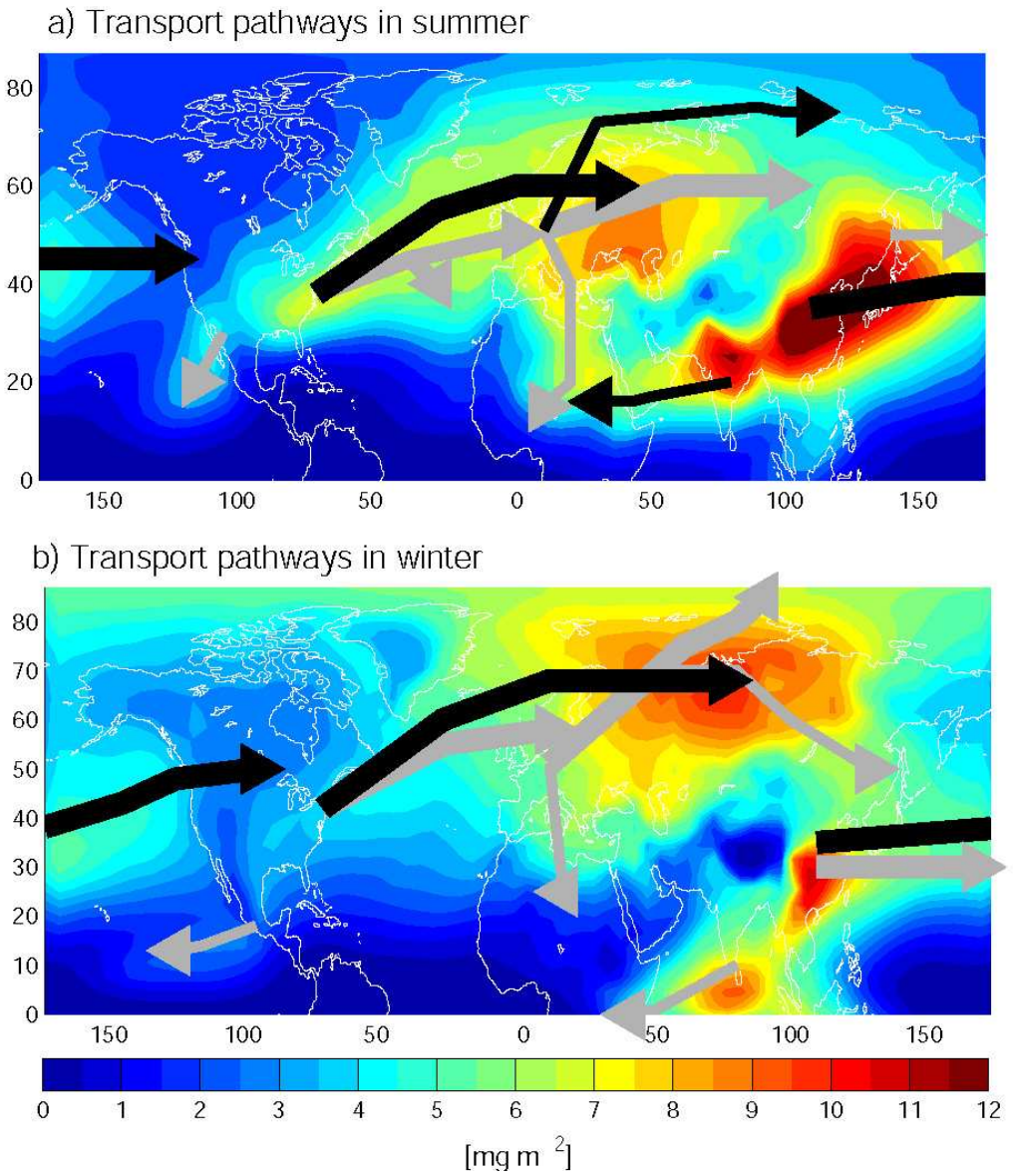

Figure 2.6 Pathways of intercontinental pollution transport in the Northern Hemisphere. Shading indicates the location of the total column of a passive anthropogenic CO tracer released over the Northern Hemisphere continents after 8–10 days of transport, and averaged over 15 years. Shown are transport pathways in summer (June, July, August – upper panel), and winter (December, January, February – lower panel). Gray arrows show transport in the lower troposphere (< 3 km) and black arrows show transport in the middle and upper troposphere (> 3 km). Image reproduced from chapter 1, figure 2, page 6, of Stohl and Eckhardt (2004), with kind permission of Springer Science and Business Media.

Only a few studies have examined the transport of European pollution to Asia (Duncan and Bey, 2004; Newell and Evans, 2000; Pochanart et al., 2003; Wild et al., 2004) Newell and Evans (2000) estimated that, on an annual basis, 24 per cent of the air parcels arriving over Central Asia have previously crossed over Europe, and some 4 per cent have originated in the European atmospheric boundary layer. Pochanart et al. (2003) have shown that average O_3 and CO concentrations in east Siberia are enhanced in air masses transported from Europe. Over Japan, impacts of North American and European emissions are similar (Wild et al., 2004)

European pollution is also exported southwards in the lower troposphere across the Mediterranean Sea to North Africa all the year round, although the strongest events occur during

summer (Lelieveld et al., 2002; Stohl et al., 2002). The transport of many species and PM from Europe to Africa during summer can be very efficient due to the diminished impact of wet removal processes (Kallos et al., 1998). A modelling study by Duncan and Bey (2004) indicates the exported European pollution is responsible for 5–40 per cent of the lower tropospheric ozone above North Africa during summer.

2.4.2 Contribution to background pollution

The combination of long transport times and the mixing of air masses from diverse source regions determines the distribution of background trace gas concentrations. The background concentrations of surface emissions tend to be oriented along potential temperature surfaces in the mid-latitudes due to the rapid mixing along these surfaces (Bowman and Carrie, 2002; Bowman and Erukhimova, 2004; Hess, 2005; Plumb and McEwan, 1978). The atmospheric circulation tends to transport the highest concentrations of constituents towards the poles (Hess, 2005). Recent studies suggest background levels of pollutants along the west coast of North America (Jaffe et al., 2003b; Parrish et al., 2004) and Europe (Derwent et al., 1997; Oltmans et al., 2006) are changing. Fiore et al. (2002) found an out-of-phase relationship between background and locally produced ozone at the surface of the United States in summer. During high pollution episodes under stagnant conditions, background ozone concentrations were small, as mixing between the free troposphere and atmospheric boundary layer was suppressed. Conversely, when the atmospheric boundary layer was more thoroughly vented, locally produced ozone was at a minimum, but background ozone concentrations were greater.

2.4.3 Feedbacks between transported air pollutants and regional climate and meteorology

Trace gases and aerosols have been shown to modify both the radiative balance of the atmosphere as well as cloud formation and precipitation. Therefore, it might be expected that pollutants from an upwind source can modify the climate and meteorology (including precipitation) of a receptor site, which may then create feedback mechanisms that modify how pollutants are transported around the globe. However, there is still a large degree of uncertainty as to the magnitude of the aerosol direct effect (scattering and absorption of solar and infrared radiation) and indirect effect (altering cloud albedo and precipitation efficiency) on the global radiative forcing of the atmosphere, with the latest report of the Intergovernmental Panel on Climate Change assigning a "level of scientific understanding" of medium-low for the direct effect and low for the indirect effect (IPCC, 2007). Here we summarize some recent studies that link changes in aerosol and ozone concentrators to climate and transport feedbacks, but point out that additional studies are needed to quantify these effects.

Long-range transport of aerosols has received much attention due to the discovery of atmospheric brown clouds (Ramanathan et al., 2001a), which form a haze layer over much of southern Asia between December and April. Regionally, the haze was highly absorbing (mainly black carbon aerosol), which decreased the surface solar radiation and increased the lower tropospheric solar heating; model simulations suggest that the additional heating significantly perturbs tropical rainfall patterns (Ramanathan et al., 2001b). The seasonality of brown clouds and their dimming effect is being investigated in the "Project Atmospheric Brown Cloud" campaign (Ramana and Ramanathan, 2006).

Rotstayn and Lohmann (2002) report a southward shift in observed tropical rainfall over land during the period 1900–1998. Their modelling work suggests that spatially varying aerosol-related forcing (both direct and indirect) can substantially alter low-latitude circulation and rainfall. Recent work has also shown that the frequency of deep convective clouds along the North Pacific wintertime storm track increased from 1984 to 2005 (Zhang et al., 2007). Modelling indicated that the increase in the deep convective clouds could be explained by aerosol effects associated with Asian outflow, and also indicated increased precipitation over the North Pacific. This suggested intensification of the Pacific storm track could lead to changes in global circulation.

Intercontinental transport of desert dust from Asia and North Africa has been well documented, (e.g. Liu et al. (2003)). A study by Dunion and Veldon (2004) suggests that Saharan

dust transport can lead to suppressed hurricane formation. They postulate that when the dust-laden Saharan air layer advects from Africa to the tropical North Atlantic, it can engulf tropical waves, or pre-existing tropical cyclones, and the associated dry air, temperature inversion, and strong vertical wind shear may inhibit the ability of tropical waves or cyclones to strengthen (Dunion and Velden, 2004). Evan et al. (2006) demonstrate a positive correlation between North Atlantic tropical cyclone activity and atmospheric dust cover in support of this hypothesis.

The efficiency of sunlight absorption in aerosol layers is greater in the Arctic than at lower latitudes due to the prevalence of large solar zenith angles and the high albedo (or reflectivity) of the underlying snow and ice. Black carbon is a minor but important component of the Arctic haze and causes heating of the haze layers (Quinn et al., 2007). In addition, deposition of black carbon onto snow and ice results in a reduction of the albedo (Clarke and Noone, 1985; Warren and Wiscombe, 1980). It has been suggested that the climate forcing due to this albedo effect is significant compared to the effect of greenhouse gases (Hansen and Nazarenko, 2004).

The radiation budget of the atmosphere is sensitive to the tropospheric ozone distribution, which is partly controlled by transport. A study by Shindell et al. (2006) suggests tropospheric ozone has caused enhanced warming (>0.5 degrees C) over polluted northern boreal zones during summer between 1890 and 1990, and between 0.4 and 0.5 C during winter and spring between 1890 and 1990. Warmer temperatures in the Arctic reduce the transport barrier between mid-latitudes and the polar regions facilitating transport of polluted European air masses into the Arctic (Stohl et al., 2007a).

2.5 Impact of climate change on future intercontinental transport patterns

Climate change will affect future intercontinental transport patterns through alteration of the large-scale circulation and regional climate and hence the transport processes described above. Predictions of changes in regional climate are generally less robust than those on larger scales, such as hemispheric (Giorgi et al., 2001). Examining experiments generated for the Intergovernmental Panel on Climate Change (IPCC) fourth assessment report, Held and Soden (2006) found that increased greenhouse gas concentrations produced a warmed climate and the following changes that were consistent across the models: a decrease in deep convective mass flux**,** and an enhancement in patterns of evaporation minus precipitation (i.e. dry locations will get dryer, wet locations moister) and its temporal variance. Changes in climate will also modify tropospheric chemistry and hence the concentrations of pollutants arriving at downwind continents. Increased water vapour in a future warmed atmosphere leading to increased ozone destruction and shorter ozone lifetimes is a prominent and robust multimodel feature (Stevenson et al., 2006). This effect may cause a reduction in the contribution of Asian emissions to background ozone over the United States in a future climate (Murazaki and Hess, 2006). On the other hand, a number of studies (Hauglustaine et al., 2005; Murazaki and Hess, 2006) have shown increased local production of ozone in high emission areas, leading to increased ozone export, at least locally. In addition, intercontinental transport of ozone may also be influenced by enhanced lightning NO_x emissions (Hauglustaine et al., 2005) and enhanced stratosphere-troposphere exchange in a warmer climate (Collins et al., 2003; Stevenson et al., 2006; Zeng and Pyle, 2003).

A number of general circulation model (GCM) studies, though not all, have reported a tendency towards fewer but more intense mid-latitude cyclones in the future (Cubasch et al., 2001; Lambert and Fyfe, 2006), a poleward shift of mid-latitude cyclones (Bengtsson et al., 2006; Cubasch et al., 2001; Lambert and Fyfe, 2006; Yin, 2005), a poleward shift of mid-latitude storms (Bengtsson et al., 2006; Yin, 2005), and an associated increase in cyclonic circulation patterns over the Arctic (Cassano et al., 2005). Chemistry transport model simulations performed under future SRES A1(b) climate scenarios also suggest a decrease in synoptic activity (intensity and frequency) over the United States (Mickley et al., 2004; Murazaki and Hess, 2006), in agreement with the GCM studies. A decrease in synoptic activity would result in less long-range pollutant transport driven by synoptic systems (Holzer and Boer, 2001), but possibly an increased role for convection and high-altitude transport.

There is also a trend towards a positive phase of the North Atlantic Oscillation (NAO) projected by the majority of GCMs (Miller et al., 2006), associated with a northeastward shift in Atlantic storm track activity, which suggests that the Arctic will become more polluted in the future

(Eckhardt et al., 2003). However, model projections vary widely in their magnitude of response (Miller et al., 2006; Osborn, 2004). These results concur with observational evidence of a poleward shift of the storm track (McCabe et al., 2001) and a positive trend in the Northern Annular Mode (NAM) or Arctic Oscillation (AO), and NAO (Harnik and Chang, 2003) during the 1970s through the 1990s. Hess and Lamarque (2007) show that a positive AO results in increased transport of United States emissions to Europe. Finally, Liu et al. (2005) relate stronger eastward transport of East Asian emissions to El Niño events; a future trend towards more El Niño-like conditions has been noted in a number of (but not all) studies (Collins and the CMIP Modelling Groups, 2005).

2.6 Outstanding issues and recommendations

2.6.1 Basic transport mechanisms

Basic large-scale mechanisms for transporting pollutants out of the atmospheric boundary layer have been reasonably well documented in both measurements and models; however, the importance of small-scale venting on intercontinental transport, such as dry convection, local circulations (valley, or land-ocean) and gravity wave mixing, has not. These smaller-scale venting mechanisms have greatest importance during summer due to diminished large-scale stirring by synoptic systems, especially for low-altitude transport events from Asia to the United States (Holzer et al., 2005). These types of events are difficult to observe (i.e. they are not associated with large-scale plume transport) and are not often resolved in large-scale model simulations.

The transport of pollutants from the free troposphere to the surface is also not well observed, either in large-scale models or in observations. This may be partly due to the fact that descent into the atmospheric boundary layer often does involves not large concentrated plumes, but background air. In addition, venting through the top of the atmospheric boundary layer often involves small-scale processes not directly simulated on the global scale.

The transport of species out of the atmospheric boundary layer is often associated with wet-deposition, as water vapour is condensed during the transport events. The extent to which soluble species are wet-deposited depends on details of the microphysics, i.e. to what extent (a) convective clouds are able to transport soluble species to the upper troposphere; (b) soluble species are retained as liquid water freezes; and (c) rain is evaporated as it falls through the atmosphere. Again, these processes are not suitably modelled on the large scale and have not been adequately measured.

Finally, transport in the tropics and subtropics has received less attention than in the mid-latitudes. With the growth of future emissions and mega-cities expected in the tropics and subtropics, these processes will gain greater importance. Furthermore, the nature of the transport barrier between the tropics and mid-latitudes is not particularly well known.

2.6.2 Modelling

While large-scale transport is reasonably well represented in global models, sub-grid scale parameterized processes are not, in particular deep convection, atmospheric boundary layer mixing, atmospheric boundary layer venting, and wet deposition (Lawrence and Rasch, 2005; Murazaki and Hess, 2006). For example, Auvray et al. (2007) find differences in the sign of net chemical tendencies (i.e. photochemical production and loss rates) in the middle troposphere in the North Atlantic and North Pacific, which they attribute to differences in model transport schemes and water vapour transport as well as lightning. These processes are critical for venting pollutants into and out of the atmospheric boundary layer. Uncertainties in the transport of water soluble pollutants are high. Processes that are influenced by sub-grid-scale orography are also a key uncertainty for models; venting and mixing into the atmospheric boundary layer by land-sea breezes, mountain-valley breezes, gravity waves and flow over mountains are not well represented or not represented at all. These issues may be partly addressed through coupling between regional and global models. Further studies to evaluate interannual and longer-term variability in intercontinental transport are also warranted. Model limitations are not restricted to transport processes. In addition to the uncertainties in the transport processes above, there also remains significant uncertainty regarding emissions and model simulation of HO_x and NO_x sources and photochemistry (e.g. Singh et al. (2007)).

Although source-receptor relationships have been extensively studied at the continental and country levels, studies to quantify the impacts of emissions at fine spatial scales such as mega-cities (Lawrence et al., 2006) are few, but increasingly important given the rapid and future projected growth of world cities. Model uncertainty associated with the methodology used for estimating source-receptor relationships also deserves attention (see section 5.1.1). Since source-receptor relationships require an accurate knowledge of the emission source, more sophisticated estimate methodologies and a better quantification of model error are also necessary. The ongoing Hemispheric Transport of Air Pollution (HTAP) model intercomparison discussed in chapter 5 should help quantify inter-model errors and sensitivities to intercontinental transport.

2.6.3 Measurements networks

To fully understand the impact of upwind emissions on downwind continents, a comprehensive in situ measurement network must be established to monitor trace gases and PM along the inflow and outflow boundaries of each continent throughout the entire tropospheric column and the lowermost stratosphere. No such network currently exists and the logistical and financial barriers to its establishment are formidable. For the time being, the most accessible observational information on pollutant import and export will have to come from remote sensing by polar orbiting instruments. The ENVISAT SCIAMACHY[2] instrument provides column nitrogen dioxide (NO_2) measurements, while the AIRS instrument on the NASA AQUA[3] satellite provides column CO measurements, and the MISR[4] instrument on TERRA[5] measures aerosol optical depth. The NASA AURA satellite can monitor O_3, nitric acid (HNO_3),CH_4, aerosol extinction, CO, and SO_2 in the upper troposphere using the HIRDLS[6] and MLS[7] limb sounders, while the OMI[8] instrument can provide column measurements of ozone, SO_2, formaldehyde (HCHO), NO_2, aerosol optical thickness and aerosol single scattering albedo. Importantly, the TES[9] instrument on Aura provides tropospheric profiles of O_3, CH_4 and CO, and upper tropospheric profiles of HNO_3 and NO_2. While these instruments provide global coverage, the frequency at which they pass over a location means the data are best suited for monthly or seasonal averages. Other limitations of these instruments are their inability to make measurements within and below cloud (although MLS can measure some trace gases in the presence of tropical cirrus), the coarse vertical resolution of the nadir-viewing instruments (with TES having the best resolution of 2 km), and the limitation of MLS in that it can only view the tropical upper troposphere.

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[2] European Space Agency Environmental Satellite, Scanning Imaging Absorption Spectrometer for Atmospheric Chartography/Chemistry.

[3] Atmospheric Infared Sounder, National Aeronautics and Space Administration; AQUA is a NASA polar-orbiting satellite.

[4] Multiangle Imaging Spectro-Radiometer.

[5] A NASA polar-orbiting satellite.

[6] High Resolution Dynamics Limb Sounder.

[7] Microwave Limb Sounder.

[8] Ozone Monitoring Instrument.

[9] Tropospheric Emission Spectrometer.

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3. OBSERVATIONAL EVIDENCE AND CAPABILITIES RELATED TO HEMISPHERIC OR INTERCONTINENTAL TRANSPORT

3.1 Introduction

For several decades it has been possible to measure aerosols, ozone (O_3) and the important aerosol and ozone precursors at the concentrations found in the most remote regions in the Northern Hemisphere. Even the earliest measurements in these remote regions consistently indicated that long-range transport exerts a strong influence on the observed concentrations. For example, dust of Asian origin has been observed throughout the North Pacific region (Duce et al., 1980; Prospero, 1979) and studies at the west coast of North America in 1985 identified the influence of Asian emissions on the sulphur budget (Andreae et al., 1988) and on the concentrations of O_3, hydrocarbons, and peroxyacetyl nitrate (PAN) (Parrish et al., 1992). It is now clear that the Northern Hemisphere "background" concentrations of many atmospheric species are elevated by emissions that circulate globally from all of the continents until they are finally removed. Several chapters in a recent monograph (Stohl, 2004) give an overview of some of the evidence for the impact of intercontinental transport between the three continents at northern temperate latitudes. The goal of this chapter is to provide a summary picture of this long-range transport that has emerged from surface monitoring, vertical profiling, intensive field campaigns and satellite data sets.

Each of these four data sources provides a different perspective on the issues related to the scope of the intercontinental or hemispheric transport of air pollutants. Satellites provide a global view of atmospheric transport patterns for a relatively few species of interest; often this view is temporally integrated over weeks or seasons, although in favourable cases the evolution of intense transport events can be followed on a daily scale. Satellite instruments primarily yield column or partial column data, with limited vertical resolution and, to a greater or lesser extent depending upon species, lower sensitivity near the surface. Intensive campaigns provide measurements of a much wider range of species, a more detailed view of the important atmospheric processes, and better spatial and temporal resolution, but these studies are generally severely limited in temporal and spatial coverage. Vertical profiling and surface monitoring yield data sets that have even more limited spatial resolution (although enough sites allow broad spatial coverage) and are usually limited to only a few species; however, these efforts make it possible to discern long-term trends when the measurements can be sustained on a decadal time scale.

Because of the spatial limitations of intensive campaigns, vertical profiling and surface monitoring, great importance is placed on the proper selection of measurement locations and sites and on the interpretation of the resulting data sets. Data must be obtained that are representative of the long-range transport of the species of interest into the region under investigation; thus, the upwind boundary (i.e. western edge at northern temperate latitudes) of the receptor region or continent is usually most appropriate. It is also important to combine boundary layer observations with vertically resolved measurements in the free troposphere, since events of concentrated pollutant transport usually occur in atmospheric layers elevated above the surface. It is these concentrated pollutant transport events that often provide the best opportunities to investigate the relationships between transported species and the processes that occur during long-range transport. Surface sites generally receive "background" inflow, which is certainly affected by long-range transport, but the effects of this transport are more difficult to discern. In addition it is often difficult to interpret data from ground-based sites because measurements are inevitably affected by local emissions or loss processes, and many sites lack measurements that can discriminate against these influences.

The following sections of this chapter discuss the long-range transport of O_3 and its precursors (section 3.2), aerosols and their precursors (section 3.3), the implications for surface air quality in receptor regions (section 3.4), our ability to determine source regions (section 3.5) and long-term trends in hemispheric transport (section 3.6). These are followed by some concluding remarks.

3.2 Long-range transport of ozone and its precursors

There are three sources of O_3 in the more remote regions of the troposphere: (a) downward mixing from the stratosphere; (b) export from regions with net photochemical O_3 production; and (c) in situ photochemical production from natural and transported anthropogenic precursors. Satellite data sets provide a large-scale view of the transport of O_3 and its precursors from the source regions, primarily on the continents. In situ measurements provide much greater detail regarding transport mechanisms and the production and removal processes that occur during transport. While our focus in this section is on O_3, additional tracers such as CO (lifetime 1–3 months), sub-micron aerosol (lifetime of hours to weeks depending upon removal processes), PAN (a product of NO_x photochemistry, with a lifetime of weeks in the upper free troposphere), mercury (Hg^0, lifetime of months) and VOCs species (lifetimes of hours to months) are all useful for interpreting in situ measurements. To fully characterize the transport of O_3, it is necessary to understand the transport of the precursors – NO_x, CO, and VOCs – that provide the raw materials to fuel the in situ photochemical production in the more remote troposphere.

3.2.1 The view from satellites

Satellite capabilities have advanced rapidly in the past decade and now provide a critical global view in cloud-free regions. Present capabilities include observations of tropospheric O_3 (GOME, SCIAMACHY, OMI, and TES[1]), CO (e.g. MOPITT[2], AIRS[3], SCIAMACHY, and TES) and NO_2 (e.g. GOME, SCIAMACHY, OMI). Figure 3.1 presents example images that provide clear evidence for intercontinental transport of each of these three species. Figure 3.1a shows significant O_3 enhancements in the regions of greatest precursor emission, i.e. the eastern United States, Europe, and East Asia, with a broad enhanced region in the mid-latitudes, associated with export of O_3 and O_3 precursors from the adjacent continental regions.

Satellites have been especially useful in detecting CO and aerosols associated with large biomass burning events and the subsequent transport of those emissions. Figure 3.1b shows the CO plumes from intense wildfires in Alaska and Canada that can be traced across North America and the Atlantic Ocean. Inverse modelling using the MOZART (Model of Ozone and Related Tracers) chemical transport model showed that the fires emitted about as much CO as did human-related activities in the continental United States during the same time period, about 30 Tg CO for June-August, 2004. Modelling and measurements also show that emissions from the 2004 North American wildfires caused ground-level concentrations of O_3 to increase by 25 per cent or more in parts of the northern continental United States and the central North Atlantic and by 10 per cent as far away as Europe (Pfister et al., 2006; Val Martin et al., 2006).

Satellite measurement of the long-range transport of tropospheric NO_2 is quite challenging, primarily due to the short lifetime of this species in the emission regions. Generally only a small fraction ($\leq$ 10%) of the NO_x originally emitted leaves the boundary layer of the continental emission region. However, as indicated in Figure 3.1c long-term averages of satellite data can reveal the long-range transport of NO_2, in this case from North America to Europe. Aircraft observations as part of ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) field study and GEOS-Chem model[4] calculations provide additional evidence for this upper tropospheric transport (Martin et al., 2006).

[1] Global Ozone Monitoring Equipment, Scanning Imaging Absorption Spectrometer for Atmospheric Chartography/Chemistry, Ozone Monitoring Instrument, and Tropospheric Emission Spectrometer.

[2] Measurements of Pollution in the the Troposphere.

[3] Atmospheric Infared Sounder.

[4] A global three-dimensional atmospeheric composition model driven by data from the Goddard Earth Observing System.

3.2.2 Evidence for ozone and precursor transport from in situ measurements

Over the past two decades, in situ measurements conducted as part of intensive field studies, surface monitoring networks and long-term aircraft and sonde programmes have begun to characterize the impact of long-range transport on the O_3 budget of the troposphere.

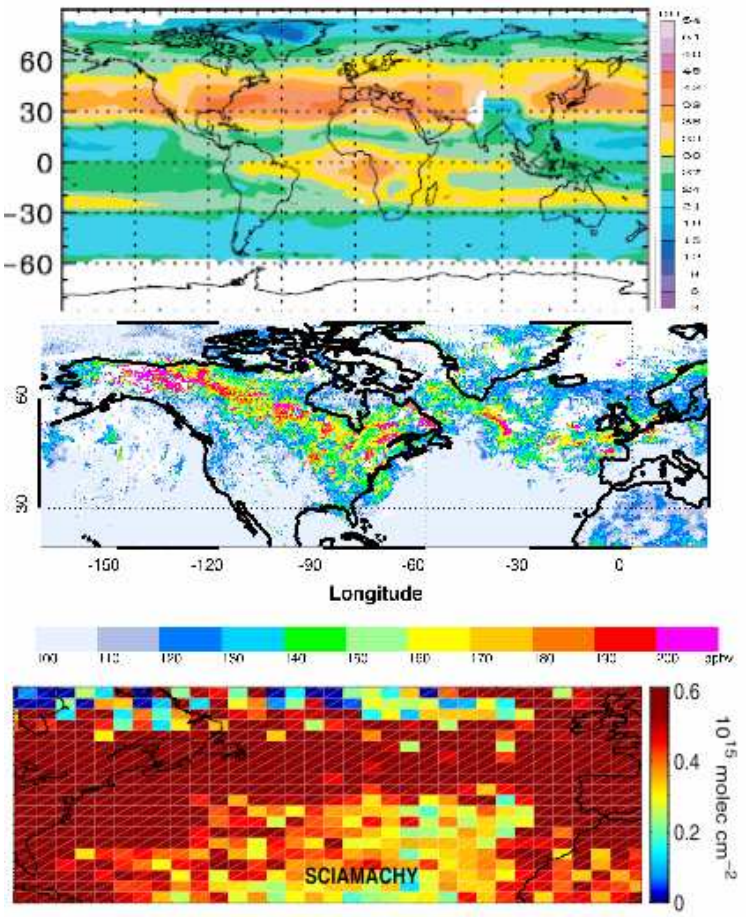

Figure 3.1 Examples of satellite measurements of (a) O_3, (b) CO and (c) NO_2. The images are (a) the seasonal average GOME tropospheric ozone columns (Dobson units) for June–August 1997 (Liu et al., 2006), (b) MOPITT 700 hPa CO mixing ratio (ppbv) for 15-23 July 2004 (Pfister et al., 2006), and (c) Tropospheric NO_2 columns (10^{15} molec cm^2) for May–October 2004, as retrieved from SCIAMACHY (Martin et al., 2006). The colour bar in (c) has been selected to emphasize outflow over the Atlantic.

Figure 3.2 illustrates a concentrated trans-Pacific pollutant transport event observed at a mountaintop site near the west coast of North America. Surface sites that receive air transported above the marine boundary layer and aircraft often observe such clearly discriminated plumes throughout the Northern Hemisphere. This event exemplifies the strong correlation often found between O_3 and emission tracers, in this case CO, mercury and aerosols. The $\Delta O_3/\Delta CO$ ratio represents the enhancement in O_3 that accompanies an enhancement of pollution transport. For this episode, this ratio is similar to previously reported episodes of transport from Asia to North America (e.g. Price et al. (2004)) and the excellent correlation suggests that O_3 and/or its precursors were transported together with the emission tracers from the Asian source region.

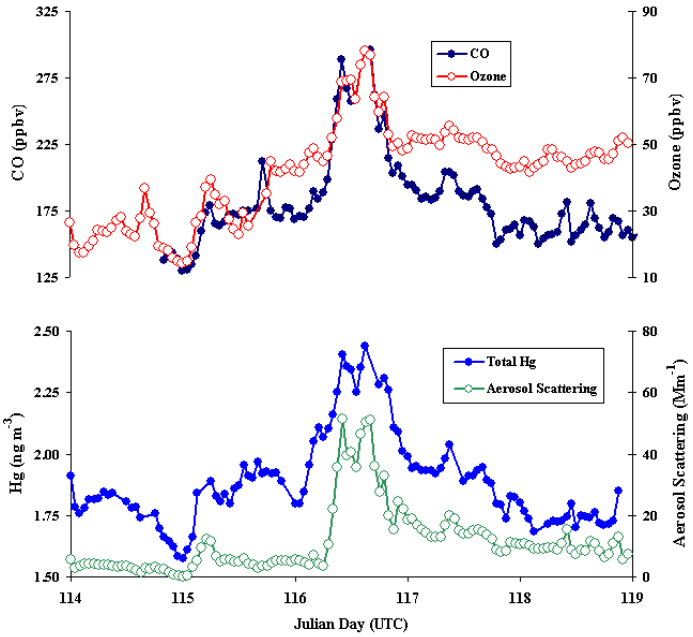

Figure 3.2 Concentrated trans-Pacific pollutant transport event observed at mountaintop site near the west coast of North America. Observations of O_3, aerosols, Hg and CO in a plume transported from Asia to the Mt. Bachelor Observatory in Oregon near the west coast of the United States in April 2004. The O_3 vs. CO linear correlation gives an R^2 value of 0.84 and a slope of 0.22 (Jaffe et al., 2005).

The commonly observed correlation between O_3 and emission tracers in transported plumes is strong evidence for the impact of long-range transport on the ozone budget, but that evidence is not unequivocal. Figure 3.3 shows the O_3-CO correlation from the ITCT-2K2 campaign conducted off the west coast of North America during spring 2002. On three flights of the National Oceanic and Atmospheric Administration (NOAA) WP-3D aircraft, plumes of anthropogenic emissions were encountered (defined by CO>150 ppbv). Modelling of the air mass transport history confirmed Asia as the source region. Two of the plumes exhibit positive O_3-CO slopes, which suggests that O_3 was indeed produced in these plumes from Asian anthropogenic sources. This production could have occurred within the Asian continental boundary layer, or during trans-Pacific transport. For the 17 May plume, the O_3-CO correlation slope is 0.19, very similar to the slope found for the plume illustrated in figure 3-2. However these springtime plumes are typically lofted from the Asian source region by warm conveyor belts associated with mid-latitude cyclones. These cyclones also contain a dry air stream that brings high concentrations of O_3 of stratospheric origin into the mid- to lower-troposphere where these plumes were encountered (Cooper et al., 2004). Ozone of stratospheric origin is certainly responsible for the concentrations above 100 ppbv observed during the May 10 flight, and could conceivably account for the observed O_3 enhancements on all three flights (Nowak et al., 2004). In contrast to these lofted Asian plumes, significantly higher O_3-CO slopes are observed in the central North Atlantic lower free troposphere (Honrath et al., 2004), a region impacted by North American emissions transported in low-level outflow of the type described in (Owen et al., 2006).

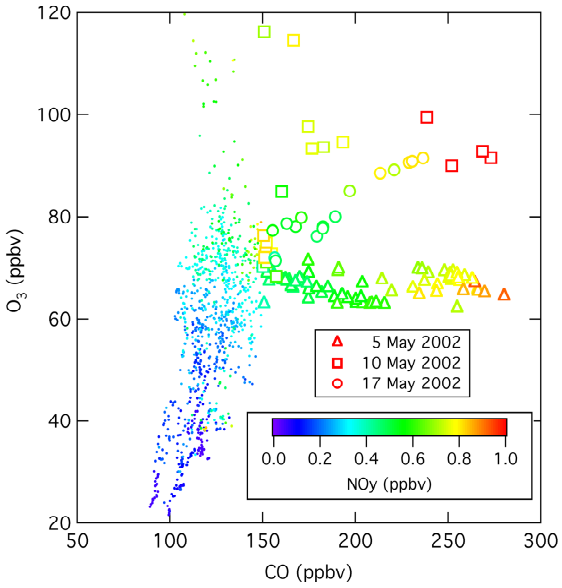

Figure 3.3 One-minute average O_3 versus CO measured during the ITCT-2K2 aircraft campaign. The figure includes all data without recent North America influence; they are colour-coded by measured NO_y concentration. On three flights, concentrated plumes of Asian emissions were encountered and these are identified by the symbols noted. (Figure adapted from Cooper et al. (2004) and Nowak et al. (2004))

Some evidence does point to the importance of in situ O_3 formation during trans-Pacific transport. All three plumes in figure 3.3 were characterized by strong correlations of CO with NO_y (Nowak et al., 2004). The average NO_y versus CO slope of these correlations is much smaller than the Asian NO_x to CO emission ratio, which implies that ≥90 per cent of the originally emitted NO_x had been removed from these plumes before or during transport. For plumes transported in at mid- to high altitudes, most of that NO_y was in the form of PAN (Roberts et al., 2004). However, the 17 May plume had subsided during the previous few days of transport and HNO_3, rather than PAN, accounted for most of the NO_y. The observed relationships among these species can be explained by the following hypothesis. Lofting of air from the moist Asian continental boundary layer produces precipitation that scavenges most NO_y as well as aerosols and other soluble gas-phase species. Consequently, the relatively insoluble PAN (the major efficiently transported NO_y species) is lofted in the air along with any O_3 that already had been produced in the boundary layer. When the pollution plumes eventually descend at far downwind distances, PAN decomposes to release NO_x, a precursor required for photochemical O_3 production. Thus, in subsiding plumes in situ O_3 production, transport of O_3 from the Asian boundary layer and input from the stratosphere may all contribute to the observed O_3 enhancements. This hypothesis is consistent with the modelling work of Moxim et al. (1996) and Hudman et al. (2004), and the observations of Kotchenruther et al. (2001). To determine the exact fraction of the O_3 enhancement that is due to each of these three sources requires observations in consort with detailed chemical transport modelling.

The International Global Atmospheric Chemistry (IGAC)-sponsored ITCT Lagrangian 2K4 experiment, which was part of the ICARTT field campaign in summer 2004, provided the opportunity to model in situ formation of O_3 and to compare that modelling with observations. That study employed four aircraft to investigate the evolution of pollutants during transport from emission regions in North America across the North Atlantic to receptor regions in Europe. Two aircraft were operated from North America, a third in the central North Atlantic from the Azores, and a fourth from Europe. Methven et al. (2006) identify four cases when the same air mass was sampled by aircraft on both sides of the Atlantic, three of which also included sampling in the central Atlantic. Trajectory calculations, temperature, humidity and hydrocarbon fingerprints were analysed to establish the successful interception of the same air masses. Figure 3.4 illustrates the measurements and model simulations of those measurements made from three aircraft during one of those cases.

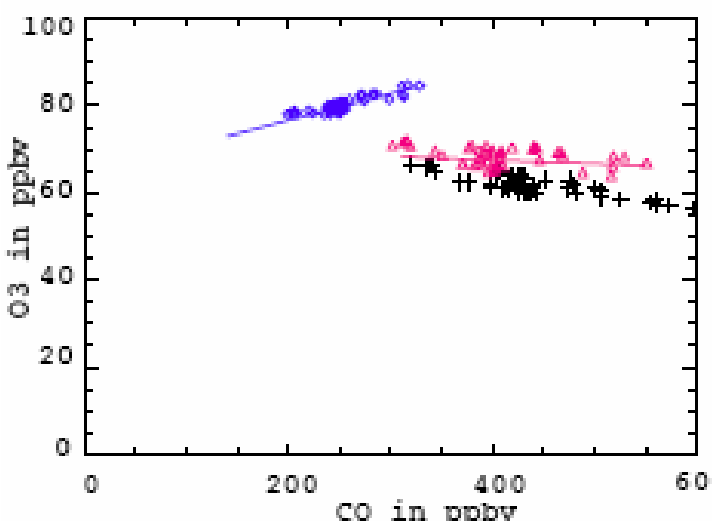

Figure 3.4 Measured and modelled O_3 versus CO from aircraft over the western North Atlantic. The first aircraft measured near the North American coast (black +). The lines give linear fits to measurements (not shown with symbols) from a second aircraft 2 days later over the Central Atlantic (red line), and a third aircraft 5 days later over Europe (blue line). The model simulations, initialized with the black pluses, gave results in excellent agreement with the measurements: the red triangles after 2 days and the blue diamonds after 5 days (Real et al., 2007).

Figure 3.4 illustrates our emerging ability to observe and model long-range transport and the evolution of O_3 and its precursors during that transport. Atmospheric transport models operated in forecast mode successfully guided three aircraft into a pollutant plume during its transport across the entire North Atlantic basin. Chemical transport models operated in a retrospective mode have provided accurate simulations of the O_3 formation, precursor evolution and plume dilution that occurred during transport. This example represents forest fire emissions from Alaska emitted three to four days before the first measurement near the east coast of North America. Measurements in the plume showed enhanced concentrations of CO, hydrocarbons and oxidized nitrogen species, mainly PAN. Maximum observed O_3 levels increased by 17 ppbv over five days, and the O_3 vs. CO linear correlation slope increased from about -0.03 to +0.08. These increases were due to photochemical production initiated by PAN decomposition, which released the NO_x ozone precursor, during descent of the plume towards Europe. Significant dilution accompanied this plume descent. These measurements and their successful reproduction by modelling demonstrate our current understanding of long-range transport of pollution plumes in the free troposphere (Real et al., 2007).

The transport examples discussed above occurred from Asia to North America (figures 3.2 and 3.3) and from North America to Europe (figure 3.4); transport between Europe and Asia is much more difficult to directly discern because it occurs predominately within the continental boundary layer over Eurasia (Stohl et al., 2002; Wild and Akimoto, 2001). The diurnal boundary layer evolution and convection processes disperse and dilute transported air masses, and deposition and photochemical destruction lead to more rapid removal of O_3 and its precursors. Nonetheless, the possible impact of European pollution on Asia has been demonstrated from ground-based observations at Mondy, a remote mountain site in east Siberia. Figure 3.5 shows that O_3 and CO mixing ratios in air masses transported from Europe are higher than those from Siberia and high-latitude regions. The residence time analysis of air masses transported from the European continent indicates that CO mixing ratios significantly decrease with longer transport time of air masses from Europe. This decrease is due to photochemical loss by hydroxl radicals (OH) and the dilution of polluted European air by continental background air during air mass transport over Eurasia (Pochanart et al., 2003).

3.2.3 Long-term trends in background ozone

Studies have documented increases over the past two decades in the "background" concentrations of O_3 in onshore flow at the west coast of Europe and North America (see figure 3.6). This increase has occurred while peak O_3 concentrations have declined in many of the more densely populated areas of the United States and Europe due to reductions in local emissions of NO_x and non-methane hydrocarbons (NMHCs) precursors (EMEP, 2005, ; U.S. Environmental Protection Agency, 2003). In Europe, observations at Mace Head, Ireland, (figure 3.6a) between 1987 and 2003 show an increase in all seasons with an annual mean increase of 0.5 ppbv yr^{-1} (Simmonds et al., 2004). A similar increase has been seen from ship-based observations in the upwind North Atlantic between 1977 and 2002 (Lelieveld et al., 2004). At the German high-altitude mountain site on Zugspitze, a significant increase in O_3 was observed between the early 1980s and 1995, with no significant change seen since then (Oltmans et al., 2006). Along the west coast of North America, three sets of observations – at marine boundary-layer surface sites (figure 3-6b), at Lassen National Park in Northern California, and from aircraft – all indicated a positive trend in O_3 of 0.4-0.7 ppbv/year, depending on season, between 1984 and 2002 (Jaffe et al., 2003a). Fiore et al. (2002) used the GEOS-Chem chemical transport model to interpret O_3 observations in North America and found that background O_3 produced outside of the North American boundary layer contributed an average of 25-35 ppbv to afternoon O_3 concentrations in surface air in the western United States and 15-30 ppbv in the eastern United States during the summer of 1995.

Without a west coast, it is much more difficult to establish long-term trends for background O_3 entering Asia. Evaluation of data at Okinawa, Japan, from 1989 to 1997 indicates an O_3 increase of 2.5 per cent/year in Asian continental air during the winter-spring period (Lee et al., 1998). At a rural station in Hong Kong Special Adminstrative Region of China, Chan et al. (2003) found an O_3 increase of 1.5 per cent/year for the period 1984–1999, which was attributed to increasing emissions from China. Tanimoto (2007) reported an O_3 trend of 0.0-0.4 ppbv/year for boundary layer sites and 1-2 ppbv/year for elevated locations from the late 1990s to 2004 based upon 12 sites in Japan and several island sites downwind of East Asia.

Several studies have investigated trends in tropospheric column O_3. Logan et al. (1999) concluded that the trend in tropospheric O_3 was highly variable and dependent upon location. The European sites studied showed a positive trend between 1970 and 1996, but this trend became less significant if the analyses were started from 1980 rather than 1970. Over Canada, the trend appears to be negative over this same time period. Over Japan, the trends are similar to those over Europe. Thus, Logan et al. (1999) concluded that it was not possible to derive a mid-latitude trend given the variability observed. Oltmans et al. (2006) performed a similar study and reached similar conclusions with regard to the large amount of variability in the trends between sites. Naja et al. (2003) investigated the European O_3 sonde launches from the Hohenpeissenberg, Germany, and Payerne, Switzerland, sites and found some suggestion of an increase in the background O_3 up to about 1990 followed by a possible decrease. Fusco and Logan (2003) identified a complex pattern of changes with a decrease in upper tropospheric O_3 over most regions and an increase in middle and lower tropospheric O_3 over Europe and East Asia. Zbinden et al. (2006) derived temporal trends from MOZAIC (Measurement of Ozone on Airbus In-service Aircraft) profiles measured over Germany, France, New York and Japan from 1995 to 2001; they find significant positive trends of 0.7 to 1.6 per cent/yr at all 4 sites with the strongest increases in winter. Based upon 25 years of TOMS (Total Ozone Mapping Spectrometer) and SBUV (Solar Backscatter Ultraviolet Radiance) satellite data, Ziemke et al. (2005) derived a consistent increase in the Pacific tropospheric O_3 column in the mid-latitudes of both hemispheres.

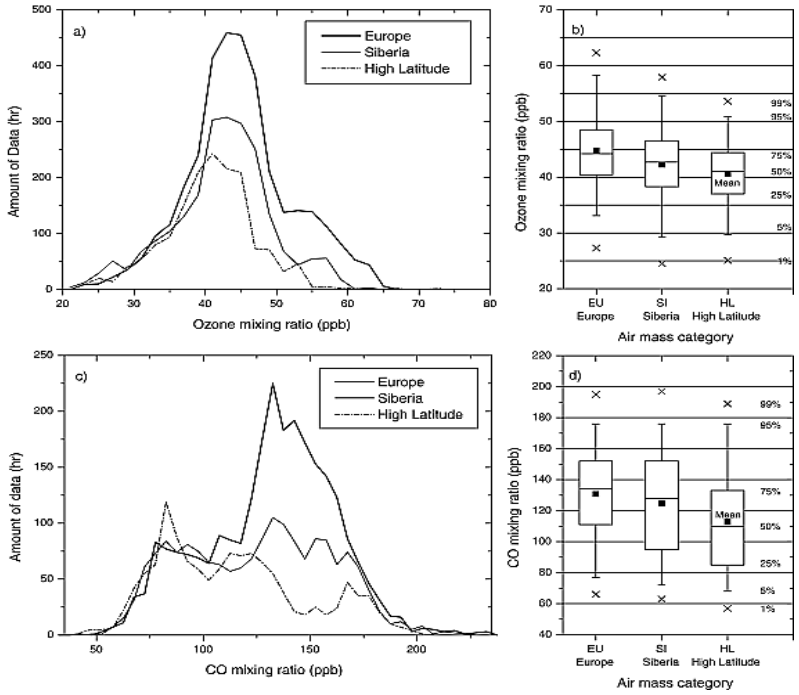

Figure 3.5 O_3 and CO mixing ratios measured at Mondy, Russian Federation. Frequency distributions of 1-hr average O_3 and CO mixing ratios in different air mass regions from July 1997 to December 1998: (a) O_3 histogram; (b) O_3 box chart; (c) CO histogram; and (d) CO box chart. Numbers in (b) and (d) indicate data percentiles (Pochanart et al., 2003).

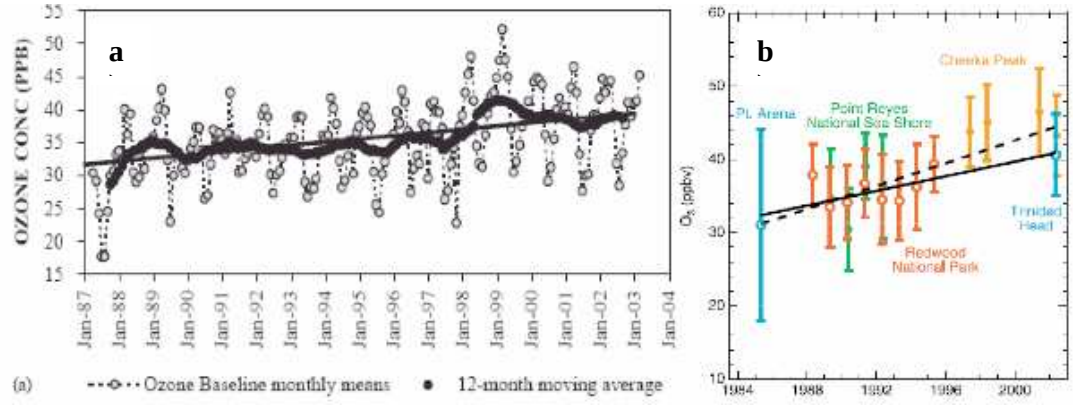

Figure 3.6 "Background" concentrations of O_3 in onshore flow. Increasing marine boundary layer O_3 trends at (a) Mace Head, Ireland (Simmonds et al., 2004) and at the (b) North American west coast (Jaffe et al., 2003a).

In summary, the majority of studies have found significant positive temporal trends in the background O_3 at northern temperate latitudes. However, there does remain significant uncertainty regarding the magnitude and exact cause of these increases that deserves further research. Nevertheless, most researchers have identified the primary cause as increasing emissions of O_3 precursors at those northern temperate latitudes (e.g. Parrish et al. (2004)), with the product O_3 and a fraction of the precursors transported hemisphere-wide. This transport is reflected in the spatial distribution of O_3 depicted in figure 3.1.

3.2.4 Summary, remaining uncertainties and future needs

Numerous surface, sonde and aircraft-based in situ observations, as well as satellite data, all show that intercontinental transport of O_3, either from direct transport or from in situ production from transported O_3 precursors, does indeed occur. Given the knowledge that intercontinental transport of O_3 can contribute to degradation of surface air quality (e.g. section 3.4) and the evidence for increasing background O_3, it is important to develop a comprehensive understanding of this transport. Satellites (e.g. figure 3.1) provide a clear indication of general O_3 transport patterns, a view of direct intercontinental transport of CO associated with specific sources, and a general picture of nitrogen oxide sources and its transport. In situ measurements from aircraft and mountaintop sites (e.g. figures 3.2–3.5) have characterized intercontinental transport events. It has even proven possible to track an identified air mass during a complete trans-Atlantic journey (e.g. figure 3.4) and to successfully model the in situ chemical transformations that occurred during that transport. It is also clear that the background O_3 in the troposphere has changed in important ways over the past two to three decades (e.g. figure 3.6). A number of scientific questions do remain that should be the focus of future work:

- What is responsible for the positive O_3 trends seen at many background sites? Can further analyses reduce the uncertainties in the derived trends? Can models successfully reproduce the observed trends?
- How can we use observed correlations of O_3 with various tracer species (e.g. CO, hydrocarbons, Hg, etc.) to quantify the O_3 transport associated with anthropogenic, biomass burning and stratospheric sources?
- How is the intercontinental transport flux of O_3 divided between “plumes” and “background” air?
- What is the optimum observation strategy for in situ measurements to characterize further the present intercontinental transport of O_3 as well as the future evolution of that transport?
- To what extent can satellite observations be used to track O_3 on a daily, weekly and monthly basis?
- How can we best integrate in situ observations and satellite data with regional and global models into a consistent picture of intercontinental transport?

3.3 Long-range transport of aerosols and their precursors

Chemical analysis has indicated that aerosols in remote regions originate from a variety of primary emissions (dust, biomass burning, sea spray and anthropogenic sources) and are produced by secondary formation within the atmosphere from precursors both natural (e.g. dimethyl sulfide (DMS), VOCs) and anthropogenic (e.g. SO_2, NO_x, VOCs). We find evidence of the long-range transport of aerosols from surface and aircraft in situ measurements, remote measurements by lidars, and satellite products. Networks of oceanic stations with measurement records that began as early as 1965 provide evidence for long-term trends. Analysis of satellite aerosol products have begun to yield quantitative estimates of pollutant aerosol transport across ocean basins, which can provide a strong constraint on model estimates.

3.3.1 In situ and lidar observation of pollutant aerosol outflow from continents

The outflow of aerosols and their precursors from mainland Asia and North America has been studied from downwind island sites. Bermuda, a site of long-term measurements from the AEROCE (Atmospheric/Ocean Chemistry Experiment) campaign (Prospero, 2001), has provided a great deal of information on the outflow of O_3, aerosols and their precursors from North America. In Asia, dust and anthropogenic aerosols transported from China have been observed in Japan; figure 3.7 illustrates one example event. Two peaks due to anthropogenic aerosol were observed at 0:00 and 15:00 on November 7, 2005. The first peak was associated with little if any dust. The second peak

just preceded the dust arrival, which persisted after anthropogenic aerosols decreased. The elemental carbon/organic carbon (EC/OC) ratio stayed at 0.2 during the whole episode, which indicates that anthropogenic aerosol was present even when dust dominated (Takami et al., 2006). Episodes of enhanced SO_2 concentrations measured at the summit of Mt. Fuji, Japan, (3776 m above sea level) provide another example of Asian outflow. Enhanced CO and radon (^{222}Rn) concentrations always accompanied the enhanced SO_2 concentrations, and backward trajectories for such events indicate an Asian continental origin (Igarashi et al., 2006).

3.3.2 Observations of pollutant aerosol in continental inflow

Measurements on the eastern sides of the Atlantic and Pacific Oceans record arrivals of pollution aerosol from North America and Asia, respectively. At Mace Head, Ireland, anthropogenic sulphate has accounted for 85–90 per cent of the total non-seasalt sulphate (nss-SO_4) during marine inflow conditions, indicating that the aerosol arrived from across the ocean (Savoie et al., 2002). Supporting evidence for that conclusion is provided by the comparison of Mace Head data with comparable concentrations at similar latitudes in the Southern Hemisphere; figure 3.8 indicates that the nss-SO_4 values at Mace Head are several times higher than in the southern hemisphere (Barrie et al., 2001). In addition, the observation of elevated levels of black carbon aerosol at Mace Head in 1998 were traced back to Canadian wildfires (Forster et al., 2001), demonstrating clearly that long-range transport of significant amounts of aerosols from North America to Europe is possible, which was confirmed by a similar case during ICARTT. However, analyses within EARLINET (European Aerosol Research Lidar Network) have shown that aerosol plumes from the North American boundary layer are mostly not similarly intense, even during years with enhanced forest fire activity in the United States (EARLINET, 2003). Measurements also clearly demonstrate that Asian industrial sources account for at least some of the elevated aerosol events that are observed at the west coast of North America. Figure 3.8 in the preceding section illustrates an episode of elevated sub-micron aerosol scattering that corresponds to approximately 20 ug/m^3 of PM_1. The observed Hg^0/CO ratio in this episode indicates that the detected aerosol was associated with Asian industrial sources, not mineral dust or biomass burning (Jaffe et al., 2005; Weiss-Penzias et al., 2007).

During the ITCT-2K2 campaign, aircraft on the west coast of North America observed additional evidence of trans-Pacific transport of anthropogenic Asian aerosols. Figure 3.9 illustrates aerosol measurements made during an aircraft profile through layered plumes marked by variations in CO concentrations. Transport modelling identified Asia as the emission source region. In the layer with the highest CO concentrations (5.3 to 6.8 km), the observed particle size distributions and sulphate mass concentration were consistent with nearly pure sulphuric acid (H_2SO_4) particle formation over the mid-Pacific from gas-to-particle conversion following long-range transport of SO_2 through a mid-latitude cyclonic system. Such cloud systems effectively scavenge most pre-existing particles, but allow transport of gas-phase precursors that, in some cases, substantially alter downstream particle microphysical and chemical properties. The hygroscopicity of H_2SO_4 suggests a larger potential climatic effect if particles are transported intercontinental distances in the form of acid droplets rather than as fully or partially neutralized sulphates (Brock et al., 2004).

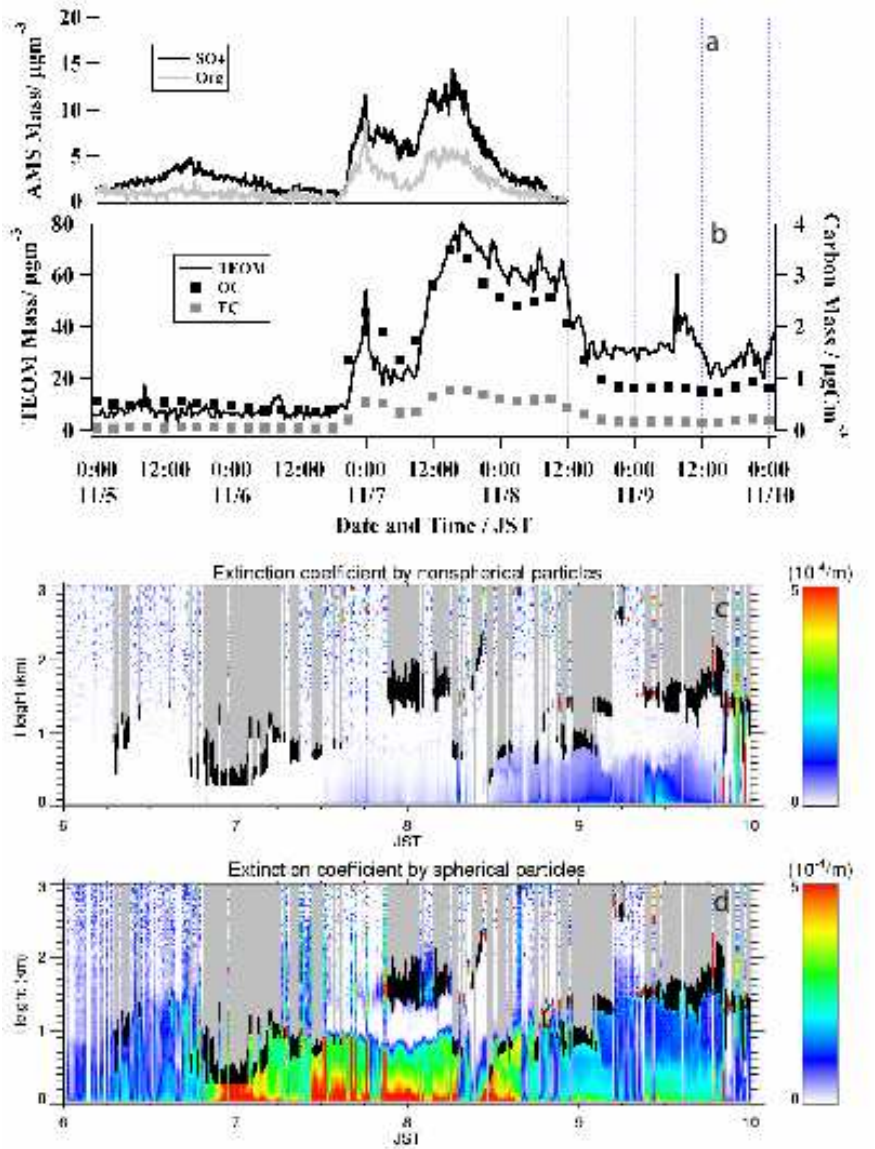

Figure 3.7 Aerosol transport event observed at Cape Hedo, Okinawa, Japan. From top: (a) sulfate and organic components; (b) organic carbon, elemental carbon and $PM_{2.5}$; and vertical distributions of c) dust (non-spherical particles) and (d) deliquesced anthropogenic aerosol (spherical particles) (Takami et al., 2006).

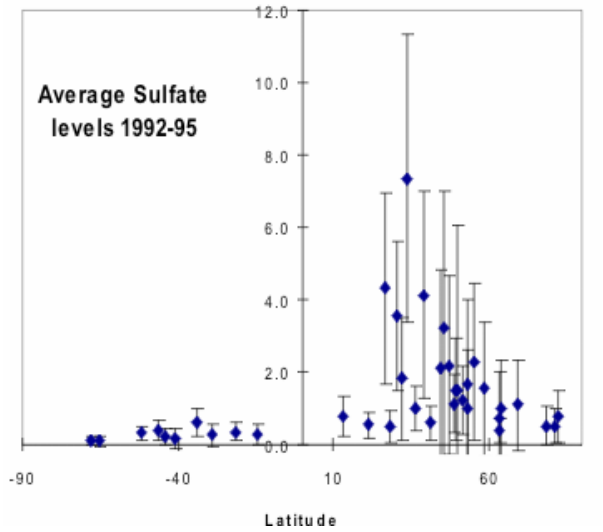

Figure 3.8 Average sulphate levels measured at oceanic sites in the Northern and Southern hemispheres (Barrie et al., 2001).

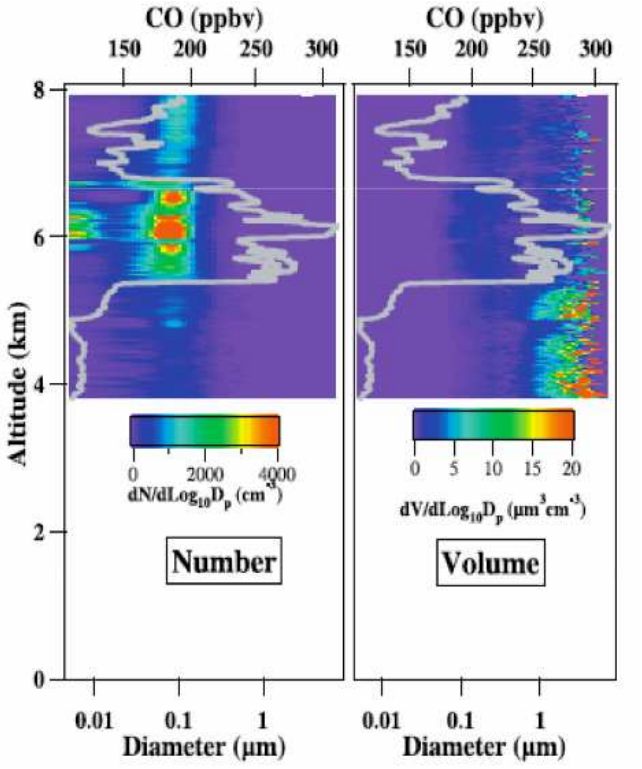

Figure 3.9 Aerosol measurements marked by variations in CO concentrations. Vertical distributions of CO with aerosol particle number size distribution (left) and particle volume size distribution (right) in a plume of emissions transported from Asia. The measurements were made near the west coast of North America on 5 May 2002. (Figure adapted from Brock et al. (2004)).

Below the CO-rich layer (3.7 to 5.3 km), a layer of enhanced particle volume with much lower CO concentrations was observed; the aerosol in this layer was dominated by mineral dust, also of Asian origin. Above the CO-rich layer (above 6.8 km) was another layer with elevated particle number concentrations; measurements of species such as acetonitrile, along with the modestly elevated CO concentrations, indicated that this layer carried biomass burning emissions.

3.3.3 Long-term record of aerosol transport across ocean basins

Strategically placed island sampling sites have provided important information about pollutant aerosol transport across ocean basins. In the North Pacific, weekly aerosol samples from Midway Island, 1981 to 2001, include nitrate and nss-SO_4 measurements. Concentrations show a strong seasonal cycle with maximum concentrations in spring, coincident with the cycle of Asian dust transport. The lowest concentrations occur in the summer, when dust transport is also at a minimum. The increased concentration of springtime nitrate and nss-SO_4 is attributed to the impact of pollution transport from Asia. Each spring, several large aerosol peaks occur at Midway that can be directly associated with large-scale pollution episodes seen in satellite images.

The impact of continental pollution sulphate sources at island sites in the North Atlantic is reflected in the presence of trace elements, e.g. antimony is found in high concentrations in emissions from smelters and from some fossil fuel combustion processes. At Bermuda, which is frequently impacted by pollution from North America, the peaks in sulphate are correlated with peaks in antimony. A similar close relationship is seen at Izaña Observatory, Tenerife, Canary Islands (figure 3.10). The high correlation between these two species affirms that pollution sources are dominant. At stations in the North Atlantic the impact of pollution sources is also evident in the relatively high concentrations of iodine, selenium, vanadium, and zinc (Arimoto et al., 1995). From comparisons of the measured concentrations of antimony and methanesulphonic acid (MSA, an oxidation product of DMS, a biogenic precursor of aerosol sulphate), Savoie et al. (2002) estimated that, on an annual

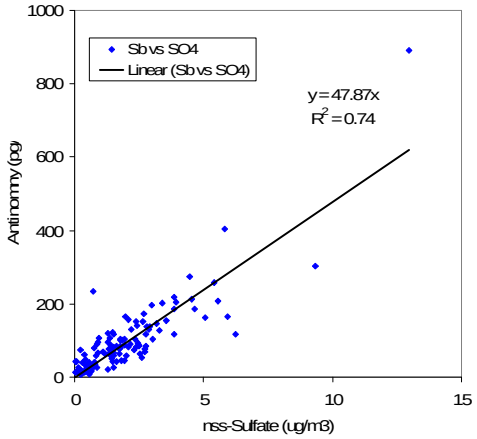

Figure 3.10 Correlation between the concentrations of antimony and nss-SO_4 at Izaña, Tenerife, Canary Islands, from 30 April 1989 to 30 April 1993. (Figure adapted from Savoie et al. (2002))

basis, anthropogenic sources account for about 50 per cent of the total nss-SO_4 in aerosols at Barbados, 70 per cent at Bermuda, about 90 per cent at Izaña, and 85–90 per cent at Mace Head. In contrast to these Atlantic sites, at remote southern ocean stations such as American Samoa, the concentrations of antimony in aerosols are at the detection limit of the analytical procedure.

3.3.4 Quantitative estimates of total pollution aerosol transport from satellite

In situ measurements provide strong evidence that intercontinental transport of anthropogenic aerosol from Asia to North America and across the Atlantic Basin is a significant and persistent occurrence. Satellites uniquely provide clear visual images of aerosol transport that considerably enhance the interpretation of in situ measurements. Images of the global distribution of aerosols clearly show the hemispheric/intercontinental transport of aerosols. Such transport has also been documented for many specific case studies; figure 3.11 shows one example. Such images provide qualitative evidence of intercontinental transport of aerosols, but generally they do not provide quantification of the transport flux, or distinguish between pollution aerosols and natural aerosols such as mineral dust, which often dominates these examples.

Following Kaufman et al. (2005), who applied a quantitative method to determine the net transport of dust from Africa to the Amazon basin, Yu et al. (2007) have used MODerate resolution Imaging Spectroradiometer (MODIS) aerosol products to determine the transport of "pollution" aerosol across the Pacific Basin. They isolate the pollution portion of the aerosol optical thickness measured by MODIS and translate the optical thickness to total column mass using relationships derived from field experiments. Analysed wind fields then allow a quantitative calculation of the amount of pollution mass leaving the Asian continent and the amount arriving at North America. Figure 3.12 shows the results. MODIS observes 15.2 Tg/yr leaving Asia within the latitude range of 30 to 60^{o}N and 4.2 Tg/yr arriving in North America. This compares well with a chemical transport model (Goddard Global Ozone Chemistry Aerosol Radiation Transport (GOCART)), which estimates 14.5 Tg/yr leaving Asia and 4.8 arriving in North America. Pollution is defined in the model as the sum of black carbon, organic material and sulphate components. The MODIS data show a seasonal cycle with a transport maximum in spring, but transport is observed in all seasons. Uncertainty analysis shows that the estimation of the aerosol transport height contributes the largest uncertainty to the satellite-based estimate. Adding information from CALIPSO (Cloud-Aerosol Lidar and Infared Pathfinder Satellite Observations), will decrease this uncertainty and make such satellite-based estimates of cross-oceanic transport a reliable constraint on model estimates.

3.3.5 Summary, remaining uncertainties and future needs

The intercontinental transport of aerosols, particularly mineral dust and smoke from forest fires, is qualitatively well documented. In situ measurements have characterized the chemical and physical aerosol properties, and in some cases illuminated secondary aerosol formation from transported precursors in a variety of case studies. Lidars, both surface-based and satellite-borne, are providing large data sets that promise to characterize the frequency of occurrence of aerosol transport events and the meteorological conditions responsible for them. Lidars also give some important information regarding the aerosol physical and chemical properties. Finally, tools are emerging to provide quantitative estimates of transport fluxes of pollutant aerosol as well as dust. This aerosol transport has important air quality implications; mineral dust can make significant contributions to PM in downwind continents. The long-range transport of aerosols also underlies two of the major drivers of regional and global climate change, the direct interaction of aerosols with solar radiation and the interaction of aerosols with clouds, which affects the radiative properties of those clouds. In

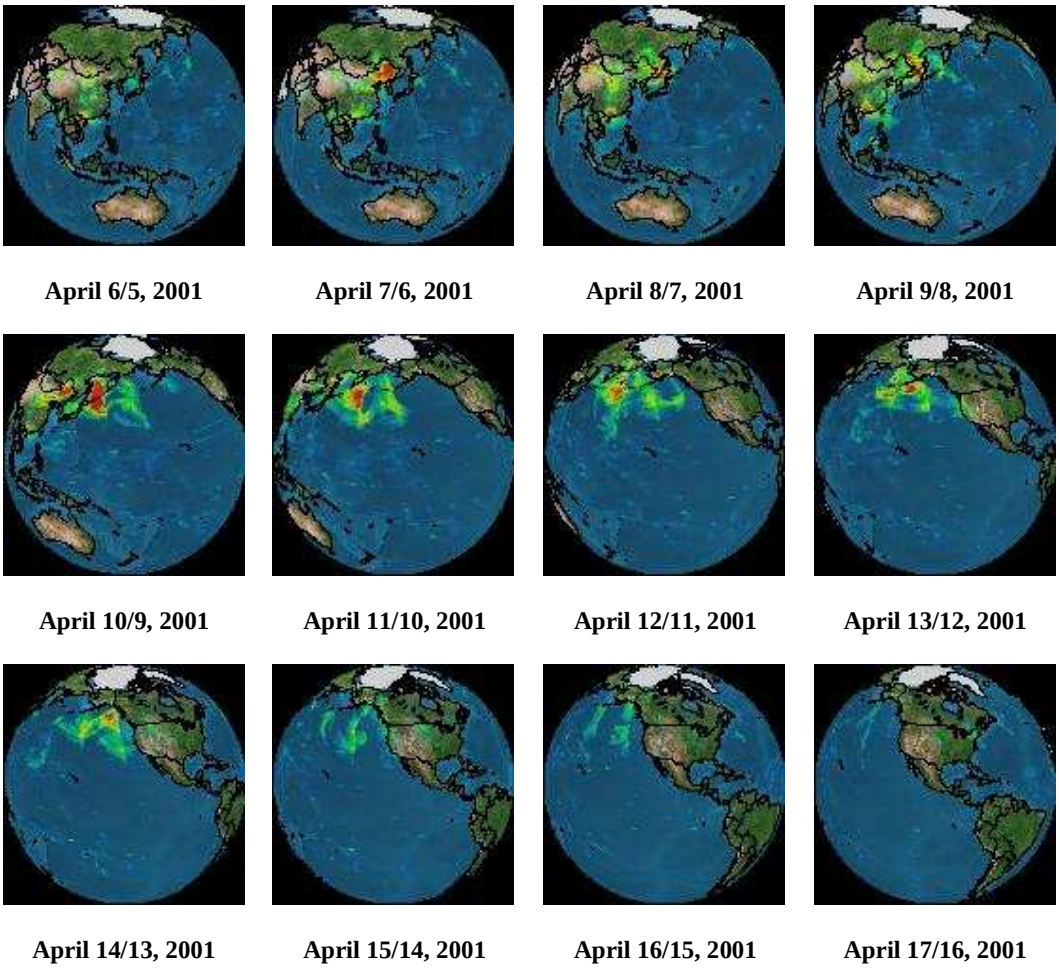

Figure 3.11 Hemispheric/intercontinental transport of aerosols. A sequence of daily (5–16 April 2001) aerosol index images from TOMS that shows the transport of a very strong aerosol event that originated in Asia and crossed the Pacific to North America. This aerosol was identified as desert dust. Two dates are shown for each image because the images straddle the international date line. (from http://jwocky.gsfc.nasa.gov/aerosols/today_plus/yr2001/asia_dust.html.)

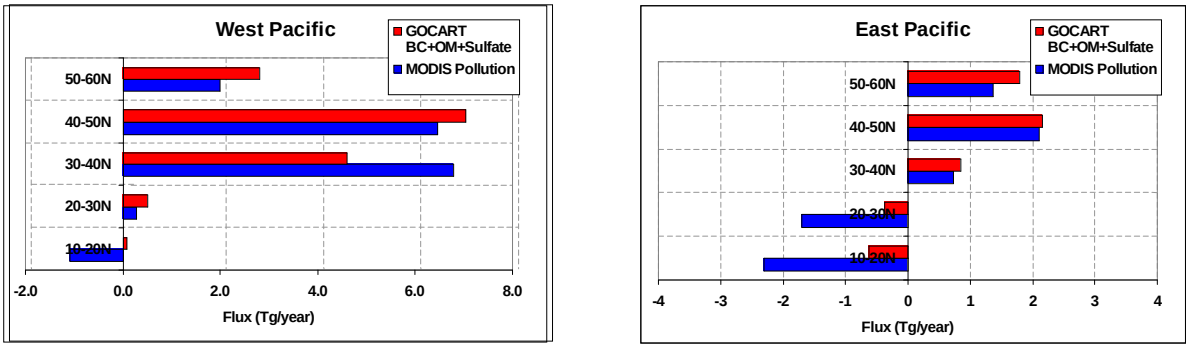

Figure 3.12 Calculated amount of pollution mass leaving the Asian continent and the amount arriving at North America. Estimates of annual pollution aerosol flux leaving Asia in the West Pacific (left) and arriving in North America in the East Pacific (right) for different latitude ranges. The blue bars are derived from MODIS aerosol products, and the red bars are estimates from the GOCART model (Yu et al., 2007).

this regard, the transport of secondary organic aerosols and black carbon are particularly important. A number of scientific questions should be the focus of future work:

- What are the sources and properties of the organic component of aerosols? Presently they are very poorly understood (Heald et al., 2005; Volkamer et al., 2006)What are the quantitative fluxes of aerosol (pollution, dust, biomass-burning emissions) transported between continents?
- What are the important source regions of black carbon? What are the relative magnitudes of these sources? How is it transported and removed from the troposphere?
- What is the optimum observation strategy for in situ measurements to characterize further the present intercontinental transport of aerosols as well as the future evolution of that transport?
- To what extent can satellite observations be used to track aerosols on a daily, weekly and monthly basis?
- How can we best integrate in situ observations and satellite data with regional and global models into a consistent picture of intercontinental transport?

3.4 Concentrations seen at downwind receptor locations and implications for surface air quality in those regions

From an air quality management perspective, we wish to know: What is the effect of long-range transport of air pollutants on the local concentrations of ozone and aerosols? The answer to this question has two aspects. First, what are the maximum concentrations transported to the receptor location during a transport event? Such maximum impacts can potentially push a local area over its air quality standard. Second, what is the effect of the transport on the average background levels? It is these background levels that determine how much local pollution can be tolerated before exceeding the local standard.

3.4.1 Ozone concentrations

Figure 3.6 illustrates the increase of O_3 in marine boundary layer air coming ashore at the west coasts of Europe and North America. The average trends observed imply an increase of approximately 10 ppbv over the preceding two decades. This increase represents a substantial fraction of the air quality standards on the respective continents (120 μg m^{-3} or about 60 ppbv, 8-hour average in Europe and 80 ppbv, 8-hour average in the United States). Clearly, less local O_3 production in continental regions receiving inflow from the upwind marine area can be tolerated before the ambient standard is exceeded. Figure 3.5 indicates that flow from the European continent also significantly impacts the background O_3 levels over the downwind Asian continent.

Episodic enhancement of surface level O_3 associated with Northern Hemisphere transport of O_3 and precursors has been observed at high-elevation locations in the western United States (Jaffe et al., 2005). Figure 3.2 shows O_3 concentrations exceeding 70 ppbv. The enhancement of the O_3 concentration above the background of about 40 ppbv was linked to transport of Asian anthropogenic emissions through back trajectory analysis and coincident chemical markers.

Model calculations are required to apportion the observed O_3 concentrations among "background," i.e. a long-range transport contribution, and local pollution. Figure 3.13 illustrates results from one such calculation for a relatively high-altitude site in California. The model results indicate that the 17 May plume of anthropogenic emissions (included in figure 3.3) impacted the site during this period. The 7–10 ppbv attributed to long-range transport is only about 10 per cent of the observed concentrations on that particular day, but that enhancement pushed the observed concentrations over the standard. Figure 3.14 illustrates an episode when O_3 concentrations exceeded the air quality standard at a lower altitude surface site (Jaffe et al., 2004). The long-range transport in this episode from Siberian forest fires increased surface level ozone values by about 15 ppbv. The

local and long-range contributions are shown stacked in different orders to demonstrate that transport and local pollution both bear responsibility for exceeding the standard.

Models also suggest that North American emissions have substantial effects on Europe. Li et al. (2002) find in their model that 20 per cent of the exceedance of the European Community ozone standard in the summer of 1997 over Europe would not have occurred in the absence of anthropogenic emissions from North America. These North American effects are relatively small and not easily detected in surface observations, but they are large enough to push the observed concentrations over the standard.

3.4.2 Aerosol concentrations

The discussion of the impact of aerosols on air quality in downwind regions has been generally limited to the episodic transport of dust. One particularly severe example of the transport of Asian dust is the April 2001 episode, which led to elevated surface levels of $PM_{2.5}$ in Salt Lake City, Utah, in the U.S. Rocky Mountain region and Atlanta, Georgia, in the eastern United States (figure 3.15). The dust was transported initially in the free troposphere, and then brought to the surface through a combination of processes, which differed from region to region. Subsequent mixing with local pollutants led to $PM_{2.5}$ levels exceeding the new daily standard of 35 $\mu g/m^3$. During this specific case, the dust passed above the Pacific Northwest (e.g. Seattle, Washington) and had greater impact at the surface in the Rocky Mountains and eastern United States. This event was diagnosed through a combination of air mass trajectory calculations, satellite imagery (episode illustrated in figure 3.11) and use of chemical markers (figure 3.16), proving a clear Asian dust signature crossing the United States. Dust transport from North Africa impacts a wide region of the Western Atlantic, the Caribbean, and the southern and eastern United States (Perry et al., 1997, ; Prospero, 1999). Figure 3.17 shows the monthly mean mineral dust concentrations measured on Barbados and at Miami, Florida, indicating levels approaching or exceeding the current United States daily standard for $PM_{2.5}$. Strong low pressure systems can sweep African dust in a northerly direction also, across the Mediterranean and into Europe as far north as England, Scandinavia (Engelstaedter et al., 2006) and Iceland (Sodermann et al., 2006).

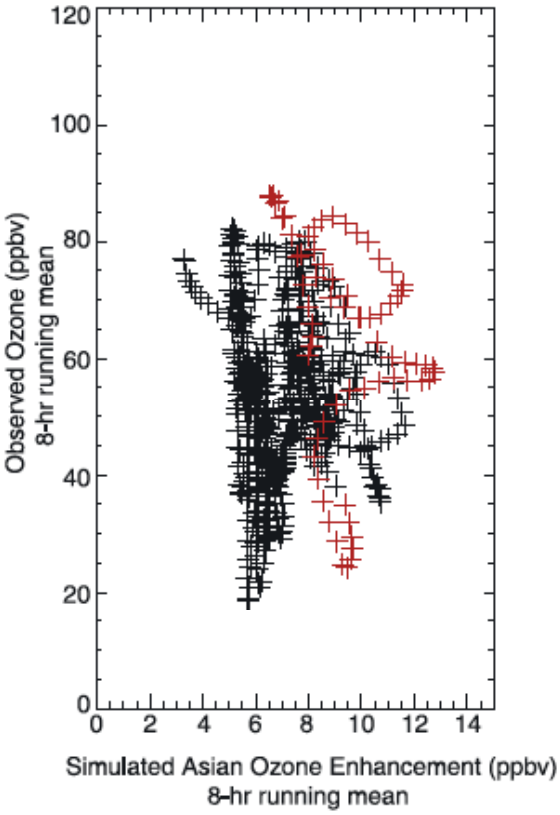

Figure 3.13 Ozone concentrations observed at Sequoia National Park, California (36 N, 118 W, 1890 m altitude) in May 2002 versus the corresponding Asian pollution enhancements simulated by the GEOS-CHEM model (Hudman et al., 2004). Red symbols indicate the period 17-20 May when observed O_3 exceeded the standard.

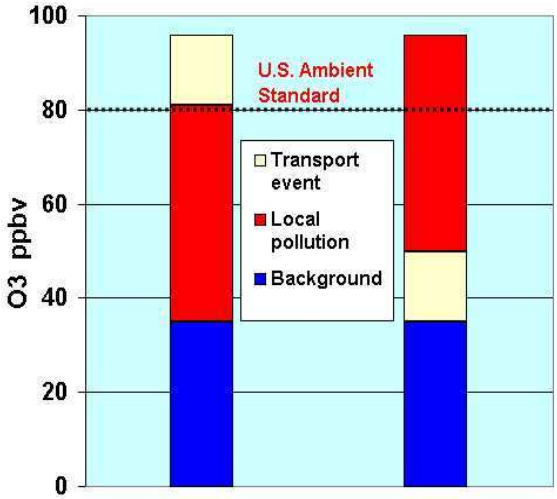

Figure 3.14 Contributions from three sources to surface O_3 for Enumclaw, Washington, on 6 June 2003 (Jaffe et al., 2004).

The impact of the long-range transport of anthropogenic aerosols on local and regional air quality is less well characterized by observations. However, this transport likely has significant effects in particular episodes. For example, the aerosol levels illustrated in figure 3.2 correspond to about one half of the United States daily $PM_{2.5}$ standard.

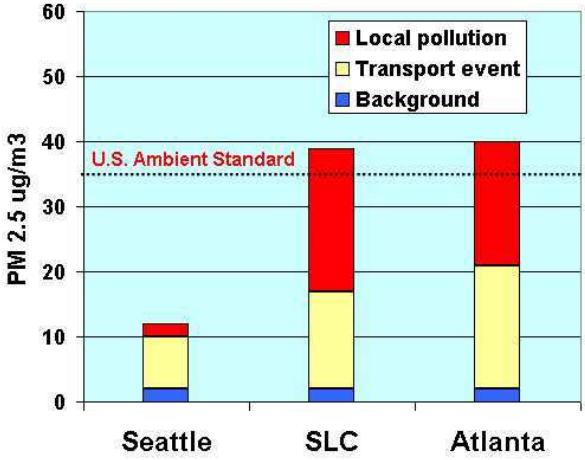

Figure 3.15 Contributions from three sources to surface $PM_{2.5}$ in three United States cities (adapted from Jaffe et al. (2003b)).

3.4.3 Summary, remaining uncertainties and future needs

Long-range transport affects the concentrations of ozone and aerosols at downwind receptor locations, but there exists a clear distinction between the effects of this transport on these two species. In situ observations document episodic elevated concentrations due to transport of both ozone and aerosols, but the latter are much more pronounced. Analyses of long-term observations also generally find that the background O_3 concentration in marine air at the west coasts of Europe and North America has increased over the past two decades. It is clear that O_3 is, in effect, primarily a hemispheric pollutant with a rising, hemisphere-wide background; only infrequent significant elevated surface levels result from long-range transport. For aerosols, spectacular enhancement events have been observed at the surface, but very clean background concentrations can still be observed throughout the globe. The episodic enhancement events do yield an average elevation of the total aerosol column over the long term.

Despite observations of the effects of long-range transport, the implications for surface air quality can only be determined from models, and these models are still rather uncertain. The future needs here lie largely in the improvement of models, both through improvement in the model parameterizations and in testing the model's ability to quantitatively reproduce both the observed episodic enhancements and the long-term increases in background concentrations.

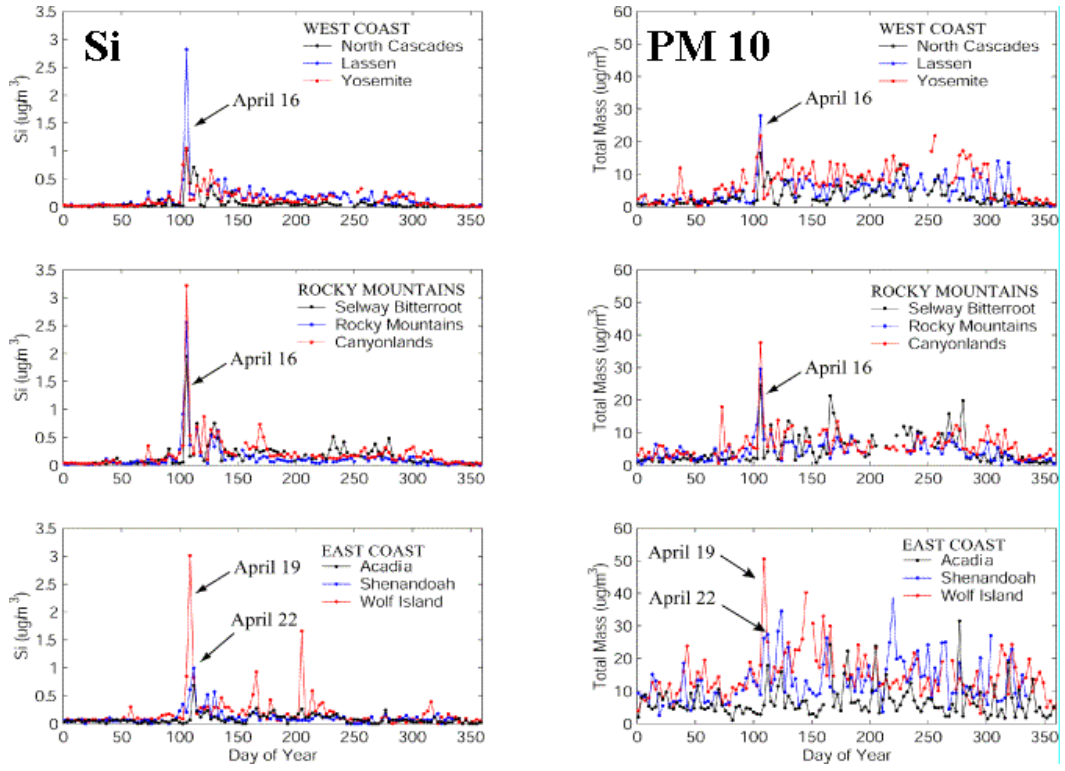

Figure 3.16 Eastern movement of Asian dust plume across the United States based on measurements from the IMPROVE (Interagency Monitoring of Protected Visual Environments) monitoring network. Silicon served as a dust tracer linking elevated PM concentrations to Asian-derived dust air mass (Jaffe et al., 2003b).

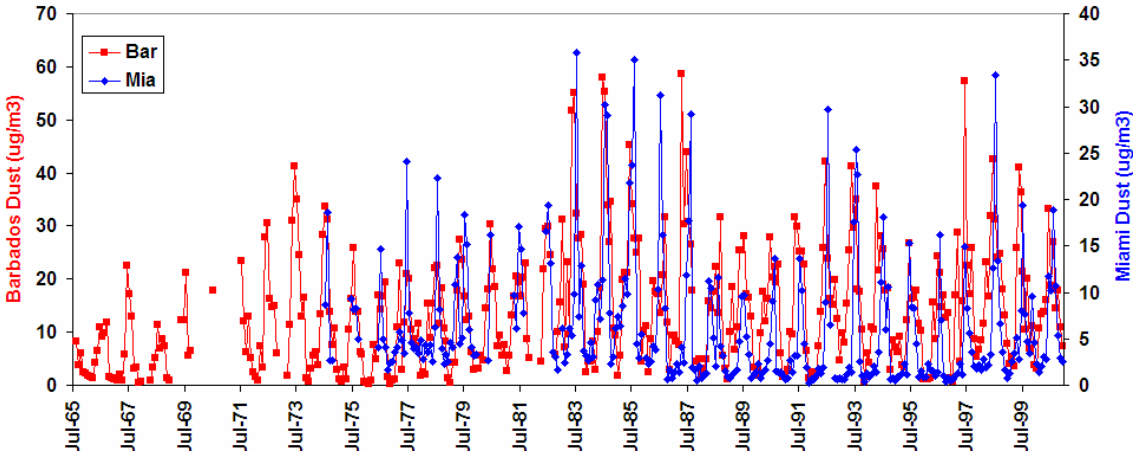

Figure 3.17 Monthly mean mineral dust concentrations measured at Barbados (red) and Miami, Florida (blue). (Prospero (1999) updated; Prospero and Lamb (2003) updated)

3.5 Observational evidence for attribution of source regions

From an air quality management perspective, we would also like to know: What are the relative contributions from different source regions to the long-range transport of air pollutants affecting the local concentrations of ozone and aerosols? The answer to this question is generally available only from model calculations and really never directly available from observations. However, there are inverse modelling approaches being developed that directly utilize observations to improve emission inventories. The emission inventories are in themselves not truly source attribution for locally observed ozone or aerosols, but they do at least identify the relative magnitudes of emissions of O_3 precursors and aerosols as a function of source region.

3.5.1 Inverse modelling of emissions from satellite data sets

Inverse modelling is a formal approach to infer pollutant source strength from observations of atmospheric concentrations. A chemical transport model is often used to calculate the emissions of various pollutants that would reproduce the observations. This "top-down" information is being used to evaluate and improve "bottom-up" emission inventories. Satellites provide a major source of observations used for inverse modelling of emissions.

The GOME (Burrows and Weber, 1999), SCIAMACHY (Bovensmann et al., 1999), and OMI (Levelt et al., 2006) satellite instruments provide the capability for global retrievals of tropospheric NO_2 and formaldehyde (HCHO) columns, as shown in figures 3.18 and 3.19, respectively. Tropospheric NO_2 columns are closely related to surface NO_x emissions due to: (a) the large increase in the NO/NO_2 ratio with altitude; and (b) the short lifetime of NO_x in the lower mixed layer. Leue et al. (2001) initially applied GOME retrievals of tropospheric NO_2 columns, together with an assumed constant global NO_x lifetime, to derive a global NO_x emissions inventory by mass balance. Martin et al. (2003) further improved on this approach by using coincident local information from a chemical transport model (GEOS-Chem) on the NO_x lifetime and the NO_2/NO_x ratio, and by combining top-down constraint with a bottom-up a priori inventory to produce an optimal a posteriori inventory. Jaeglé et al. (2005) developed a method for global partitioning of satellite-derived NO_x sources into contributions from fossil fuel combustion, biomass burning, and soil emissions. Subsequent improvements to NO_x inversions include developing an adjoint method for inference of emissions (Muller and Stavrakou, 2005), applying regional models at higher resolution (Kim et al., 2006; Konovalov et al., 2006), and better accounting for free tropospheric NO_2 in the inversion (Wang et al., 2007).

HCHO columns are closely related to surface VOCs emissions since (a) HCHO is a high-yield intermediate product from the oxidation of reactive non-methane VOCs and (b) reactive VOCs have a short lifetime. Palmer et al. (2003) first developed a method for deriving emissions of VOCs using GOME retrievals of HCHO over North America, by applying an inversion based on the relationship between the HCHO column and the sum of VOC emissions scaled by their HCHO yields. Abbot et al. (2003) found that the seasonal variation in GOME HCHO columns over North America is broadly consistent with the seasonal cycle of isoprene emission. Fu et al. (2007) applied GOME satellite measurements of HCHO columns over East and South Asia to improve regional emission estimates of reactive non-methane VOCs, and found the need for a 25 per cent increase in anthropogenic VOC emissions and a five-fold increase in biomass burning VOCs emissions in order to be consistent with the satellite observations.

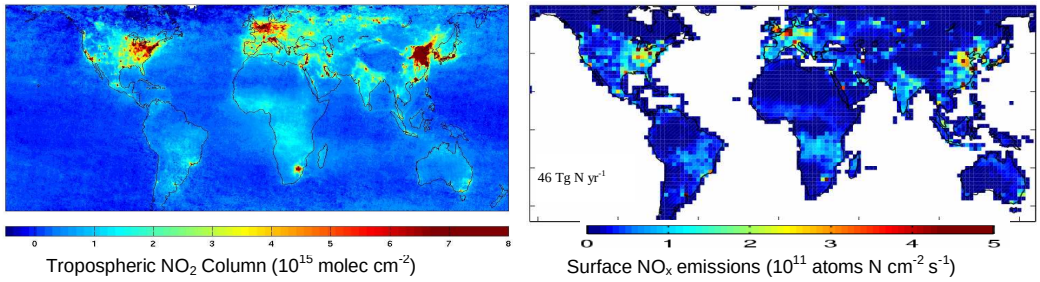

Figure 3.18 Tropospheric NO_2 columns captured with various satellite instruments (Left) Tropospheric NO_2 columns for 2004-2005 determined from the SCIAMACHY satellite instrument. (Right) Surface NO_x emissions for 2004-2005 determined through inverse modelling of the SCIAMACHY observations using a chemical transport model (GEOS-Chem). ICARTT Aircraft measurements support the SCIAMACHY inventory (Martin et al., 2006).

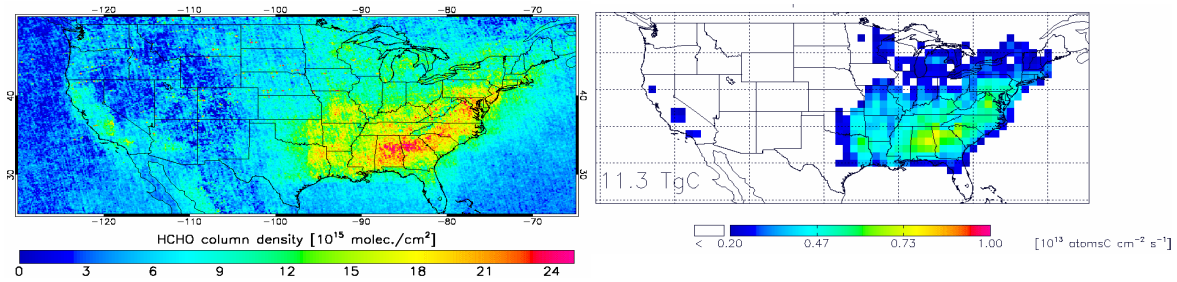

Figure 3.19 Tropospheric HCHO columns captured with various satellite instruments (Left) HCHO columns over the United States determined from the OMI (Ozone Monitoring) satellite instrument for summer 2006. (Right) Isoprene emissions determined through inverse modelling of the OMI observations using a chemical transport model (GEOS-Chem). Aircraft measurements show that isoprene is the dominant source of variability in column formaldehyde over North America (Millet et al., 2006).

The availability of near-global and long-term CO observations from space, in conjunction with global chemical transport models, has allowed the use of inverse methods in constraining regional CO sources (Arellano Jr. et al., 2004; Arellano Jr. et al., 2006; Heald et al., 2004; Petron et al., 2004; Pfister et al., 2006; Stavrakou and Muller, 2006), as shown in figure 3.20. This has provided insight into our understanding of present-day fossil-fuel and biofuel use and on the patterns of biomass burning activity. For instance, inverse modelling studies using MOPITT CO retrievals consistently report that emissions from fossil-fuel and biofuel use in Asia are significantly larger than previously estimated. This finding contributed to recent efforts to revisit and update the bottom-up emission inventory in Asia (Streets et al., 2006). In addition, these inverse modelling studies provide important constraints on the magnitude and spatio-temporal variability of biomass burning emissions, which are poorly constrained in current global models, particularly in southern Africa, South America, Southeast Asia and the boreal regions. Such information has important implications for better characterizing the distribution of CO and other chemical and radiatively-active atmospheric constituents that are not observed but are also products of anthropogenic combustion activities such as CO_2 and aerosols (Randerson et al., 2006).

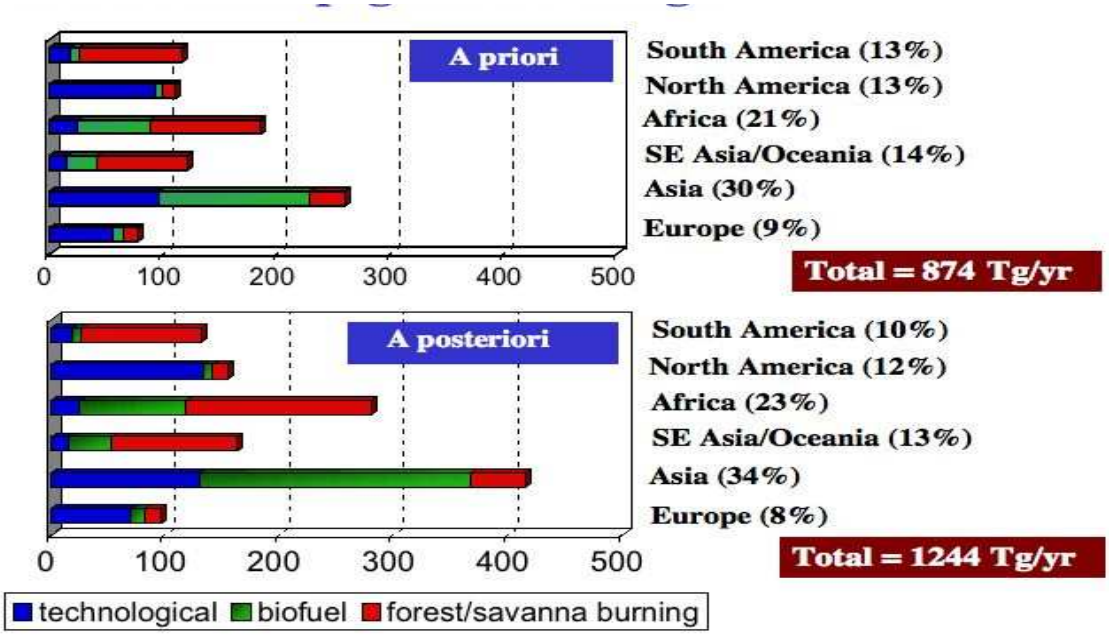

Figure 3.20 Anthropogenic CO emissions by different sources and geographical regions from previous inventory estimates, a priori (above), and after performing a linearized Kalman filter inversion of the MOPITT CO data at 700 hPa in conjunction with the MOZART-2 chemical transport model, a posteriori (below). (Petron et al., 2004)

Dubovik et al (2006) demonstrated inverse modelling techniques applied simultaneously to both pollutant and naturally occurring aerosols using MODIS aerosol optical thickness data with the GOCART model. The authors identified and quantified the major aerosol sources during a one-week period in August 2000 (figure 3.21). Sources were identified in the southeast United States, the Amazon basin, Europe, West Africa, southern Africa, the Indo-Gangetic Basin and East Asia. Emission strengths in these primary source regions exceed 0.2 x 10^7 kg mass/day. Inversions for specific aerosol types such as pollution or biomass burning are also possible. Using such inversion techniques, we can improve emissions inventories and produce emission estimates for highly variable emissions sources, such as biomass burning.

3.5.2 Summary, remaining uncertainties and future needs

The current satellite database offers a useful constraint on evaluating and improving the inventories of O_3 precursors and aerosols in current models. Primary weaknesses include a lack of in situ observations for use in evaluating satellite retrievals of short-lived trace gases such as HCHO and NO_2, a lack of information on vertical profiles of trace gases, and the inability of polar-orbiting satellites to observe diurnal variation in emissions. Additional aircraft measurements of reactive nitrogen species and speciated VOCs in highly polluted regions would be particularly valuable for evaluation of both the retrieved columns, and of the chemical transport model inversions to relate tropospheric NO_2 and HCHO columns to emissions. Such aircraft measurements should be closely coordinated with ground-based measurements to ensure adequate coverage of the lower mixed layer. Satellite observations at higher temporal resolution, such as from a geostationary platform, would be particularly valuable in constraining the diurnal variation of emissions.

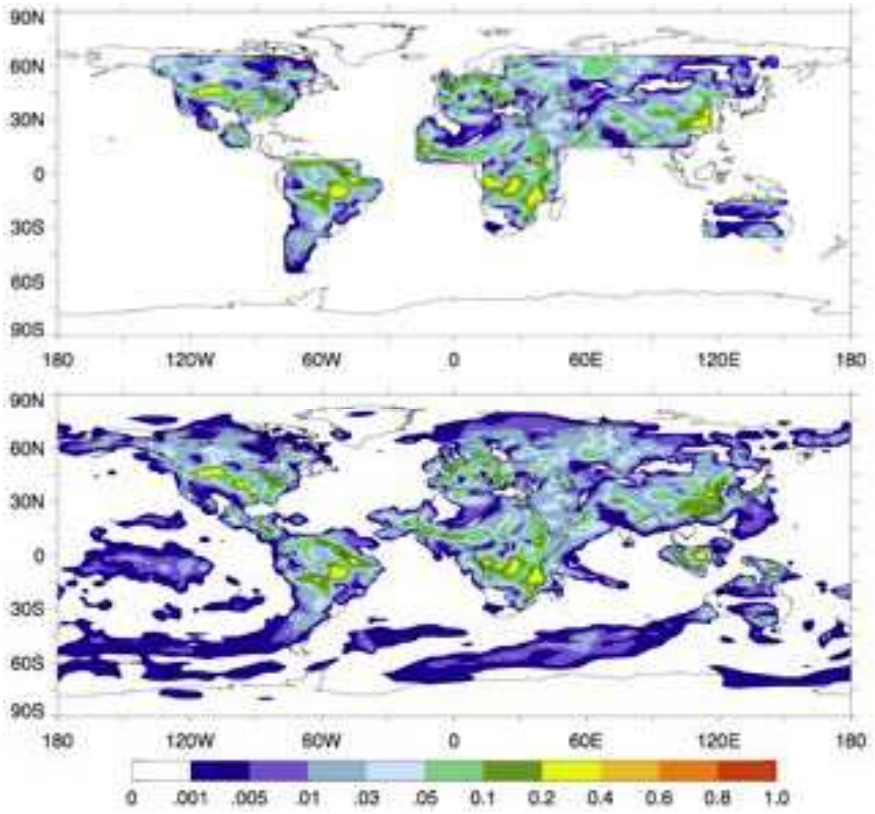

Figure 3.21 Averaged (20-28 August, 2000) fine aerosol sources (10^7 kg mass/day) retrieved from MODIS aerosol optical depth data. Upper panel: retrieval with emission constrained to land. Lower panel: retrieval with emission not constrained to land (Dubovik et al., 2006).

Obtaining concentrations of air quality relevant trace gas species in the planetary boundary layer (PBL) has been identified as a priority measurement in a number of studies (Edwards, 2006; IGACO Theme Team, 2004; National Research Council, 2007). This is essential for determining emissions estimates and characterizing urban pollution. The alternatives are total column or free troposphere satellite measurements, both of which require model assumptions during inverse modeling to estimate a PBL (Planetary Boundary Layer) concentration and derive sources. The problem here is that PBL descriptors (venting and height) and convection parameterizations are among the least certain modeling elements. In addition to source determination, a measure of PBL concentration in conjunction with free troposphere profile information allows local production to be separated from transported pollution. Coincident multi-spectral observations of CO are probably the best candidate satellite measurements to address this issue.

3.6 Ability to track long-term trends in hemispheric transport from existing surface observations

Due to great year-to-year variability, long-term monitoring (typically more than a decade) at carefully chosen sites is essential to assess trends in intercontinental transport from surface observations. The interannual variability at receptor sites is largely driven by the temporal and spatial variability of upwind emissions and of the weather and climate patterns that determine transport processes. The characterization of trends in hemispheric transport is especially difficult because local effects, with possibly confounding temporal trends, often obscure the influence of that transport. Measurements intended to characterize intercontinental transport are best carried out at remote sites that are relatively free of local and regional impacts. Island sites are ideal, especially those that are

sparsely inhabited or minimally developed. Coastal and mountain sites may also be suitable so long as attention is given to local and regional sources that might impact on the site and to the vagaries of local circulations, e.g. sea-breeze cycles or mountain-induced circulations induced by diurnal heating and cooling cycles and regional-scale forcing induced by the wind field interacting with the mountain mass. Unfortunately, there are very few data sets that meet these requirements. In the following sections we present some examples.

3.6.1 Characterization of ozone trends

A number of studies and assessments have been made in an effort to elucidate large-scale trends in O_3 concentrations using data from continental networks. The EMEP Assessment (EMEP, 2004) is typical. It shows that trends across Europe are dominated by changes in regional emissions, thus making it difficult to elucidate the impact of long-range transport. Nonetheless, at some relatively remote continental sites it is possible to extract information on trends by the careful selection of samples based on air mass trajectories or other criteria. Examples are shown in figures 3.6 and 3.22, which all show positive temporal trends. However, a critical uncertainty exists regarding the validity of the O_3 concentration trends presented in these figures. Oltmans et al. (2006) have recently reviewed evidence for temporal trends in tropospheric ozone from surface and ozone sonde sites. They generally find increasing trends in surface O_3 over Europe in agreement with the results presented here in figures 3.22 and 3.6a. However, they find no significant trend in western North America, a conclusion in clear disagreement with the result presented here in figure 3.6b. This disagreement, unless it can be resolved by further analysis, strongly suggests that existing surface observations are not adequate to track conclusively the trends in transport of ozone in the lower troposphere.

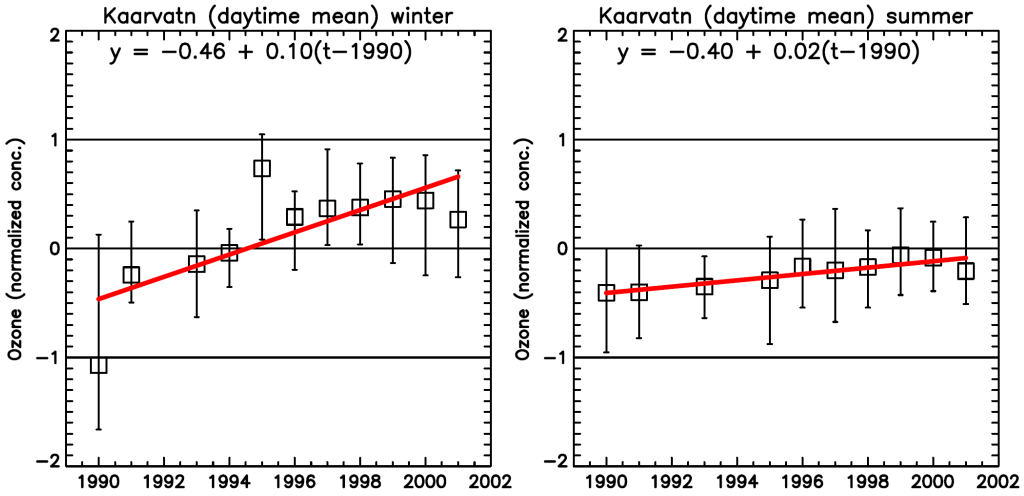

Figure 3.22 Trends determined by samples based on air mass trajectories. Linear trends in median, normalized ozone concentrations at Kårvatn, mid-Norway, based on data filtered by back trajectory for only the background transport sectors. The boxes mark the annual medians, and the error bars the 25- and 75-percentiles (EMEP, 2005).

3.6.2 Characterization of trends in dust transport

Mineral dust is a major and sometimes dominant aerosol component over many ocean regions (Cakmur et al., 2006; Mahowald et al., 2005). North Africa is the world's most active dust source (Engelstaedter et al., 2006; Prospero et al., 2002) with strong transport to the Atlantic, the Caribbean, North America and Europe (Perry et al., 1997; Prospero, 1999). Winds also carry large quantities of dust out of Asia and over large areas of the North Pacific (Prospero et al., 1989) and across North America (Husar et al., 2001; VanCuren, 2003). The resulting dust concentrations downwind from

these two regional sources exhibit strong seasonal cycles with the maximum in boreal summer (African source) or spring (Asian source). In addition, there is considerable interannual and long-term variability that is linked to weather and climate processes in the continental source regions.

The intercontinental transport of African dust is perhaps the best-documented example of long-range transport on intercontinental scales because of the long-term surface-level measurements made in a network of island and coastal stations in the Atlantic as well as in Miami, Florida. The longest record is from Barbados, West Indies, where daily measurements began in 1965 and continue to the present (figure 3.17) (Prospero and Lamb, 2003). Most notable in that record is the sharp increase in dust transport in the early 1970s in response to the onset of prolonged drought in the Sahel-Soudano region. Drought, and the corresponding increase in dust transport, appears to be linked to large-scale climate processes. Although concentrations are somewhat lower in Miami than Barbados and the dust transport season is shorter, the longer-term variability is similar at both sites. Measurements at Bermuda between 1987 and 1998 also show the strong and persistent impact of African dust comparable to that in Miami.

Measurements of Asian dust made on Midway Island from 1981 to 2001 show that there were periods in the mid-1980s and in 1998 when dust transport increased sharply (Prospero et al., 2003). There had been considerable speculation that the increase in dust activity in the late 1990s was due to increased impacts on land use resulting from the rapid economic expansion in China. The record on Midway argues against this explanation because there was no evidence of a long-term trend, only a sharp increase in the late 1990s that is now attributed to drought that occurred at that time. This conclusion is now generally accepted based on dust studies in China.

Temporal trends in dust transport, at least from these two major global source regions, have been reliably documented from surface observations. However, there is considerable uncertainty regarding our ability to extend these measurement records over the coming decades due to a lack of resources committed to such efforts.

3.6.3 Characterization of trends in transport of sulphate and nitrate aerosol

Measurements of NO_3 and nss-SO_4 concentrations on island sites in the Pacific and the Atlantic can provide temporal trends in the long-range transport of these pollutants from the Asian and North American source regions. The trend in pollution SO_4 can be estimated by computing the "natural" non-sea-salt sulphate based on the MSA concentrations and subtracting it from the total nss-SO_4.

On Midway, NO_3 and pollution SO_4 concentrations show a strong seasonal cycle with maximum concentrations in spring, coincident with the increased dust transport noted previously. This cycle is attributed to the impact of pollution transport from Asia. Figure 3.23 shows that anthropogenic SO_4 concentrations approximately doubled from 1981 to the mid-1990s, a trend that closely matches the increase in SO_2 emissions from China over that period (Streets et al., 2000). Nitrate concentrations (not shown) yield a similar trend over the same time period. The data also suggest that anthropogenic SO_4 and NO_3 decreased in the late 1990s, as did emissions. However the data become less reliable because of the effects of the very strong El Niño in 1997–1998 and because of operational difficulties that led to the termination of the observations in 2001.

The annual mean concentrations of nss-SO_4 and nitrate NO_3 measured in Bermuda (figure 3.24) show coherent patterns. Aerosol nss-SO_4 decreased steadily from the start of the measurements until the mid-1990s, then stabilized and finally increased slightly thereafter. The emissions of SO_2 from United States sources decreased substantially over this time period by an amount that roughly corresponds to that observed at Bermuda, yet the timing of the decreases are markedly different— the United States SO_2 emissions decreased sharply between 1994 and 1995 when more stringent controls were mandated. In contrast, there is no substantial change in nitrate (NO_3) over this period, in agreement with the trend in NO_x emissions, which show only a very slight increase over the period. The substantial differences between the aerosol record and the emissions record demonstrate the challenges in attempting to monitor long-term trends at distant receptor sites.

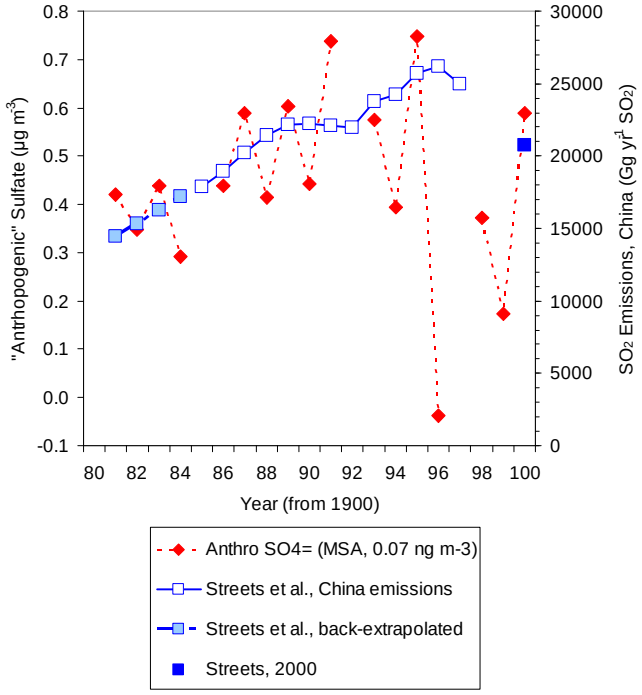

Figure 3.23 Anthropogenic sulphate concentrations on Midway Island compared to the emissions of SO_2 from China (Prospero et al., 2003).

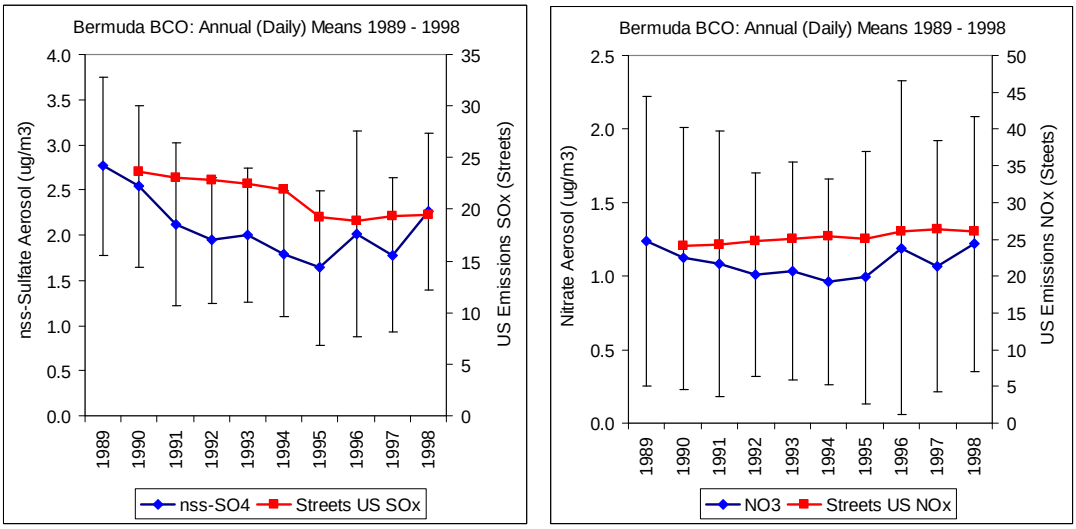

Figure 3.24 Annual mean nss-sulphate and nitrate concentrations on Bermuda during onshore winds compared to the eastern United States SO_2 (Tg S/yr) and NOx emissions (Tg N/yr). (Based on data discussed in part in Savoie et al. (2002)

Aerosol SO_4 and NO_3 measurements have also been made continuously on Barbados since 1989. This record suggests that NO_3 remained unchanged and that nss-SO_4 decreased by about 20 per cent. Approximately half of the NO_3 and nss-SO_4 at Barbados is attributable to anthropogenic sources (Savoie et al., 2002), predominately from European sources (Hamelin et al., 1989). If we assume that the change in nss-SO_4 concentration is due to the transport of pollutants and if we take into consideration that about half of the nss-SO_4 is natural (from DMS), then the actual decrease in pollutant SO_4 is roughly 40 per cent, a change that is consistent with the sharp drop in European SO_2 emissions over this time period (EMEP, 2004). However the absence of a discernable change in Barbados NO_3 concentrations is puzzling in the light of the substantial reduction of European emissions of NO_x, roughly 25per cent (EMEP, 2004).

3.6.4 Summary, remaining uncertainties and future needs

There are analyses that derive trends in the long-range transport of O_3, dust and anthropogenic NO_3 and SO_4 aerosol. However, there are critical uncertainties. The disagreement over the validity of derived trends in O_3 is of great concern, and limits their utility for the testing of models. The available data for O_3 and aerosols are limited to a small number of sites, and for O_3 generally to only the past two decades. In some cases (especially for dust), large interannual variability obscures more systematic trends. The derived trends in NO_3 and pollutant SO_4 aerosol transport are usually consistent with estimated trends in the precursor emissions, but there is at least one disagreement (the NO_3 trend in Barbados). There is considerable uncertainty regarding our ability to extend these measurement records over the coming decades.

Long-term monitoring data are important for the development and testing the wide variety of models currently being used to address aerosol transport issues (Textor et al., 2005). Such models currently play an important role in addressing environmental issues on regional scales and these models are being extended to larger scales. However, we will only be able to rely on the models when they are thoroughly tested against long-term data sets acquired on continental and global scales. It is notable that in the IPCC 2001 assessment, the only aerosol data available for model evaluation over the oceans was that obtained in the University of Miami aerosol network that was in operation during the 1980s and 1990s. That network no longer exists. Thus there is no data available for future model development.

3.7 Concluding remarks

Two conclusions are clear from the preceding discussion. First, observations from surface sites, vertical profiling, intensive campaigns and satellites provide a wealth of evidence that long-range transport of O_3, its precursors, aerosols and their precursors profoundly impact the distribution of these species throughout the troposphere; sections 3.2 and 3.3 present a small sample of these observations. Second, global chemical transport models are required to provide the policy-relevant information needed in sections 3.4 and 3.5 (implications for surface air quality at downwind receptor locations, and attribution of source regions). Such models can provide answers to all questions that may be raised about long-range transport, or indeed about any issue concerning the chemistry or transport in the atmosphere. Unfortunately, however, the models do not accurately represent all relevant aspects of the atmosphere, and there remains a great deal of uncertainty in the answers provided by the models. One major goal of ongoing research in atmospheric chemistry and transport can be succinctly stated – to increase the accuracy of the model representations and reduce the uncertainty of the answers they give. This model improvement can only come through a close collaboration between the model development and the observation communities. The goal of this section is to discuss the future observational capabilities needed to improve our understanding of hemispheric transport of O_3 and aerosols within this close model-observation collaboration.

For O_3, a combination of surface-based networks, vertical profiling (by aircraft flights, sondes and ground-based lidars) and satellite sensors measure complementary aspects of the tropospheric O_3 distribution. Reproducing these measured distributions has constituted the primary tests of model simulations that have been conducted up to this point. Reproducing the long-term trends potentially available from surface measurements is an additional challenge for models. This challenge has not

been widely addressed in the past, but that promises to change; the WCRP-SPARC/IGBP-IGAC Atmospheric Chemistry and Climate Initiative intends as their first activity, a 20- to 25-year hindcast of the tropospheric distribution of O_3 and aerosols. Intensive field campaigns advance our understanding of atmospheric processes and provide characterizations of the interrelationships between O_3, its precursors, other photochemical products, as well as other species. Incorporation of this process understanding into models is critical for their improvement. Reproducing the observed interrelationships is another, largely unaddressed challenge for models; the model intercomparison currently being conducted by the Task Force will address some aspects of this challenge.

The long-range transport of air pollution is an international problem, with each continent the receptor as well as the source of the emissions. Likewise, understanding the issue will require international measurement and modelling efforts. Following is an outline of required measurement programmes. Recommendations emerging from this assessment, as well as future policy agreements, should include provisions for long-term maintenance of these measurement programmes.

3.7.1 Surface site needs

Over the coming decades, significant changes are expected in the long-range transport of air pollutants due to changing emissions, most probably increases from rapidly developing economies and continued decreases from the more mature economies. Figure 3.25 and table 3.1 describe a surface measurement network suitable for following the evolution of long-range transport of air pollution in the Northern Hemisphere. Both high elevation and near sea-level sites are included. Each can typically serve multiple uses, including assessments for climate change, stratospheric ozone depletion, as well as long-range pollutant transport and model development. Coincident measurements of O_3 and aerosol components (NO_3, SO_4, organic and elemental carbon, trace metals), precursors of O_3 and aerosol (total reactive nitrogen, peroxyacetyl nitrate, and VOCs, SO_2) and atmospheric tracers (such as CO and CO_2 and Hg) will strengthen long-range transport assessments and model evaluation efforts.

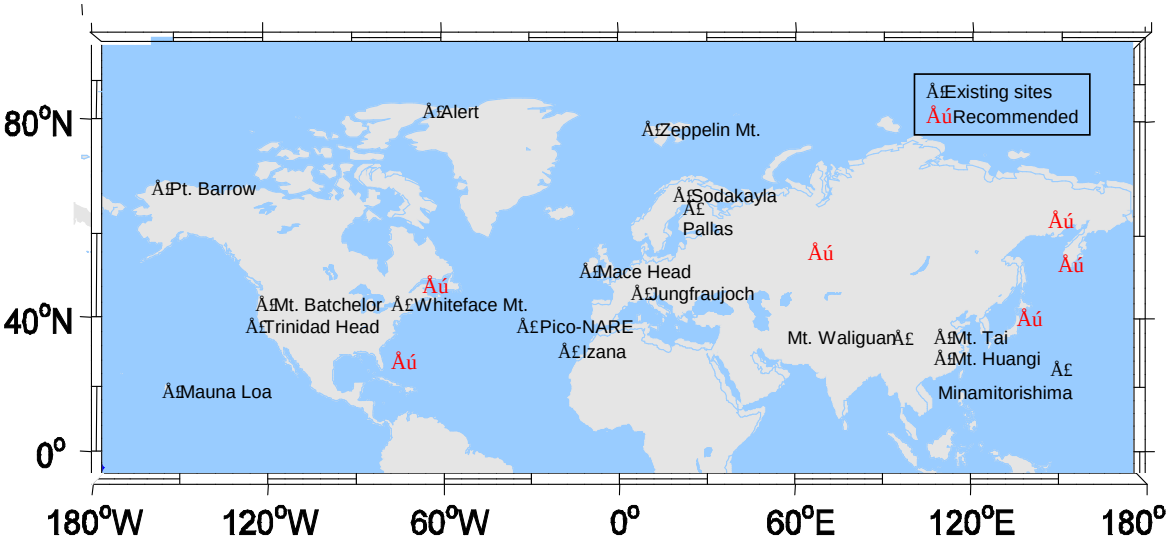

Figure 3.25 Surface site network for monitoring long-range transport of air pollutants.

Table 3.1 Long-term Northern Hemispheric trace gas and aerosol surface stations.

Station	Location		Notes	Information
Alert	**Canadian Arctic (210 m asl)**	**82.450°N 62.517°W**	**GAW global site**	**http://www.empa.ch/gaw/**
Izana	**Spain (Tenerife Is., west coast of Africa) 2360m asl**	**28.300°N 16.500°W**	**GAW global site**	**http://www.empa.ch/gaw/**
Jungfraujoch	**Switzerland 3580 m asl**	**46.548°N 7.987°E**	**GAW global site**	**http://www.empa.ch/gaw/**
Mace Head	**West coast, Ireland, 5m asl**	**53.326°N 9.899°W**	**GAW global site**	**http://www.empa.ch/gaw/**
Mauna Loa	**Hawaii, U.S. 3397 m asl**	**19.539°N 155.578°W**	**GAW global site**	**http://www.empa.ch/gaw/**
Minamitor-ishima	**Japan (2000 km SE Tokyo) 8m asl**	**24.300°N 153.967°E**	**GAW global site**	**http://www.empa.ch/gaw/**
Mt. Batchelor	**Oregon, U.S. 2700m asl**	**43.979°N 121.687°W**	**University of Washington**	**http://research.uwb.edu/jaffegroup/modules/MBO/**
Mt. Waliguan	**China 3810 m asl**	**36.283°N 100.900°E**	**GAW global site**	**http://www.empa.ch/gaw/**
Pallas	**Finland 560m asl**	**67.974°N 24.116°E**	**GAW global site**	**http://www.empa.ch/gaw/**
Trinidad Head	**West coast, CA, U.S., 107m asl**	**41.050°N 124.15°W**	**NOAA, U.S.**	**http://www.cmdl.noaa.gov/obop/thd/**
Pt. Barrow	**Alaska, U.S. 8 m asl**	**71.323°N 156.609°W**	**GAW global site**	**http://www.empa.ch/gaw/**
Pico Mountain	**Azores, Portugal 2225m asl**	**38.82°N 28.242°W**	**University of Azores**	**http://www.cee.mtu.edu/~reh/pico/**
Sodankylä	**Finland 180m asl**	**67.367°N 26.650°E**	**GAW global site**	**http://www.empa.ch/gaw/**
Whiteface Mt.	**New York, U.S. 1500m asl**	**44.4° N 73.9° W**	**ASRC SUNY, Albany**	**http://www.asrc.cestm.albany.edu/research/whiteface.htm**
Zeppelin Mt.	**Norway 474m asl**	**78.908°N 11.881°E**	**GAW global site**	**http://www.empa.ch/gaw/**

The network comprises a limited number of presently operating and proposed sites. The limits are a consequence of the pragmatic issues associated with servicing remote sites and a general lack of long-term funding commitments that compromise not only a needed expansion of adequate stations, but also the sustainability of existing platforms. Most of these stations are part of the World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) programme, and share data information systems and quality assurance protocols. While GAW enhances data consistency and access, funding for these sites is dependent on agency sponsorship, which is subject to changing research and programme priorities and budget levels. Tracking long-term changes is a major advantage of surface stations and requires sustained support over decadal time periods. In addition, there are important locations (e.g. Central and East Asia, east coast of Asia, central North America and North American east coast) that represent significant gaps in key transport corridors requiring additional support.

3.7.2 Vertical profiling needs

Vertical profiling is necessary to complement measurements at surface sites to help characterize the venting of pollutants from surface sources to the free troposphere and investigate the long-range transport that often occurs in layers that are elevated above the surface. Sondes, lidars and aircraft profiles can provide this profiling. The existing MOZAIC and other proposed programmes can provide at least some of this required complement.

The MOZAIC programme has made more than 20,000 flights between 1993 and 2004 that provided O_3 and water vapour measurements. The last three years of the programme also included CO and NO_y measurements. Most of the data have been collected in the upper troposphere or lower stratosphere, where significant pollution transport events are occasionally observed. More

importantly, two vertical profiles are obtained each flight (i.e. >40,000 total vertical profiles). It is proposed to continue and expand the programme to 20 aircraft for the next 20 years as part of IAGOS[5]. The expansion will include the addition of aerosol, CO_2 and NO_x measurements. One limitation of the programme is that the profiles are measured only over the locations of major airports, which are predominately localized in Western Europe and the east coasts of North America and Asia.

Cooper et al. (2006) have demonstrated the scientific results that are potentially available from a coordinated network of daily ozone sonde releases. Figure 3.26 illustrates a proposed network of launch sites. Such a network would be invaluable for characterizing the tropospheric O_3 distribution and for improving our understanding of the O_3 budget of the troposphere. The sites are positioned to provide data that will allow the quantification the intercontinental transport of O_3. The estimated cost for daily launches from such a network is about US$ 20 million per year.

Aerosol lidars provide detailed information regarding aerosol transport events. The European community has been particularly active, deploying several lidar systems capable of aerosol backscatter measurements up to at least 5 km; some of these systems also provide O_3 vertical profiles.

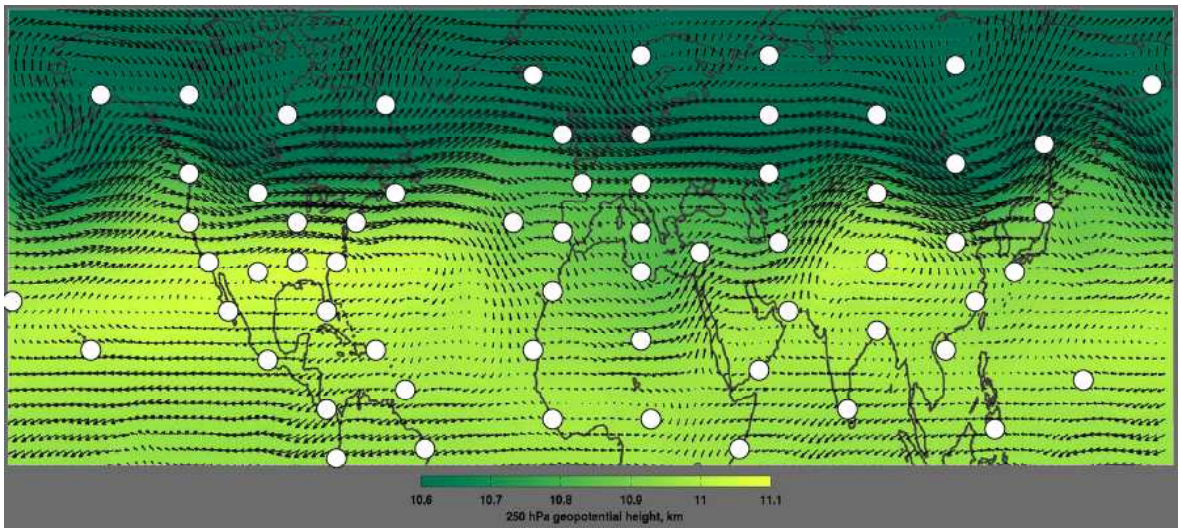

Figure 3.26 Launch site locations for proposed O_3 sonde network.

3.7.3 Satellite data needs

There is great near-term potential for advancing our understanding of long-range transport from satellite observations. Unprecedented satellite-borne measurement capability (IGAC, 2007) is presently in orbit. Those instruments will be joined by a few more in the coming years. Also, great progress has been made in developing retrieval algorithms that yield quantitative results. Presently, two factors limit the use of satellite data sets: development of a clear understanding of the accuracy and precision of the retrieved results (i.e. satellite validation); and resources available for the analysis of the satellite data sets, and indeed for the validation process itself. Making all satellite data available for free in an easily accessible and timely manner would increase the value of the measurements. The development and launching of the satellite instruments is so expensive that it is imperative that resources be made available to take full advantage of the resulting data sets.

In the longer-term, potential advances from satellite observations may be much more limited. Many of our current missions are maturing and already past their design lifetimes. Other missions have just been launched, but there is no comprehensive plan for maintaining capability for observations of atmospheric composition. To quote the recently published United States National Research Ccouncil (NRC) Decadal Survey recommendation “The United States Government, working in concert with the private sector, academe, the public, and its international partners, should renew its investment in Earth observing systems and restore its leadership in Earth science and

[5] Integration of routine Aircraft measurements into a Global Observing System.

applications." Within the international atmospheric composition community, there have been a number of recent initiatives to define priorities for future missions. These initiatives have been in the form of reports such as IGACO 2004, the United States National Research Ccouncil (NRC) Decadal Survey, community workshops, and competitive agency opportunities to begin initial studies. Proposals for future missions are recognizing the complementary nature of measurements of trace gases and aerosols from both low Earth orbit, which provides wide spatial coverage, and geostationary orbit for investigating pollution events and transport and atmospheric composition processes at different altitudes and across spatial and temporal scales. Few proposals exist for missions with significant capability before the 2015 time frame. Very high priority should be given to moving these proposals forward as quickly as possible.

3.7.4 Intensive campaign needs

Several important intensive campaigns focusing on long-range transport have been conducted in the last few years, e.g. ICARTT in 2004, INTEX[6]-A and B in 2004 and 2006, MILAGRO[7] and AMMA[8] in 2006. Several more will be conducted in 2007 and 2008 as part of the POLARCAT[9] component of the International Polar Year. Unprecedented data sets have been and will be collected during these campaigns. The primary need in this regard is similar to the primary short-term satellite need, i.e. the resources invested to allow full evaluation of the resulting data sets.

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4. EMISSION INVENTORIES AND PROJECTIONS FOR ASSESSING HEMISPHERIC OR INTERCONTINENTAL TRANSPORT

4.1 Introduction

Gridded global, regional, and national emission estimates exist for many of the pollutants that are important for assessing the hemispheric transport of air pollution (SO_2, NO_x, NMVOCs, NH_3, CH_4, organic carbon (OC), black carbon (BC), PM, and CO). Some of these are publicly available, whereas others are used by individual research groups or government agencies to study specific aspects of emissions or atmospheric processes. Most inventories are developed by combining emission factors, in units of mass of emissions per unit of activity, with activity levels or proxies thereof. The quality of emission inventories varies widely, however, and is difficult to assess objectively. For developed countries, the inventories for some pollutants from some sectors are considered of high quality, as they have been cross-checked by field studies and laboratory tests and through air quality modelling. Examples of high-quality inventories would be the SO_2 emissions from power generation in North America and Europe. For other pollutants and sectors, the quality of inventories may be considerably lower. For developing and newly industrializing countries, the quality of emission inventories is generally poor, due to a lack of actual emissions measurements and intensive ambient observations, an incompleteness of the activity data and an absence of test-based emission factors. A shorter history of inventory development in these regions also means a lack of expertise and institutions. Many such developing and newly industrializing regions fall within the Northern Hemisphere, and they provide challenges to the compilation of a complete and reliable emission inventory of all species.

Major uncertainties in emission inventories are associated with inadequate knowledge of open biomass burning (e.g. forest fires, agriculture waste burning), biofuel use (e.g. heating and cooking), artisanal industry, residential combustion of coal and agricultural production systems. These propagate into higher uncertainties in emissions for the pollutants that are mainly associated with these activities, such as CO, PM, OC, BC, CH_4 and individual NMVOCs species. Due to a lack of comprehensive activity data, there is a tendency to underestimate the emissions of these pollutants. Also, there are some source types that are relevant for the intercontinental transport of air pollution but less relevant for local air quality management, including marine emissions (though ship emissions are becoming of major importance in some European cities) and aviation emissions, natural emissions (including various methane sources, soil and lightning NO_x, volcanoes, and windblown mineral dust), agricultural emissions and biomass burning in remote areas. These source types may need greater attention from the Task Force than they have received thus far from national governments. In recent years, some new tools have become available to address the uncertainties in emission inventories, including direct (forward) and inverse modelling of air quality observations (from ground-based monitors, aircraft or satellites) and laboratory tests of combustion and similar processes. These new techniques may not only improve the accuracy of emission inventories, but they also offer the possibility of providing the basis for more rapid updates of emissions than can presently be obtained using statistics-based methods. Bringing together the scientific communities in these areas could be a valuable function of the Task Force.

This chapter first reviews the status of present-day inventories at global, regional, and national scales. Next, it identifies sources of uncertainty, methods of quantifying uncertainty, and pathways to reduce uncertainties by making use of new satellite and modelling capabilities. Third, methods to project future emissions are presented, together with examples of what has been achieved thus far. The major sources of natural emissions are introduced. Finally, the chapter discusses opportunities to harmonize the presently available inventories and makes recommendations to the Task Force on how to make progress in improving the emission inventories that are the foundation of hemispheric transport determinations.

4.2 Present-day emission inventories

4.2.1 Global inventories and databases

Currently available global emission inventories differ in the compounds included, emission sources covered, and spatial and temporal resolution. This is often related to the different purposes for which they were developed. Most inventories are based on an emission factor approach. Emission factors are identified for specific sectors, considering fuels, production or combustion technology, and the presence and effectiveness of abatement. Some inventories explicitly define control measures and technologies, while in others emission factors are derived from reported or measured emission data (implied factors). Table 4.1 summarizes relevant global emission inventories that can be used today for assessing intercontinental transport and hemispheric pollution. Some natural sources (wildfires, soil and lightning NO_x, etc.) may be included in some of these inventories, but the emphasis is usually on anthropogenic emissions.

Table 4.1 Overview of global, gridded anthropogenic emission inventories with compounds included that are relevant for studies of hemispheric transport of air pollutants

	Individual studies	Project-based calculations	Emission databases	Inventory compilations
CO	-	RETRO, QUANTIFY, POET	EDGAR, RAINS	GEIA
NH_3	Bouwman et al. (1997)	-	EDGAR(v2)	GEIA
NO_x	-	RETRO, QUANTIFY, POET,	EDGAR, RAINS	GEIA
NMVOCs (total)	-	RETRO, QUANTIFY, POET	EDGAR	GEIA
NMVOCs (speciated)	-	RETRO, QUANTIFY, POET	EDGAR(v2)	GEIA
SO_2	Stern (2005)	QUANTIFY	EDGAR, RAINS	GEIA, AEROCOM
BC	Bond et al. (2004)	QUANTIFY	EDGAR, RAINS	GEIA, AEROCOM
OC	Bond et al. (2004)	QUANTIFY	EDGAR, RAINS	AEROCOM
CH_4	-	QUANTIFY	RAINS	UNFCCC

All of the global inventories draw heavily on individual studies of a particular pollutant or source type that, in the ideal case, apply a consistent methodology across all included regions. Global emissions of a number of individual species have been studied in such special inventories, e.g., NH_3 (Bouwman et al., 1997), BC and OC (Bond et al., 2004) and SO_2 (Stern, 2005), as well as particular source types, e.g. biomass burning (van der Werf et al., 2003) and shipping (Corbett et al., 1999). However, these special inventories are prepared for different years, often make use of different activity data sources and vary in level of detail. For some pollutants, there are no global studies of this type, e.g. PM. Consequently, there are a number of projects where compilations of specific inventories, enhanced with regional work and attempts to draw on consistent activity databases, have led to the development of large-scale databases for use in global modelling studies. While the projects listed below strive to maintain internal consistency, they often produce incomparable results due to, for example, differences in principal assumptions about the evolution or importance of some parameters (driven by the objective and spatial resolution of the project), source coverage, underlying activity data, etc. The latter is of vital importance, especially for projects that strive for finer temporal and spatial resolution and therefore need to make use of local activity data that are often not compatible among regions and with the available international data sets (see also discussion in section 4.2.2).

EDGAR (Emission Database for Global Atmospheric Research) presents global emissions of air pollutants and greenhouse gases from anthropogenic and biomass burning sources distributed by country and on a 1° × 1° degree grid using a variety of proxy and actual spatial distributions (Olivier

et al., 1996). Emissions are calculated using an emission factor approach. EDGAR emissions have been used as the basis of various other inventory projects (e.g. GEIA, POET, and QUANTIFY). The latest data set is for the year 2000 (Olivier et al., 2005; van Aardenne et al., 2005).

The **RAINS** (Regional Air Pollution Information and Simulation) integrated assessment model has been developed by International Institute of Applied Systems Analysis (IIASA). Although largely used in regional studies for Europe (Amann et al., 2004; Kupiainen and Klimont, 2007) and Asia (Cofala et al., 2004; Klimont et al., 2001), recently it has been extended to estimate global emissions for several compounds for anthropogenic sources (Cofala et al., 2007). Emissions are calculated for the period 1990-2030 by country or larger region using an emission factor approach, where penetration of abatement technologies is explicitly included. Recently, emissions have been allocated to 1° × 1° grid cells based on the EDGAR methodology (Olivier et al., 2005).

To study precursors of ozone and their effects in the troposphere, the **POET** project developed an emission inventory that included anthropogenic and biomass burning emissions (Granier et al., 2005). For anthropogenic sources, emissions were estimated for 1990, 1995 and 1997 relying on EDGARv3 data, while biomass burning emissions were calculated for 2000 based on satellite fire counts data and vegetation maps (Tansey et al., 2004a; Tansey et al., 2004b).

For analysis of the tropospheric composition over the past 40 years, the **RETRO** (Reanalysis of the troposheric chemical composition) project developed an anthropogenic and biomass burning emission inventory covering the period 1960–2000 with monthly emissions of CO, NO_x, and NMVOCs on a 0.5° × 0.5° grid . Anthropogenic emissions are calculated using **TEAM** (the TNO Emission Assessment Model) (TNO, 2005), based on an emission factor approach with explicit assumptions about technologies applied.

To quantify the climate impact of global and European transport systems, the **QUANTIFY**(Quantifying the Climate Impact of Global and European Transport Systems) project currently develops global, gridded emission inventories and scenarios up to 2100 for anthropogenic emissions of greenhouse gases and air pollutants. In particular, transport (road, shipping and aviation) emissions are calculated, and non-transport emissions are taken from the EDGAR 2000 data set. The results are not yet published, but data sets may be available on request (http://www.pa.op.dlr.de/quantify/).

Although not providing the most recent emissions data, the **GEIA** (Global Emissions Inventory Activity) project has compiled global emission inventories for many of the pollutants of interest on a 1° × 1° grid, by assembling material from a variety of databases (e.g. EDGAR), studies and projects. Dentener et al. (2006) compiled global emission sets for 2000 for the **AEROCOM** (Aerosol Comparisons between Observations and Models) project to study aerosols and aerosol precursors. Data originate from various sources, e.g. anthropogenic emissions of BC and particulate organic matter (POM (Bond et al., 2004), SO_2 (Cofala et al., 2006)), international shipping and spatial (1° × 1° grid) distribution patterns from EDGAR (Olivier et al., 2005), and biomass burning from the **GFED** (Global Fire Emissions Database) (van der Werf et al., 2003).

The relative importance for global emissions of different sectors and fuel types is presented in figure 4.1. The contributions shown in this chart can be markedly different, however, for individual countries and regions. The estimates are based on EDGAR FT2000 (Bond et al., 2004; Olivier et al., 2005), EDGARv2 (Olivier et al., 1996), and Bouwman et al. (1997).

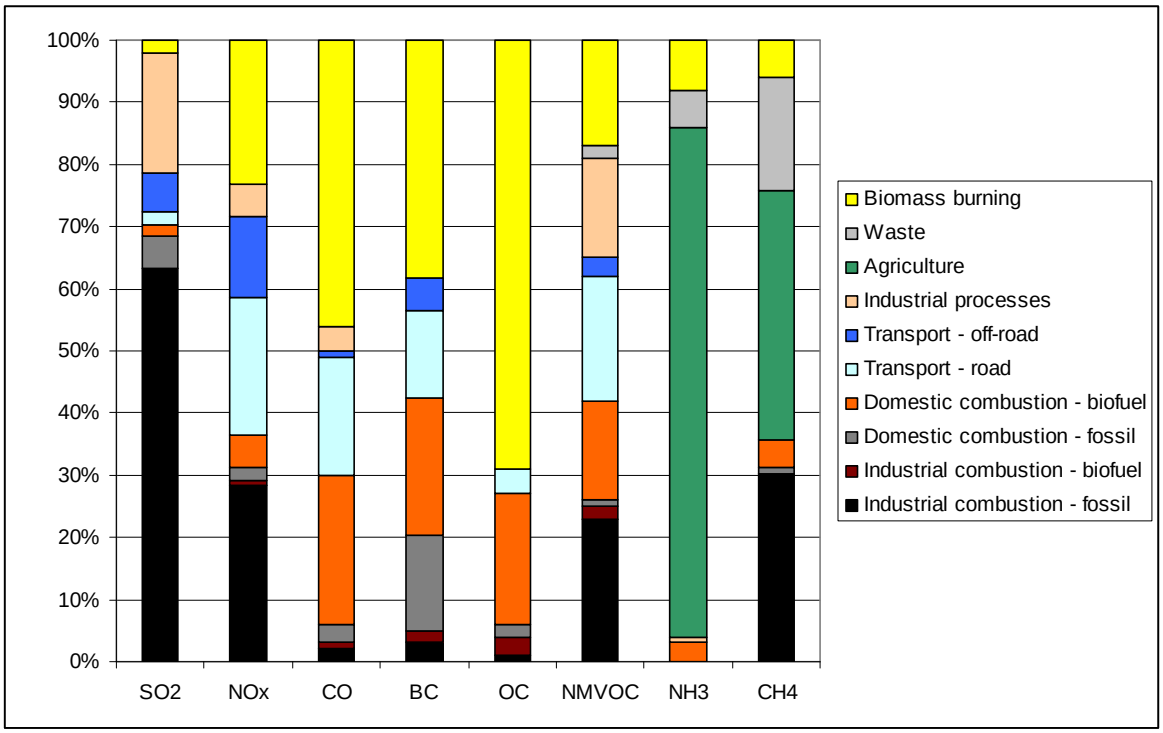

Figure 4.1 Relative importance of different sectors to global emissions (% of total emissions). For NMVOCs and CH_4, the sector "Industrial combustion – fossil" includes emissions from exploration and distribution of oil and gas.

An illustration of the geographical distribution of global emissions is shown in figure 4.2, which presents gridded emissions of CO and NO_x from anthropogenic sources (excluding aviation) and biomass burning from the EDGAR database.

In addition to the emission factor calculations, some post-processing of the resulting emission inventories is usually performed to allocate emissions to grid cells or to include more accurate data from specific point sources (e.g. stack-measured emissions data from, for example, the U.S. Environmental Protection Agency (EPA) power plant data set or the EU EPER (European Pollutant Emission Register) system in Europe). Allocation of national emissions to grid cells defined in the atmospheric models is, in addition to the allocation of large point source emissions to the location of the facility, based on auxiliary data sets such as population density maps, road network data, ship tracking data, or animal density maps. Allocation to a vertical release height is necessary for modelling, but is one of the weakest aspects of present-day inventories. In principle, the stack height of large point sources could be used as an indication of the model layer into which the emissions are released. However, due to both data limitations (no stack height information, for example, in the EPER system) and the fact that one should also take into account physical stack parameters such as heat and volume, vertical resolutions are often not provided by the emission inventory community.

The allocation of annual emissions to a temporal resolution as used by the model (monthly, weekly, or diurnal pattern) is often performed through time profiles based on, for example, monthly oil consumption data, traffic counts for those activities that follow specific patterns (e.g. fuel used for space heating or rush-hour/weekend traffic). For some sectors, defining a temporal resolution is less straightforward. For example, power plants follow a time pattern defined by demand (peak demands in warm summer months due to air conditioning needs), planned maintenance, and plant outages. These are difficult to capture on a global scale and rarely applied in the inventories.

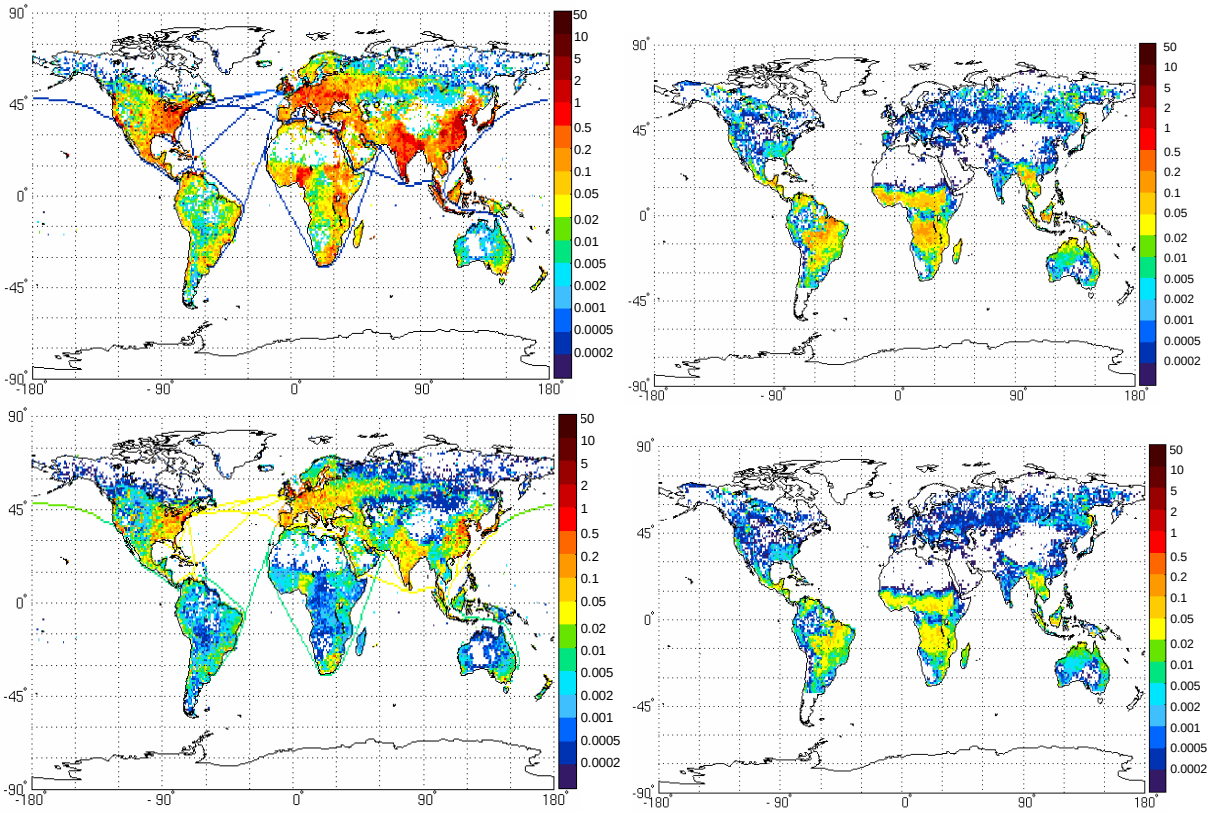

Figure 4.2 Geographical distribution of global emissions. Global emissions of CO (top panels) and NO_x (bottom panels) from anthropogenic sources (left panels) and biomass burning (right panels), gridded at 1° × 1° resolution, taken from the EDGARv32FT2000 data set (units 10^9 kg m^{-2} s^{-1}).

4.2.2 Regional and national inventories and databases

Regional and national inventories are developed as part of research projects or as official data for regulatory purposes or international reporting. Similarly to global work, special inventories play an important role in compiling regional emission databases. The inventory methods are similar to those used in the global data sets, but typically contain more information on plant measurements and individual facilities. While these assessments cannot be described here in any detail, some important ones need to be mentioned. Also, the scope of such studies has been different among the continents. While in Europe and North America more sector-specific studies have been produced (e.g. transport, power and agriculture), in Asia more pollutant-oriented work has been published (e.g. studies of SO_2 and NO_x). In Europe and the United States, emission inventory guidelines have been developed, such as the EMEP/CORINAIR Guidebook and the U.S. EPA AP-42 database, which have become part of legislation or regulations asking for annual national emission estimates for regulated pollutants.

The **EMEP** database is the main European data set (http://www.emep.int). It includes a range of pollutants (no greenhouse gases, BC or OC) for past, present and future years (up to 2020) and is distributed both by source and on a 50 km × 50 km grid. The origin of the data is official national submissions from Parties to the Convention on Long-range Transboundary Air Pollution. Where submissions are missing or incomplete, gaps are filled by EMEP (EMEP, 2006b).

The **RAINS** model has been independently developed for Europe (Amann et al., 2004; Kupiainen and Klimont, 2007) and Asia (Cofala et al., 2004; Klimont et al., 2001) and contains detailed sectoral emission assessments for countries (Europe) and for countries, states, and provinces (Asia). The RAINS model calculates emissions of SO_2, NO_x, NH_3, NMVOCs, PM, BC, OC, CO, CO_2, and CH_4 for the period 1990–2030, based on an emission factor approach, and its databases for Europe have been subject to review by national experts. The results for historical years have been

found to be in good agreement with the EMEP database and several special inventories.

The **CEPMEIP** (Coordinated European Programme on PM Emission Inventories, Projections and guidance) project (Berdowski et al., 2002) studied European PM emissions for 1995. The data and results are available from the dedicated website (http://www.air.sk/tno/cepmeip/). An updated PM inventory (by country) for Europe for the year 2000, building on the CEPMEIP project, was completed in 2006 and results are expected to be available in the near future.

EPER (http://eper.ec.europa.eu), established in 2000, includes emissions to air and water. The database covers approximately 9,200 industrial facilities in the EU15, Norway and Hungary for 2001 and approximately 12,000 facilities in the EU25 and Norway for the year 2004. EPER will be further developed to include more facilities in the future.

The United States, Canada and Mexico all prepare and maintain emission inventories. **NEI** (the United States National Emission Inventory) includes data on all criteria pollutants, important precursors and hazardous air pollutants by detailed source categories. Data are based on state, local and tribal submittals for point sources, supplemented by EPA analysis for area sources (http://www.epa.gov/ttn/chief/index.html). Updated versions of the NEI are released every three years, with 1999 and 2002 being the latest versions. A helpful summary of North American emission inventories can be found in a recent **NARSTO** (North American Research Strategy for Tropospheric Ozone) assessment (NARSTO, 2005).

Several regional emission inventory tools are now available for Asia, including the RAINS-Asia model referred to above, the NASA **TRACE-P** (Transport and Chemical Evolution over the Pacific) inventory (Streets et al., 2003), and the Japanese Regional Emission Inventory in Asia (**REAS**) (Ohara et al., 2007). REAS presents emissions for several pollutants for the period 1980-2020. For some Asian countries, there are inventories available of similar quality to those in Europe and North America—particularly for Japan, the Republic of Korea and Taiwan Province of China. Elsewhere, inventories are available but based on less stringent requirements and not fully verified (e.g. China, India, Thailand). For the rest of Asia, national inventories are weak or non-existent.

For the rest of the Northern Hemisphere, information from local inventories is seriously lacking (Central Asia, the Russian Far East, the Middle East, and relevant parts of Africa and Central and South America). Emission estimates for these regions tend to be focused on specific cities where air quality problems are known to exist. Though emissions tend to be low in many of these regions, recent energy-related activities such as oil and gas extraction are growing fast and may make them more important in the future.

Within Europe and North America, several countries have established emission inventory programmes with ongoing reporting and updating activities, primarily as a response to requirements in international and national legislation. Japan has established a similar emission inventory programme. Where such programmes have become well-established institutions, they provide regular, consistent and transparent emission inventories that are often of great value to compilers of regional and global inventories. Comparable institutions do not exist yet in most of the rest of the world.

4.3 Uncertainties and verification of present-day emission inventories

An uncertainty estimate is one of the quality indicators of an inventory and can be used to prioritize efforts to improve the inventory. Verification has been defined as *the collection of activities and procedures conducted during the planning and development, or after the completion of an inventory that can be used to establish its reliability for the intended application of the inventory* (IPCC, 2006). Verification methods include comparisons of different inventories, comparisons of results of alternative methods and comparisons with atmospheric measurements. These methods are complementary.

4.3.1 Quantification of uncertainties

Statistical approaches to estimate uncertainties in emission inventory levels and trends have been developed at the large scale by IPCC (2006) and in more specific applications (e.g. Frey and

Zheng (2002)). Two approaches are typically used, simple error propagation and Monte Carlo simulations. The main challenges in estimating inventory uncertainties are, however, uncertainty in the input data and developing methods to quantify systematic errors. For most inventory applications the random component of an uncertainty estimate will be small compared to the systematic component. IPCC (2006) lists the following sources of uncertainties to consider: lack of completeness, inventory model (estimation equation), lack of data, lack of representativeness of data, statistical random sampling error, measurement error, misreporting or misclassification, and missing data. Systematic expert judgments can be used to complement other sources of information on uncertainties. The usual metric for expressing uncertainty estimates is two standard deviations as a percentage of the mean.

4.3.2 Intersection of inventories with observations and modelling

Since 2000, there have been a number of new analytical tools applied to the elucidation of emissions emanating from sources in the Northern Hemisphere. Techniques include improved direct (forward) modelling and inverse modelling, making use of improved ground-station monitoring networks, and aircraft observations during large-scale field campaigns. Also, a new generation of satellites has provided trends based on column data that have been compared with emission trends. More often than not, the observation-based methods have suggested that emission estimates obtained from inventories, particularly for developing and newly industrializing countries, are too low. Bergamaschi et al. (2000) were the first to apply inverse modelling techniques to CO, finding that their estimate of the CO source strength in the Northern Hemisphere (~800 Tg CO/yr) was considerably larger than inventory-based estimates of 550 Tg CO/yr (IPCC, 1996) and the EDGAR value of 478 Tg CO/yr (Olivier et al., 1996). Since then there have been many more inverse modelling studies. Streets et al. (2006) reconciled inventory and inverse modelling estimates of China's CO emissions following the NASA TRACE-P mission and subsequent data evaluation studies (Heald et al., 2004; Kasibhatla et al., 2002; Palmer et al., 2003), with a resulting 36 per cent increase in China's CO emissions. In contrast, from analysis of ambient measurements and fuel-based calculations, CO emissions from on-road vehicles in the United States are thought to be overestimated by perhaps a factor of two in current inventories (Parrish, 2006), and inaccuracies in NO_x and NMVOCs inventory estimates have also been noted. Other recent studies include Muller and Stavrakou (2005) on CO and NO_x and Meirink et al. (2006) on CH_4. Similarly, there are an ever-growing number of satellite-based studies of emission trends, of which Richter et al. (2005) on China's NO_x emission trends was the ground-breaking work that showed faster growth in NO_x emissions between 1995 and 2004 than estimated in emission inventories, though reconciliation attempts are ongoing (Zhang et al., 2007). More recently, the Richter/Burrows group presented results for SO_2 changes, and although these are more uncertain, the growth of China's SO_2 in recent years was also shown to be much faster than anticipated. The use of satellite observations to inform emission inventories is discussed further in section 3.5.1. From the perspective of improving emission inventories, it is clear that there are potentially large benefits and opportunities to be gained if emission inventory compilers can work together with the atmospheric science and remote sensing communities to identify deficiencies in inventory estimates.

4.3.3 Important and uncertain sources

Uncertainties in inventories will vary by region, source, pollutant and inventory year. Uncertainty estimates for all world regions are not available. Generally it is expected that regions with the longest experience in compiling inventories and with well-developed statistical systems (e.g. Western Europe, North America and Japan) compile inventories with lower uncertainties than other regions.

The primary reasons for differences in uncertainties between sources are: (a) activity statistics are missing or weak; (b) emission factors and technologies are known better for some sources than for others; and (c) emissions depend on natural and variable factors such as temperature and precipitation. Usually, emissions related to the household sector, agriculture, and waste disposal are

more uncertain than for transportation and large stationary sources. Natural sources and semi-natural sources (e.g., forest fires) are more uncertain than anthropogenic sources.

Uncertainties for individual pollutants differ also with the level of experience of compiling an inventory, and these uncertainties typically can be reduced over time. SO_2 inventories have a long history in Europe and North America and are considered relatively reliable in those regions. For other world regions, inadequate information about the sulphur content of fuels and sulphur removal efficiencies may add to the uncertainty. NO_x inventories are generally regarded as less certain than SO_2 inventories, while NMVOCs and CO inventories carry high uncertainties. Due to the short experience in compiling PM, BC, and OC inventories and the lack of data on the distribution of technology types in key regions, these are even more uncertain. BC and OC inventories have uncertainty ranges of -25 per cent to a factor of two (higher for open burning) (Bond et al., 2004). Typical reported ranges of uncertainty estimates for Europe are: SO_2: ±5 per cent; NO_x: ±14 per cent; NMVOCs: 10–39 per cent; and CO: ±32 per cent (EMEP, 2006a). For the TRACE-P inventory, Streets et al. (2003) estimated uncertainties in Asian emissions that ranged from ±16 per cent for SO_2 and ±37 per cent for NO_x to more than a factor of four for BC and OC. Within Asia, there was wide variation among countries and regions, with emission uncertainties in Japan being similar to those in Europe, and emissions in South Asia having high uncertainty.

4.4 Projection of future emissions

The development of emission projections typically requires assumptions about economic growth, population growth and the emission characteristics of new production technologies. These are building blocks for the development of more detailed sets of assumptions and parameters, such as energy use projections, livestock developments, production of goods, changes in environmental legislation leading to different emission factors over time, etc.

To consider changes in the spatial distribution of emissions, additional factors may need to be projected, e.g. the pace of economic and demographic development in the considered regions (also at subnational level). Important shifts in spatial distribution of emissions could occur, for example, if the scenario of opening up of the Arctic region for shipping would be realized. Likewise, emissions associated with oil and gas exploration and development in Siberia and Central Asia could alter the patterns of emissions of hydrocarbons and other species.

4.4.1 Driving forces

The most important factors determining future emission levels are activity, level of technology development and penetration of abatement measures. Activity changes are strongly linked to economic growth, population growth and energy growth, but they are also dependent on the geopolitical situation, trade agreements, level of subsidies, labour costs, etc. While production technology improvements (with respect to emission levels) are also related to economic growth, a far more important factor is environmental legislation. The latter can be a key factor in determining the penetration of abatement measures and consequently the apparent emission factors. Comparison of historical per-capita NO_x emissions in the United States and Europe shows a strong relationship to per capita income, in which growth beyond about $5,000 leads to a strong increase in car ownership. Recently, several developing countries reached such income levels, and a rapid increase in traffic-related emissions and worsening of air quality, especially in megacities, has occurred. Historically, however, societal acceptance of measures to improve local air quality has also grown with increasing economic wealth. Consequently, it is unlikely that global air pollutant emissions will grow substantially in the future.

As one example, figure 4.3 shows the varying stages of automobile emission restrictions in Asian countries, compared to the stage of application in the EU (Euro 1-4). Traditionally, national legislation drove the installation of control technology, but in some regions international (regional or global) agreements have become the key drivers. Examples include the Kyoto Protocol, the protocols to the UNECE Convention on Long-range Transboundary Air Pollution, and EU Directives. At the national level, the economic projections are frequently updated, as are as some key activity factors, e.g. population and energy use. Regional or global projections of drivers are updated less frequently,

and such work is often driven by policy needs, e.g. the global SRES (Special Report on Emission Scenarios), EU energy or agricultural projections and the work of international agencies such as the International Energy Agency (IEA), the Organisation for Economic Co-operation and Development (OECD), and the Food and Agriculture Organization of the United Nations (FAO).

Figure 4.4 demonstrates the estimated impact of already committed legislation on NO_x emission estimates for the SRES scenarios compared to the original SRES scenario results. Cofala et al. (2007) assessed the technically feasible emission reduction potential within the next 20 years. Such potential varies for various pollutants and regions, but on a global scale emissions of NO_x or SO_2 could be reduced by about 50–70 per cent compared to the current levels, while those of BC could probably be reduced by no more than 30–40 per cent. The major reason for the lower reduction potential for BC is the significantly different emission source structure; over 60 per cent of emissions originate from small scale domestic combustion, a sector where few add-on measures exist and the most efficient reduction strategy is to replace stoves and boilers with new ones. It is expected, however, that many of the traditional appliances will remain in use until 2030, especially in the developing world. One additional aspect of environmental legislation is consideration of the actual level of compliance. Typically, historical inventories take into account information on compliance both in terms of the timely introduction of respective laws (emission standards) and the actual performance of the installed control equipment. For projections, it is assumed that the technical abatement measures will be installed in a timely manner to comply with the law. As far as performance of the equipment is concerned, approaches vary between studies: some assume that emission factors equal emission standards, while others make explicit assumptions about the probability of failure, e.g. the percentage of "smokers" among the vehicle population. The latter assumptions most often rely on the experience with existing equipment that might not necessarily be representative for new and future technologies. For a good understanding of the projections, it is of utmost importance to state these assumptions explicitly.

Figure 4.5 demonstrates the importance of careful consideration of potentially rapid changes of emission factors, especially in developing countries. In China, the building of new, large coal-fired power plants to replace or augment the set of older, smaller plants has dramatically altered the mix of plants, and in just 10 years has cut the average emission factor of the ensemble of plants by about 16 per cent.

Last but not least, there is strong interdependence among different air pollutant species, such that many species can be mitigated at the same time by certain kinds of environmental policies, i.e. ambitious CO_2 reduction targets will also result in significant reduction of air pollutants. The IIASA GAINS model showed such results for a number of European scenarios (Amann et al., 2007; IIASA, 2004).

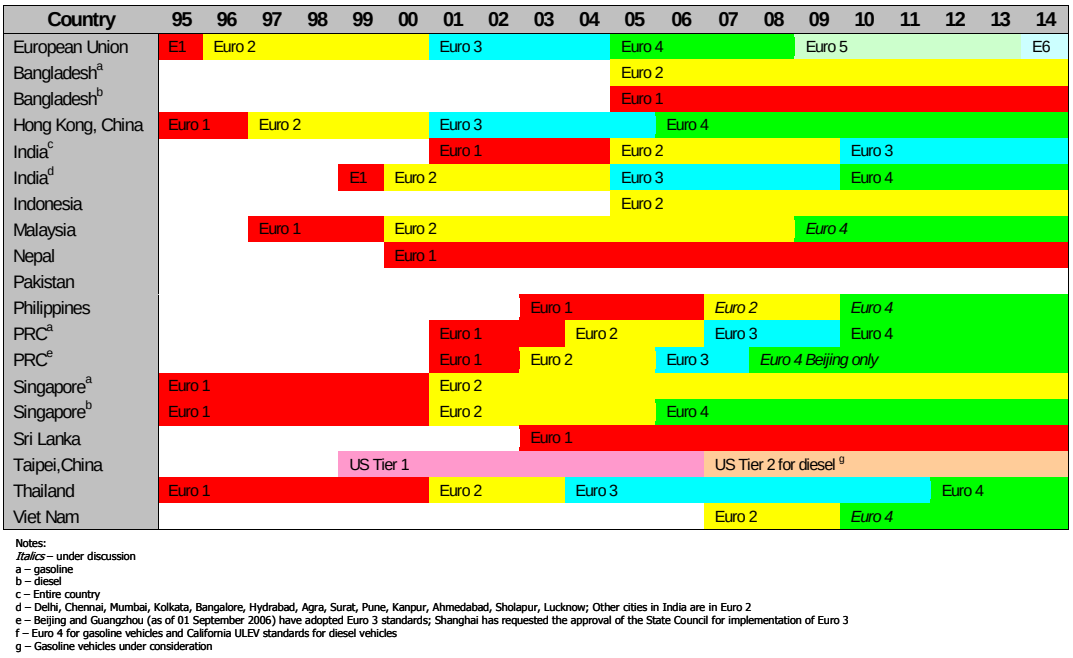

Figure 4.3 Level of automobile emission limits in Asian countries, compared with the European Union. *Source*: Clean Air Initiative for Asian cities, updated courtesy of May Ajero.

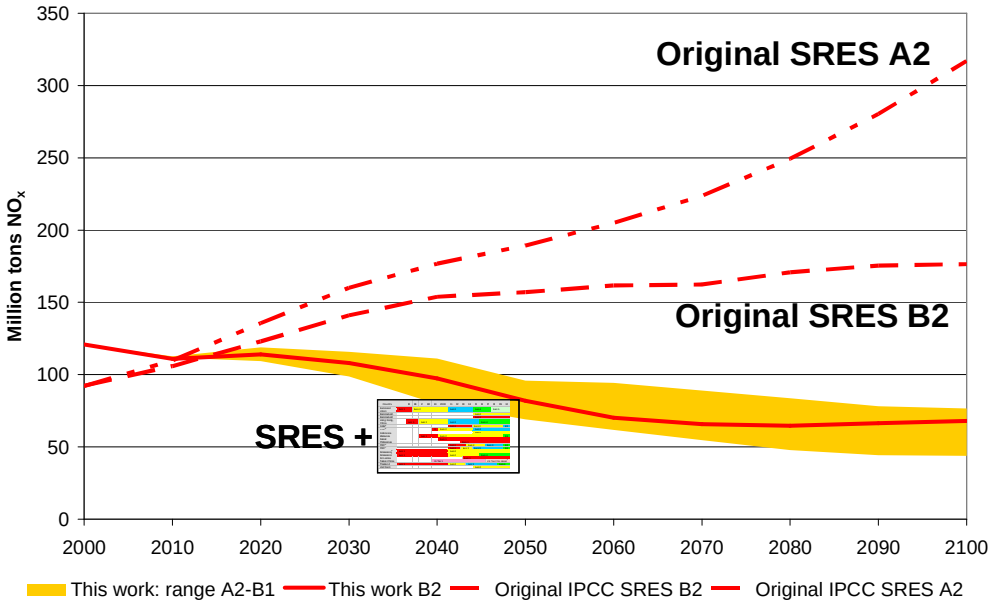

Figure 4.4 NO_x emissions in the SRES scenarios. *Source:* Cofala et al. (2006).

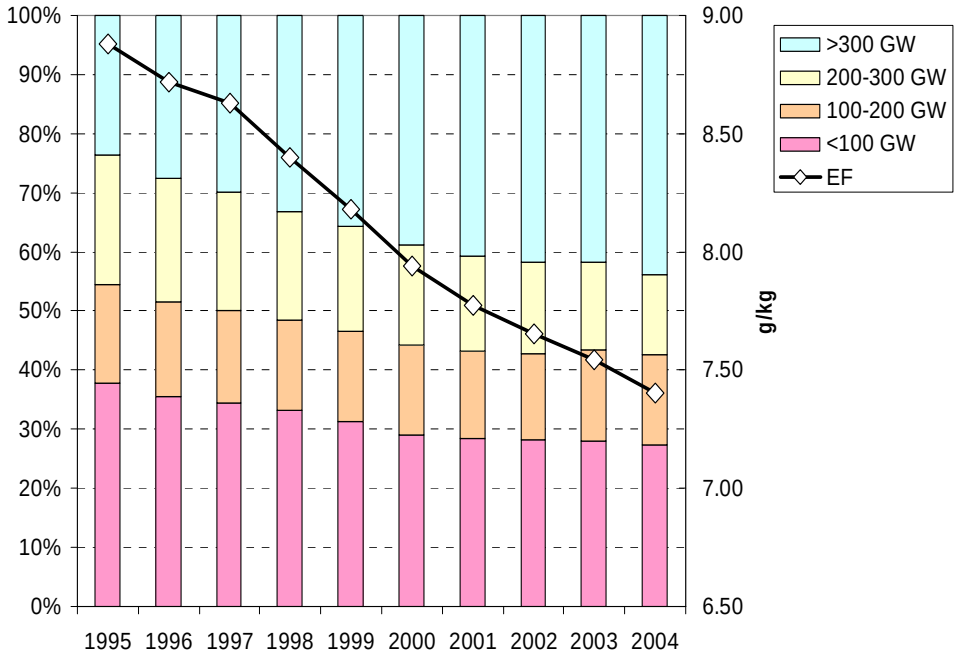

Figure 4.5 Illustration of how technology renewal in a rapidly industrializing country (China) can change the mix of plants and the net emission factor in a relatively short time.

4.4.2 Methods

There are two principal categories of approaches used to develop views of future emissions:

- Projections of activities that generate emissions (e.g. energy use, fertilizer use, livestock and production of goods) together with production technology development and penetration of abatement measures, based on existing and forthcoming legislation and autonomous improvement of technology over time, e.g. Streets et al. (2004).
- Projections of proxies, such as population or economic growth, to change emissions over time, assuming little or no change in unit emissions; an enhancement of this approach is to use elasticity against emissions to account for improvements in production technology or increased penetration of abatement measures, but this approach requires much historical data.

It is important to carefully consider consistency when compiling projections from different sets of data where the underlying methods may differ or the assumptions are not well known or documented.

4.4.3 Future emission inventories

There are a number of key studies and papers that provide important information on future emission levels, globally and in certain world regions and countries.

The **IPCC SRES** (Special Report on Emission Scenarios) scenarios (IPCC, 2000a) are well known and reflect a large, global, long-term effort, and so cannot be updated very often (the last scenarios were developed in the mid-1990s). Although the SRES scenarios assume improvements in production technology, they do not include some of the expected changes in the future penetration of abatement measures (the impacts of existing legislation); also, they do not include some of the aerosols and PM species and are available only for aggregated regions rather than countries.

There are a number of global projections that have been published in the peer-reviewed literature. For example, Streets et al. (2004) developed a forecast of future BC and OC emissions, drawing on SRES activity data and incorporating the evolution of production and control technology, specifically for non-industrial sectors. Cofala et al. (2007) developed global projections for air pollutants (excluding NMVOCs, NH_3, and PM) and methane up to 2030. The spatial resolution varies by continent. A longer-term projection (up to 2100) for BC and OC but also taking into account CO_2 abatement options and policies was prepared by Rao et al. (2005); the activity data draw on the SRES scenarios. As part of its Clean Air Interstate Rule (CAIR), the U.S. EPA has developed near-term emission forecasts of SO_2 and NO_x (http://www.epa.gov/cair/index.html).

For Europe, the RAINS model includes projections of air pollutants and greenhouse gases up to 2030, developed in consultation with national experts (Amann et al., 2006a), and the EMEP database contains official projections (up to 2010) for several European countries. The EMEP database, however, often lacks the supporting data that would allow for reconstruction and verification of emissions.

For Asia, several studies looked at particular pollutants, while the RAINS-Asia model has been used to prepare a consistent set of projections drawing on national energy data and international studies for other drivers. The work on global projections (Cofala et al., 2007) includes updates for Asia (reflecting changes in legislation) and new projections for the Russian Federation.

Figure 4.6 presents examples of SO_2 and NO_x emission projections up to 2030 for OECD countries, Asia, and the rest of the world. Also shown are trends in three of the main driving forces of emissions: population, GDP, and energy use.

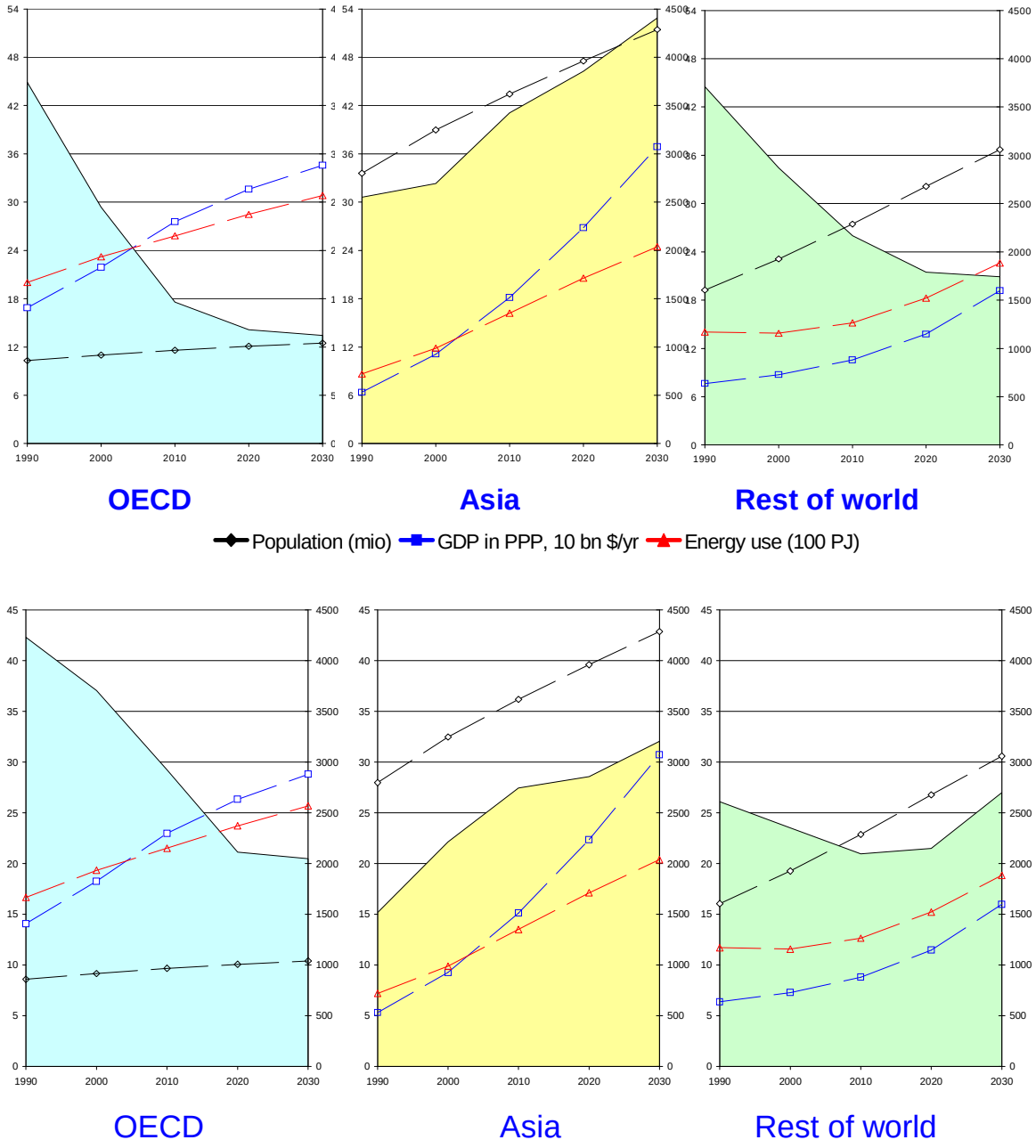

Figure 4.6 Examples of SO_2 and NO_x emission projections up to 2030 for OECD countries, Asia, and the rest of the world. Projections of emissions of SO_2 (upper frames) and NO_x (lower frames) and their major driving forces in three world regions (Amann et al., 2006b).

4.5 Natural emissions

Natural sources of atmospheric gases and particles include living and dead organisms, soil, lightning, and volcanoes. Natural emissions occur in the absence of people, but human activities can substantially alter these emissions. Methods have been developed for estimating global emissions of trace gases and particles from all major natural sources, including plant foliage VOCs (Guenther et al., 2006), mineral dust (Mahowald et al., 2006), volcanic SO_2 (Andres and Kasgnoc, 1998), lightning NO_x (Price et al., 1997), soil NO_x (Lee et al., 1997), wetlands methane (Fung et al., 1991), and wildfires (van der Werf et al., 2003). The resolutions of these models range from hourly and 1 km × 1

km for plant foliage VOCs to monthly and 1° × 1° for wetlands methane. The uncertainties associated with natural emissions are substantial and are highly dependent on the spatial and temporal scales considered. For example, the annual global isoprene emission is known to within a factor of two, but the uncertainty associated with the isoprene emission at a particular hour and location can exceed a factor of five (Guenther et al., 2006). In addition, uncertainties vary greatly for the various compounds emitted from vegetation foliage and wildfires. For example, the uncertainties associated with emissions of sesquiterpenes from foliage and NH_3 from wildfires are much higher than those associated with isoprene from foliage and CO_2 from fires.

A high resolution (10 km × 10 km) inventory of NO_x, SO_2, NH_3, PM, NMVOCs, CH_4, CO, and DMS emissions from natural sources in Europe will be completed in 2007 (**NATAIR** (Natural and biogenic emissions and Assessment of Impacts on Air Quality, http://natair.ier.uni-stuttgart.de)). Emission sources included as "natural" comprise vegetation (especially forests, forest and agricultural soils), primary biological aerosol particles, wild animals, humans, anoxic soil processes in wetlands, macro- and micro-seepages from geothermal and non-geothermal sources, wind-blown dust and Saharan dust, volcanoes and lightning. Emissions from pets, biomass burning and forest fires are also dealt with, even though they are often considered to be anthropogenic activities. The methodology developed to estimate emissions from these sources will be used to update the EMEP/CORINAIR Guidebook.

Uncertainty assessments of natural emission sources have focused on comparisons of available input databases (e.g. Guenther et al. (2006); Ito and Penner (2004); Hoelzemann et al. (2004)). The uncertainties associated with emission factors and emission algorithms are more difficult to quantify. Comparisons of different emission estimates for any of these sources tend to agree within about a factor of two on annual global scales. However, the models are generally based on at least some of the same emissions data and so are not independent estimates. Global satellite observations are beginning to provide a valuable tool for assessing emissions of, among others, foliar isoprene (Shim et al., 2005), wildfires (Pfister et al., 2005), lightning (Boersma et al., 2005; Martin et al., 2007), methane (Frankenberg et al., 2005), and dust (Mahowald et al., 2003). These observations are valuable both for providing some confidence in natural emission estimates and for indicating regions and seasons of major discrepancies.

As estimates of present-day natural emissions have improved, research efforts have focused more on how these emissions will respond to climate and landcover change. Natural emissions of mineral dust, wetland methane, foliar VOCs, and wildfires are all very sensitive to changes in landcover (e.g. vegetation type and density) and soil moisture (e.g. Mahowald et al. (2006); Guenther et al. (2006); Flannigan et al. (2005)). Foliar VOCs emissions are also sensitive to ambient temperature and solar radiation. Emissions could vary by a factor of two or more on time scales of years to decades. An improved understanding of the processes controlling these variations is required for accurate predictions of future natural emissions.

4.6 Harmonization of different inventories

There is no global or common standard for air pollutant emission inventories. Standards and principles for greenhouse gas inventories (e.g. source classification, sources to be included in national totals, definitions of national territories and pollutant definitions) have been developed by IPCC (IPCC, 1997; 2000b; 2006). The IPCC Guidelines also include ozone precursors (NO_x, NMVOCs, and CO) and SO_2. The EMEP inventory has, as far as practical, been adopting the principles of the IPCC Guidelines, but has recently extended the source classification. IPCC is referring to the EMEP/Corinair Guidebook (http://reports.eea.europa.eu/EMEPCORINAIR4/en/page002.html), used for reporting under the UNECE Convention, as a source of methodology information. The EMEP/Corinair Guidebook provides both simple and more advanced methods for most anthropogenic and natural sources of air pollutants. This Guidebook is currently undergoing a major restructuring and update and should be finalized by mid-2008. A manual for air pollutant inventories directed at developing countries to complement the EMEP/Corinair Guidebook has been developed under the Global Atmospheric Pollution Forum (The GAP (Global Atmospheric Pollution) Forum Air Pollutant Emissions Inventory Manual) (http://www.sei.se/gapforum/). The EDGAR global inventory is also

now building on the principles of IPCC, especially for the agriculture and waste sectors. The U.S. EPA AP-42 emission factor database (http://www.epa.gov/ttn/chief/ap42/index.html) has been used extensively both in North America and for application to developing countries. Although the EMEP/Corinair Guidebook and the U.S. EPA AP-42 contain similar information, there has not been any serious attempt to harmonize them.

All the developments listed in the preceding paragraph led to significant improvements in quality, consistency and transparency of global inventories. However, we must not forget about other important reasons for the observed improvements that are discussed in detail elsewhere in this chapter, e.g. more measurements, increased computing power leading to fine resolution atmospheric modelling and new satellite platforms and retrieval methods.

We suggest that further efforts for harmonization of air pollutant inventories use the 2006 IPCC Guidelines, the EMEP/Corinair Guidebook, the GAP Forum manual, and the U.S. EPA AP-42 as a starting point. Additional work is necessary to define sources specific for key air pollutants, and the extended source list developed by EMEP may need further extension to cover particular sources in developing countries. Furthermore, additional work is needed to define natural sources and to distinguish anthropogenic and natural sources. Methods to estimate emissions may need more development to fully take on board the range of activities and technologies in use in all world regions and the results of recent research. For example, BC and OC are not covered by any of the available guidance – while PM_{10} and $PM_{2.5}$ are.

4.7 Recommendations

The Task Force on Hemispheric Transport of Air Pollution should make use of the existing emission inventories of EMEP, EDGAR and other organizations and projects (national governments, GEIA, UNFCCC, etc.). Other research activities can contribute global emission data on topics of special importance to the Task Force, e.g. shipping and aviation, lightning and other natural sources. The Task Force should reach out to other organizations and research programmes (e.g. the Convention's Task Force on Emission Inventories and Projections, GAINS[1], the GAP Forum, EANET[2], and CAI-Asia[3]) to facilitate the incorporation of other emission inventories with local knowledge into global emission inventories. Such efforts are especially needed to improve the inventories in regions where emission factors and activity data are poorly known.

Modelling efforts should try to identify those emission estimates (including temporal and spatial resolution) and uncertainties that are most important for understanding intercontinental transport and hemispheric pollution. To evaluate the appropriate attributes of emission estimates, it is necessary to compare the absolute values, ratios, and trends of estimates contained in inventories to values, ratios and trends derived from both ambient observations (surface, in situ, and satellite-based) and atmospheric models, as part of an iterative process. The Task Force can be an advocate for capacity-building in these areas.

The Task Force should take into account other efforts to develop future emission projections, including efforts by national governments, the Task Force on Emission Inventories and Projections, UNFCCC (i.e. greenhouse gas emission reporting and national communications), IPCC (i.e. AR5 preparation), GAINS, OECD, QUANTIFY, and others. From these emission projections, efforts should be supported to identify the magnitudes and distributions (spatial, vertical, temporal, and chemical) of expected future emissions changes and to evaluate how these types of changes will change estimates of source-receptor relationships on intercontinental and hemispheric scales.

[1] Greenhouse gas and Air pollution Interactions and Synergies.

[2] Acid Deposition Monitoring Network in East Asia.

[3] Clean Air Initiative-Asia.

Improvement of emission inventories and development of projections is of special importance for Asia, because anthropogenic emissions are already larger than in Europe and North America today and they will continue to increase in the future. To achieve this, the Task Force should make efforts to improve collaboration with Asian organizations and programmes (national governments, EANET, CAI-Asia, etc.).

Finally, we recommend that the Task Force assists in raising awareness of transboundary and intercontinental air pollution in regions where the concept is less well known and in linking this awareness to the need for improved knowledge of contributing emissions and the importance of building high-quality national and regional emission inventories. The Task Force can assist in creating crucial links between institutions (including national focal points, regulatory bodies, and research groups) both within countries and across regional and hemispheric scales. These linkages are an important step towards meeting the need for increased capacity for developing good emission inventories throughout the Northern Hemisphere.

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5. REGIONAL, HEMISPHERIC AND GLOBAL MODELLING

Chemical transport models (CTMs) are important tools used to explore pollution transport pathways and to assess the impact of long-range transport on air pollutant concentrations in specific regions. There is growing interest in estimating the contribution of emission sources both near and far on ambient pollution levels. However, quantification requires methods for the diagnosis and analysis of intercontinental transport and dispersion, as well as procedures to track the contributions of specific source regions to pollution levels over regions of interest (i.e. the source-receptor (S/R) relationships). Such analyses are being done with regional and global scale CTMs.

In this chapter we discuss the various modelling methods and approaches being used to predict pollution levels over regional and global scales, with a focus on issues related to estimating S/R relationships. Previously published estimates of the hemispheric transport of O_3, aerosols and subsequent deposition of various aerosol components are presented, as well as preliminary results from the model intercomparison organized under the Task Force, hereinafter referred to as the HTAP intercomparison. In addition, the capabilities and limitations of current models are analysed, along with the sensitivity of S/R relationships to future changes in emissions and climate. Further activities needed to improve the modelling capabilities and the estimates of hemispheric transport of pollutants are identified.

5.1 Modelling methods for diagnosing or quantifying transport

In identifying effective measures to reduce pollution loads at a specific place, information on the breakdown of observed concentration or deposition fields into their respective contributions is useful. It seems intuitive to be able to attribute observed concentration or deposition fields to natural versus man-made sources or to local, trans-boundary or intercontinental sources. However, seldom are we confronted with the situation where the observed concentration or deposition fields at a particular location are due solely to emissions from a particular source. Generally speaking, the further we are from source regions and the longer the atmospheric lifetime of the pollutant, the more likely it is that several sources of the pollutant in question contribute to observed levels. Since observations generally cannot answer such questions on their own, models are widely used to provide the required information.

Atmospheric dispersion models have been widely used to quantitatively describe the downwind ground-level concentrations and deposition fields generated by point, line and area sources of atmospheric pollutants over the short-range distance scales (50 km or smaller). Numerous field experiments have been conducted over different environments using a wide range of different tracers to verify dispersion model performance and to study dispersion processes at these scales. Model comparison and harmonization exercises have been completed so that atmospheric dispersion models can be used with confidence in scientific, regulatory and policy applications. Tracer experiments are more difficult to perform over the long-range (100 km or greater) transport scale but there have been two notable studies. CAPTEX[1] and ETEX[2] have quantitatively examined atmospheric transport and dispersion over distance scales of ~1000 km, and have been used to assess skills of ensembles of models for dispersion forecasting (Galmarini et al., 2004). The accident at the Chernobyl nuclear plant stimulated quantitative intercomparison exercises involving a number of long-range transport and dispersion models (Klug et al., 1992).

The situation with respect to quantifying atmospheric dispersion and transport on the intercontinental scale presents a new set of challenges. First, there is no equivalent to the large body of tracer studies for model validation on the intercontinental scale. Second, intercontinental transport models are so different in formulation to simple atmospheric dispersion models that the issue of diagnosing or quantifying transport and dispersion has been given scant attention until now. Finally,

[1] Cross-Appalachian Tracer experiment.

[2] European Tracer Experiment.

there have not been pressing needs to motivate the evaluation and harmonization of the description and parameterization of the transport and dispersion processes involved in hemispheric pollution transport.

Most studies of intercontinental transport and dispersion performed up until now have employed global-scale or hemispheric-scale Eulerian grid CTMs. These models generally require large volumes of meteorological data to drive all the relevant processes, including emissions, advection, dispersion, scavenging, and chemical production and loss. Grid resolution has improved rapidly with the increasing power of computer systems, so that most CTMs achieve the resolution of a few degrees or finer, with tens of vertical levels between the Earth's surface and the tropopause. Commonly, the CTMs operate "off-line" from the driving numerical weather prediction model or "free-running" global circulation models initialized from fields of observed sea surface temperatures (SSTs); there is no coupling between the atmospheric chemistry back to the meteorology which determines the transport and dispersion of air pollution. In almost all published studies, it is tacitly assumed that as long as the meteorological databases from numerical weather analysis and prediction or global circulation models can meet the levels of spatial and temporal resolution described in the above paragraph, then they are suitable for the quantification of intercontinental transport. However, there are aspects of the representation of small- and large-scale convection, boundary layer processes, wet scavenging and chemistry that are difficult to handle in current numerical weather prediction models. These aspects may limit the capabilities of current CTMs to quantitatively represent intercontinental transport and dispersion.

An illustrative example of the use of global CTMs to investigate transport patterns of pollutants is shown in figure 5.1. In this study, artificial tracers representative for atmospheric pollutants with a lifetime of 10 days were simultaneously emitted from the world's 36 largest mega-cities and simulated for one year. Shown are annual mean tracer concentrations at the surface and column amounts above 5 km. These results illustrate the transport processes discussed in chapter 2 and the fact that these processes vary from location to location and with altitude. Several key features of the export characteristics of pollution emerge: (a) long-range near-surface pollutant export is generally strongest in the middle and high latitudes, especially for source locations in Eurasia; (b) large-scale extra-tropical cyclones and deep convection in the tropics provide efficient pollutant transport to the middle and upper troposphere (Eckhardt et al., 2004; Hess, 2005); (c) not only are there order of magnitude interregional differences, such as between low and high latitudes, but also often substantial intraregional differences; (d) efficient long-range export does not necessarily correspond with a more significant dilution of pollutants near their source, rather the amount of low-level, long-range export (e.g. below 1 km and beyond 1000 km) is well-correlated with high surface density on regional scales near the source (e.g. within ~1000 km); this implies that pollutant build-up to high densities in the surface layer of the region surrounding the source location is more strongly influenced by vertical rather than horizontal transport. Further details can be found in Lawrence et al. (2006).

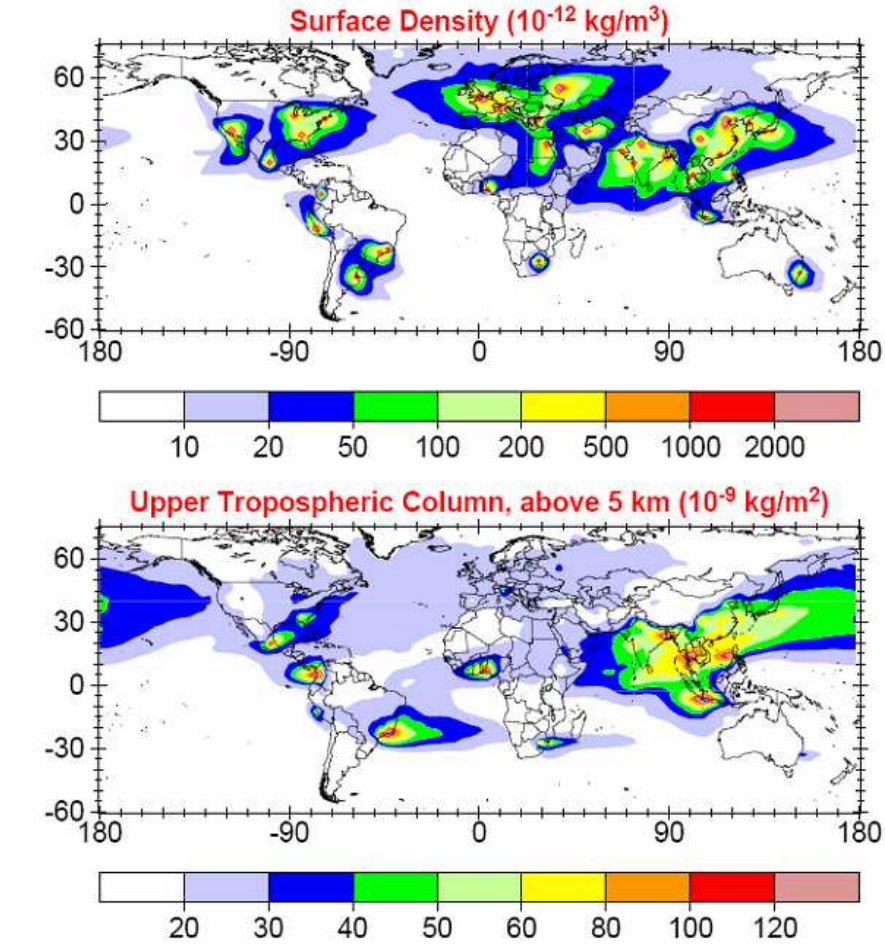

Figure 5.1 Annual mean tracer concentrations at the surface and column amounts above 5 km. Annual mean plots of the sum of all of the (10 day) major population centres' tracers for the model surface layer density (10^{-12} kg/m^3) and the column above 5km (10^{-9} kg/m^2). From Lawrence et al. (2006).

5.1.1 Methods for calculating source-receptor relationships

Source attribution or source apportionment is the process by which an observed concentration or deposition is split up into a number of components or fractions that represent a source contribution. In principle, the sum of all of the components should add up to unity or the observed concentration. A source-receptor relationship is the means by which the contribution of an emission source A to the concentration or deposition of a specific pollutant at a receptor point B can be quantified. S/R relationships are different in concept to source attributions (Venkatram and Karamchandani, 1986), and imply a response at a particular receptor location to a change in the emissions at a particular source and as such are model constructs. Source attributions are observation-based analyses where information on chemical composition is decomposed into or mapped according to source profiles. These approaches would give a similar impression of the importance of different sources for the simple case of an inert tracer or for the components of an exactly linear chemical system (Seibert and Frank, 2004). For nonlinear chemical systems, however, source attribution under one emissions scenario may be different from the source attribution after emissions have changed. The attribution of distant sources to local O_3 levels is particularly difficult, owing to strong non-linearities in the response of O_3 production to changes in NO_x emissions.

In analysis of S/R relationships, it is useful to treat pollution levels as composed of a component that responds to changes on time scales of several months, and a component that responds

to changes in emissions over longer time scales (over a number of years), such as that associated with the slower response of atmospheric methane. Unless stated otherwise, the results presented in the discussions that follow relate to the pollution component that responds on time-scales of less than one year, although some of the work in this chapter also addresses longer time scale effects.

Calculations of S/R relationships can be classified into source-oriented and receptor-oriented approaches. The source-oriented approach is the most commonly used method, where emissions from individual source regions are perturbed, and these perturbations are propagated forward throughout the modelling domain at future times. In the receptor-oriented approach the perturbation in the concentrations at the receptor location with respect to emissions are traced backwards in time. S/R relationships are model constructs, and therefore, there is no simple means of verification or validation. The accuracy and validity of a S/R relationship depends on the adequacy and completeness of the atmospheric models from which they have been derived. This topic is discussed later in section 5.7.

Most published studies describing and quantifying long-range transport S/R relationships have used a source-oriented approach. The techniques being used to characterize these relationships on an intercontinental scale are largely being adopted from regional scale analysis. For example, Arndt and Carmichael (1995) and Arndt et al. (1998) describe S/R relationships for sulphur deposition and SO_2 emissions over Asia; Gebhart et al. (2000) for visibility at Big Bend, Texas; Stohl (1996) for particulate sulphate and European SO_2 emissions; and Charro et al. (2000) for acidity in France. For more than two decades, EMEP has been generating source-receptor matrices which estimate the contribution of the emissions in any European country to the depositions or concentrations of any acidifying or photochemical pollutant in any other European country to inform the development of policies under the Convention on Long-range Transboundary Air Pollution (for example, Bartnicki (2000), EMEP (2007)). One direct measure of the relationship between a source and a receptor is simply the effect of an additional imaginary source on the concentration or deposition at the receptor. An alternative is to perturb (e.g. switch off) an emission source and estimate the impact at the receptor. Sensitivity to emissions can also be addressed using the direct decoupled method in which extra differential equations are added into the atmospheric model allowing a more elegant and mathematical analysis. A consideration in source-receptor calculations has to do with how sensitive the calculations are to the size of the perturbation. In general, CTMs represent non-linear systems and thus, in principle, the response should be dependent on the size of the perturbation. This is of practical importance when selecting the size of the change in the perturbation approach. The direct sensitivity approach assumes an infinitesimal perturbation. Comparison of results from direct sensitivity analysis and perturbation calculations provides a means to estimate the sensitivity of the S/R relationship to the emission change of interest. Other approaches use small perturbations (e.g. pulse experiments, see Wild et al. (2001); Stevenson et al. (2004)) to approximate the small (infinitesimal) perturbation. Alternatively, tagging methods where molecules from specified source regions are "tagged" and explicitly tracked in the model (using extra variables) are also used to evaluate S/R relationships (Auvray and Bey, 2005; Derwent et al., 2004; Sudo and Akimoto, 2007; Wang et al., 1998).

Receptor-oriented source-receptor calculations can be carried out using adjoint sensitivities as demonstrated in Hakami et al. (2006), or by running air-parcel models backwards in time (see Stohl et al., (2002)). The application of adjoint sensitivities are currently limited by the number of CTMs for which adjoint models are available (as this requires significant additional model development activities). However, this is expected to change in the future as more adjoint models are developed for application in chemical data assimilation (see Carmichael et al., (2007), and references therein).

5.1.2 The role of model intercomparisons

While there is an expanding literature to draw upon for analysing pollution transport pathways and S/R relationships, it is difficult to quantitatively compare the results from the published papers due to a lack of consistency in the methods and metrics being used by different researchers to describe or quantify intercontinental transport or to evaluate and characterize model performance. One method for developing comparable modelling results is to design a collaborative multimodel

study or intercomparison. A host of model intercomparisons relevant to hemispheric transport issues has been performed in the past (e.g. NASA Models and Measurements Study (Prather and Remsberg, 1993), and the World Climate Research Program (WCRP) transport model intercomparison of 1993 (Jacob et al., 1997). Recent intercomparisons include: for O_3 air pollution, ACCENT-PhotoComp (Dentener et al., 2006b) on a global scale and EuroDelta on the European scale; for aerosol, the global AeroCom, the European EuroDelta, and the Asian MICS-Asia[3] intercomparisons; for mercury on the European to hemispheric scales, the "Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury"; and for POPs on European to global scales, the ongoing intercomparison study involving both box and spatially resolved atmospheric and multi-compartment models. Further, we mention the TRANSCOM[4] phase 3 intercomparison (Baker et al., 2006; Gurney et al., 2002; Gurney et al., 2003; Gurney et al., 2004; Law et al., 2003), which focuses on large-scale transport issues in global models of the carbon cycle, and the ACCENT experiment A (Gauss et al., 2006), which focused on changes in O_3 between the pre-industrial and present day in the troposphere and lower stratosphere. There are also ongoing intercomparisons in the WCRP-SPARC (Stratospheric Processes and Their Role in Climate) community (e.g. Eyring et al., (2007)).

Past intercomparisons have differed in complexity, scope, participation, duration and analysis. The geographic scale has also differed, being based on pollutants or issues of interest; for example, O_3 and PM are mostly regional problems that have a significant hemispheric component and are sensitive to transport mechanisms on synoptic scales. Nevertheless, aspects of the models can be compared, including transport trajectories or patterns, wet deposition patterns, oxidant fields, etc.

An advantage of the multimodel approach is that the estimates from the individual models can be further analysed to provide an ensemble (mean) estimate as well as to provide estimates of the differences between the model predictions (e.g. standard deviation among the models, etc.). Such information, when combined with model evaluation using observational data, conveys valuable information with respect to model uncertainty and confidence in the estimates. To facilitate the analysis of S/R relationships for intercontinental transport a model evaluation and intercomparison activity was organized under the Task Force. Details are presented in the following section.

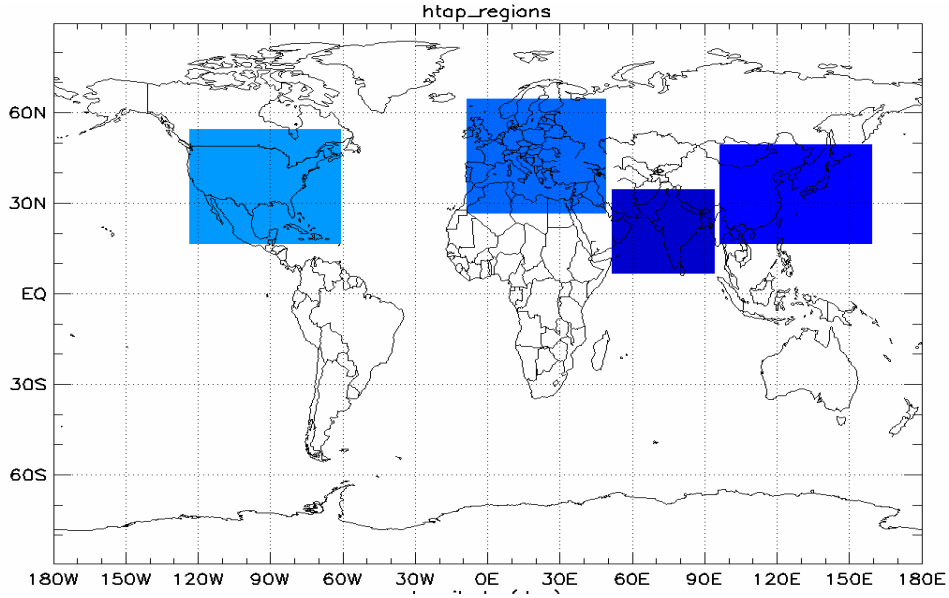

Figure 5.2 The source-receptor regions used in the HTAP intercomparison. The regions are: NA- North America; EU- Europe; EA- East Asia; and SA- South Asia.

[3] Model Inter-Comparison Study for Asia.

[4] Atmospheric Tracer Transport Model Intercomparison Project.

5.1.3 The HTAP intercomparison

The HTAP intercomparison is designed to eliminate several key uncertainties that hinder efforts to draw consistent conclusions from the existing literature on hemispheric transport of O_3 and aerosols. These uncertainties include the divergent assessment methods, spatial extent of source and receptor regions, and reported metrics resulting from the various objectives addressed by individual studies. We adopt here a coherent framework to restrict the diversity across models to that associated with the combination of model differences in transport, emissions, chemistry and other boundary conditions. The inter-model diversity gives some indication of the real uncertainty associated with model calculation of hemispheric transport. Four sets of coordinated model studies are planned in the framework of the HTAP intercomparison (more extensive information can be found at: http://aqm.jrc.it/HTAP/). Experiment Set 1 (2006–2007) S/R relationships among four world regions. Experiment Set 2 (2007) consists of artificial tracer experiments that will help to interpret the model diversity found in Experiment Set 1. Experiment Set 3 will focus on dedicated experiments for mercury, O_3, aerosol, and POPs, and will link closely to measurements. Finally, in 2008–2009 a revised set of S/R simulations will inform the Task Force's 2009 assessment report.

In Experiment Set 1, participants were requested to use three-dimensional tropospheric chemistry models at 4° x 5° horizontal resolution or finer that cover at least the Northern Hemisphere, 2001 meteorological fields, and each individual modelling group's best estimates for year 2001 emissions. Methane abundances were fixed throughout the model domain to the observed global average value of 1760 ppb. For each of the source regions depicted in figure 5.2, anthropogenic NO_x, NMVOCs and CO were decreased by 20 per cent individually. A subset of experiments focused on aerosol S/R relationships were also performed, where aerosol precursor (SO_2, NH_3, NO_x) and primary emissions (BC, particulate organic matter (POM) that consists of both primary and secondary components, and $PM_{2.5}$) were simultaneously reduced with O_3 precursors by 20 per cent. An additional sensitivity simulation in which atmospheric methane abundance was reduced by 20 per cent (to 1408 ppb) was conducted. In total, 17 sensitivity simulations were conducted. An overview of models participating in the HTAP intercomparison Experiment Set 1 is given in table 5.1.

The simulations in Experiment Set 1 were used to identify the sensitivity of tracer concentrations in the surface layer, vertical columns ("loads"), and deposition in the receptor regions (designated in figure 5.2) to the 20 per cent decreases in emissions within the four source regions. A region-wide 20 per cent emission perturbation was chosen to be within the range of anticipated regional emission changes expected over the next decade or so. Significant non-linearities exist when comparing 100 per cent (i.e. completely switching off regional anthropogenic emissions) and smaller perturbations (e.g., Doherty et al., (2005)), which may explain some discrepancies between existing S/R studies for O_3 (table 5.2). Perturbations in the range 0–20 per cent respond in a more linear fashion (Wild, 2007), also supporting the perturbation magnitude used here. Publications of the HTAP intercomparison results are planned; the results presented below should be considered as provisional.

To illustrate the variability of NO_x emissions used by the various models, we present in figure 5.3 the global and regional total (anthropogenic and natural) NO_x, VOCs, and CO emissions used in Experiment Set 1. The variation of emissions among models (~10-20 per cent of the mean) is probably a lower limit of the real uncertainty of current emission inventories. Emissions of NO_x are roughly equal in North America, East Asia, and Europe, and half of that in South Asia. The anthropogenic emissions typically represent 85–90 per cent of all emissions in Europe, East Asia, and North America, and 75 per cent in South Asia.

In the sections that follow, we discuss in detail modelling analysis of the hemispheric transport of O_3, aerosols, and deposition of various components, drawing on the published research, as well as preliminary results from the HTAP intercomparison. We use the following nomenclature to discuss the results for the emission perturbation studies with regard to S/R relationships.

1. The response is the absolute change of concentrations, columns, or deposition in receptor regions from changes in foreign and domestic source regions. In the following discussion,

domestic pertains to the impact of a region on itself, whereas foreign refers to the influence of one or more external regions.

2. The relative response due to imported pollution versus domestic emissions, or "import sensitivity", is the response to the sum of 20 per cent anthropogenic emission reductions in the foreign regions divided by the response to a 20 per cent anthropogenic emission reduction in the region itself.

Table 5.1 Overview of models currently participating in the HTAP intercomparison. Details of models are available at: http://www.mi.uni-hamburg.de/List-classification-and-detail-view-of-model-entr.567.0.html#classification.

Model/version	Institute	Simulations
GEOSChem-v07	Harvard University, Cambridge, MA, USA	Photochemistry Aerosol
MOZARTGFDL-v2	Geophysical Fluid Dynamics Laboratory, Princeton, NJ, USA	Photochemistry Aerosol
STOCHEM-v02	Hadley Centre, Met Office, United Kingdom	Photochemistry Aerosol
CAMCHEM-3311m13	National Center for Atmospheric Research, Boulder, CO, USA	Photochemistry Aerosol
INCA-vSSz	IPSL, Paris	Photochemistry
LLNL-IMPACT-T5a	Lawrence Livermore National Laboratory, Livermore, CA, USA	Photochemistry Aerosol
MSCE-HM	Meteorological Synthesizing Centre-East, Moscow.	Heavy Metals
MSCE-POP	Meteorological Synthesizing Centre-East, Moscow	POPs
EMEP-rv26	Norwegian Meteorological Institute, Oslo	Photochemistry Aerosol
OsloCTM2	Oslo University, Norway	Photochemistry Aerosol
FRSGCUCI	Centre for Atmospheric Science, University of Cambridge, United Kingdom	Photochemistry
UM-CAM-v01	National Centre for Atmospheric Science, University of Cambridge, United Kingdom	Photochemistry
TM5-JRC-cy2-ipcc-v1	European Commission, Joint Research Centre, Italy	Photochemistry Aerosol
MOZECH	Research Centre Juelich, Juelich, Germany	Photochemistry
GEOSChem-v4.5[5]	CIEMAT, Spain	Photochemistry
GOCART	NASA, USA	Aerosol
GEMAQ-v1p0	AMDAL/CRESS York University, Toronto, Canada	Photochemistry
GEMAQ-EC	Environment Canada, Canada	Photochemistry aerosol
ULAQ	University of L'Aquila, Italy	Aerosol
ECHAM5-HAMMOZ	LMCA, EPFL, Lausanne, Switzerland	Photochemistry Aerosol
SPRINTARS-v356	RIAM, Kyushu University, Japan	Aerosol
STOC-HadAM3-v01	University of Edinburgh, Edinburgh, United Kingdom	Photochemistry Aerosol
INCA-v2MS	IPSL, Paris	Aerosol
GISS-PUCCINI-modelE	NASA GISS Goddard Institute for Space Studies, New York, NY, USA	Photochemistry Aerosol
GISS-PUCCINI-modelA	NASA GISS Goddard Institute for Space Studies, New York, NY, USA	Photochemistry Aerosol
GMI-v02a	NASA GSFC (Goddard Space Flight Center), Greenbelt, MD, USA	Aerosol

[5] This is an instance of GEOSChem v07 run at 4ox5o resolution, as opposed to 2ox2.5o resolution.

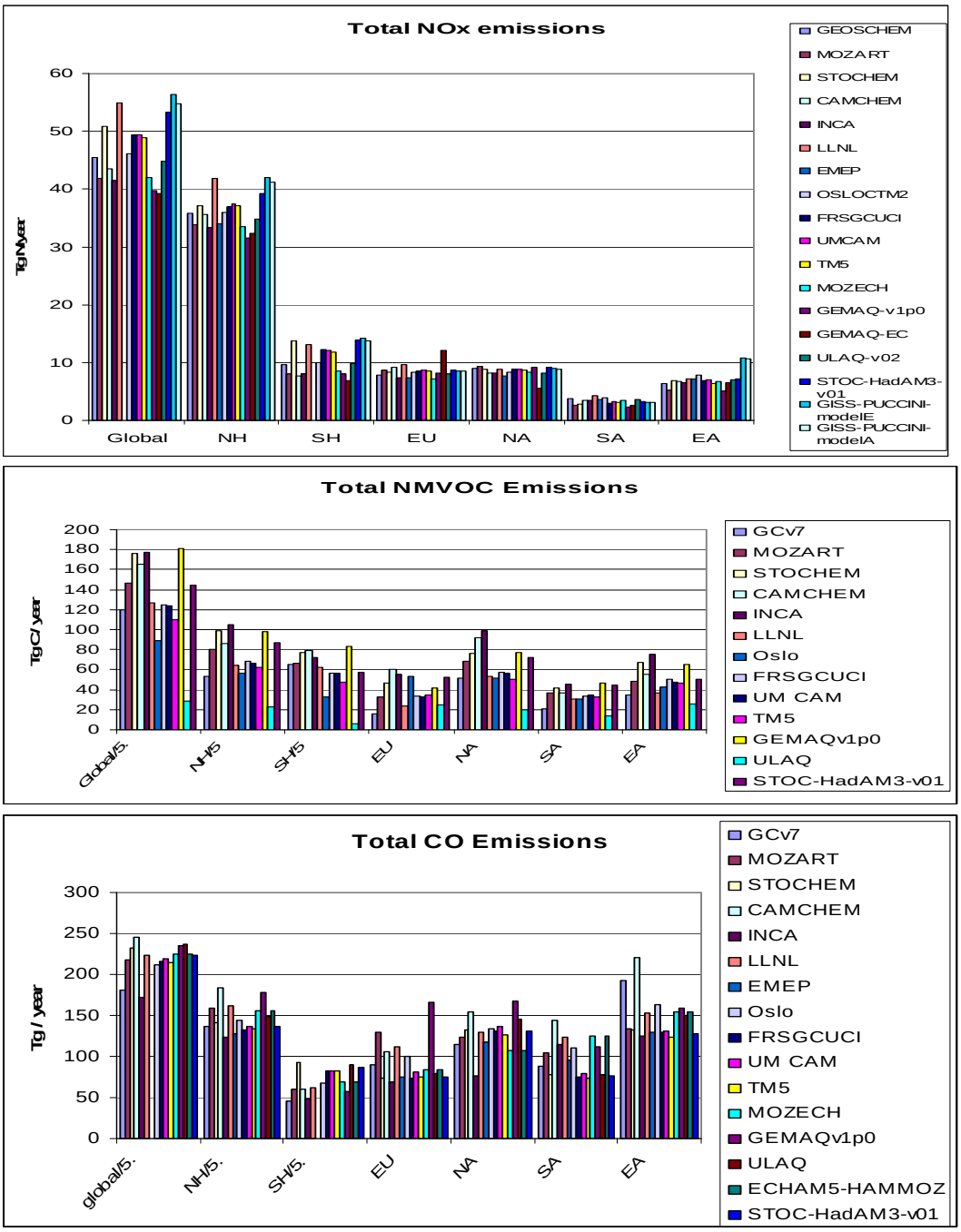

Figure 5.3 Global and regional total (anthropogenic and natural) NO_x, VOCs, and CO emissions used in Experiment Set 1. Global, hemispheric and regional NO_x (Tg N/year), NMVOCs (Tg C/year) and CO (Tg CO / year) as used by individual models in the HTAP intercomparison**.** Note that global, Northern Hemisphere (NH), and Southern Hemisphere (SH) emission totals for NMVOCs and CO have been divided by a factor of 5 for visualization purposes, and not all models reported all emissions.

5.2 Estimates of ozone transport and its precursors

5.2.1 The global tropospheric ozone budget and surface ozone

The IPCC-AR4 ACCENT/PHOTOCOMP study evaluated the computed O_3 budgets and surface O_3 concentrations of about 25 models. Many of these models also contributed to the simulations presently performed for the HTAP intercomparison. Results for the global O_3 budget suggest a tropospheric O_3 burden of 340±40 Tg, with sources governed by chemical production (5100±600 Tg/yr), an influx from the stratosphere (550±170 Tg/yr), and removal by chemical destruction (4670±730 Tg/yr) and deposition (1000±200 Tg/yr) (Stevenson et al., 2006). Model studies of pre-industrial conditions demonstrate that anthropogenic emissions have had a large impact on tropospheric O_3, contributing about 100 Tg (9 DU) to the global burden (Gauss et al., 2006; Prather et al., 2001), about 40 per cent of which is due to changes in CH_4 and the rest to changes in emissions of NO_x, CO, and NMVOCs (Shindell et al., 2005).

In the ACCENT-PhotoComp study, comparison of surface O_3 concentrations with observations showed that, despite large inter-model differences, the model ensemble mean tended to fall within 5-10 ppbv of the observed values, except for South Asia where the models exhibited a systematically high bias compared to the limited measurements available (Dentener et al., 2006(b). Figure 5.4 shows the comparison of the mean model with sets of measurements in specific regions, combined with the standard deviation of the measurements.

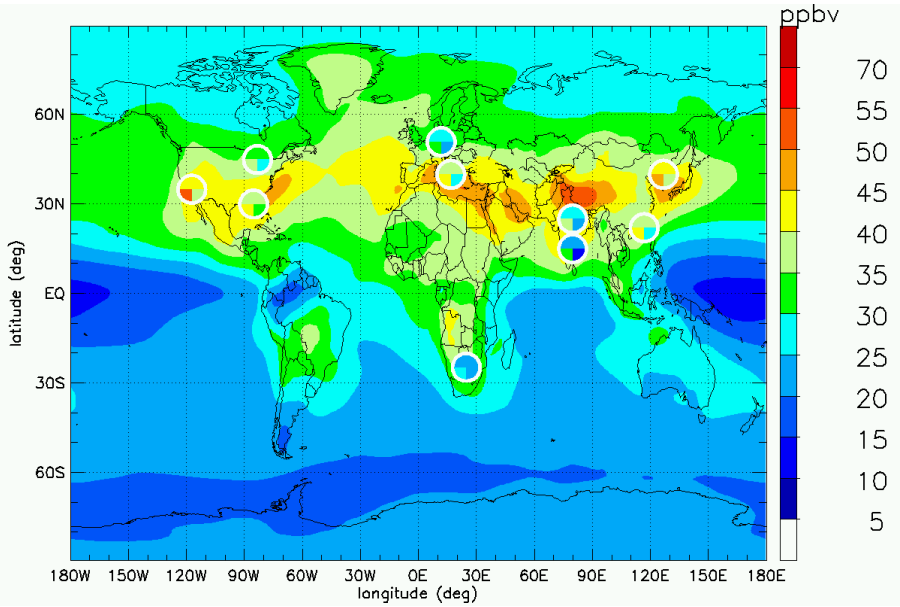

Figure 5.4 Comparison of the annual average ensemble mean ACCENT-PhotoComp model with measurements (circles). The colour of the upper part of the circle represents the average of a set of measurements; colours of the lower part show the mean plus (left) and minus (right) one standard deviation of the measurements. Global model results are difficult to compare with measurements that may display local features [Dentener, 2006 #1642].

5.2.2 Previous studies of ozone and precursor transport

Estimated surface O_3 enhancements from published studies of intercontinental transport at northern mid-latitudes are presented in table 5.2. These regional contributions have been calculated using different methods, and cover annual or seasonal mean enhancements, typically 1–5 ppbv, and individual transport episodes where enhancements are often 10 ppbv and may exceed 30 ppbv under some assumptions of future conditions. S/R relationships are strongest for North American effects on Europe due to large NO_x sources and relatively short cross-Atlantic transport. Most studies show a

distinct seasonality to transport with the greatest effects in late spring. While these studies paint a broadly consistent picture, they are not directly comparable because of the different choices of methodology, region and metric used.

As discussed in chapter 3, background O_3 levels in the lower troposphere have been rising at many locations in the Northern Hemisphere over the last several decades (Jaffe et al., 2003; Naja and Akimoto, 2004; Parrish et al., 2004a). One of the possible interpretations is that this is due in part to increasing emissions of O_3 precursors from Asia. In fact, socio-economic activities are rapidly expanding in many countries of Asia. In particular, the tropospheric NO_2 column over China may have increased about 150 per cent in the past decade as revealed by satellite sensors, likely as a consequence of increasing emissions of NO_x from anthropogenic sources (Richter et al., 2005). Such rapid increases in emissions of O_3 precursors are expected to have a large impact on air quality at the regional as well as hemispheric scale. The contribution in Japan of regionally produced O_3 from East Asian sources was estimated to be 5–15 ppbv in spring, contributing up to 10–20 per cent towards the violation of air quality standards of 60 ppbv (Tanimoto et al., 2005). In addition to anthropogenic sources, emissions of O_3 precursors from boreal forest fires can affect surface air quality in the Northern Hemisphere. Large hemispheric enhancements of CO due to boreal forest fires in 1998, 2002, and 2003 were reported by several groups (Yurganov et al., 2004; Yurganov et al., 2005). Although O_3 production off the coast of the Eurasian continent is rather weak in relatively fresh plumes (Tanimoto et al., 2000), substantial O_3 production can occur during transport towards the western United States and during descent over the Eastern Pacific (Heald et al., 2003; Hudman et al., 2004) and contribute to the violation of the United States NAAQS (National Ambient Air Quality Standards) under certain meteorological conditions (Jaffe et al., 2004).

While most of these model studies have focused on mid-latitude transport in the Northern Hemisphere, a number of recent studies have explored the transport of O_3 and its precursors in other parts of the world. Of particular interest is the transport of O_3 and its precursors from biomass burning and other sources in Africa, which are shown to contribute several ppb to surface O_3 over wide areas of the tropics and Southern Hemisphere (Aghedo et al., 2007). The easterly transport of pollutants in the upper troposphere from southern and southeastern Asia in summertime has been shown to have a significant influence on tropospheric O_3 over the Mediterranean (Lelieveld et al., 2002; Roelofs et al., 2003)

5.2.3 Present-day source-receptor relationships for ozone

In figure 5.5 we present HTAP intercomparison results for monthly mean O_3 concentrations averaged over the continental-scale source-receptor regions. The seasonal cycle in surface O_3 concentrations is similar across the 20 models contributing to the O_3 experiments, although the monthly mean values vary by as much as 10–30 ppbv in some regions and seasons. This model divergence is similar to the results from the previously discussed ACCENT-PhotoComp study. In Europe (EU), surface O_3 is highest in summer due to photochemical production; in South Asia (SA), the winter dry-season conditions favour O_3 formation, whereas during the summer monsoon season the high humidity and efficient mixing with and venting from the boundary layer reduce O_3. In East Asia (EA) and North America (NA), most models show a spring enhancement, which reflects a combination of stratospheric influx of O_3, long-range transport, and chemical production.

Table 5.2 Estimated surface ozone enhancements in receptor regions from emission in specific source regions (drawn from published studies of intercontinental transport at northern mid-latitudes). The coloured areas designate the different source-receptor areas.

Receptor Region	Source Region	O_3 enhancement (ppbv)	Method	Reference
Europe	United States	2	a)	Wild and Akimoto (2001)
Europe	North America	2-4	b)	Auvray and Bey (2005)
Europe	North America	5.8-12.2	c)	Derwent et al. (2004)
Europe	North America	3-5 (summertime avg); 10-12 (events)	b)	Guerova et al. (2006)
Europe (summer)	North America	2-4 (daytime mean); 5-10 (events)	b)	Liu et al. (2002)
Europe (yearly mean)	North America	18 (Atlantic fringes); 10-15 (Central Europe)	c)	Derwent et al. (2002)
Europe (high altitudes)	North America	10-12 (events)	d)	Huntrieser et al. (2005)
Mace Head, Ireland	North America	0.4 (winter); 0.2 (spring); -0.3 (summer); -0.9 (fall)	e)	Derwent et al. (1998)
Europe	Asia	0.8	a)	Wild and Akimoto (2001)
Europe	Asia	9 (Atlantic fringes); 5-7 (Central Europe)	c)	Derwent et al. (2002)
Europe	Asia	3.5-6.6	c)	Derwent et al. (2004)
Europe	Asia	1.5-4	b)	Auvray and Bey (2005)
United States	Asia	1	a)	Wild and Akimoto (2001)
Northwestern United States. (spring)	Asia	4 (mean); 7.5 (max)	b)	Berntsen et al. (1999)
Northwestern United States (spring)	Asia	4	f)	Jaeglé et al. (2003)
Western United States (spring)	Asia	3-10 (range during Asian pollution events)	g)	Yienger et al. (2000)
United States	Asia (future)	2-6 (Western U.S.); 1-3 (Eastern U.S.); highest April-June	h)	Jacob et al. (1999)
Western United States (spring)	Asia (future)	30-40 (max during Asian pollution events)	i)	Yienger et al. (2000)
United States	Europe	0.9	a)	Wild and Akimoto (2001)
United States (summer)	Asia + Europe	4-7 (typical afternoon range); 14 (max)	b)	Fiore et al. (2002a; 2002b)
United States	Background (1980-1998)	3-5 (spring, fall)	j)	Lin et al. (2000)
United States (Mar-Oct 2001)	Background (hemispheric pollution)	4-12 (typical range in afternoon mean)	k)	Fiore et al. (2003)
United States (summer)	Background (anthropogenic methane)	6 (afternoon mean)	l)	Fiore et al. (2002a; 2002b)
West Coast United States	Background (1984-2002)	10	m)	Jaffe et al. (2003)
Asia	Europe	1.1	a)	Wild and Akimoto (2001)
East Asia (spring)	Europe	3 (daytime mean)	b)	Liu et al. (2002)
Japan	Europe	0.2-2.5	b)	Wild et al. (2004a)
East Siberia (1997-1999)	Europe	2 (annual); 3 (spring-summer)	n)	Pochanart et al. (2003)
Asia	United States	0.8	a)	Wild and Akimoto (2001)

a) Annual mean enhancements from sensitivity simulations with 10 per cent increases in emissions from source region; results are multiplied by 10 to estimate total effect of anthropogenic emissions from the source continent. b) Sensitivity simulation with anthropogenic emissions turned off (typically not including CH_4 emissions). c) O_3 produced in tropospheric column over source region. d) Observed difference in plume of North American origin vs. monthly mean. e) Mean observed difference in O_3 concentrations 1990–1994 for air masses originating from United States and Canada vs. from Iceland and Greenland. f) Tagged O_x tracer simulations sampled at Cheeka Peek Observatory. g) Sensitivity simulation with all source emissions turned off. h) Sensitivity simulation with tripled Asian NO_x and NMVOCs emissions. i) tagged CO tracer and parameterized O_3 chemistry; scenario with quadrupled Asian emissions. j) Observed trend in the lower quartiles of the O_3 frequency distribution at rural sites. k) Sensitivity simulation with quadrupled Asian emissions. l) Sensitivity simulation with anthropogenic CH_4 emissions reduced globally by 50 per cent; results are multiplied by 2 to estimate total enhancement from anthropogenic CH_4. m) Observed trend at surface sites and from aircraft missions (1984–2002). n) Difference between median observed O_3 concentrations for air masses originating from Europe vs. Siberia and high latitudes.

The annual mean S/R relationships among the four regions for the surface O_3 response to 20 per cent reductions in anthropogenic NO_x emissions are given in table 5.3a. In general, the standard deviations of the model responses indicate that the results are statistically significant.

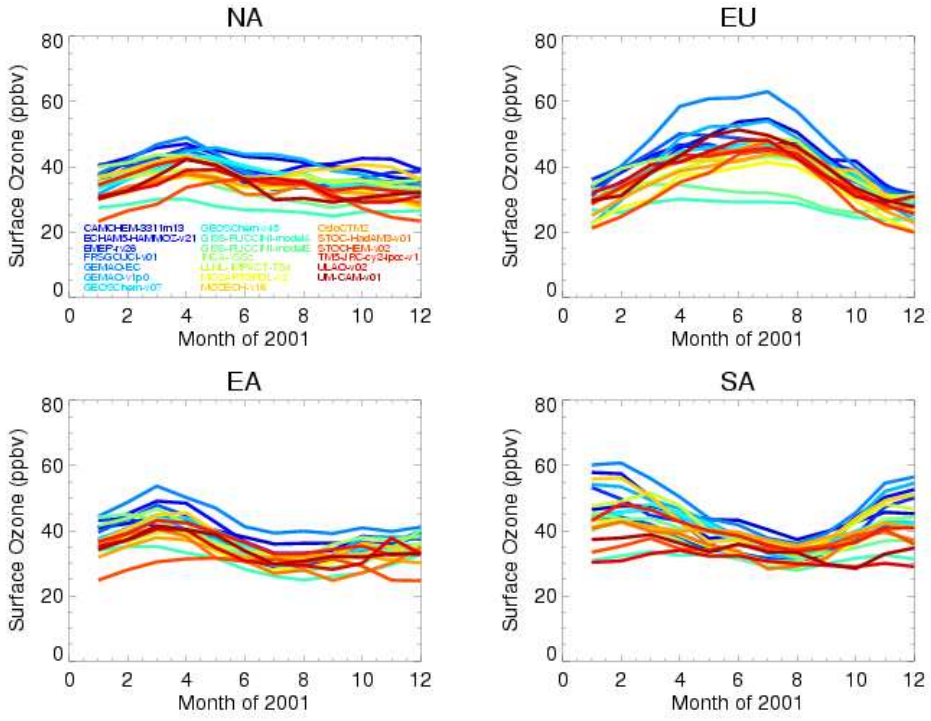

Figure 5.5 HTAP intercomparison results for monthly mean surface ozone averaged over the continental-scale source-receptor regions: Monthly mean surface O_3 volume mixing ratios (ppbv) averaged over the HTAP intercomparison source-receptor regions for the unperturbed base case model simulations (coloured lines; 20 models)

Table 5.3 Surface ozone responses to NOx perturbations in the hTAP intercomparison: Multi-model mean (20 models) response of (a) annual mean and (b) seasonal mean surface O_3 (ppbv) in receptor regions (columns) to 20 per cent reductions of anthropogenic NO_x emissions in the source regions (rows). Contributions of a region on itself (termed "domestic") are shown in bold. Inter-model standard deviations are shown in parentheses. Seasonal means are reported for the season of highest sensitivity to domestic NO_x emission perturbations.

(a)

Annual mean decrease of surface O_3 (ppbv) due to 20 per cent emission reduction of anthropogenic NO_x

	Receptor Region	EU	NA	EA	SA
Source Region	EU	**0.46 (.21)**	0.08 (.03)	0.12 (.04)	0.15 (.04)
	NA	0.22 (.06)	**0.75 (.21)**	0.12 (.03)	0.10 (.03)
	EA	0.08 (.02)	0.12 (.04)	**0.61 (.18)**	0.09 (.03)
	SA	0.04 (.02)	0.04 (.02)	0.09 (.03)	**1.08 (.22)**

(b)

Mean decrease of O_3 (ppbv) during the season of maximum sensitivity to domestic NO_x emission perturbations

	Receptor Region/Season	EU/JJA	NA/JJA	EA/JJA	SA/SON
Source Region	EU	**1.26 (.30)**	0.05 (.02)	0.07 (.03)	0.15 (.04)
	NA	0.21 (.08)	**1.35 (.35)**	0.05 (.02)	0.10 (.03)
	EA	0.04 (.02)	0.07 (.05)	**1.01 (.23)**	0.14 (.05)
	SA	0.02 (.02)	0.02 (.01)	0.08 (.03)	**1.28 (.27)**

In all regions, the annual average response to individual foreign source regions is at least 5 per cent of the response to similar domestic emission perturbations. The maximum influence found is that of NA on EU, with an annual mean response of nearly 50 per cent of that due to the effect of EU NO_x reductions. It is important to note that the responses shown in table 5.3 can be also thought of in terms of sensitivities. The EU, NA, and EA regions emit similar quantities of NO_x, with SA contributing about half as much (figure 5.3). Thus the domestic sensitivities (response in the region per unit emitted in the region) are of similar magnitude for the EU, NA and EA regions, while the sensitivities for SA are ~60 per cent higher.

The import sensitivity, or response due to the combined influence of the three foreign source regions on a receptor region compared to the domestic response, is 30-70 per cent, depending on the receptor region. These annual mean numbers, however, mask large seasonal changes in the contribution of hemispheric transport of surface O_3 to the receptor regions, including compensating effects of O_3 response in winter (titration effects) and summer in the EU source region. For example, table 5.3b also shows the O_3 response to NO_x emissions perturbations during the season of highest local O_3 production, when the multimodel seasonal mean surface O_3 concentrations respond most strongly to the 20 per cent decrease in domestic NO_x emissions (summer for EU, NA, EA and autumn for SA). In table 5.3b, the import sensitivity is much smaller than in the annual mean, ranging from 10-30 per cent (minimum for NA; maximum for SA). NA NO_x alone still exerts the largest influence of a single source region on EU, with a response of roughly 20 per cent of that from EU NO_x emissions on surface O_3 in summer.

Previous studies have indicated that enhanced ventilation of the Asian continent and subsequent long-range transport contributes to a springtime maximum in the Asian contribution to air pollution over North America (Bey et al., 2001; Jaffe et al., 1999; Weiss-Penzias et al., 2004; Yienger et al., 2000), as well as over each of the major mid-latitude source regions (Wild and Akimoto, 2001). The enhanced contribution of intercontinental transport to surface O_3 concentrations during spring is due to the relatively long lifetime of O_3 during this season (Wang et al., 1998) and to efficient transoceanic transport associated with the strengthening storm tracks and strong westerly flow at mid-latitudes in the free troposphere (Stohl et al., 2002). In the following analysis, we discuss HTAP intercomparison results for spring (March-May). Figure 5.6 shows the multimodel mean impact of 20 per cent reductions of the O_3 precursor emissions (NO_x, NMVOCs, and CO), applied individually and combined, on mean surface O_3 over the receptor regions. Each block of bars thus represents four perturbation experiments, plus the sum of the perturbation responses for the three foreign source regions. This fifth bar helps to illustrate the import sensitivity, enabling comparison of the total response to foreign influence (as represented by the source regions considered here) relative to the domestic (i.e. source region on itself) response. The whiskers depict the full range of results across individual models. Given the consistent definition of source-receptor regions and emission perturbations used in the HTAP intercomparison, the range of estimates of S-R relationships narrows substantially from that in the literature (table 5.2). However, the spread among individual models often encompasses a factor of two and is particularly large for anthropogenic NMVOCs emission perturbations (figure 5.6). The latter result is probably due to the much larger spread in anthropogenic NMVOCs emissions compared to NO_x emissions used in the models (figure 5.3b). In spring, the model ensemble mean O_3 response to 20 per cent decreases in O_3 precursors in the foreign source regions (fifth bar in ALL categories) is more than 50 per cent of the response from the domestic O_3 precursors perturbations in all regions.

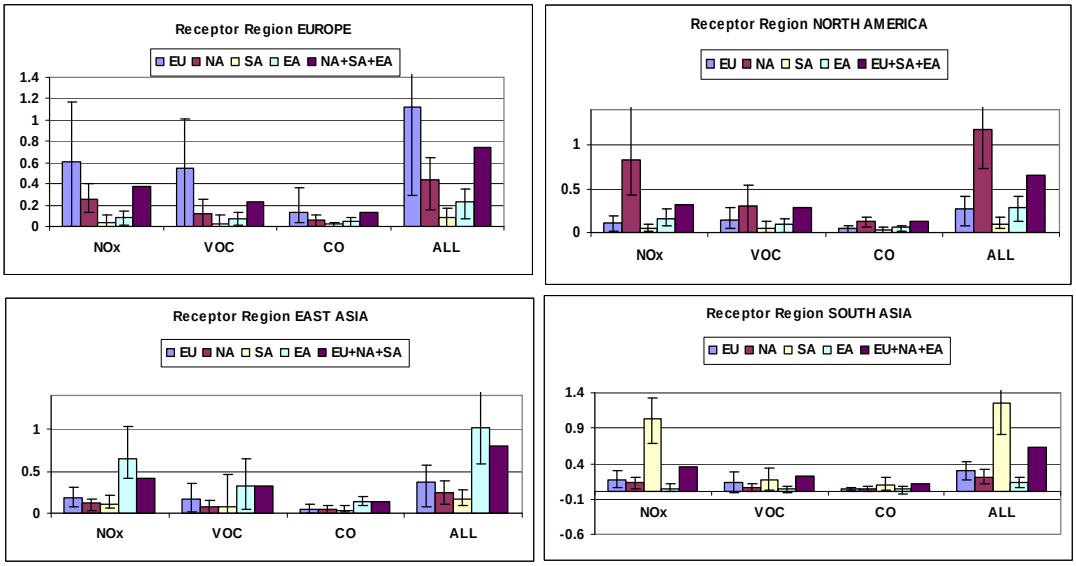

Figure 5.6 Multi-model mean impact of 20 per cent reductions of the ozone precursor emissions (NO_x, NMVOCs, and CO) on mean surface ozone over the receptor regions. Decrease in springtime (March-April-May) mean surface O_3 (ppbv) in the HTAP receptor regions resulting from 20 per cent emission reductions of O_3 precursors NO_x, NMVOCs, and CO individually within the source regions, and applying all perturbations together (ALL category). Each group of five bars includes the four perturbation experiments; the fifth bar is the sum of the three "foreign" impacts. The bars denote the multimodel mean response (coloured by source region) and the whiskers span the full range of the individual model responses. Results were submitted from 9–14 models for the sensitivity simulations included here.

While the seasonal maximum in intercontinental foreign influence (i.e. the contribution from 20 per cent reductions in all O_3 precursors (NO_x+CO+NMVOCs) in all three foreign source regions) generally occurs in spring over all the HTAP receptor regions, the model ensemble seasonal means show that some individual source regions exert maximum influence in winter (EA on SA and NA on EA). The multimodel mean results indicate that anthropogenic emissions from EU and NA often contribute more to EA and SA surface O_3 than EA and SA contribute to each other.

The multimodel seasonal mean surface O_3 response over the receptor regions is generally somewhat less-than-linear (typically within 15 per cent, with a larger deviation of 30 per cent for SA on EA) when the precursors are reduced individually vs. simultaneously. Caution is needed, however, to draw definitive conclusions as not all models have conducted all the simulations, and all available model data are included in figure 5.6, rather than excluding models that did not conduct all 16 sensitivity simulations. Future work will assess linearity across species by analysing only those models that have conducted all simulations, as well as conducting additional simulations to examine how the 20 per cent reductions analysed here are expected to scale to perturbations of other magnitudes.

Figure 5.6 indicates that the hemispheric response to NO_x emission perturbations generally outweighs that from CO or NMVOCs. The model results, however, neglect a long-term feedback of these emission perturbations on O_3 that occurs through the hydroxyl radical (OH) and CH_4 (the major sink for CH_4 is reaction with the hydroxyl radical in the lower troposphere). Specifically, NO_x emission reductions will decrease OH, causing CH_4 oxidation rates to decline and the CH_4 abundance to rise (Fuglestvedt et al., 1999). Because CH_4 is also an O_3 precursor, there will be a small increase in O_3 that occurs on the decadal time scale of the CH_4 lifetime; this O_3 increase will follow the spatial pattern of O_3 produced via CH_4 oxidation rather than that of NO_x. This long-term met CH_4-driven O_3 increase could potentially offset the NO_X emission-reduction driven O_3 decrease; this has been demonstrated specifically for aircraft NO_x emissions (Wild et al., 2001). A recent study by West et al. (2007) showed that 20 per cent decreases in global anthropogenic NO_x emissions yielded long-term

global increases in population-weighted surface O_3 that counteracted the short-term decrease by 6–14 per cent. In contrast, 20 per cent reductions in global anthropogenic CO emissions cause OH to increase, thereby further decreasing O_3 produced from CH_4, leading to a global 16–21 per cent enhancement to the short-term population-weighted surface O_3 decreases. Reduction of anthropogenic NMVOCs emissions had little impact on O_3 generated by CH_4 oxidation (West et al., 2007). On the basis of the West et al. (2007) results, we estimate that the results presented here, which neglect the long-term effects of CH_4, should be decreased by about 5–15 per cent for NO_x, and increased by 15-20 per cent for CO. Given the opposing effects that CO and NO_x have on the long-term O_3 response, the results from the simulations with the combined NO_x, CO, and NMVOCs reductions ("ALL" in figure 5.6) are expected to be closer to representing the net effect on surface O_3 if the long-term feedback through CH_4 were included.

The results in figure 5.6 provide insights into differences in regional chemistry. Ozone in EU is more sensitive to domestic anthropogenic NMVOCs perturbations than in the other regions. While the EU, NA and EA regions all emit similar quantities of NO_x (figure 5.3), biogenic NMVOCs emissions in EU are less than half of those in the other two regions, and may contribute to a more NO_x-saturated (VOCs-limited) O_3 production regime. Consequently NMVOCs reductions in EU yield larger changes over the receptor regions, often of similar magnitude to those achieved with decreases in EU NO_x. In contrast to the NO_x emissions reductions, the contribution from anthropogenic NMVOCs emissions on domestic surface O_3 in all regions peaks in winter and early spring. When NO_x, CO, and NMVOCs are reduced together, however, the seasonality of the O_3 reductions most closely follows those of NO_x alone. Overall, the results in figure 5.6 show that the EU region is much more sensitive to domestic NMVOCs emissions (NO_x-saturated) than the other three regions, where the O_3 sensitivity to domestic NO_x reductions is much stronger (NO_x-limited).

Methane acts mainly on the large-scale background O_3 distribution. Since the global CH_4 abundance was decreased uniformly by 20 per cent, the results from this simulation are not directly comparable with those from the 20 per cent regional reductions of the other O_3 precursors. The uniform 20 per cent reduction in global CH_4 abundances yields a ~1 ppbv decrease (0.7–1.8 ppbv range from individual models) in annual mean surface O_3 over the receptor regions (figure 5.7). When examined in the context of previous work, the magnitude of the surface O_3 response to CH_4 indicates a fairly linear relationship between the surface O_3 response and changes in CH_4 (Dentener et al., 2005; Fiore et al., 2002b; Fiore et al., 2006; West et al., 2006). The multimodel mean results indicate that surface O_3 responds more strongly to CH_4 in summertime over EU and late spring over SA (0.7 and 0.4 ppbv seasonal amplitude, respectively), with little seasonality over NA and EA (0.2 and 0.1 ppbv seasonal amplitude, respectively).

In order to compare these results with those in table 5.3 and figure 5.6, we use the O_3 response from the global CH_4 perturbation simulation along with estimates of anthropogenic CH_4 emissions to approximate the surface O_3 response that would result from individual 20 per cent reductions of CH_4 anthropogenic emissions in the source regions. We first account for the feedback of CH_4 on its own lifetime (through OH) using the "feedback factor" recommended by IPCC of 1.4 (Prather et al., 2001) to translate the 20 per cent decrease in CH_4 abundances to a 14.3 per cent decrease in total global CH_4 emissions. If we assume, as recommended by IPCC, that anthropogenic CH_4 emissions are 60 per cent of the total CH_4 emissions, then the 14.3 per cent decrease in total global CH_4 emissions is equivalent to a 23.8 per cent decrease in global anthropogenic CH_4 emissions.

We then use the EDGAR 2.3 FT2000 emission inventory for anthropogenic CH_4 emissions (Olivier et al., 2005) to determine the regional distribution of anthropogenic CH_4 emissions, and find that NA, EU, SA, and EA each contribute 16.6 per cent, 16.0 per cent, 17.3 per cent, and 19.0 per cent, respectively, to total global anthropogenic emissions (298 Tg CH_4 yr^{-1} in 2000). The percentage contribution of individual 20 per cent decreases in anthropogenic CH_4 emissions within these regions to the total response in the global CH_4 perturbation simulation (where abundances are reduced globally by 20 per cent) is thus: 14.0 per cent, 13.5 per cent, 14.5 per cent, and 16.0 per cent for NA, EU, SA, and EA, respectively.

The multimodel mean decrease in surface O_3 resulting from the 20 per cent decrease in global CH_4 abundances is fairly uniform at ~1.2 ppbv over all of the receptor regions (figure 5.7). Based on the analysis above, we estimate that the corresponding surface O_3 response is 0.17, 0.16, 0.17 and 0.19 ppbv for a 20 per cent decrease in anthropogenic CH_4 emissions within NA, EU, SA, and EA, respectively. Comparing these values with table 5.3a and figure 5.6 indicates that 20 per cent reductions in domestic NO_x emissions are substantially more effective than domestic CH_4 emissions at decreasing domestic surface O_3, but for most regions, the CH_4 emission reductions are likely more effective than NO_x at decreasing surface O_3 over the foreign receptor regions. As discussed above, if we were to account for the feedback of changes in NO_x emissions on O_3 through CH_4, then the relative impact of CH_4 would be even larger.

Future work should consider the role of CH_4 in a more coherent framework, considering also the interaction of the timescale of emission reductions and CH_4 equilibrium lifetimes.

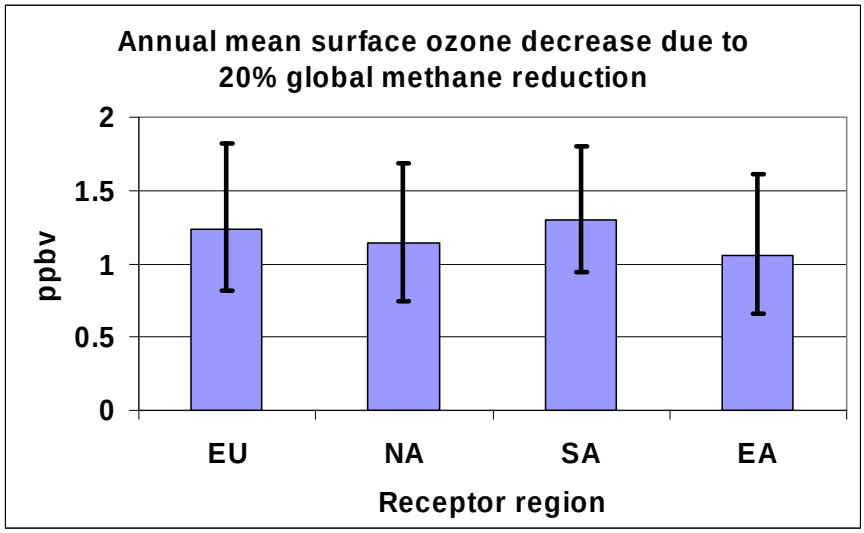

Figure 5.7 Reduction in mean surface O_3 resulting from CH_4 reduction. Decrease in annual mean surface O_3 over the receptor regions resulting from a 20 per cent decrease in global CH_4 concentrations. The bar depicts the 14-model mean, and the whiskers span the full range of individual models. Note that these results cannot be directly compared to the NO_x, NMVOCs, and CO experiments, since a decrease of 20 per cent CH_4 concentrations implies a 20-30 per cent reduction of global anthropogenic CH_4 emissions.

5.2.4 Ozone exceedance

It has been suggested that hemispheric emission changes may influence the frequency of pollution events (Fiore et al., 2002a; Fiore et al., 2002b; Li et al., 2002). As a first analysis of this issue, we used a threshold-based statistic: the number of days with 8 hours running average O_3 concentrations in excess of 60 ppbv (DAYS60). Figure 5.8 illustrates the large spread among models in estimating DAYS60. This threshold statistic is inherently sensitive to differences in model grid resolution and will accentuate systematic model biases. Nevertheless, as examined here for the CH_4 perturbation simulation, the sign and magnitude of the change in this threshold statistic is fairly robust among the models, suggesting that the models can provide useful information in estimating the relative improvement in air quality as represented by such a threshold metric.

Based on analysis of the NO_x and CH_4 perturbation simulations, the response of monthly mean daily maximum 8-hour O_3 concentrations exhibits identical seasonality, and a similar magnitude reduction over the receptor region, as the monthly and yearly averaged 24-hour mean results discussed above (to within a few tenths of a ppbv). This result suggests that the response of the seasonal means averaged over the continental regions reported here are relatively insensitive to the choice of averaging statistic.

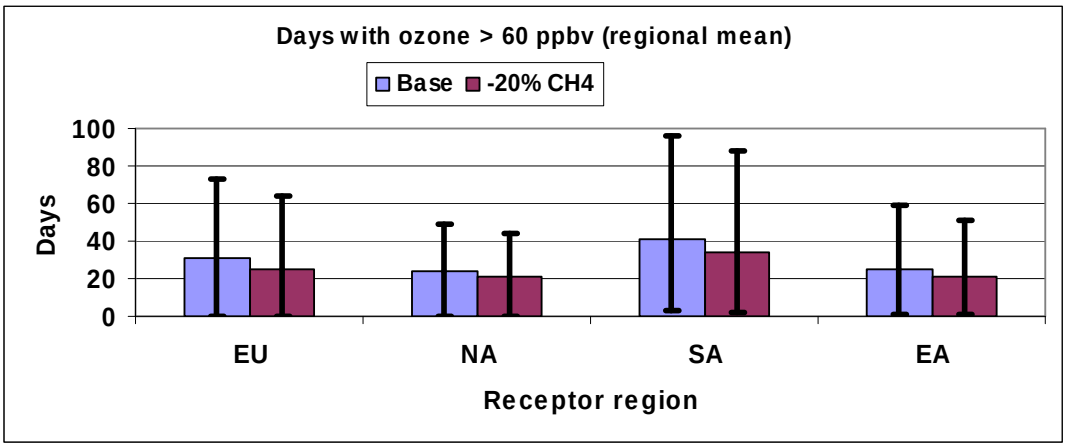

Figure 5.8 Variation among models estimating DAYS60. Ensemble mean (13 models) number of days per year where O_3 concentrations exceed 60 ppbv, spatially averaged over the receptor regions for the base simulation (light purple bar) and the simulation with global CH_4 concentrations reduced by 20 per cent (red bar). The whiskers span the full range of individual models.

5.2.5 Present-day source-receptor relationships for ozone columns

Substantial O_3 transport takes place above the boundary layer in the free troposphere. In this section, we show the perturbation to tropospheric O_3 columns resulting from lowering NO_x emissions by 20 per cent in individual regions. We compare the change in O_3 columns resulting from 20 per cent decreases in foreign NO_x emissions to that from a 20 per cent decrease in domestic NO_x emissions. We calculated the O_3 columns up to 150 hPa (~13 km altitude). This altitude is somewhat above the tropopause at middle and high latitudes and somewhat below in the tropics. In figure 5.9, we show the tropospheric O_3 column perturbations resulting from 20 per cent reductions in NO_x emissions in each region. Over EU, the tropospheric O_3 column response to NO_x perturbations from NA is as large as the response to domestic emissions. The response of O_3 columns to foreign (=NA+EA+SA) emissions perturbations is nearly twice as large as the domestic perturbation. North American column O_3 is less strongly impacted by foreign emission perturbations, with an import sensitivity of roughly 60 per cent that of the domestic sensitivity. In EA, the impact of EU NO_x emissions is particularly strong, of the same magnitude as the impact of EU emissions on the EU O_3 column, and the foreign (=EU+NA+SA) contribution to EA O_3 columns is larger than the domestic one by ~50 per cent. Finally, over SA, the domestic impact on the O_3 column is larger than that from foreign (=EU+NA+EA) NO_x emissions, with about equal contributions from the other three regions. We note large differences from the impacts of 20 per cent decreases in regional NO_x emissions on surface O_3 in figure 5.6. In EU and EA, the domestic impacts on surface O_3 are relatively stronger than on column O_3. The model spread in the sensitivity of the tropospheric column O_3 to NO_x emissions is lower than for surface O_3.

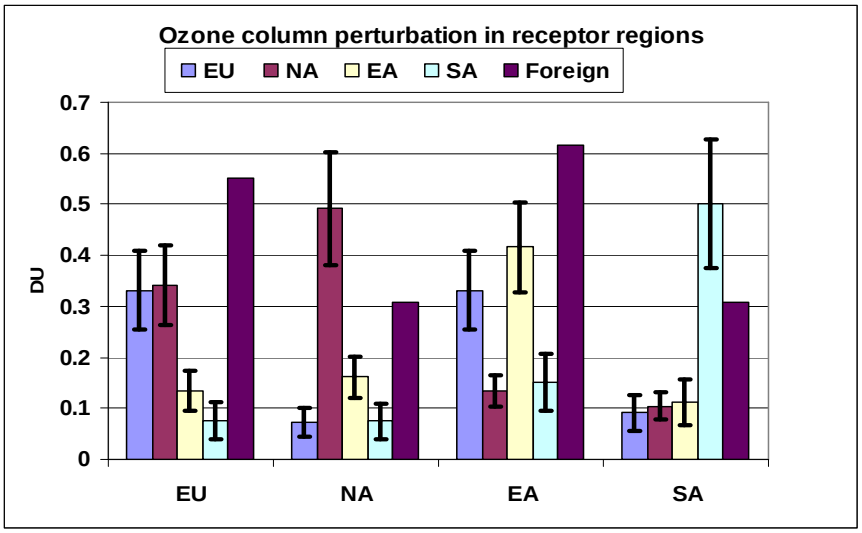

Figure 5.9 Reduction in O_3 columns resulting from NO_x emission reductions. Decrease in annual mean O_3 columns (DU=Dobson Unit=2.69×10^{16} O_3 molecules cm^{-2}; integrated from surface to 150 hPa) over the receptor regions resulting from a 20 per cent decrease in regional NO_x emissions. The bar depicts the 14-model mean, and the whiskers span the ± 1 standard deviation of the models.

5.3 Estimates of transport of aerosols and their precursors

5.3.1 Introduction

Long-range transport of aerosol particles is documented by satellite and surface observations of dust outbreaks and plumes from large fires. Occasionally, such aerosol clouds can be followed many days and around the globe by means of repeated satellite observations (Damoah et al., 2004). Dust has been shown to cross regularly the Pacific Ocean, the Atlantic Ocean and the Mediterranean Sea (Darmenova et al., 2005; Moulin et al., 1998; Wilkening et al., 2000). Backward trajectory analysis and particle transport modelling explained enhanced lidar signals measured at European Aerosol Research Lidar Network (EARLINET) sites in Europe by dust arriving from the Sahara as well as from Gobi and Taklamakan deserts reaching the French Alps (Amiridis et al., 2005; Grousset et al., 2003; Mattis et al., 2004). A Canadian forest fire plume in August 1998 crossed Greenland and the North Atlantic into Europe as shown with the space-borne Total Ozone Mapping Spectrometer (TOMS) and several lidar instruments (Forster et al., 2001; Hsu et al., 1999). These aerosol plumes appear as distinct aerosol layers in the free troposphere (Wandinger et al., 2002). In spring and summer 2003, intense fires in southeast Russia produced long-range transported aerosols and CO that could be observed through satellite observations (Damoah et al., 2004; Edwards et al., 2004).

In consequence, surface PM concentrations can be enhanced by intercontinental transport of both natural and anthropogenic aerosol. Prospero (1999) showed that PM levels in the south-eastern United States are being influenced by Saharan dust transport. Southern European PM levels may exceed EU limit values through episodic import of Saharan dust (Raes and Hjorth, 2006). Heald et al. (2006) showed export of Asian pollution may enhance sulphate levels by 0.16 $\mu g/m^3$ in the north-western United States in spring, contributing to an exceedance of visibility standards in wilderness areas.

It is commonly believed that aerosols are removed from the atmosphere within hours to several days (Warneck, 1988), but the examples above show that there are many exceptions to this averaged timescale. In contrast to episodic hemispheric scale transport, the role of continuous transport is poorly explored. Yet this may be an important mechanism for anthropogenic aerosol (precursor) transport and may lead to a rise of hemispheric background PM concentration levels. For example, a recent study based on MODIS satellite data, estimates annual transport of pollution PM from Asia to North America of 4 Tg/year (Yu et al., 2007). However, even if the transport is episodic,

it occurs regularly all year long. Most of the column aerosol transport occurs in spring (~1.7Tg), but ~ 0.9 Tg in winter, 0.7 Tg in summer and ~0.9 Tg in autumn. This is about 20–25 per cent of the non-dust aerosol that leaves Asia. In comparison, 240 Tg/year of dust is estimated, based on satellite-observed aerosol optical thickness data, to leave the Sahara with 50 Tg/year deposited in the Amazon basin and another 50 Tg/year deposited in the Caribbean (Kaufman et al., 2005).

Only a few studies have explored S/R relationships for aerosol emissions and burdens on continental scales (Park et al., 2004), and there are large discrepancies among them. In a modelling study to estimate the impact of transboundary transport of pollutants on the visibility in the United States, Park et al. (2004), reported that trans-Pacific transport of Asian pollution accounts for 30 per cent of the sulphate levels in the absence of United States pollution sources at the surface in both the western and eastern United States and suggested that meeting the U.S. EPA visibility objectives requires a combination of domestic and international emission controls. However, Chin et al. (2007) pointed out that even though the Asia pollution may make significant contribution to the background sulphate level, its influence is about 6 times smaller in the western United States and 60 times smaller in the eastern United States than North American regional sources. In addition, only 2–6 per cent of the annual average surface fine aerosol mass in the United States is estimated to be from the intercontinental transport of aerosol pollutants, including SO_4, BC, and OC (Koch et al., 2007). Estimating the total column export amount from major pollution source regions, Koch et al. (2007) suggest large export of aerosols from source regions; about 70-80 per cent by mass of most aerosol species are exported from Europe, Asia, and North America to other regions. South and East Asia contribute about 15 per cent of global sulphate and 30 per cent of global black carbon (BC) pollution loads, and Europe and North America each contribute about 5 per cent of global BC and sulphate pollution loads. These results are in contrast to Chin et al. (2000), which reported that less than 25 per cent of sulphur emitted in the major source regions is transported out from the regional column, and Park et al. (2004), which showed negligible amount of BC and OC over the United States that were from other pollution regions. Recently, Liu et al. (2007) performed tagging and perturbation experiments of sulphur emissions and showed that these techniques produced almost similar results over North America.

Results from the European Union project PHOENICS[6] (Kanakidou and Dentener, 2005) designed to evaluate import and export budgets of European aerosol are visualized in figure 5.10, showing the importance of natural dust and sea salt entering Europe, and the substantial export of anthropogenic elemental carbon (EC), particulate organic matter (POM), SO_4 and NO_3. Results from the PHOENICS study indicated that, depending on the aerosol component, European anthropogenic emissions contributed 40-80 per cent to European aerosol column loadings, and 1–5 per cent to the aerosol load in the rest of the world. Reddy and Boucher (2006) calculated that sources from East and South Asia together contribute more than 50 per cent to the global radiative forcing by BC. The trans-Pacific transport of BC and $PM_{2.5}$ into North America has recently been studied using aircraft and surface observations from the Cloud Indirect Forcing Experiment (CIFEX) (Roberts et al., 2006) and the IMPROVE network, and a CTM (Hadley et al., 2007). This study found that the value of the trans-Pacific flux of BC into the western boundary of North America in April 2004 was ~75 per cent of the North American BC emissions. Furthermore, ~80 per cent of the BC mass transport occurs above 2 km over the Pacific Ocean. BC of Asian origin was estimated to comprise ~20 per cent of the observed BC at IMPROVE measurement sites at 1 km elevation, and over 50 per cent at sites at 2 km elevation and higher.

[6] Particles of Human Origin Extinguishing Natural solar radiation in Climate Systems.

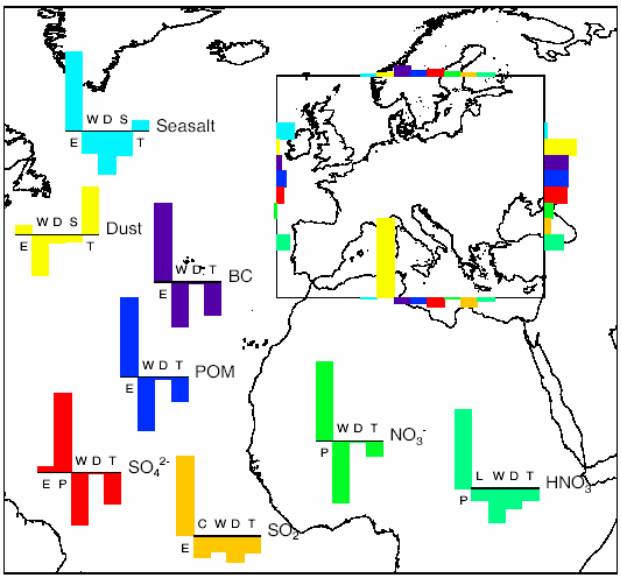

Figure 5.10 Annual average aerosol budget over Europe as modelled in PHOENICS**.** The bars on the bounding box around Europe (similar to the HTAP intercomparison EU region) denote advective fluxes **entering and leaving** Europe. The other bar-graphs denote for eight aerosol (precursor) components the production and loss fluxes **within** Europe, where E=emission, W=total wet deposition, D=dry deposition, S=sedimentation, T=total transport in/out Europe, P=chemical production, L=Chemical loss. For graphical representation the bars are normalized according to the maximum fluxes (Sea salt: 87.2 Tg yr-1; Dust: 34.2 Tg yr^{-1}; BC (or EC): 0.55 Tg C yr^{-1}; 1.72 Tg POM yr^{-1}; 10.6 Tg SO_4 yr^{-1}; 1.81 Tg NO_3 yr^{-1}, 22.8 Tg SO_2 yr^{-1}, 1.61 Tg HNO_3 yr^{-1}.

5.3.2 Analysis of results from AeroCom relevant for hemispheric transport of air pollution

The AeroCom model intercomparisons initiative was designed to evaluate the main uncertainties and performance of 20 global aerosol models (Kinne et al., 2006; Schulz et al., 2006; Textor et al., 2005). To describe the chemical and physical character of aerosol distributions, global aerosol models have become more complex in recent years. Many models now describe the aerosol as composed of several chemical species which are distributed among particles of diameter spanning from tenths to several microns. Since the focus of AeroCom was on climate issues, the most extensive comparison with measurement was focused on global data sets of aerosol optical depth from satellites and sun photometers. Interestingly, the total aerosol optical depth in most models is comparable to observational based estimates (Kinne et al., 2006) , whereas the underlying aerosol fields of sulphate, organic matter, dust and sea salt showed a much larger variation. The reason for the diversity in the individual aerosol fields is due to differences in the treatment of the important processes (i.e. dry and wet deposition, secondary aerosol formation, aerosol aging, transport and mixing, aerosol hygroscopic growth, interaction with radiation, aerosol-cloud-interactions), as well as to differences in emission estimates. The AeroCom experiments found that harmonization of emissions did not sufficiently reduce the diversity in aerosol column loads. Model-specific transport, removal, chemistry parameterization, and parameterisation of aerosol microphysics in the different models still were the major drivers for the diversity among the models (Textor et al., 2007). With respect to long-range transport, an example of model diversity is the estimate of transport to polar regions. Figure 5.11 shows that a considerable, but highly variable, fraction of global aerosol mass can be found in polar regions, where anthropogenic emissions are almost absent. Europe is the largest contributor to BC in northern high latitude regions (>70N).

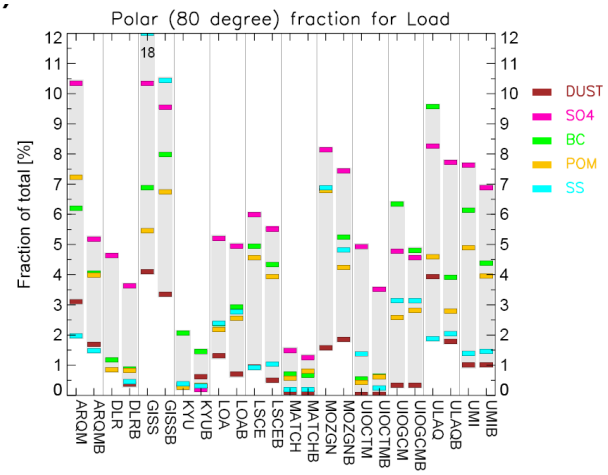

Figure 5.11 Annual fraction of global aerosol burden reaching the Polar Region (north of 80N and south of 80S). Five aerosol species are differentiated (Textor et al., 2007). The gray bars frame the range for each model.

An important conclusion from AeroCom was that the mean or median model compared better to observations than any of the individual models (Textor et al., 2007).

5.3.3 HTAP model simulations of hemispheric transport

In the HTAP intercomparison, compared to the previous AeroCom intercomparisons, new models and model versions have been used. Of the 17 HTAP intercomparison models with aerosol diagnostics, five participated in AeroCom. Results presented below are for the model simulations for HTAP Experiment Set 1. A 20 per cent reduction in all anthropogenic emissions of aerosol precursors and aerosol primary emissions was introduced into the perturbation experiments for each of the four target regions. The result of this perturbation was compared to the reference experiment with the original emissions.

Table 5.4, supported by figures 5.12 to 5.14, shows the results of the perturbation experiments. The difference between the base and perturbation simulation for four regions has been used to estimate the relative response to changes in imported pollution as compared to domestic pollution for each region, and to trace the fate of the different chemical species. Shown in table 5.4 are the annual mean import sensitivities (see section 5.1.2.1) for surface concentrations, deposition, and column loadings for the four HTAP regions. In general, the smallest import sensitivity is for surface concentrations, with the largest for the column loadings. The import sensitivities also vary by region. For example, the import sensitivity of surface concentrations varies from 5 per cent for EU to 25 per cent for SA. The largest import sensitivities for the SA region can be explained by several peculiarities of the HTAP experiment set-up:

(a) The SA region is close to and downwind of the EU region;

(b) The calculation of the import sensitivities is related to the area averages of the PM concentration, deposition, or column load originating from the SA region itself. This domestic impact is distorted to the lower end because the main emission sources in the SA region are in the eastern part of the box defined for the HTAP experiments.

c) The limited knowledge of the global spatial distribution of the anthropogenic emission sources for anthropogenic PM may produce high import sensitivities if models were using similar but incorrect emission estimates.

Table 5.4 Annual mean import sensitivities for surface concentrations, deposition, and column loadings for four HTAP regions. Multimodel derived import sensitivities for surface concentration, deposition and column load of anthropogenic BC, POM, SO_4 and all sulphur species (SO_2 + SO_4). Import sensitivities are calculated for each model as the response in a receptor region to the combined influence of 20 per cent emission reductions in the three intercontinental source regions compared to the response to 20 per cent emission reductions in the region itself. The mean sensitivities ± 1 standard deviation are derived from individual model results as displayed also in figures 5.12 to 5.14.

Receptor region:	EU	NA	SA	EA	# of models
Surface Concentration Import Sensitivity					
PM	5% ±4%	7% ±6%	25% ±12%	10% ±9%	9
Deposition Import Sensitivity					
SO_4	10% ±7%	9% ±5%	32% ±11%	13% ±9%	11
Sulphur (SO_2 + SO_4)	3% ±2%	3% ±3%	19% ±8%	5% ±3%	11
BC	1% ±1%	4% ±2%	13% ±24%	3% ±1%	8
POM	1% ±1%	3% ±2%	12% ±24%	4% ±1%	8
Column Load Import Sensitivity					
SO_4	33% ±23%	33% ±23%	59% ±19%	31% ±21%	11
BC	21% ±14%	32% ±23%	19% ±3%	17% ±5%	7
POM	27% ±21%	30% ±24%	13% ±5%	24% ±6%	7

The import sensitivities also vary by species. For example, deposition import sensitivity for sulphate is 3 times larger than for total sulphur (SO_2+SO_4). The spatial dispersion of total sulphur deposition is dominated by sulphur dioxide deposition, which occurs close to the sources. The fraction of total emitted anthropogenic sulphur from a given region that reaches a foreign receptor region is thus small. The total anthropogenic sulphur (SO_x=SO_2+SO_4) deposition import sensitivity varies between 3 to 20 per cent between regions. In contrast, the sulphate aerosol produced from SO_2 emissions has a longer lifetime. Thus the impact from foreign regions in terms of sulphate deposition and sulphate aerosol column load is considerable as shown in figure 5.12. For example, the import sensitivity of SO_4 column load varies from 31 to 59 per cent. This indicates the substantial role of aerosol transport above the planetary boundary layer.

Preliminary analysis with a smaller number of models also shows that the emissions of POM and BC have a larger impact on the source region itself; the import sensitivity of the anthropogenic soot column load is 17 to 32 per cent and that of particulate organic matter is 13 to 30 per cent. Diversity among models is in part due to whether the models do or do not take into account secondary organic aerosol formation.

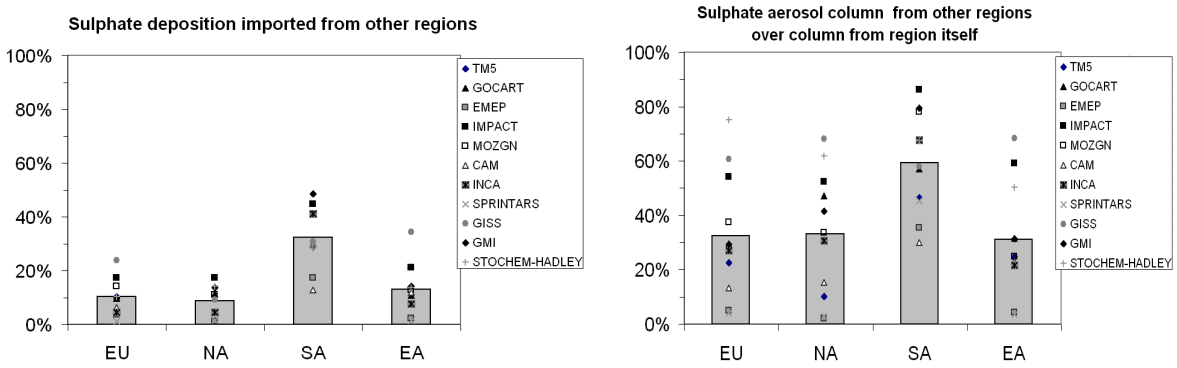

Figure 5.12 Sulphate deposition and sulphate aerosol column load import sensitivity in four HTAP regions. Average import sensitivity (as bars) of total sulphate deposition (left panel) and sulphate aerosol column burden or "load" (right panel) from other regions into target region. Individual model results are superimposed as symbols.

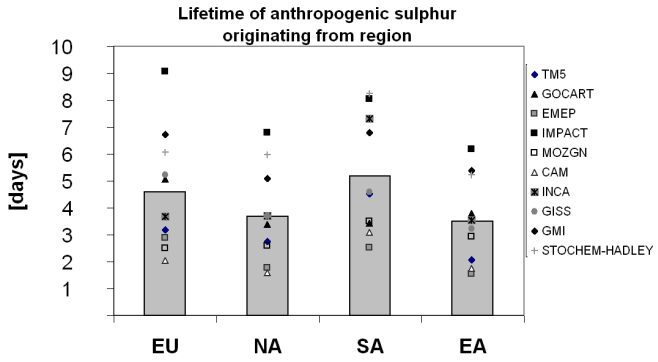

Figure 5.13 Lifetime of anthropogenic sulphate originating from different regions in different models (symbols) and its average (bars). Lifetime is computed from the anthropogenic sulphate burden response and total deposition response due to perturbation of emissions in each region.

The HTAP intercomparison allows for the first time an investigation of the differences among models of the specific aerosol life cycles experienced by an aerosol originating from different regions. Figure 5.13 illustrates the diversity of sulphate lifetimes in the models. The impact of one region on another depends on the transport pathways, their length, and the intensity of the removal processes that the aerosol is experiencing along its way. In general, models with the longer lifetimes predict larger import sensitivities. The differences in lifetimes between regions are consistently described by the individual models. Emissions from North America and East Asia appear to be slightly more efficiently removed than those from Europe and South Asia. As in AeroCom, the model diversity of sulphate lifetimes in absolute terms for a given region is considerable (a factor of four), and reflects differences in the process level model formulations.

As stated earlier, a preliminary analysis of the surface aerosol concentrations reveals that the import sensitivity for the annual average anthropogenic $PM_{2.5}$ varies between 5 and 25 per cent (see tables 5.4 and 5.5). There is consistency among the models with respect to the relative import sensitivities of the different regions, but appreciable diversity between the model predictions in terms of the absolute contributions (see figure 5.14). As discussed above, the SA region is estimated to receive the highest PM load from abroad. However, the limited number of models with similar complexity, especially with regard to organic aerosols, makes the results rather uncertain (see also the

high standard deviations in tables 5.4 and 5.5). Furthermore, the import sensitivity depends on the method of averaging. For table 5.4, import sensitivities are first established for each model and then averaged, giving 25 per cent PM import sensitivity for SA. In table 5.5, imported PM concentrations are averaged across model results and the fraction is established on the basis of the annual mean average response, resulting in 18 per cent PM import sensitivity for SA.

Table 5.5 Import sensitivity for four HTAP regions calculated from the multimodel mean response. Multi-model (9 models) response of (a) annual mean surface PM (ng/m^3); (b) column load of sulphate ($\mu g/m^2$) and (c) column load of particulate organic matter ($\mu g/m^2$) in receptor regions to 20 per cent reductions of anthropogenic gas and aerosol emissions in the source regions (rows). Domestic contributions are shown in bold. Inter-model standard deviations are shown in parentheses. PM is computed from sulphate and particulate organic matter surface concentrations.

(a) Annual mean response of PM (ng/m^3)

	Receptor region	EU	NA	SA	EA
Source region	EU	**-711 (313)**	-8 (5)	-76 (20)	-22 (16)
	NA	-18 (9)	**-352 (116)**	-9 (7)	-6 (3)
	SA	-4 (6)	-1 (2)	**-627 (225)**	-19 (6)
	EA	-7 (5)	-10 (7)	-27 (18)	**-738 (338)**
Total import sensitivity		4%	6%	18%	6%

(b) Annual mean response of column load SO_4 ($\mu g/m^2$)

	Receptor region	EU	NA	SA	EA
Source region	EU	**-964 (271)**	-53 (46)	-266 (92)	-129 (94)
	NA	-173 (137)	**-587 (195)**	-48 (47)	-48 (49)
	SA	-47 (40)	-35 (36)	**-696 (213)**	-103 (61)
	EA	-102 (93)	-122 (104)	-100 (78)	**-950 (303)**
Total import sensitivity		33%	36%	59%	30%

(c) Annual mean response column load POM ($\mu g/m^2$)

	Receptor region	EU	NA	SA	EA
Source region	EU	**-180 (211)**	-4 (4)	-25 (24)	-12 (16)
	NA	-16 (19)	**-95 (86)**	-4 (3)	-4 (4)
	SA	-13 (9)	-9 (8)	**-410 (283)**	-47 (26)
	EA	-13 (18)	-16 (21)	-25 (20)	**-293 (258)**
Total import sensitivity		23%	30%	13%	21%

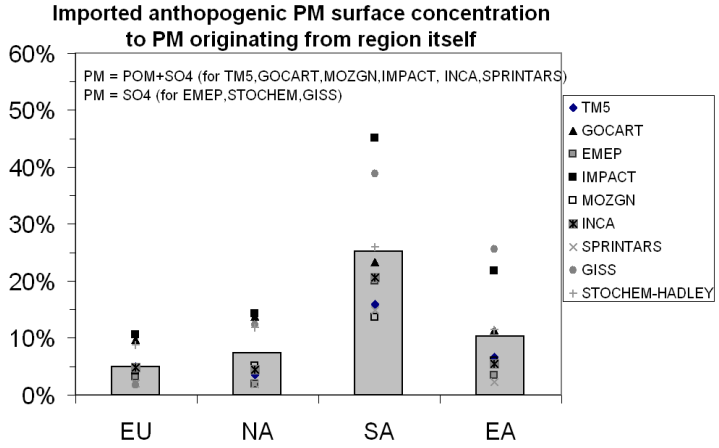

Figure 5.14 Surface PM concentration import sensitivity in different models (symbols) and its average (bars).

5.4 Impact of hemispheric transport on nitrogen deposition

Reactive nitrogen, consisting of oxides of nitrogen and ammonia in gaseous and condensed forms (referred to here as Nr = (NO_y + NH_x)), plays a central role in the chemistry of the atmosphere as well as being a key nutrient of marine, freshwater and terrestrial ecosystems. There is a host of evidence that increasing human activities seriously disturb the natural nitrogen cycle (Galloway et al., 2004; Phoenix et al., 2006; Vitousek et al., 1997). Nr enters the environment through a number of processes related to fertilization, waste discharge, and atmospheric emissions. A recent multimodel study (Dentener et al., 2006a) evaluated the impact of 3 different emission scenarios on future ecosystem nitrogen loads. Figure 5.15a shows the global Nr deposition for the year 2000, which often exceeds a threshold of 1000 mg(N) m^{-2} yr^{-1} (also called "the critical load", above which adverse changes in ecosystem functioning may occur, and for which values for specific ecosystems can be very low, e.g. as low as 300 mg (N) m^{-2} yr^{-1} for heathland). Figure 5.15b shows that Nr deposition, according to the IPCC-SRES A2 scenario in 2030, typically increases by 20–100 per cent over most of the Northern Hemisphere, with the patterns of growth suggesting that long-range transport plays an important role.

However, from the Dentener et al. (2006a) study it is not clear to what extent the individual regions used in the HTAP intercomparison export Nr and contribute to this long-range transport. Figure 5.16 shows results from the HTAP experiments where anthropogenic NO_x emissions are reduced by 20 per cent in each of the four regions. The sensitivity of Nr deposition, expressed as a fraction of the emission perturbation, is shown for each of the regions, as well as for the Arctic (AR) (70–90°N). Most (74–80 per cent) of the Nr deposition impacts occur within the emission perturbation regions, but there is some export through long-range transport of NO_y. The largest impacts from one region on another are 6.0 per cent (SA on EA); 2.4 per cent (EU on AR); 2.0 per cent (NA on EU); and 1.4 per cent (EA on NA). Thus altogether, regions export between 20 and 25 per cent of the Nr; of which 2–8 per cent deposits in other HTAP regions.

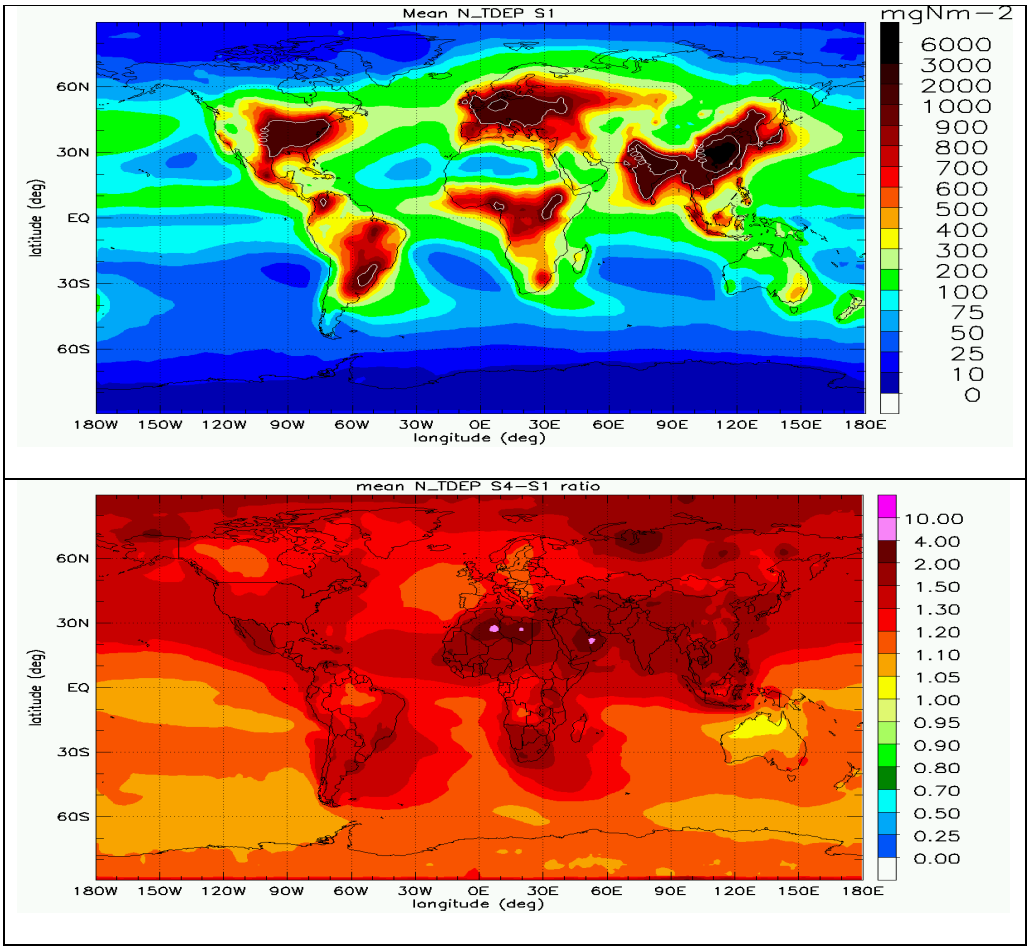

Figure 5.15 Nr deposition flux for 2000 and 2030. (a) Annual deposition flux of Nr for year 2000 (mg(N) m^{-2} yr^{-1}); (b) Ratio of the annual deposition flux of Nr in 2030, under the SRES A2 scenario, relative to the year 2000 flux. Results are multimodel means from 23 models described in Dentener et al. (2006a).

In table 5.6, we present a receptor-oriented analysis of how much a 20 per cent decrease in NO_x emissions from each HTAP region contributes to the overall change in deposition in each receptor region. The results show that for the receptor region EU, 96 per cent of the change in deposition is due to emissions changes in EU, while 4 per cent of the change in deposition arises from changes in emissions elsewhere. It should be noted that in this model analysis no changes in emissions outside the HTAP regions were considered. When comparing the fractional Nr changes in table 5.6 with the import matrix of aerosol components displayed in table 5.4, relatively similar numbers are found for Nr deposition changes compared to, for instance, sulphur deposition.

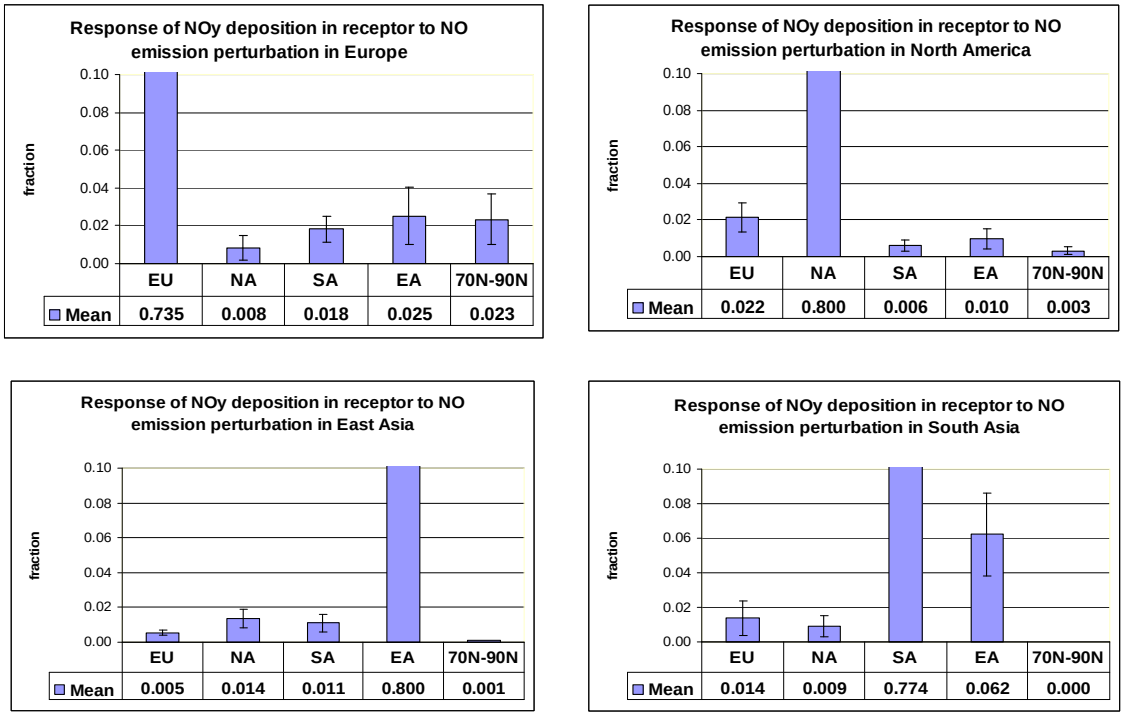

Figure 5.16 Sensitivity of Nr deposition (response normalized to the emission perturbation), following a 20 per cent anthropogenic NO_x emissions reductions in each of the regions. Bars are multimodel means (10 models); whiskers are inter-model standard deviations. Note that the bars in the source regions are "off-scale".

Table 5.6. Overall change in deposition in each receptor region resulting from a 20 per cent decrease in NO_x emissions from all HTAP regions. Each entry represents the fractional contribution to total Nr deposition changes over each receptor region, resulting from anthropogenic NO_x emission perturbations of 20 per cent in each of the four source regions. (Note columns add up to 1.)

	Receptor region			
Source Region	**EU**	**NA**	**SA**	**EA**
EU	**0.961**	0.009	0.076	0.033
NA	0.028	**0.974**	0.020	0.013
SA	0.005	0.003	**0.881**	0.025
EA	0.006	0.014	0.033	**0.929**

5.5 Source-receptor relationships using idealized Carbon Monoxide tracers

Differences in model results may result from various sources. In particular, models may use different emission data sets, chemical schemes, parameterizations for transport and deposition processes, and different meteorological fields (i.e. the European Centre for Medium-range Weather Forecasting (ECMWF), NCEP[7], or other weather analysis centres or output from general circulation models). To understand better inter-model differences, as part of Experiment Set 2, a multimodel experiment was set-up with passive tracers that show CO-like behaviour. Specifically, all models participating in this experiment used identical emission data and the same chemical loss rate, parameterized as a globally uniform constant decay rate, with a lifetime of 30 days, comparable to CO. As a consequence, all differences in the model results can be attributed solely to differences in

[7] National Centers for Environmental Protection (United States).

the driving meteorology and model parameterizations of transport processes. Tracers were tagged for the four source regions displayed in figure 5.2.

Figure 5.17 compares some of the CO results of the full chemistry experiments in Experiment Set 1 to the CO results of the constrained passive tracer experiments in Experiment Set 2. The left panel shows the seasonal cycle of CO transport from SA to EA using results from the full chemistry simulations. Since the chemical lifetime of CO is longer than the typical transport time between SA and EA, different representations of the loss processes (e.g. reaction with OH) should be of minor importance in explaining these inter-model differences. However, from the set-up of these experiments, it remains unclear whether differences in emissions (for example the seasonality of biomass burning emissions) or differences in transport are dominating. The right panel of figure 5.17 displays the results from the better-constrained passive tracer experiment. The model spread is strongly reduced, which indicates that there are large differences in the various emission data sets used in the full chemistry simulations for the SA region, including, for example, the seasonality of biomass burning emissions.

In figure 5.18, an evaluation of the boundary layer mixing of the different models in the constrained passive tracer experiment is shown for the NA region. Boundary layer mixing is deduced from analysing the ratio of CO concentrations in the boundary layer (integrated over the lowest two kilometres) to the modelled CO concentrations at the surface. We note that this analysis may include some bias due to different surface layer thicknesses in the individual models, although we have not seen a systematic effect. While the models agree reasonably well in winter (mixing coefficients between 0.35 and 0.55), the inter-model differences in summer are much larger (0.4 to 0.8). It is likely that this can be attributed to the increased convective activity during summer and the large uncertainties related to the sub-grid scale parameterization of convective mass fluxes in the models. Similar results (with somewhat smaller numbers) have been found for the EU region (not shown). These results are consistent between the full chemistry simulations and the passive tracer runs, which indicate that inter-model variability in this case is due to differences in transport parameterizations and possibly also the meteorological datasets used.

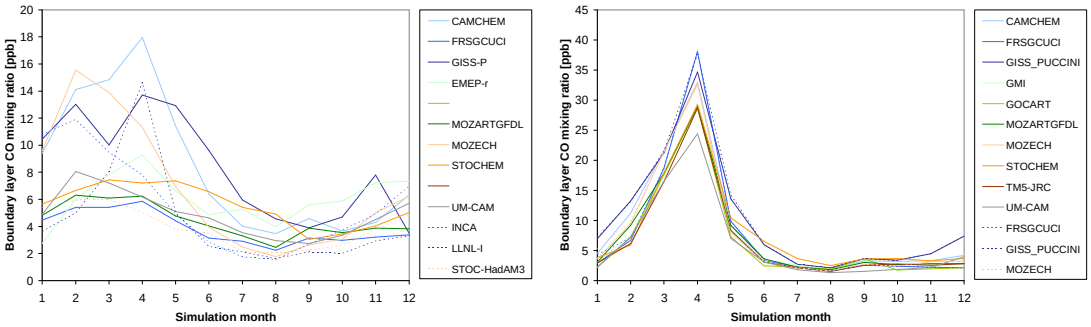

Figure 5.17. Seasonal cycle of simulated CO concentrations from emission sources in South Asia in the boundary layer (0-1 km) over East Asia. Left: results from the full chemistry simulations performed in HTAP Experiment Set 1 (unconstrained emissions and chemistry), right: results from the passive tracer runs in Experiment Set 2 (identical emissions and fixed tracer lifetime)

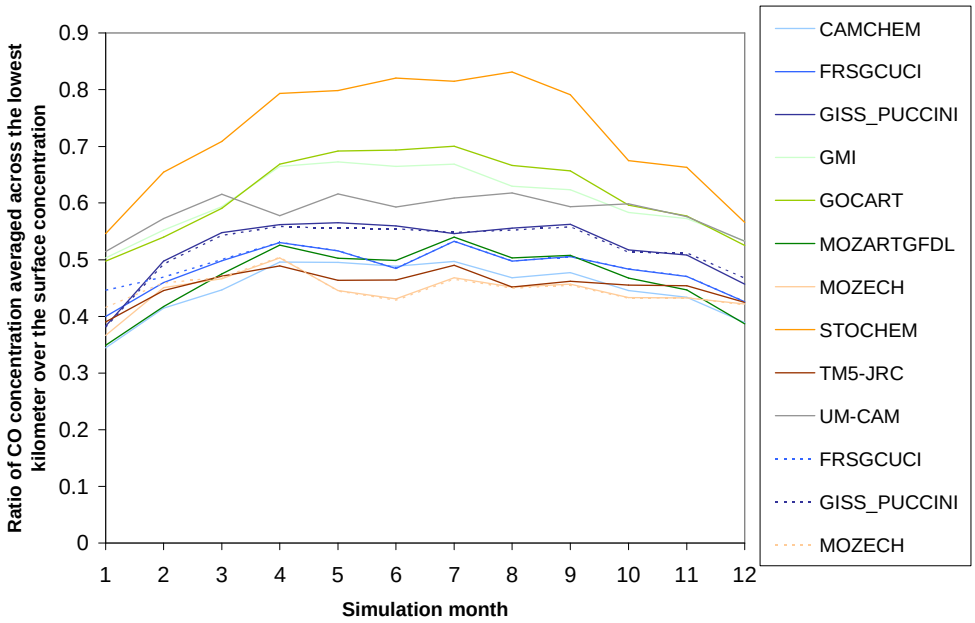

Figure 5.18. Ratio of North American CO concentrations in the boundary layer (0-1 km) to the surface concentrations (lowest model layer) in the source region. Models with larger ratios exhibit more active boundary layer mixing. Results are from the constrained passive tracer experiments in HTAP Experiment Set 2.

5.6 Regional source-receptor studies

Global CTMs provide the framework for which intercontinental scale S/R relationships can be estimated. However, these global models often utilize fairly coarse spatial and temporal resolutions and simplified physical and chemical parameterizations because of computational limitations. Thus, it is important to evaluate how sensitive the predictions of pollution import and export for a region are to model resolution, and how predicted S/R relationships vary within the region.

S/R relationships calculated with regional CTMs were compared for a study domain similar to the EA domain used in the HTAP intercomparison in MICS-Asia (Carmichael et al., 2002). S/R relationships estimated by the models showed a high degree of consistency in identifying the main S/R relationships, as well as in the relative contributions of wet and dry pathways for removal. But at some locations estimated deposition amounts were found to vary by a factor of five. A sample result is shown in figure 5.19, where S/R relationships for two different receptor sites are shown. The influences of model structure and parameters on model performance were evaluated. The S/R relationships were found to be most sensitive to uncertainties in the sulphur emission inventory and, secondarily, to the driving meteorology. Both factors were found to be more important than the uncertainties in the model parameters for wet removal and sulphate production. A major limitation of this study was the lack of coherent data sets of sulphur ambient concentrations and deposition measurements over EA with which to evaluate model performance. The interannual variability of the country-to-country S/R relationships was evaluated using a single model run for 25 years (1975 to 2000) using NCEP meteorology (Calori et al., 2001). Shown in figure 5.20 are the variations in annual S/R relationships (country-to-country) over the 25 year period due to the interannual variability in meteorology (with emissions held constant). The variation in S/R relationships for nearby sources is in the order of 10 per cent, however, as the distance between source and receptor increases, the inter-annual variability also increases, reaching values in some cases of greater than 50 per cent.

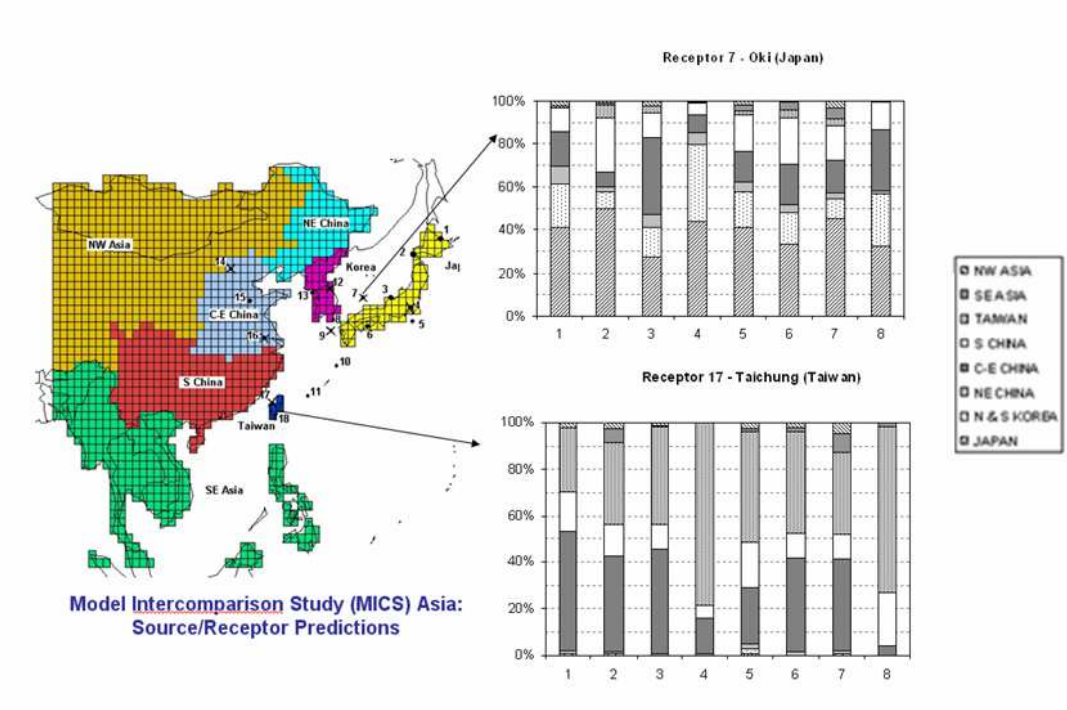

Figure 5.19 S/R relationships for two different receptor sites. S/R relationships for total sulphur deposition calculated from eight models for two different receptor sites. The source regions are NW China, NE China, CE China, S China, SE Asia, Korea and Japan. Results are from the MICs study (Carmichael et al., 2002).

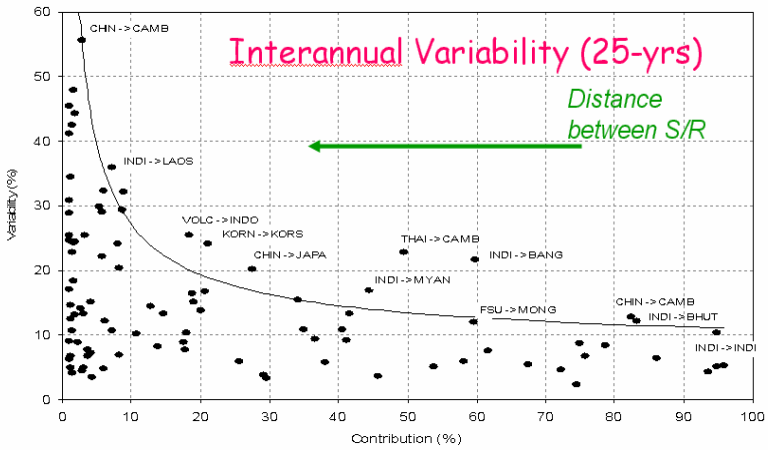

Figure 5.20 Variations in annual source-receptor relations over the 25-year period due to the inter-annual variability in meteorology. Variation in the Asian source receptor relationships (country to country) due to inter-annual variations in meteorology. Each point represents one set of S/R relationships, with the contribution referring to the 25-year mean impact of that source region to the total deposition at the receptor. The variability is with respect to the differences between the annual S/R relationships for the 25 different years. In general, the contribution of a source to a receptor region increases as the distance between them decreases. For example, the far right point marked Indi->Indi indicates that, for the 25 years of calculation, the emissions from India account for 95 per cent of the deposition on India (the x-axis) and that the inter-annual variability in this relationship is 10 per cent. In contrast, the influence of emissions from China on deposition in Cambodia (upper left point) has a mean value of 3 per cent with an inter-annual variability of 55 per cent.

In 2003, the MICS study was expanded to include nitrogen compounds, O_3, and aerosols – key species for regional health and ecosystem protection, and pollutants of growing concern throughout Asia. This broader collaborative study – MICS-Asia, Phase II – examined four different periods, encompassing two different years and three different seasons (i.e. March, July, and December in 2001, and March in 2002). An additional feature unique to Phase II was the inclusion of global inflow to the study domain. Nine different regional modelling groups simulated the chemistry and transport of O_3, precursors, sulphur dioxide and secondary aerosols, using common emissions and boundary conditions derived from the global Model of Ozone and Related Tracers (MOZART, v. 2.4). The inclusion of a global model along with the regional models in the intercomparison, provided a means to assess the impact of model resolution. Details are presented in Holloway et al. (2007). In terms of prediction of ground level O_3, when compared to observations from the Acid Deposition Monitoring Network for East Asia (EANET), the global model predictions were comparable to the regional models, with an r-value for monthly means for all four seasons in the upper 50 per cent of the individual regional model predictions, but the mean bias error (MBE) was greater than most of the MICS models. To assess the magnitude of import from other major emission areas in the Northern Hemisphere to the East Asia domain, additional simulations with the global models were made where anthropogenic emissions from North America and Europe were separately shut off. Strong seasonal cycles in the contributions of these sources to surface O_3 in East Asia were calculated, with monthly contributions ranging from 1–4 ppbv for each source region.

In the next phase of MICS, it is intended that the regional models will perform experiments in support of the HTAP intercomparison.

Regional models have also been used in long-term model simulations to elucidate the impact of increasing emissions from East Asia on long-term trends of lower tropospheric O_3. For example, model simulations were performed by a CMAQ[8]-based regional CTM[9] (with initial and lateral boundary conditions supplied from a global CTM), with year-dependent and constant emissions inventory for eight years from 1996 to 2003. The modelled long-term O_3 trends in East Asia with year-dependent emissions were rather small for both boundary layer and lower free troposphere over Japan, in contrast to the observed increasing trend (~1 ppbv/year) at several mountainous sites in Japan (Tanimoto, 2007). This suggests that current bottom-up estimates of NO_x emissions from East Asia may still be underestimated as suggested by recent observations from space (Richter et al., 2005) and that contributions from other sources such as soil emissions by land-use changes may be substantial. However, the observation-based trends may be overestimated due to limited numbers of monitoring stations.

5.7 Suitability of current models

The suitability of the current generation of models for quantifying the effects of the intercontinental transport of oxidants and aerosols relies on an adequate representation of the controlling chemical and physical processes. To some extent, models can be evaluated using model intercomparison studies, but low diversity in the predictions does not ensure that the predictions are realistic. Direct comparisons of the predictions with observations are needed to assess the accuracy of the predictions. A challenge in assessing the suitability of current CTMs for simulating hemispheric transport is that the S/R relationships of interest are model constructs, and there is, therefore, no simple means of evaluation. At present, the accuracy and validity of a S/R relationship depends on the adequacy and completeness of the atmospheric models from which it has been derived. Below we summarize major findings from studies that have evaluated the strengths and weaknesses of current CTMs with respect to predicting ambient distributions, with an emphasis on the relevance for assessing model skill at representing hemispheric transport of air pollution.

Model intercomparison studies with evaluation against observations have been conducted in most of the source regions used in the HTAP intercomparison using regional CTMs. Results from

[8] Community Multiscale Air Quality Model.

[9] Chemical Transport Model.

these studies evaluate how well current CTMs are able to capture observed O_3 and PM features in the source regions, and provide insight into how well we expect global CTMs to estimate pollution export and to quantitatively predict surface O_3 values. For example, the EuroDelta project was designed to evaluate the regional responses to emission reduction scenarios, in support of the European Commission Clean Air for Europe (CAFE) Programme and of work under the Convention. Long-term (year-long) O_3 simulations from seven regional air quality models were analysed for O_3 prediction metrics related to human and ecosystem health for the European domain. In general, daily O_3 maxima were slightly better simulated than daily averages, and summertime concentrations were better simulated than wintertime concentrations. Ensemble average concentrations were found to almost always exhibit a closer proximity to observations than any of the models, and further analysis shows that the spread of the model ensemble is representative of the uncertainty in the simulations. Further details can be found in van Loon et al. (2006) and Vautard et al. (2007). Similar results have emerged from a study in North America, where results from eight models were compared with surface O_3 and $PM_{2.5}$ observations (McKeen et al., 2005). Analysis of the ensemble predictions demonstrated that, for a variety of skill measures, the ensemble usually had greater skill than each of the individual models and the ensemble of the bias-corrected models had the highest skill of all. Further details can be found in Pagowski et al. (2005) and Wilczak et al. (2006). At the global scale, CTMs can reproduce the magnitude and seasonal variations in observations of O_3 and its precursors from O_3 sondes and ground-based measurement stations (Hauglustaine et al., 1998; Prather et al., 2001; Stevenson et al., 2006; Wang et al., 1998), but have more difficulty representing long-term trends.

Several field measurement campaigns over the past decades have focused specifically on export of pollutants from the continental boundary layer and their subsequent transport, e.g. PEM-West B(Hoell et al., 1997), TRACE-P (Jacob et al., 2003), ACE-Asia (Huebert et al., 2003), and ICARTT (Fehsenfeld et al., 2006). These experiments allow for the evaluation of predictive capabilities of models in relation to the export of pollutants from a continent. In general, the predictive skill for the chemical species decreases with distance above the surface, reflecting the large uncertainty associated with vertical transport processes. An intercomparison of four global-scale and three regional-scale CTMs, using TRACE-P CO observations, found substantial differences in spatial distributions and column amounts due to differences in the representation of meteorological processes (Kiley et al., 2003). Model differences in the treatment of planetary boundary layer dynamics, vertical convection and lifting in frontal zones were found to result in differences in modelled column amounts along specific flight paths of a factor of two (Kiley et al., 2003). Correlations between tracers are typically captured correctly, providing some evidence that the models adequately represent pollutant export (e.g. Li et al. (2004), Parrish et al. (2004b)). However, reproducing specific features in the observations (e.g. plume encounters) have met with mixed success because of their sensitivity to the timing and location of meteorological events.

Less is known about the capability of CTMs to predict the intercontinental transport of pollutants at receptor locations. For species such as CO and aerosols, where it is possible to observe intercontinental transport episodes directly, a combination of space-based, aircraft and ground-based measurements have been used to evaluate global tropospheric chemistry models (Guerova et al., 2006; Heald et al., 2003; Kiley et al., 2003; Pfister et al., 2005; Pfister et al., 2006). However, direct hemispheric transport of O_3 is difficult to observe (Goldstein et al., 2004; Price et al., 2004), and thus previous studies have often evaluated models with measurements of total O_3 and related species at receptor locations and then used the models for source attribution (Auvray and Bey, 2005; Derwent et al., 2004; Fiore et al., 2002b; Hudman et al., 2004; Jaeglé et al., 2003; Li et al., 2002; Sudo and Akimoto, 2007; Weiss-Penzias et al., 2004; Wild et al., 2004a; Yienger et al., 2000). These studies have shown that CTMs are able to capture much of the day-to-day variability in observed surface O_3 concentrations at continental and downwind sites, often to within 5 ppbv. Comparison of models to measurements during field campaigns in the north-western United States and north-eastern Pacific revealed model skill at reproducing the temporal variability in observed CO concentrations (Goldstein et al., 2004; Jaeglé et al., 2003).

While a number of studies indicate that global tropospheric chemistry models can resolve synoptic-scale transport events, plumes are often diluted more quickly than observed due to numerical

diffusion and poor resolution (Heald et al., 2003; Pfister et al., 2006). The dilution of model plumes makes it especially difficult to quantitatively model episodic transport. Therefore, the relative importance of episodic vs. background transport for O_3 and aerosols has not been clearly characterized (Wild et al., 2004a; Yienger et al., 2000). Furthermore, transport on scales smaller than the synoptic are represented in global models with less certainty, or not represented at all. For example:

(a) Globally, the vertical mass flux of air due to deep convection is comparable to the mass flux due to extra-tropical cyclones (Cotton and Pielke, 1995). Yet, convective parameterizations are generally tuned to match the thermodynamic transport due to deep convection, but are not as constrained when applied to tracer transport (Yano et al., 2003). Large discrepancies in upwards mass transport between different models may exist. The tracer transported is sensitive to the convective parameterization used (e.g., Mahowald et al., (1995)). Ozone distributions respond to convection differently in different models (Doherty et al., 2005; Lawrence et al., 2003).

(b) The representation of the boundary layer and boundary layer venting is problematic in global models. This includes substantial uncertainties in the diurnal cycle of boundary layer depths and the venting between the boundary layer and the free troposphere. While pollutants can be vented from the boundary layer in observable plumes on the synoptic scale, there are very few measurements of episodic transport into the boundary layer. For example, Derwent et al. (1997) found only five probable cases of North American emissions influencing Mace Head on the west coast of Ireland, but the pollutant concentrations were quite low.

(c) Other processes shown to be important for venting the boundary layer, such as topography and mountain valley circulations (Henne et al., 2004) or land-ocean breezes can only be represented in global CTMs to a very limited extent.

Quantifying the contribution of hemispheric pollution in driving the longer-term changes (decadal trends) in the hemispheric background levels of pollutants is an outstanding scientific question. Addressing this question will require a closer integration of measurements and models, and more detailed evaluation of the capabilities and limitations of models to reproduce decadal trends. Insights into the capabilities of models to reproduce long-term trends will come from the RETRO Project (Reanalysis of the Tropospheric chemical composition over the past 40 years). The primary objective of RETRO is to understand, detect and assess long-term changes and interannual variability of the tropospheric chemical composition over the last 40 years, thereby providing the essential framework for understanding possible future changes. Results from five models are now being analysed. Further details can be found at: http://retro.enes.org/.

A final consideration is the fact that predictions of pollution distributions and trends, as well as S/R relationships, are inherently uncertain due to the large uncertainty in emissions and their changes over time. Emissions are generally poorly constrained on a regional basis. Bottom-up uncertainties in emission estimates are often not available, although Bond et al. (2004) suggests the uncertainty range for black carbon is often a factor of five and Yevich and Logan (2003) suggest uncertainties in estimates of biofuel burning of a factor of two. Arellano and Hess (2006) show that even under idealized circumstances top-down emission estimates via inverse modelling often differ by more than 50 per cent depending on the transport model used.

In summary, the analysis of hemispheric transport of pollutants and an assessment of the contribution from emissions from various sources depend critically on the use of CTMs. While current regional and global models remain far from ideal for reproducing the magnitude and timing of specific transport episodes, they are able to capture the general behaviour of well-mixed background conditions adequately, and are therefore suitable for estimating mean S/R relationships. Additional model comparison and harmonization exercises are needed to further increase confidence in scientific, regulatory, and policy applications of CTMs to hemispheric transport problems. Several recent studies have identified pressing research needs to improve pollution predictions (Dabberdt et al., 2004; Moussiopoulos and Isaksen, 2007). Furthermore, more critical tests of the ability of models to reproduce the processes governing intercontinental transport may be possible in future by

combining model and measurement techniques. Weiss-Penzias et al. (2007) showed that the enhancement ratio of total airborne mercury to CO can be used to identify plumes containing Asian fossil fuel effluents reaching Mt. Bachelor Observatory, Oregon, United States. Concurrent space-based measurements of O_3 and CO from the TES instrument may prove useful for assessing model estimates of anthropogenic contributions to global O_3 (Zhang et al., 2006). Wild et al. (2004b) and Auvray et al. (2007) have evaluated modelled chemical tendencies with observation-derived O_3 production and loss rates and demonstrated how this may be used to test chemical timescales during pollutant export.

5. 8 Future changes in source-receptor relationships

5.8.1 Influence of anthropogenic changes in emissions

As the distribution and magnitude of trace gas emissions changes in the future, so will downwind impacts on receptor regions. Multimodel studies have considered future changes in tropospheric O_3 under a variety of scenarios (Dentener et al., 2006b; Gauss et al., 2003; Prather et al., 2003; Stevenson et al., 2006).

It is likely that, in the coming decades, strong regional emission changes can be anticipated, as discussed in chapter 4. These changes in emissions will impact the S/R relationships since the altered concentrations of many species (e.g. OH, HO_2) will influence chemical lifetimes. If the emission locations change, the physical lifetime (i.e. dry and wet deposition) may also be affected, although the effect of this may be markedly different for O_3 and its precursors and for aerosols. However, past global modelling studies have focused on responses to changes in global emissions, rather than isolating individual source regions, so it is difficult to quantify changes in specific S/R relationships based on the literature.

There have been a few studies explicitly considering source apportionment of projected future changes (Stevenson et al., 2002; Szopa et al., 2006). A general finding is that for regions with future projected emission reductions (e.g. Europe, North America, Japan), increases in background O_3 associated with long-range transport are likely to become a proportionately more important component of surface O_3.

5.8.2 Influence of climate change

Specific meteorological processes (e.g. convection, frontal passage, subsidence) affect export of pollution from a region. These processes display natural variability, but in addition, if emission locations change, and/or climate changes, the intersection of pollution source regions and meteorology may also change, and this in turn may change export and import processes and magnitudes. Pollution export is dependent upon location (as shown in figure 5.1): Export from Southeast Asia is more efficient than from North America, and Europe is the least efficient exporter. This is due to the amount of convective activity and also downwind frontogenesis over oceans (Stohl, 2001; Stohl et al., 2002). Future emissions are set to shift towards the tropics (IPCC, 2000), tending to favour enhanced export.

A number of robust changes in transport patterns under climate change scenarios have emerged from model, theoretical, and observational studies. Hess and Lamarque (2007) and Li et al. (2002) show S/R relationships in the Northern Hemisphere are modulated by the North Atlantic Oscillation (NAO), with increased transport of United States emissions to northwest Europe, and increased transport of European emissions to the Arctic during the positive phase of the NAO. Most models used in the IPCC Fourth Assessment report an increasing trend in the positive phase of the NAO in climate change scenarios (Miller et al., 2006). A future decrease in synoptic activity in the Northern Hemisphere mid-latitudes has been noted in a number of observational (Agee, 1991; McCabe et al., 2001; Zishka and Smith, 1980) and modelling studies (Dai et al., 2001; Mickley et al., 2004; Murazaki and Hess, 2006). This is likely to lead to less frequent large-scale venting of the boundary layer (Mickley et al., 2004). Holzer and Boer (2001) show that less vigorous atmospheric flow associated with a warmer climate leads to larger concentrations of an emitted species near its source and smaller concentrations away from the source. Finally, Held and Soden (2006) show that

discrepancies between the future trend of water vapour and precipitation as climate warms, imply a decrease in the convective mass flux in the tropics.

Once exported from a source region, the extent of long-range transport depends upon the lifetime of the pollutant. For O_3, this is determined by chemical loss and dry deposition; for some O_3 precursors (and their reservoirs), wet deposition may also be important. The mean lifetime of tropospheric O_3 decreases with rising global emissions; a further decrease occurs when future climate change is also considered (Johnson et al., 1999; Johnson et al., 2001; Liao et al., 2006; Murazaki and Hess, 2006; Racherla and Adams, 2006; Stevenson et al., 2005; Stevenson et al., 2000; Stevenson et al., 2006). For the case of aerosols, the lifetimes are heavily dependent on the wet removal processes (i.e. the lifetimes decrease as wet removal rates increase). Changes in cloud properties and precipitation patterns and quantity resulting from climate change will impact the transport distances of aerosols. Depending upon the future evolution of emissions and climate change, changes in O_3 and aerosol lifetimes may be large enough to cancel or override increases in export from source regions.

5.9 Summary and recommendations – the road forward

5.9.1 Current status

The published studies summarized in this chapter (table 5.2) regarding O_3 transport on intercontinental scales all indicate that precursor emissions on one continent contribute to surface O_3 enhancements over other continents. Estimates of these enhancements, however, span a large range, up to an order of magnitude. Intercontinental transport of aerosols has been mainly demonstrated by measurement-based approaches, focusing on episodic transport of dust and biomass burning plumes. Little effort has been devoted thus far to evaluate aerosol S/R relationships on the intercontinental scale.

Due to the use of different models, methods, metrics, and definitions of regional boundaries, it is difficult to draw coherent conclusions from the model- and measurement-based studies in the literature. The preliminary results of the HTAP intercomparison begin to give a coherent picture of S/R relationships for O_3, aerosols and deposition, and are summarized below. Twenty-six (as present in the HTAP database on 1 August 2007) global and hemispheric models have performed up to 16 simulations where pollutant emissions were decreased by 20 per cent relative to a year 2001 reference simulation. The choice of 20 per cent emission reductions was a compromise between the near-linear responses of models due to emission change, and the larger (both positive and negative) anticipated changes in emission expected in these regions in the coming years. We note here that a 20 per cent reduction implies different amounts per component and per region.

Preliminary results of the S/R relationships estimated with the HTAP intercomparison Experiment Set 1 are summarized below.

(a) *Ozone.* Anthropogenic emissions of NO_x, CO and NMVOCs were perturbed individually and combined (along with aerosols) by 20 per cent in four Northern Hemispheric regions (North America, Europe, East Asia, and South Asia). The models also evaluated the effect of a 20 per cent reduction of CH_4 abundances. Results indicate that:

- The annual mean surface O_3 import sensitivity varies from 30–70 per cent, where import sensitivity is the response to the sum of 20 per cent anthropogenic emission reductions in three foreign regions divided by the response to a 20 per cent anthropogenic emission reduction in the region itself.
- The annual average S/R relationship may mask large seasonal variations.
- Intercontinental influences on O_3 are generally strongest in the Northern Hemisphere spring season, but surface O_3 is still sensitive to foreign influences (10–30 per cent import sensitivity) in the (three-month) high O_3 season.
- NO_x emission perturbations generally exert a stronger influence on surface O_3 than NMVOCs and CO, except in Europe where NMVOCs emission perturbations are equally

important, implying that the European O_3 production regime is more NO_x-saturated than in the other regions.

- Sources in North America and Europe tend to contribute as much to surface O_3 over East Asia and South Asia as the Asian sources contribute to each other.
- A reduction of global CH_4 abundance by 20 per cent would lead to a surface O_3 decrease of approximately 1.2 ppbv; note that such a reduction implies a 20–30 per cent decrease of global anthropogenic CH_4 emissions.
- Hemispheric emissions may alter the frequency of O_3 pollution events defined relative to a threshold concentration.
- The sensitivity for O_3 column changes due to foreign emission perturbations is stronger and different from those for O_3 surface concentrations.
- Depending upon the future evolution of emissions and climate change, changes in O_3 and aerosol lifetimes may be large enough to offset increases in export from source regions.

(b) *Aerosol.* Anthropogenic aerosol precursor (SO_2, NO_x and NH_3) and primary (elemental carbon (EC), POM and $PM_{2.5}$) emissions were perturbed by 20 per cent in four source regions. The results have been analysed for surface PM concentrations, deposition, and column loadings. Results indicate that:

- The import sensitivity for annual mean surface anthropogenic $PM_{2.5}$ varies from 5 per cent to 10 per cent in the European, North American and East Asian regions, and 25 per cent for South Asia.
- The import sensitivity of total column aerosol load is significantly larger than that for the surface concentrations. Regional column load import sensitivities for sulphate aerosol are 31 to 59 per cent, and for BC are 13 to 30 per cent. These have significant, but not yet quantified, implications for regional aerosol radiative forcing and climate change.
- The import sensitivities of aerosol are similar in the European, North American and East Asian regions, but are much larger for the South Asian region. The large sensitivities for the South Asian region can be explained in part by several peculiarities of the HTAP experiment set-up, and these require further study.
- The import sensitivity of aerosol deposition varies from 10 to 30 per cent in the case of sulphate, and 1 to 13 per cent for carbonaceous aerosols.
- The impact of one region on another depends on the modelled aerosol lifetimes, which vary significantly (a factor of four) between models. In general, models with longer lifetimes predict larger import sensitivities. The diversity in modelled lifetimes reflects differences in the process-level model formulations, which require further evaluation and analysis.

(c) *Nitrogen deposition.* Results were analysed for the effect of a decrease of anthropogenic NO_x emissions on total reactive nitrogen deposition (including aerosol nitrate and gaseous forms) in the four source regions. It was found that:

- Regions export 20–25 per cent of their anthropogenic NO_x emissions to the global troposphere, and 2–8 per cent of these emissions are deposited in the other HTAP regions.
- The import sensitivity of nitrogen deposition is between 3 and 12 per cent.
- The contributions of hemispheric transport of pollutants to nitrogen deposition are similar to those calculated for carbonaceous aerosol deposition. In addition, the largest imported contribution for reactive nitrogen deposition is estimated for South Asia.

Many modelling results in the HTAP intercomparison database have not been explored or explored only superficially. Small groups of authors could further explore this rich set of results and

funding agencies should support such further analysis of the results. Priorities for further analysis based on Experiment Set 1:

- Further analysis of the O_3 budgets, emission and deposition budgets, and aerosol optical parameters.
- Analysis of results for the Arctic based on the results of Experiment Set 1, along with tracer results from TP1.
- Extend the analysis of the seasonality of impacts and include more tracers in the analysis.
- Assess the scalability of the 20 per cent emission reductions to perturbations of different magnitudes (particularly for O_3).
- Examine the impact of the large variation across models in both anthropogenic and biogenic NMVOCs emissions on S/R relationships for O_3.
- Evaluate the ability of models to resolve observed frequency distributions of surface O_3 and the contribution of intercontinental transport under clean versus polluted conditions.
- Identify the relative role of different NO_y component species in long-range transport, as well as contributions from NH_3 and NH_4.
- Include additional models in the aerosol analysis to reduce uncertainty in the S/R relationships thus far estimated with a limited number of models that exhibit a wide range of results.

5.9.2 Priorities for future model studies

(a) Coordinated studies to identify processes contributing most to uncertainties in S/R relationships

As indicated in previous sections, there are poorly quantified uncertainties in the ability of coarse resolution global and hemispheric scale models to describe intercontinental transport. Of particular concern are the small-scale processes such as convection and boundary layer mixing that govern continental outflow and inflow of pollutants. Uncertainty in our understanding (and subsequent implementation in current models) of O_3 chemistry, particularly regarding the role of hydrocarbon emission, oxidation efficiency and the scavenging of soluble precursors and aerosol, requires evaluation.

In HTAP intercomparison Experiment Set 2, idealized tracer studies are used to assess several model sensitivities associated specifically with the physical parameterizations. Likewise in the context of AeroCom, and the new joint IGBP-IGAC/WCRP-SPARC initiative in Atmospheric Chemistry and Climate (Ravishankara, 2006), experiments are planned that further assess the processes that lead to variability among models.

As was indicated in section 5.8.2, climate change can influence transport processes in various ways. However, a consistent quantification is lacking. Therefore in HTAP intercomparison Experiment Set 3, dedicated studies on the role of climate change in hemispheric transport of air pollution are foreseen.

A link between the HTAP intercomparison and regional-scale model intercomparisons may further assess the uncertainties associated with calculation of import-export budgets of O_3, aerosol, and their precursors. When using boundary conditions from global models for regional experiments, special attention should be given to inconsistencies in boundary conditions, e.g. emission inventories.

As was the case in previous studies, there are no generally accepted “benchmark” tests that can be used for model evaluation. The HTAP intercomparison should strive to establish some tests and datasets that can be used to establish a model baseline against which future improvements can be demonstrated. Collaboration with the measurement community is needed to choose benchmark datasets.

Credibility and improvement of the reliability of the estimates of S/R relationships rely on comparison with atmospheric measurements. One focus of the HTAP intercomparison Experiment Set 3 will be to evaluate the representation of pollutant export and chemical evolution in models by comparison with observations from the TRACE-P and ICARTT field campaigns in 2001 and 2004.

With regard to aerosols, substantial evaluation of aerosol optical properties (AOD) has been performed during AeroCom, but less attention has been paid to surface aerosol, including the role of periodic aerosol transport. For instance, an evaluation of surface $PM_{2.5}$ concentrations of global models is currently not available. A further critical issue for aerosol is large model discrepancies in the vertical distributions. With limited observational constraints available to date, CALIPSO satellite data should be explored as a potential observational constraint. In the framework of hemispheric transport of air pollution, the AeroCom community is performing dedicated experiments to further explore S/R relationships.

More generally, the Group on Earth Observations (GEO), in its 2007–2009 workplan, is advocating for a stable and improved in situ and space-based observing system of global air quality in line with the Integrated Global Atmospheric Composition Observations (IGACO) recommendations. GEO also supports the efforts of WMO to increase spatial and temporal resolution of observations. A priority action for the GEO community is to evaluate and recommend strategies for an integrated sampling frame for air pollution. It is also noteworthy in this context that the GEO workplan calls for member countries and participating organizations to coordinate the construction of a high spatial and temporal resolution monitoring and forecasting system including atmospheric, terrestrial and oceanic observations, modelling and chemical data assimilation for global and local air quality. In addition, and of relevance to aerosol measurements in particular, GEO supports the development of international systems for both sand and dust-storm warning and biomass-burning monitoring.

(b) Alternative methods of S/R assessment

Adjoint models are currently under development in regional and global CTMs, and provide an interesting alternative to the forward model sensitivity simulations that are heavily relied upon at present to estimate S/R relationships. Adjoint models can estimate sensitivities within a model to specific chemical and physical processes, helping to identify the most important processes controlling S/R relationships. However, the adjoint model results remain model dependent, and there needs to be intercomparison between adjoint models and other methods.

A further development is chemical data assimilation in numerical weather prediction models, where analysis of specific episodes of intercontinental transport of NO_x, CO, and aerosols may be combined with surface-based measurements to better quantify the magnitude and efficiency of O_3 and aerosol production and transport. Future assessments of S/R relationships may benefit from chemical data assimilation in models.

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6. INITIAL ANSWERS TO POLICY-RELEVANT SCIENCE QUESTIONS

In 2005 at its first meeting, the Task Force on Hemispheric Transport of Air Pollution identified seven policy-relevant science questions (see chapter 1, box 1) to guide its work. This interim report is a first attempt to address these questions as they relate to tropospheric ozone, fine particles and their precursors. Below we summarize information presented in the preceding chapters, organized as initial answers to the guiding questions.

1. How does the intercontinental or hemispheric transport of air pollutants affect air pollution concentration or deposition levels in the Northern Hemisphere for ozone and its precursors, fine particles and their precursors, and compounds that contribute to acidification and eutrophication?

Observations from the ground, aircraft and satellites provide a wealth of evidence that ozone and fine particle concentrations in the UNECE region and throughout the Northern Hemisphere are influenced by intercontinental and hemispheric transport of pollutants. In some instances, the impacts of intercontinental transport are profound. Observations from mountaintop sites or from aircraft can track distinct plumes of polluted air with elevated levels of O_3, PM, CO and other trace components of specific sources, such as Hg from coal combustion. These plumes are often confined in altitude and can be traced backward using chemical transport models to specific source regions. Also satellite-borne instruments provide direct observations of the general patterns of O_3 transport, NO_x emissions and direct observations of the intercontinental transport of dust and CO. Ground- and space-based lidars are providing new information on the vertical structure of aerosol transport events and the meteorological conditions that drive them. Striking observations of intercontinental transport have been documented associated with forest fires in Siberia and North America and dust storms carrying PM from the Asian deserts over the Pacific Ocean and from the North African desert across the Atlantic and across the Mediterranean Sea.

While there is clear observational evidence of intercontinental transport on an episodic basis and at high altitudes, intercontinental transport also impacts surface air quality and background air pollution concentrations. These impacts, however, are harder to discern. Long time series of tropospheric O_3 indicate an increase of background O_3 in many places in the Northern Hemisphere. At the same time, the emissions of O_3 precursors and peak O_3 pollution levels have been reduced in North America and Europe. The observed increase of background O_3 is consistent with the more general increase of O_3 precursor emissions across the Northern Hemisphere. For PM, large inter-annual variability in dust transport tends to obscure long-term trends. The few available trends from isolated islands for nitrate and anthropogenic sulphate are generally consistent with estimated trends in emissions.

The processes that determine the overall patterns of transport at this scale are relatively well understood and our ability to quantify the magnitude of transport is improving. Our improved understanding comes from an increasing body of observational evidence, including new information from intensive field campaigns and satellite-borne instruments, improved emissions inventories and improved global and regional chemical transport models.

Together, models, observations and emissions inventories suggest that intercontinental and hemispheric transport are determined by the location and timing of emissions, the chemical and physical transformation of pollutants in the atmosphere and the prevailing patterns of meteorology and atmospheric circulation. The dominant meteorological processes that drive transport differ by geographic region and season.

In the mid-latitudes, prevailing winds are from the west to east creating stronger west-east flows compared to north-south flows. Wind speeds increase with height, so processes that lift pollutants out of the boundary layer near the ground and into the free troposphere increase the

potential for rapid intercontinental transport. Two mechanisms for such lifting are cyclonic weather systems and deep convection, which are primarily responsible for the transport of pollution from North America to Europe and from Asia to North America. In mid-latitude cyclonic systems, lifting takes place in the air stream known as the Warm Conveyor Belt (WCB), which can loft pollutants into the middle and upper troposphere, transporting them from North America to Europe in two to four days and from Asia to North America with slightly longer transport times. Cyclonic systems, which track from west to east, occur throughout the year although they are weakest in the summer. The formation of WCBs within cyclonic systems is most common along the east coasts of North America and Asia, where there are also heavy concentrations of emissions. Deep convection, which is strongest in the summer over the middle of North America and Asia, is also an important mechanism for lofting pollutants into the free troposphere. In both WCBs and deep convection, as the air rises and cools, water vapour condenses and wet scavenging removes many water soluble pollutants. The pollutants that are transported out of the boundary layer are subject to chemical reactions, physical transformations and mixing with stratospheric air as they are transported downwind. Eventually, the lofted pollutants are mixed back down to the surface in areas of atmospheric subsidence, such as over western North America and Europe, particularly over the Mediterranean. The subsidence process tends to disperse the pollutants and dilute significantly the pollutant concentrations reaching the surface. Subsidence can also have important effects on the chemical and physical transformations of the transported pollutants, such as when transported peroxyacetyl nitrate (PAN) decomposes as it subsides, contributing to the formation of O_3.

Topography can also have a strong impact on the transport of pollutants into the free troposphere, such as when winds associated with mountains and valleys carry pollution out of the boundary layer. This mechanism may play an important role in the flow of pollution from Europe to Asia. Topography may also be important in the entrainment of dust from high elevation deserts, such as the Gobi.

Long-range pollutant transport may also occur near the ground without significant lifting. Such transport may occur in association with passing weather fronts or may occur in a stable atmospheric layer, such as a residual layer that forms after the collapse of a daytime boundary layer. This sort of transport dominates the flow of pollutants out of Europe and into the Arctic, in winter, and across the Mediterranean, in summer. Such processes may also play an important role in the transport of Saharan dust across the Mediterranean to Europe and the transport of North American emissions over the Atlantic Ocean.

In the Arctic region, there are few anthropogenic emissions sources and transport into and out of the region are dominated by low-level flows near the ground. The lack of vertical transport is mainly due to the lack of strong convention and the low input of solar radiation, which also has important implications for the chemical and physical transformation of pollutants. Recent studies have suggested that anthropogenic pollution (both black carbon and O_3) transported to the Arctic have significant effects on atmospheric warming in the region, which further enhances the potential for transport from the mid-latitudes.

Transport in the tropics is less well understood than transport in the mid-latitudes. In the tropics, prevailing winds are generally from the east to the west. The deep convective clouds of the shifting inter-tropical convergence zone (ITCZ) lift air pollution into the free troposphere, with subsidence occurring elsewhere in the tropics. The Asian monsoon circulation can have important impacts on the transport of pollutants from South Asia, carrying pollutants westward from India to Northern Africa and Europe. The majority of biomass burning globally occurs in the tropics with activity peaking in different regions at different times of the year. The exchange of air and the pollutants it carries between the tropics and the mid-latitudes is limited, such that air in the mid-latitudes tends to recirculate in the mid-latitudes and air in the tropics tends to recirculate in the tropics.

2. More specifically, for each region in the Northern Hemisphere, can we define source-receptor relationships and the influence of intercontinental transport on the exceedance of established standards or policy objectives for the pollutants of interest?

Long-range transport affects the concentrations of ozone and aerosols at downwind receptor locations, but the effects of transport on these two species are distinctly different. In situ observations document episodic elevated concentrations due to transport of both O_3 and aerosols, but the latter are much more pronounced. Analyses of long-term observations also generally find that the background O_3 concentration in marine air at the west coasts of Europe and North America has increased over the past two decades.

It is clear that O_3 is, in effect, primarily a hemispheric pollutant with a rising, hemisphere-wide background. The influence of one continent on another is small, but is significant from a management perspective when compared to levels of O_3 decrease that can be achieved through local and regional emission controls. The benefits of measures to decrease intercontinental transport of O_3 would be distributed across the hemisphere.

For aerosols, spectacular enhancement events, especially those associated with major emission events such as fires or dust storms, have been observed at the surface. However, very clean background concentrations can still be observed throughout the globe. The episodic enhancement events do yield an average elevation of the total aerosol column over the long term. The intercontinental transport of both O_3 and fine particles has substantial impacts on total atmospheric column loadings, which have significant implications for climate change.

Despite observations of the effects of long-range transport, source-receptor (S/R) relationships, defined as the sensitivity of concentrations or deposition at a "receptor" location due to a change in emissions at a "source" location, can only be determined from models. Previously published modelling studies for O_3 have estimated annual or seasonal mean influences of one continent on the other in the range of 1 to 5 ppbv, with enhancements due to individual transport episodes around 10 ppbv. For fine particles, previous modelling studies suggest large fractions of aerosols, 40 to 80 per cent, are exported from source regions through intercontinental transport. The impact of this aerosol export on long-term average surface concentrations or deposition in other regions has not been extensively examined. Reported estimates of annual or seasonal intercontinental influences on total aerosol column loadings have ranged from 1 per cent to 50 per cent, depending on the region, season and chemical fraction of interest. However, it is difficult to directly compare previously published studies because of differences in the methodologies, definitions of regions and metrics used.

To provide comparable results, examine and improve the ability of models to represent transport processes, and estimate uncertainty in current estimates of S/R relationships, the HTAP intercomparison was organized to conduct a series of coordinated model experiments. In the first set of experiments, emissions of NO_x, CO, and NMVOCs were perturbed individually and combined by 20 per cent in four Northern Hemispheric regions (North America, Europe, East Asia, and South Asia). The HTAP intercomparison also examined the effects of a 20 per cent reduction of global methane concentrations. Similar 20 per cent perturbation analyses were conducted for emissions of aerosol precursors (SO_2, NO_x, and NH_3) and primary aerosol (elemental carbon (EC), POM and $PM_{2.5}$).

For each of the four regions and each pollutant, the relative sensitivity to imported pollution versus domestic emissions, or "import sensitivity", was calculated as the change in concentration (or column loading or deposition) within a receptor region to the combined influence of the 20 per cent emission reduction in the three other source regions divided by the change in concentration due to the 20 per cent emission reduction in the region itself. The preliminary results for O_3, fine particles and nitrogen deposition are summarized below.

For the 20 models contributing to the O_3 experiments, the seasonal cycle in surface O_3 concentrations is similar across the models, although the monthly mean values vary by as much as 10 to 30 ppbv in some regions and seasons. The mean results from the models suggest that a 20 per cent decrease in anthropogenic NO_x emissions will decrease annual mean surface O_3 concentrations within

the same region by 0.46 to 1.08 ppbv, depending on the region. Combined emission reductions in the three foreign source regions will decrease annual mean surface concentrations in the fourth receptor region by 0.24 to 0.34 ppbv, depending on the receptor region. Thus, the import sensitivity of annual mean surface O_3 varies between 30 per cent and 70 per cent, depending on the receptor region.

These annual average S/R relationships may mask large seasonal variations, which will require further analysis to identify. Intercontinental influences on O_3 are generally strongest in the Northern Hemisphere spring season, not when O_3 is highest. But surface O_3 is still sensitive to foreign influences (import sensitivity of 10 to 30 per cent) in the (3-month) high O_3 season. The initial results further suggest that changes in emissions on a hemispheric scale may change the frequency of O_3 pollution events defined relative to a threshold concentration.

The largest influence of a single source region on O_3 in another receptor region is associated with the impact of North American NO_x emissions on Europe. Changes in North American NO_x emissions account for roughly 20 per cent of the response of European summertime surface O_3 to changes in European NO_x emissions. Sources in North America and Europe tend to contribute as much to surface O_3 over East Asia and South Asia as the Asian sources contribute to each other.

The initial results suggest that hemispheric response to NO_x emission perturbations generally outweighs that from CO or NMVOCs. The modelling experiments, however, neglect a long-term feedback of these emission perturbations on O_3 that occurs through changes in the hydroxyl radical and methane. The net result of neglecting this long-term feedback is that these results overestimate the impact of NO_x reductions by 5 to 15 per cent and underestimate the impact of CO reductions by 15 to 20 per cent. Initial results suggest that the combined effect of NO_x, NMVOCs, and CO perturbations may be slightly less than sum of their individual effects, however more analysis is needed to assess the linearity of the reductions.

The initial results of the 20 per cent decrease in global methane concentrations, which implies a 20 to 30 per cent decrease in global anthropogenic emissions of methane, suggest an annual mean surface O_3 decrease of approximately 1 ppbv across all of the regions. Such a decrease in the global methane emissions may have as large an effect on intercontinental transport of O_3 as 20 per cent decreases of other O_3 precursors, as well as decreasing the climate forcing of both methane and O_3.

Substantial O_3 transport takes place above the boundary layer in the free troposphere. The sensitivity of the O_3 column to changes in foreign emissions is stronger and different from those for O_3 surface concentrations. Over Europe, the tropospheric O_3 column response to NO_x perturbations from North America is as large as the response to domestic emission perturbations. North American column ozone is less strongly impacted by foreign NO_x emission perturbations, with an import sensitivity of roughly 60 per cent that of the domestic sensitivity. In East Asia, the impact of European NO_x emissions is particularly strong, and the import sensitivity of East Asian O_3 columns is larger than the domestic sensitivity by approximately 50 per cent. Finally, over South Asia, the sensitivity of the ozone column to domestic emission perturbations is larger than the sensitivity to perturbations in foreign NO_x emissions, with about equal sensitivity to emission changes in the other three regions.

For fine particles, results from 8 to 11 models, depending on the chemical component of interest, are available from the HTAP intercomparison. The mean results suggest that a 20 per cent decrease in anthropogenic gas and aerosol emissions will decrease annual mean surface PM concentrations within the same region by 0.35 to 0.74 $\mu g/m^3$, depending on the region. Combined emission reductions in the three foreign source regions will decrease annual mean surface concentrations in the fourth receptor region by 0.02 to 0.1 $\mu g/m^3$, depending on the receptor region. Thus, the import sensitivity of annual mean surface PM concentrations varies between 4 per cent and 18 per cent.

The import sensitivity of annual deposition of sulphate, reactive nitrogen, and carbonaceous aerosols are similar to those for surface concentrations. The import sensitivity of annual mean deposition varies from 10 to 30 per cent for sulphate to 1 to 13 per cent for carbonaceous aerosols and 3 to 15 per cent for total reactive nitrogen.

The contribution to the aerosol column loading is significantly larger than that for the surface concentrations or deposition. The import sensitivity of the regional sulphate annual mean aerosol column loadings is 30 to 59 per cent. The import sensitivity of POM annual mean column loadings is somewhat less, 13 to 30 per cent. The impacts on total column loadings have significant, but not yet quantified, implications for regional aerosol radiative forcing and climate change. The contributions of imported aerosol are similar in Europe, North America, and East Asia, but are much larger for the South Asia region. The large response due to imported aerosol for the South Asia region can be explained in part by several peculiarities of the HTAP experiment design, and these require further study.

The impact of one region on another depends on the modelled aerosol lifetimes, which vary significantly (a factor of four) between models. In general, models with longer lifetimes predict larger import sensitivities. The diversity in modelled lifetimes reflects differences in the process-level model formulations, which require further evaluation and analysis.

The HTAP intercomparison has provided the first set of comparable estimates of intercontinental S/R relationships from multiple models. The initial analyses of results have focused on annual and seasonal means. Additional analyses will be conducted to explore the impacts on other metrics more closely related to existing ambient standards and environmental policy objectives. Further experiments and additional analysis will explore the differences between the models, as well as potential biases introduced by the design of the experiments. This continuing effort will enable us to assess, and ultimately reduce, the variability and uncertainty in model estimates of intercontinental S/R relationships in terms of impacts on the achievement of environmental policy objectives.

3. How confident are we of our ability to predict these S/R relationships? What is our best estimate of the quantitative uncertainty in our estimates of current source contributions or our predictions of the impacts of future emissions changes?

Current models can reproduce much of the observed spatial and seasonal patterns of intercontinental transport. The ability of models to adequately describe continental outflow of pollution and individual transport events has been demonstrated through comparisons with aircraft and surface observations. However, current models, with existing emissions inventories, have not been able to reproduce some of the trends derived from the relatively sparse observations available from remote locations. The average of an ensemble of models, as used in this work, has been shown in previous studies to have greater predictive skill than the individual models in the ensemble, though low diversity in the predictions does not necessarily warrant that the predictions are realistic. To ensure confidence in the ability to predict S/R relationships, model predictions must continue to be evaluated with observational data and compared with other models.

To continue to enhance the ability of models to estimate quantitative S/R relationships, both models and emission inventories must be improved. Current models need better representation of some physical and chemical processes in the atmosphere that are important for assessing intercontinental transport, and more robust emissions inventories must be developed for many important source categories and regions of the world.

Direct comparisons with observations are one key to understanding and assessing the accuracy of model predictions. Both short-term and long-term data sets are valuable for evaluating model representations of transport events and trends in background concentrations. A number of recent intensive field studies have produced rich data sets for short periods of time (days to weeks) that can be used to evaluate model representations of specific processes or events. Future efforts under the HTAP intercomparison will focus on comparisons with the observations from several such field studies. In addition to intensive field studies, ongoing long-term monitoring programmes provide data that are useful for evaluating models, although the data are sparse in some key regions. Analyses of such data have derived trends in the long-range transport of O_3 and aerosols such as dust and anthropogenic nitrate and sulphate. While comparison of model predictions is generally straightforward, there is no simple means of directly evaluating S/R relationships, as they are model constructs. Therefore, a suite of short-term and long-term measurements and model results must be

carefully assessed to understand S/R relationships, as is planned as part of the HTAP intercomparison.

In estimating the effects of future emissions changes, the predictions used for future pollution distributions, trends and composition contribute large uncertainties to the estimates. To reduce these uncertainties, more efforts are needed to better characterize current emissions and understand potential future emissions scenarios. Better characterization of emissions should include incorporating local knowledge in regions where emission factors and activity data are poorly known and characterizing emissions from sources that are of special importance with respect to intercontinental transport, such as shipping, aviation, lightning and other natural sources. Additionally, confidence in predictions of effects of future emissions can be improved through an iterative process to compare absolute values, ratios and trends of emissions estimates with values, ratios and trends derived from both ambient observations (surface, in situ and satellite-based) and atmospheric models.

4. For each country in the Northern Hemisphere, how will changes in emissions in each of the other countries in the Northern Hemisphere change pollutant concentrations or deposition levels and the exceedance of established standards or policy objectives for the pollutants of interest?

The initial results of the HTAP intercomparison presented above describe the sensitivity of annual and seasonal mean pollutant concentrations, column loadings and deposition to changes in emissions over large regions. These estimates will be further refined in future phases of the HTAP intercomparison, but calculating S/R relationships on the geographic scale of individual countries has not yet been attempted.

The impact of intercontinental transport on exceedance of established standards or policy objectives has not yet been explored in detail. However, initial results for O_3 suggest that the models will provide useful information about exceedance of threshold values. The annual mean results, however, show that intercontinental transport may have much greater impacts on total column loadings than on surface air quality or deposition values associated with existing standards or policy objectives.

5. How will these S/R relationships change due to expected changes in emissions over the next 20 to 50 years?

Profound changes in intercontinental transport are likely due to changes in the magnitude and spatial distribution of anthropogenic emissions caused by the continuing implementation of pollution control measures, regional differences in the pace of economic development, the growth in shipping and aviation emissions, and the implementation of climate change mitigation measures. One future projection to 2030 that has assumed compliance with existing legislation suggests that NO_x and SO_2 emissions will continue to decline in OECD countries, but will continue to increase in Asia.

The most important factors determining future anthropogenic emission levels are activity, level of technology development and penetration of abatement measures. Activity changes are strongly linked to economic growth, population growth and energy growth, but they are also dependent on the geopolitical situation, trade agreements, level of subsidies, labor costs, etc.

Natural emissions may also change due to changes in climate and land use. Emissions of mineral dust, wetland methane, foliar VOCs and wildfires are all very sensitive to changes in landcover (e.g. vegetation type and density) and soil moisture. Natural emissions could vary by a factor of two or more on time scales of years to decades due to the intrinsic variability. These natural emissions influence O_3 and aerosol concentrations and interact with anthropogenic emissions. Hence, the future effects of natural emissions on hemispheric transport can vary and also influence the transport of anthropogenic emissions.

6. How will these S/R relationships be affected by changes in climate or climate variability?

The intercontinental and hemispheric transport of O_3 and fine particles has important effects on global and regional climate change. Tropospheric O_3 and aerosols have been shown to modify both the radiative balance of the atmosphere, as well as cloud formation and precipitation. However, there is still a large degree of uncertainty as to the magnitude of these effects.

In the Arctic, transported black carbon is a minor, but important, component of the Arctic Haze and causes heating of the haze layers. Moreover, deposition of black carbon onto snow and ice results in a reduction of the albedo, which may create a climate forcing that is significant compared to the effect of greenhouse gases.

Likewise, future changes in temperature, humidity, precipitation, the strength of convective systems, and the paths and frequency of cyclonic systems are likely to change emissions from natural sources as well as the atmospheric lifetimes and intercontinental flows of anthropogenic pollutants. With increasing greenhouse gas concentrations, most climate models predict less convective transport in the tropics and enhancement of current patterns of precipitation and evaporation (i.e. dry locations will get dryer and wet locations will get wetter). In previous studies, a number of models have predicted increases in water vapour leading to increased O_3 destruction and shorter O_3 lifetimes. However, some models have shown also increased O_3 production in high emission areas, enhanced lightning NO_x emissions, and increased stratosphere-troposphere exchange. Some climate models have shown changes in the frequency, intensity and paths of mid-latitude cyclones. These changes suggest a possible decrease in transport associated with mid-latitude storm systems and a possible increase in transport into the Arctic.

The combined effects of emissions changes and climate change on the absolute and relative impacts of intercontinental transport on air quality and deposition is unclear.

7. What efforts need to be undertaken to develop an integrated system of observational data sources and predictive models that address the questions above and leverages the best attributes of all components?

The assessment of the current observational evidence (chapter 3), emission inventories (chapter 4) and chemical transport modelling (chapter 5) indicates that these three aspects of understanding hemispheric transport are interdependent as shown schematically in figure 6.1.

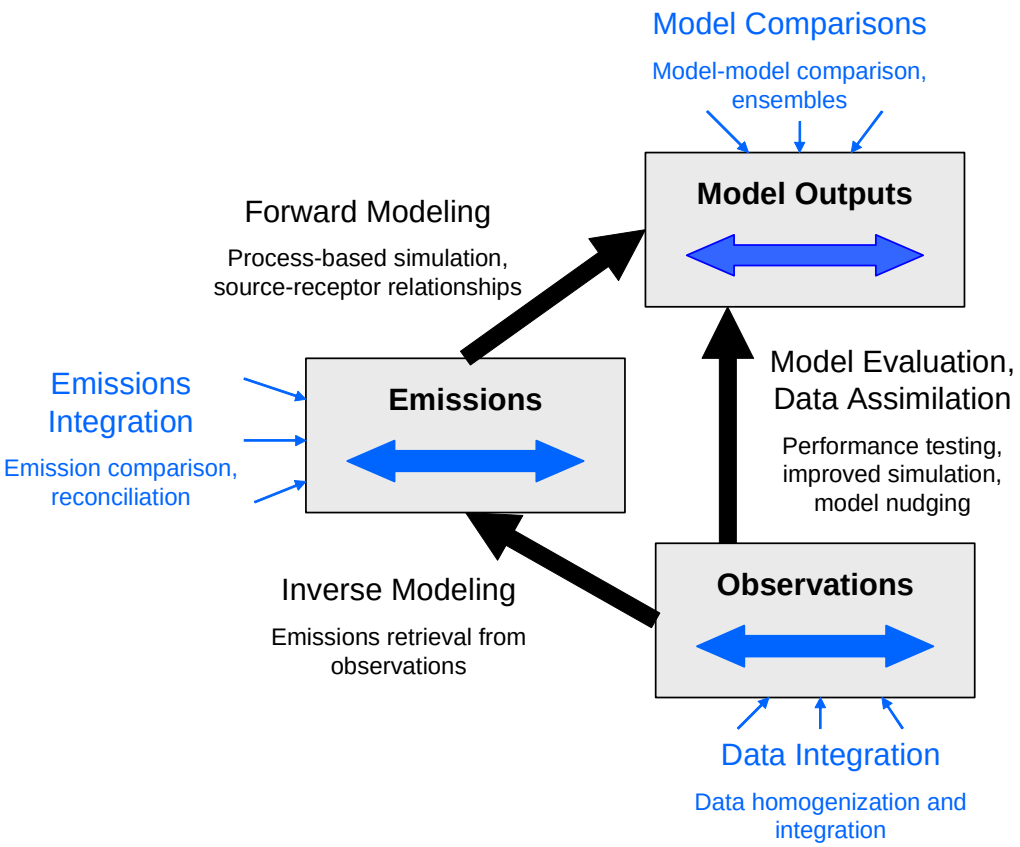

Figure 6.1. Interdependence of emissions, models and observations.

The boxes represent information resources from observations, emission inventories, and modelling, whereas arrows indicate connections and operations within and between these aspects. Operations within aspects or areas of activity (in blue) include intercomparisons, reconciliation and homogenization of data. The interconnecting arrows (in black) represent the major connections and operations that link observations, emission inventories and modelling. Observations are an essential foundation, providing the data for model evaluation as well as for data assimilation into the models. Through inverse modelling, observations also allow estimation of emissions.

Improving our assessment of intercontinental and hemispheric transport will require an integrated approach where the best available knowledge from observations, emissions and models is combined. Furthermore, reconciliation of the three areas or aspects (observations, emission inventories, models) requires considerable iteration until the deviations are minimized.

The following sections identify what further efforts are needed in the various relevant areas of work.

Improving the modelling of transport processes using existing and new field campaign data

There are a number of chemical and physical processes that could have important effects on intercontinental transport that are poorly represented in current models. These include boundary layer mixing and small-scale venting processes that allow pollutants to leave the boundary layer, such as dry convection, local circulations (valley or land-ocean) and gravity wave mixing. Likewise, subsidence and other processes that transport pollutants from the free troposphere back to the boundary layer are not well quantified in large-scale models or in observations.

The transport of species out of the boundary layer is often associated with wet deposition as water vapour is condensed during the transport events. The extent to which soluble species are wet deposited depends on details of the microphysics, i.e. to what extent (a) convective clouds are able to transport soluble species to the upper troposphere; (b) soluble species are retained as liquid water

freezes; and (c) rain is evaporated as it falls through the atmosphere. Again, these processes are not suitably modelled on the large scale and have not been adequately measured.

Finally, transport processes in the tropics and subtropics have received far less attention than in the mid-latitudes. With the growth of future emissions expected in the tropics and subtropics, these processes may gain greater importance for intercontinental transport of air pollution.

Improving our ability to model these various processes requires careful comparison of models to the rich observational data provided by intensive field campaigns. Much information can be mined from the existing integrated data sets collected by past field campaigns. One of the foci of the HTAP intercomparison Experiment Set 3 will be to evaluate the representation of pollutant export and chemical evolution in models by comparing with observations from the TRACE-P and ICARTT field campaigns in 2001 and 2004. It may also be necessary to design new field campaigns specifically to explain key processes and evaluate model parameterizations. Several new intensive field campaigns are being conducted in 2007 and 2008 as part of the POLARCAT component of the International Polar Year. For the full value of the observations collected in past and future field campaigns to be realized, further efforts must be made to facilitate access to the integrated data sets.

Improving emissions inventories using local information and inverse modelling

Major uncertainties exist in the emission inventories for many regions of the world and some important source categories, such as shipping, aviation and lightning. To improve the global emissions inventories used in the assessment of intercontinental transport, existing emissions information from national and subnational governments, and other research activities focused on specific source categories, need to be incorporated into the global inventories. Such efforts are especially needed to improve the inventories in regions where emission factors and activity data are poorly known.

Models can be used to identify those emission estimates and uncertainties that are most important for understanding intercontinental transport and hemispheric pollution as well as the temporal and spatial resolution of emission estimates needed to understand intercontinental transport. To improve confidence in emission estimates and identify potential weaknesses, comparisons of the absolute values, pollutant ratios and trends of emission estimates contained in emissions inventories can be compared with emission estimates, pollutant ratios and trends derived through inverse modelling from ground-based, aircraft and satellite observations.

Identifying and explaining long-term trends by filling gaps in the observing system and improving model descriptions

Increasing trends of background pollution concentrations, particularly for O_3, have been observed at a number of remote locations across the Northern Hemisphere consistent with increases in emissions. However, these trends are not consistent with other available observations (such as data from O_3 sondes), and are not well reproduced by chemical transport models. To explain these observed trends, limitations of the current observational system and limitations in current models (and possibly emissions inventories) must be addressed.

Although some important regional data sets are available for aerosol composition and gas phase concentrations, and valuable long-term data sets are available for a few remote locations, there are many regions of the world that are poorly covered by current observational systems. Improved information about the vertical distribution of pollutants in the atmosphere is needed.

Over the coming decades significant changes are expected in the intercontinental transport of air pollutants due to changing emissions globally. The present observational system, however, is not sufficient to capture these changes. A surface measurement network suitable for following the evolution of long-range transport of air pollution in the Northern Hemisphere has to include monitoring in regions which are now poorly covered, such as over the seas and in the interior of the Asian continent. Both high elevation and remote island sites should be included. Surface networks must be complimented by vertical profiling (such as from sondes, lidar and aircraft) and measurements from satellite sensors to characterize the tropospheric pollution distribution. Coincident measurements of O_3, aerosol components and their precursors as well as atmospheric tracers such as

CO and Hg will strengthen the utility of the observations for model evaluation and assessments.

Unprecedented satellite-borne measurement capability is presently in orbit, but the utility of the measurements is limited by a clear understanding of the accuracy and precision of the retrieved results (i.e. satellite validation). Facilitating broader access to satellite data sets for analysis and comparison to other observations and model results, and appropriate collaboration between the satellite, modelling, and emissions communities, would increase the value of these measurements. Furthermore, new satellites will be required to maintain this current measurement capability into the future.

Further experiments under the HTAP intercomparison will generate insights into the differences between current models and sensitivities to model parameterizations. Likewise, in the context of AeroCom and the new joint IGBP-IGAC/WCRP-SPARC[1] initiative in atmospheric chemistry and climate, experiments are planned that will further assess the processes that lead to variability among models and examine the ability to reproduce observed trends.

Developing a robust understanding of current S/R relationships using multiple modelling techniques and analyses of observations

The initial results of the HTAP intercomparison provide some useful information about the significance of intercontinental transport from an air quality management perspective. However, further analyses are needed to develop a robust understanding of current S/R relationships. These include addressing issues of: (a) the scalability of the sensitivities; (b) the large variability across the models of the sensitivity of O_3 to NMVOCs emissions changes; (c) the ability to reproduce observed frequency distributions of pollution; (d) the contribution of intercontinental transport under clean versus polluted conditions; (e) the S/R relationships for other aerosol components; and (f) the influence of interannual meteorological variability.

A link between the HTAP intercomparison and ongoing regional-scale model intercomparisons may inform the assessment of the uncertainties associated with the calculation of import-export budgets of O_3, aerosols and their precursors.

There are no generally accepted “benchmark” tests that can be used for model evaluation at the intercontinental scale. Therefore, the HTAP intercomparison should strive to establish some standard tests and a well-documented reference set of observations drawn from different sources that can be used to establish a baseline of model performance against which future improvements can be demonstrated.

Alternative methods for calculating mass fluxes and sensitivities to emission changes, including direct calculations from observations, should be explored and compared with the results from global CTMs. Adjoint models are currently under development in regional and global CTMs, and provide an interesting alternative for the forward model sensitivity simulations. Adjoint models need to be compared to one another and to other methods for calculating sensitivities. Future assessments of S/R relationships may also benefit from the use of chemical data assimilation to improve the analysis of specific episodes.

Estimating future S/R relationships under changing emissions and climate

Future changes in the spatial distribution and magnitudes of emissions associated with future development and control, combined with changes in the atmospheric lifetimes of pollutants and transport processes associated with climate change, will alter intercontinental S/R relationships. Scenarios of future emissions and climate change should be assessed with the available modelling tools – the typical time horizon could be both an intermediate one aiming at 2020 and 2030 as well as the longer-time perspectives of 2050 and 2100. The development of such scenarios should be coordinated with efforts under IPCC and the IGBP-IGAC/WCRP-SPARC initiative in atmospheric chemistry and climate.

[1] IGBP – International Geopshere-Biosphere Programme (part of WMO); IGAC– International Global Atmospheric Chemistry; WCRP– World Climate Research Programme (part of WMO); SPARC– Stratospheeric Processes and Their Role in Climate (part of WMO).

Improving organizational relationships and information management infrastructures to facilitate necessary research and analysis

Recent developments in information technology, observational capabilities, data assimilation, and modelling and analysis tools, have made it possible to begin to combine the best available knowledge from observations, emissions and models to inform assessments of intercontinental transport. However, further organizational efforts and investments are needed to put the pieces of an integrated observations/modelling system into place. A strategy for Integrated Global Atmospheric Chemistry Observations (IGACO Theme Team, 2004) has been formulated and is being implemented at many different scales ranging from regional to hemispheric to global, contributing an atmospheric component of the Global Earth Observing System of Systems. Building upon the existing WMO Global Atmospheric Watch programme, the proposed IGACO system is intended to provide an observations, modelling and information infrastructure for atmospheric chemistry analogous to the global infrastructure currently in place for the meteorological community. In addition to the needs for additional observations and model improvements, the strategy also addresses issues of data quality assurance, data distribution and data archiving. Such an integrated infrastructure would greatly facilitate the assessment of intercontinental transport. Efforts organized by the Task Force should build upon and/or work to further, the implementation of this strategy.

Addressing the challenges outlined above and in the preceding chapters will require the combined effort of many individual scientists, research organizations and governmental authorities. In this combined effort, the Task Force can continue to play several important roles.

The Task Force can continue to serve as a forum to identify scientific consensus concerning the current understanding of intercontinental and hemispheric transport and priorities for future research and development through the organization of workshops, meetings and assessment writing activities.

By organizing cooperative research and analysis efforts such as the HTAP intercomparison and by fostering networks for data exchange and delivery, the Task Force can serve as a forum for information exchange and collaboration.

Finally, the Task Force can assist in raising the awareness of transboundary and intercontinental air pollution in regions where the concept is less well known and in linking this awareness to the need for building robust national and regional emission inventories and observational systems. The Task Force can assist in creating crucial links between institutions both within countries and across regional and hemispheric scales. Creating these linkages could be an important step in increasing the capacity to manage the sources of air pollution in these regions.

References

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Appendix A
Activities of the Task Force on Hemispheric Transport of Air Pollution

The UNECE Convention on Long-range Transboundary Air Pollution currently has a total of 51 Parties, covering almost all of Europe and North America and extending into Central Asia. Under the Convention, eight protocols have been developed which have all entered into force. One protocol addresses the long-term funding for air pollution monitoring and modelling activities. The other protocols set air pollution reduction targets that address sulphur, nitrogen oxides, ammonia, volatile organic compounds, mercury, lead, cadmium, and persistent organic pollutants (see http://www.unece.org/env/lrtap). The most recent protocol, the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, recognized the potential for intercontinental transport of air pollution and the need for its further study. The Executive Body for the Convention is considering the issue in the first review of the Protocol since its entry into force.

A.1 Formation of the Task Force

The Task Force on Hemispheric Transport of Air Pollution was established by decision of the Convention's Executive Body at its twenty-second session in December 2004. In its decision, the Executive Body charged the Task Force:

(a) To plan and conduct the technical work necessary to develop a fuller understanding of the hemispheric transport of air pollution for consideration in the reviews of protocols to the Convention;

(b) To plan and conduct the technical work necessary to estimate the hemispheric transport of specific air pollutants for the use in reviews of protocols to the Convention and prepare technical reviews thereon for submission to the Steering Body of EMEP[1].

The United States and the European Community were designated the lead Parties for the Task Force and they appointed two Co-Chairs, Dr. Terry Keating (U.S. Environmental Protection Agency) and Dr. André Zuber (European Commission, Directorate General Environment). The Co-Chairs were encouraged to invite participation of relevant experts from countries outside the Convention across the Northern Hemisphere. In April 2005, the Convention secretariat sent invitations to about 75 Governments with known interests in air pollution, as well as to a number of accredited international organizations, inviting them to nominate experts to take part in the Task Force's work.

The Task Force reports to the Steering Body of EMEP, the Convention's atmospheric monitoring and modelling working group, but the Task Force also collaborates with other relevant bodies of the Convention. It also has engaged with other international organizations of relevance such as the World Meteorological Organization (WMO), regional organizations such as the United Nations Environment Programme (UNEP) regional office in Asia, as well as with networks including the World Bank-funded programme Clean Air Initiative-Asia and the Acid Deposition Monitoring Network in East Asia (EANET).

[1] Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.

A.2 Task Force meetings

The first meeting of the Task Force, held from 1 to 3 June 2005 in Brussels, was hosted by the European Commission and attended by than 60 participants from 17 Parties to the Convention and 4 non-UNECE countries. The meeting's main objectives were to identify the scope of the hemispheric transport issue and to set out a plan for future work leading up to an assessment of intercontinental transport of air pollution. The Task Force selected as a target date for its assessment report. To guide its work, it adopted a number of key policy relevant science questions (see box 1, chapter 1). In addition, the Task Force proposed a workplan for 2006, later adopted by the Executive Body.

The second Task Force meeting, held from 6 to 8 June 2006 in Moscow, was co-hosted by the Russian Federation and EMEP Meteorological Synthesizing Centre-East (MSC-East). More than 70 participants from 17 Parties and 7 non-UNECE countries attended the meeting. The second meeting's main aims were to take stock of the existing information on intercontinental transport of Hg and POPs, the intercontinental transport of air pollution into the Arctic, and the role of methane as a precursor for tropospheric ozone. In addition, the Task Force prepared its second workplan for consideration by the Executive Body.

The third meeting of Task Force took place from 30 May to 1 June 2007 in Reading, United Kingdom. It was hosted by the United Kingdom; more than 60 participants from 17 Parties and 4 non-UNECE countries attended. The main focus of this meeting was to discuss and agree the Task Force's interim assessment report for the first review of the 1999 Gothenburg Protocol, which was to be concluded by the end of 2007. The Task Force also held initial discussions on the relationship between climate change and hemispheric air pollution and prepared a third workplan for submission to the Executive Body.

A.3 Scientific workshops

To address its policy-relevant science questions, the Task Force organized a series of specific scientific workshops. Summary reports and material presented at each of the workshops are available at www.htap.org.

The U.S. Environmental Protection Agency (EPA) hosted a workshop on the organization of a model intercomparison exercise (the HTAP model intercomparison) on 30 and 31 January 2006 in Washington, D.C.; more than 100 experts participated. The main goals were to organize the model intercomparison and evaluation exercise and to identify the key model experiments to be done by different models. The workshop included presentations of about 30 posters on recent research results related to the intercontinental transport of air pollution. Further details on the design and progress of the HTAP model intercomparison are presented below.

A workshop on emission inventories and projections for assessing intercontinental transport of air pollution was held from 18 to 20 October 2006 in Beijing. The event was hosted by Tsinghua University. About 80 experts took stock of what information was available on emission inventories that could be used for the study of intercontinental transport of air pollution and identified major gaps in knowledge.

A workshop on integrated observations was held from 24 to 26 January 2007 in Geneva. It was co-hosted by the Task Force and WMO in cooperation with the Group on Earth Observations (GEO) secretariat. Some 120 experts participated. The workshop

examined the current state of observations relevant to hemispheric transport studies for the Task Force assessments, as well as for WMO weather, climate and environmental prediction applications and for the societal benefit areas of GEO. In addition, the workshop identified gaps in observations for priority air pollutants and in data management and made recommendations on how to fill those gaps, taking into account ongoing efforts under regional networks such as the Global Atmosphere Watch (GAW) programme, Integrated Global Atmospheric Chemical Observations (IGACO) and the development of the Global Earth Observation System of Systems (GEOSS).

A workshop to discuss progress and plan future phases of the HTAP intercomparison took place in Jülich, Germany, from 17 to 19 October 2007. It was hosted by the Forschungszentrum Jülich (Research Centre Jülich).

A.4 Websites and listservers

The Task Force has made efforts to provide most of the information of its planned and ongoing work through its website (www.htap.org), which is supported by the U.S. EPA. All of the reports and presentations from the meetings and workshops, as well as the draft and final versions of the assessments reports, are publicly available on the site. A major objective of the open and transparent information is to facilitate the outreach activity of the Task Force. The websites of the lead Parties as well as the Convention's website have links to the HTAP website.

Additional public and private websites to support specific aspects of the work of the Task Force have also been established by other institutions, including the European Commission's Joint Research Centre, Ispra, the Research Centre Jülich, Washington University (St. Louis, Missouri, United States) and the Norwegian Institute for Air Research (NILU). These efforts facilitate collaboration among the experts contributing to the work of the Task Force; in the future, further efforts will be made to coordinate the evolving network of websites and information and communication portals.

The U.S. EPA has also established several electronic mail listservers for the Task Force to help distribute general information and announcements about Task Force activities and to facilitate communication among Task Force subgroups, including the HTAP model intercomparison participants and the authors of this report. Subscription information for these listservers can be found on the Task Force website.

A.5 Assessment reports 2007 and 2009

The main purpose of this, 2007 interim assessment, report is to inform the Parties to the Convention in preparation for their review of the 1999 Gothenburg Protocol. This report therefore focuses on those air pollutants relevant for tropospheric O_3, PM, acidification, and eutrophication. The Task Force plans to produce a more comprehensive assessment for 2009. The 2009 report will update the interim report in the light of new information from ongoing research, in particular the HTAP model intercomparison. In addition, the 2009 report will address other atmospheric pollutants of interest for the Convention, such as Hg and POPs.

A.6 HTAP model intercomparison

This report presents some of the initial results of the ongoing series of model evaluation and intercomparison experiments organized by the Task Force, beginning at its

January 2006 workshop, to:

- Produce estimates of intercontinental source-receptor (S/R) relationships;
- Improve our understanding of the variability and uncertainty in current model estimates;
- Guide future model developments to decrease uncertainties in S/R relationships.

The model intercomparison and evaluation is open to all interested participants and has so far attracted contributions from about 30 models. The European Commission's Joint Research Centre (http://aqm.jrc.it/HTAP/) has established a website to provide experts with more detailed information on the complete set of model experiments, including guidance for inputs and information on the requested outputs. In addition, the Research Centre Jülich has set up a data server and discussion wiki has been established by the Research Centre Jülich (http://icg-ii-wikis.icg.fz-juelich.de/HTAPWiki/).

Based on the January 2006 workshop, a series of four sets of experiments has been identified:

- Experiment Set 1: S/R emission sensitivity studies. The simulations consist of a reference simulation (relevant for 2001) and simulations reducing anthropogenic emissions by 20 per cent in Europe, North America, East Asia, and South Asia. Provisional results are presented in this report.
- Experiment Set 2: Process and tracer studies. These experiments will develop a simple set of diagnostics that can be used to understand the model differences that occurred under Experiment Set 1. The studies will try to isolate the relative roles of mixing/transport and chemical processes using analyses of carbon monoxide (CO), coloured tracers, radon (Rn^{222}) and non-methane volatile organic compounds (NMVOCs).
- Experiment Set 3: Detailed process studies. These experiments will explore specific process relevant for intercontinental transport. Potential topics include pulsed (nitrogen oxides) NO_x versus tagging of NO_x, Hg fate, aerosols (in conjunction with AeroCom), comparison of model results to campaigns (TRACE-P and ICARTT), the role climate change and variability, and vertical mixing of O_3.
- Experiment Set 4: Improved estimates of S/R relationships. Building on the previous sets of experiments, this set would generate improved estimates and uncertainty estimates for inclusion in the 2009 assessment report.

A.7 Support by Convention bodies

The Task Force is organized under the Steering Body of EMEP. Under EMEP, there are four centres, all of which provide support to the Task Force. MSC-West has now extended its modelling domain to include the entire Northern Hemisphere and actively participates in the HTAP model intercomparison for O_3 and PM. MSC-East participates in the HTAP model intercomparison for Hg and POPs and has organized discussions on these substances at Task Force meetings and workshops. The Chemical Coordinating Centre (CCC) has provided input to Task Force meetings, workshops and assessments related to the observational evidence of intercontinental and hemispheric transport. Likewise, the Centre for Integrated assessment Modelling (CIAM) has been active in activities related to emissions inventories and projections. In addition, active cooperation has been initiated with the EMEP Task Force on Emission Inventories and Projections and its Task Force on Measurements and Modelling. The EMEP centres and Task Forces will continue to play an important role in the future to assess intercontinental and hemispheric transport and to link

the work of the Task Force on Hemispheric Transport of Air Pollution to the ongoing analytical efforts focused on the geographic scope of EMEP.

A.8 Outreach activities

Reaching out to experts in countries outside the UNECE region and building trust in the scientific underpinnings of the assessments is an important part of the work of the Task Force. To date, experts from 12 non-UNECE countries have participated in Task Force meetings or workshops: Cambodia, China, Egypt, India, Japan, Malaysia, Mexico, Nigeria, Pakistan, the Philippines, the Republic of Korea and Thailand.

A number of bodies in the United Nations system, for example WMO and the UNEP Regional Resource Centre for Asia and the Pacific, have been active in the Task Force. Other organizations such as the International Maritime Organization (IMO), the International Civil Aviation Organization (ICAO) and the United Nations Framework Convention on Climate Change (UNFCCC) receive the announcements and reports of the Task Force meetings and workshops. The GEO Secretariat has also been an active participant in the meetings and workshops.

The Task Force has been able also to establish important links with other regional initiatives on air pollution, e.g. the Acid Deposition Monitoring Network in East Asia (EANET) and the Clean Air Initiative-Asia, as well as with programmes focused on building capacity for air quality management and regional cooperation, e.g. the Regional Air Pollution in Developing Countries (RAPID-C) project and the Global Atmospheric Pollution Forum. These international organizations and cooperative efforts are important in that they provide both insight into air pollution issues at different scales and also a possible future basis for joint action on policy initiatives.

Appendix B

Glossary of transport-related terms and definitions

The following list of transport-related terms and definitions is included to help understand some of the more technical terms used by atmospheric scientists when describing air pollution transport and chemistry. The definitions are written in layman's terms and in many cases have been simplified from the more rigorous and complete scientific definitions, many of which appear in Glickman (2000). As such, the definitions provided here should not be considered completely comprehensive.

albedo – The fraction of solar radiation reflected from a surface.

background concentration (or mixing ratio) – The concentration (or mixing ratio) of a pollutant that cannot be attributed to specific local emissions or to concentrated and coherent plumes of pollution from some upwind source. Background concentrations are typically aged and can have a variety of sources.

boreal region – A biogeographical region that extends from roughly 45º N latitude to the Arctic Circle.

atmospheric boundary layer – The layer of the troposphere that is in contact with the surface. It can range in depth from tens of metres under highly stable conditions to several kilometres in convective conditions over deserts. Over land, the atmospheric boundary layer depth has a diurnal cycle; it is deepest during the daytime and relatively shallow at night.

column loading – The mass of a gas or aerosol in a vertical column of the atmosphere of some specified depth.

concentration – The mass of a trace gas or type of particulate matter per volume of air, often expressed as micrograms per cubic metre.

deposition – Processes by which trace gases or particles are transferred from the atmosphere to the surface of the earth. Wet deposition is the result of water soluble gases or particles being incorporated into water droplets and then transferred to the surface via precipitation. Dry deposition occurs when a trace gas or particle is transported to ground level followed by adsorption to a surface.

domestic – With respect to the HTAP intercomparison results, domestic relates to the impact of a region on itself, whereas foreign refers to the influence of one or more external regions.

export – In terms of the intercontinental transport of air pollution, export refers to pollution leaving the horizontal confines (i.e. crossing the shoreline) of a particular continent in either the upper or lower troposphere. Export can also refer to the upward transport of air pollution out of the atmospheric boundary layer.

free troposphere – The portion of the troposphere above the atmospheric boundary layer which is not directly impacted by the surface of the earth.

gravity wave – A wave disturbance in the atmosphere in which an air parcel's buoyancy acts as a force to restore the air parcel to its equilibrium vertical location, dictated by its density and pressure.

Hadley cell – A thermally driven circulation in the tropics and subtropics involving the equatorward movement of the trade winds between roughly 30º N latitude and the equator, rising air near the equator, poleward flow in the upper troposphere and, finally, descending air near 30º N (see chapter 2, figure 3).

import – In terms of the intercontinental transport of air pollution, import refers to pollution from some upwind source entering the horizontal confines (i.e. crossing the shoreline) of a particular continent in either the upper or lower troposphere.

import sensitivity – With respect to the HTAP intercomparison results, import sensitivity is defined as

the relative response due to imported pollution versus domestic emissions. It is the response to the sum of 20 per cent anthropogenic emission reductions in the foreign regions divided by the response to a 20 per cent anthropogenic emission reduction in the region itself.

Intertropical Convergence Zone (ITCZ) – The dividing line between the Northern and Southern Hemisphere trade winds, co-located with the ascending branch of the Hadley cell.

jet stream – Relatively strong winds concentrated in a narrow stream in the upper troposphere of the mid-latitudes, with wind speeds in excess of 25 m s^{-1} (50 knots).

latent heat of condensation – The heat released when water vapour condenses into water droplets.

lifetime – The time required for the concentration of a trace gas to decrease to 37 per cent of its original value.

meridional – A flow, average or functional variation along a line of longitude, or oriented north-south.

mid-latitude cyclone – Also referred to as an extratropical cyclone, this synoptic scale weather system consists of a migratory low pressure region in the mid-latitudes that rotates cyclonically (counter-clockwise in the Northern Hemisphere), with one or more surface frontal boundaries and typically producing a large cloud shield and precipitation.

mixing ratio – In a given volume of air, the ratio of the number of molecules of a particular trace gas to the number of molecules of all the other gases present.

North Atlantic storm track – A region over the North Atlantic Ocean with relatively intense synoptic-scale cyclone activity. In winter, this feature is located near 45º N latitude and corresponds to the mean trajectory of the mid-latitude cyclones crossing the ocean.

potential temperature – The temperature of an air parcel if it were compressed or decompressed from its actual pressure to a pressure of 1000 hPa (hectopascals), which is the approximate atmospheric pressure at sea level.

ppbv – Parts per billion by volume. A unit commonly used to quantify trace gases in the atmosphere.

radiative cooling – The process by which the temperature of an air mass decreases because it emits more radiation than it absorbs.

response – With respect to the HTAP intercomparison results, the response is the absolute change of concentrations, columns or deposition in receptor regions from changes in foreign and domestic source regions.

scavenging – Removal of pollutants from the air by either rain or snow.

sedimentation – The process by which particles fall through the atmosphere or to the surface due to gravity.

sensible heating – The transfer of enthalpy (a thermodynamic quality equivalent to the total heat content of a system) from one body to another by conduction (transfer from the Earth's surface to the overlying air) or convection (transfer by the rising and mixing of warm air with colder air aloft). These processes occur in addition to any heat absorbed or released due to the evaporation or condensation of water.

source contribution – A quantitative measure of the amount of a particular pollutant at a receptor that was emitted from a source. The source contribution depends upon (a) the transport source-receptor relationship, (b) the emission flux of the pollutant from the source region, and (c) any loss of the pollutant as it is transported from the source to the receptor.

source-receptor relationship – A term that describes the sensitivity of a "receptor" location to an upwind "source". In general, source-receptor relationships are dependent on the emissions strength of the source, the transport pathway from the source to receptor as well as the pollutant transformation, production and loss processes that occur along the pathway. The transport component of a source-receptor relationship can be expressed, independently of the strength of emissions from the source region, as a measure of (a) the quantity of air from the

source that reaches the receptor over a given time period and (b) the amount of time the air spends over the source region.

stirring – The inter-mingling of air masses caused by large-scale eddies. A distinct process from the molecular scale mixing of different air masses.

subtropics – The indefinite belts in each hemisphere between the regions of tropical and temperate climates, with the polar boundary being roughly 35º N and 35º S latitude.

synoptic – A specific scale of atmospheric motion with a typical range of many hundreds of kilometres, including features such as tropical and mid-latitude cyclones.

Tg – Teragram, or 1,000,000,000,000 grams. Equal to 1,000,000 metric tons.

trade winds – The wind system occupying most of the tropics, which blows from the subtropical highs toward the equator. In the Northern Hemisphere, the flow is generally from the north-east towards the south-west.

transport event – A loose expression applied to any instance of pollution transport from a source location to a receptor location.

troposphere – The layer of the atmosphere where most weather occurs, that is 10-20 km deep and located between the surface of the earth and the tropopause (the boundary between the troposphere and the overlying stratosphere, see chapter 2, figure 3).

warm conveyor belt – One of the major airstreams of a mid-latitude cyclone. It is located on the eastern side of the cyclone, ahead of the surface cold front. The air originates at low altitudes in the warm sector of the cyclone and travels poleward, ascending into the mid- and upper troposphere, above the cold conveyor belt. The warm conveyor belt is a key mechanism for lofting pollutants from the atmospheric boundary layer into the mid and upper troposphere (see chapter 2, figure 5).

warm sector – The region of warm air in a mid-latitude cyclone located between the cold front and the warm front, which feeds into the warm conveyor belt and can also give rise to deep convection, especially in summer (see chapter 2, figure 5).

zonal - A flow, average or functional variation along a line of latitude, or oriented east-west.

References

Glickman, T. S. (Ed.) (2000), *Glossary of Meteorology*, 2nd ed., American Meteorology Society, Boston.

For more information, consult the Convention's website, or contact:

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