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EURODELTA-Trends, a multi-model experiment of air quality hindcast in Europe over 1990-2010.

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Abstract

The Eurodelta-Trends multi-model chemistry-transport experiment has been designed to facilitate a better understanding of the evolution of air pollution and its drivers for the period 1990-2010 in Europe. The main objective of the experiment is to assess the efficiency of air pollutant emissions mitigation measures in improving regional scale air quality.

The present paper formulates the main scientific questions and policy issues being addressed by the Eurodelta-Trends modelling experiment with an emphasis on how the design and technical features of the modelling experiment answer these questions.

The experiment is designed in three tiers with increasing degree of computational demand in order to facilitate the participation of as many modelling teams as possible. The basic experiment consists of simulations for the years 1990, 2000 and 2010. Sensitivity analysis for the same three years using various combinations of (i) anthropogenic emissions, (ii) chemical boundary conditions and (iii)

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meteorology complements it. The most demanding tier consists two complete time series from 1990 to 2010, simulated using either time varying emissions for corresponding years or constant emissions.

Eight chemistry-transport models have contributed with calculation results to at least one experiment tier, and three models have – to date - completed the full set of simulations (and 21-year trend calculations have been performed by four models). The modelling results are publicly available for further use by the scientific community.

The main expected outcomes are (i) an evaluation of the models performances for the three reference years, (ii) an evaluation of the skill of the models in capturing observed air pollution trends for the 1990-2010 time period, (iii) attribution analyses of the respective role of driving factors (emissions/boundary conditions/meteorology), (iv) a dataset based on a multi-model approach, to provide more robust model results for use in impact studies related to human health, ecosystem and radiative forcing.

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1 Introduction

Air pollution is a crucial environmental concern because of its detrimental impacts on health, ecosystems, the built environment and short term climate forcing. Whereas it was originally regarded as an urban issue, in the late 1970s the large scale acidification of precipitation made it clear that at least part of the problem could only be solved through international cooperation (OECD, 1977). This was the background for the establishment of the Convention on Long Range Transboundary Air Pollution (CLRTAP) in 1979. The main vehicles of the LRTAP Convention are the Protocols that aim to reduce the emission of various compounds (sulphur in 1985, nitrogen oxides in 1988, volatile organic compounds in 1991, heavy metals and persistent organic pollutants in 1998, and the multi-pollutant multi-effect Gothenburg Protocol to abate acidification, eutrophication and ground level ozone in 1999 (and subsequent revision in 2012). The design of such mitigation strategies was largely supported by the development of models (chemistry-transport and integrated assessment tools) and monitoring networks.

After several decades of international cooperation, it is timely to take stock of the evidence available to assess the efficiency of the LRTAP Convention and the corresponding emission ceilings protocols. The Executive Body of the Convention has therefore requested an assessment of the evolution of air pollution and subsequent effects from its two scientific and technical bodies (i) the European Monitoring and Evaluation Programme (EMEP) and (ii) the Working Group on Effects (WGE). As a result, the Task Force on Measurement and Modelling (TFMM) of EMEP published an assessment of air pollution trends (Colette et al., 2016), whereas the WGE published an assessment of corresponding effects on health and ecosystems (De Wit et al., 2015), and an overall assessment report encompassing all the activities undertaken under the Convention was also released (Maas and Grennfelt 2016).

The effects of emissions on the concentrations is rather complex due to (i) the non-linearity of atmospheric chemistry, (ii) the presence of inflow of air pollution due to the intercontinental transport of air pollutants, and (iii) the meteorological variability. This is where Chemistry-Transport Models (CTMs) come into play with the multi-model air quality trend experiment introduced in the present paper.

The LRTAP convention relies in part on the results of the EMEP/MSC-W chemistry-transport model (Simpson et al., 2012). Since the beginning of the 2000s, the Joint Research Centre of the European Commission initiated a number of multi-model assessments to provide a benchmark for the EMEP/MSC-W model through its comparison with the modelling tools being used by the States-Parties to the Convention (Bessagnet et al., 2016;van Loon et al., 2007;Thunis et al., 2008). The Eurodelta-Trends (EDT) exercise builds upon this tradition, focusing on the specific context of air quality trends modelling. Its main goal is to assess to what extent observed air pollution trends could be related to emission mitigation, although this overarching question can only be addressed after having assessed the confidence we can have in the models, in particular in their capacity to reproduce the trends.

Over the recent past, a few attempts have been made to address the issue of the long term evolution of European-scale air quality by means of modelling studies. First using only one model as in (Jonson et al., 2006;Vautard et al., 2006;Wilson et al., 2012). A first ensemble was proposed through the European Project CityZen which relied on 6-models (Colette et al., 2011). While these studies were limited to about 10-yr time periods, a 20-yr hindcast study was presented in (Banzhaf et al., 2015), relying however again on a single model. It is therefore timely to engage in a multi-model hindcast of air quality over two decades.

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The purpose of the present paper is to define the science and policy questions that are addressed by the EDT exercise, and introduce the experimental setup designed to answer these questions. The models participating in the experiment will also be presented as well as the project database of model results.

2 Participating models

Eight European modelling teams submitted their calculation results to the EDT database for at least one tier of experiment (see the experiment design in Section 3) using state-of-art air quality models: Chimere (Menut et al., 2013), CMAQ (Byun and Schere, 2006), EMEP/MSC-W (Simpson et al., 2012), LOTOS-EUROS (Sauter et al., 2012;Schaap et al., 2008), MATCH (Robertson et al., 1999) MiNNI (Mircea et al., 2016), Polyphemus[1](Mallet et al., 2007), and WRF-Chem (Grell et al., 2005;Mar et al., 2016). The main specifications of the eight participating models are summarized in Table S1 (note that they can differ from the public release of the various models according to the elements provided in the table).

The representation of physical and chemical processes differs in the models. The vertical distribution of model layers (including altitude of the top layer and derivation of surface concentrations at 3m height in the case of EMEP, LOTOS-EUROS and MATCH) is not prescribed either. However, as further explained in the article, the other features of the model setup are largely constrained by the experiment input data such as forcing meteorology, boundary conditions, emissions and by the experiment characteristics such as horizontal domain and resolution. Only one of the participating models included online coupled chemistry/meteorology (WRF-Chem), while all the other models are offline CTMs.

3 Experimental design

The main policy focus being addressed in EDT analysis is the assessment of the role of European air pollutant emission reductions in improving air quality over the past two decades. Subsequent questions include assessing (1) the role of changes in global air pollution as well as (2) the role of inter-annual meteorological variability. Before addressing such issues, it will be essential to quantify the CTMs' capability in (1) reproducing observed air pollutant concentrations (processes determining air quality: chemistry, physics, transport processes, emissions, meteorology), and (2) capturing the long term evolution of air quality.

The time period covered by the experiment is 1990-2010. The year 1990 has been chosen as the beginning of the period because that year serves as reference for the Gothenburg protocol. The end of the period is 2010 because of the availability of underlying forcing data (emissions, boundary conditions and meteorology) required for model calculations at the time the work was initiated.

The EDT model experiment is divided into three tiers, targeting various science and policy questions. The tiers also differ in terms of computational demand that allowed involving as many modelling groups as possible. The tiers of experiments are summarized in

[1] http://cerea.enpc.fr/polyphemus/

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Table 1. They differ in terms of the number of modelled years to be addressed in the 1990-2010 period and in terms of forcing data used in model calculations for the anthropogenic emissions, the chemical boundary conditions, and the meteorological year. Most of the experiments consist of variations in one or two of these three factors in order to disentangle the role of each forcing. The role of chemical boundary conditions constitutes one notable exception since two sources of forcing are used: either a global CTM simulation or an observation-based climatology (further details are provided on boundary conditions in Section 8).

The first simulation of the EDT experiment is a reference for the year 2010 using the meteorology (M), the boundary conditions (B) and the emissions (E) for that year, named as M10B10E10, with two digits corresponding to the last two digits of the year. They are complemented with simulations for the years 1990 and 2000 (using corresponding meteorology, boundary conditions and emissions: M90B90E90 and M00B00E00 respectively) to form tier 1A. Tier 1A will allow testing the accuracy of all CTMs in simulating pollution changes for the near past (1990, 2000 and 2010), at a lower computational cost than running the full 21-yr period.

Tier 1B is dedicated to the first two sensitivity experiments, for which the meteorology and the boundary conditions are those of the year 2010, but the emissions correspond to 1990 and 2000 (M10B10E90 and M10B10E00). They will allow assessment of the individual impact of emission changes alone (E10 versus E90 and E10 versus E00) by comparison with Tier 1A (specifically M10B10E10).

In Tier 2A, two more sensitivity simulations are performed for the meteorological year 2010, using emissions and boundary conditions of 1990 and 2000 (M10B90E90 and M10B00E00, respectively). By comparison with Tier 1B, they will allow the assessment of the impact of global chemical background changes on European air quality between the years 1990 and 2010, and also for the sub-periods 1990-2000 and 2000-2010 (B10 versus B90 and B10 versus B00).

Tier 2B is an alternate set of reference simulations for 1990, 2000 and 2010, in which boundary conditions provided by a global model (C) instead of the observation-based boundaries (B) are used (M90C90E90, M00C00E00, M10C10E10). It will allow assessment of the uncertainty related to the large scale chemical forcing by comparison with Tier 1A.

Tier 2C is a complement to Tier 2A using the meteorology of 2000 and two combinations of 1990 and 2000 boundary conditions and emissions (M00B90E90, M00B00E90). These additional simulations are required to perform the attribution analysis for the concentration changes between 1990 and 2000, whereas the simulations required for the attribution of driving factors between 1990 and 2010 and between 2000 and 2010 are dealt with in tiers 1A, 1B, and 2A.

Tier 3A consists in 21-year simulations covering 1990-2010, using meteorology, boundary conditions and emissions for the respective years (MyyByyEyy, with yy being the 2-digits year between 1990 and 2010). It will be used to assess the capability of the models to capture observed trends in air quality by means of comparisons with available measurements. Fewer modelling teams delivered results for this higher tier of experiments, therefore model uncertainty will be put in perspective with the spread of the whole ensemble in modelling Tier 1A (1990, 2000, 2010).

Tier 3B is the last sensitivity experiment in which 21-year simulations are performed using the 2010 emissions for the complete period (MyyByyE10). By comparison with Tier 3A, it will allow the determination of the role of inter-annual meteorology and chemical boundary condition changes versus the role of European emission changes.

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Thus, the complete series of model runs included for each air quality model is 5 annual simulations for Tier 1, 7 more simulations for Tier 2, and 39 (2x21 minus one overlap for 2010, and two annual simulations belonging to Tier 1A: M90B90E90 and M00B00E00) more simulated years for Tier 3.

Figure 1 provides the schematics of the various combinations of simulations required to perform the attribution analysis for any period of time between the three reference years (1990, 2000 and 2010). The simulations labelled in black are covered by the above simulation plan. They are needed for the assessment of the relative role of emission, meteorology and boundary condition changes.

The main limitations of the simulation plan are (i) that the three selected meteorological years may be not representative, or atypical, for the full period, and (ii) the lack of interaction by considering 2^2 combinations instead of the 2^3 combinations required to cover the whole space of factors (Stein and Alpert, 1993). In the forthcoming attribution study these limitations will be explored by (i) comparing trend (tier 3A) and sensitivity (tier 1&2) tiers, and (ii) including additional simulations for the 2^3 possible combinations from one of the models (Chimere).

4 Modelling domain

The modelling domain is displayed in Figure 2. The domain follows a regular latitude-longitude projection with increments of 0.25° and 0.4° in latitude and longitude, respectively, which is about 25 km x 25 km. The total coverage extends from 17W to 39.8E and from 32N to 70N. Only one of the participating models could not be implemented on the exact same grid: CMAQB uses a Lambert Conformal Conic projection map with 25 km resolution and it delivered its results on the common grid, although the south-easternmost part of the domain was not covered by that model.

5 Meteorology

The horizontal resolution of available global meteorological reanalyses over the 1990-2010 period is considered too coarse to drive regional scale CTMs. Therefore, dynamically downscaled regional climate model simulations using boundary condition from the ERA-Interim global reanalyses (Dee et al., 2011) were used to force the CTMs involved in EDT. Most CTMs used the same meteorological driver, with a couple of exceptions.

One of the meteorological drivers was produced using the Weather Research and Forecast Model (WRF) (Skamarock et al., 2008) at 0.44 degrees of resolution. In the framework of the EuroCordex climate downscaling programme (Jacob et al., 2013) an evaluation of the regional climate models downscaled with perfect boundary conditions (ERA-Interim reanalyses instead of free climate runs) was reported by (Kotlarski et al., 2014). One of the WRF realisations in the EuroCordex ensemble was subsequently further optimized as described in (Stegehuis et al., 2015), so that we could identify an optimal WRF setup for our purpose (row #7 of Table S1 in their supplementary material). The model was re-run using grid nudging towards the ERA-Interim reanalyses (above the planetary boundary layer) in order to improve temporal correlations compared to the regular free-running Cordex hindcast simulations. This WRF simulation was interpolated on the 25km resolution EDT grid and used to drive Chimere, EMEP, Polyphemus, and Minni. In EMEP model, the interpolation of the meteorological fields from 0.4x0.4° to EDT grid was performed online.

The CMAQ model, which runs on a Lambert Conformal Conic projection, could not use the meteorological data provided on the EuroCordex grid, so that WRF was re-run in a Lambert Conformal projection at 25 km horizontal resolution using identical WRF setup and version (3.3.1). A similar

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strategy was used for WRF-Chem, which is an online model that simulates meteorology and chemistry simultaneously, but again an identical setup was used. The CTMs LOTOS-EUROS and MATCH have been meteorologically forced by ERA-Interim series further downscaled with respectively RACMO2 (van Meijgaard, 2012) and HIRLAM (Dahlgren et al., 2016). RACMO2 was also included in the EuroCordex studies by Jacob et al. (2013) and Kotslarski et al. (2014). The main features of the mesoscale meteorological models are synthesized in Table 3.

6 Biogenic and natural emissions

There were no specific constraints imposed to biogenic emissions (including soil NO emission) which are represented by most CTMs using an online module. Forest fires were ignored and each modelling team could decide whether they would include lightning as well as natural and road resuspension of dust emissions (see also the synthesis in Table S1).

7 Anthropogenic emissions

7.1 Annual totals

National annual emissions, distributed by SNAP (Selected Nomenclature for reporting of Air Pollutants) sectors, were estimated with the GAINS (Greenhouse gases and Air pollution INteractions and Synergies) model (Amann et al., 2011). The calculation was performed for 1990, 1995, 2000, 2005, and 2010 for SO_2, NO_x, NMVOC, CO, NH_3, and PM including PM_{10}, $PM_{2.5}$, BC, and OC. To derive emissions for intermediate years, sectorial results for five-year periods were linearly interpolated.

The key activity data originates from Eurostat[2] and International Energy Agency (IEA, 2012) for energy use and from Eurostat, UN Food and Agriculture Organization (FAO)[3], International Fertilizer Association (IFA) for agriculture. For the transport sector, additionally the results of the COPERT model for the EU-28 countries were used (Ntziachristos et al., 2009); this data includes detailed transport sources, fuel distribution, mileage, and level of penetration of control measures. The emission calculation considers impact of existing national and international source specific emission limits and air quality legislation, e.g., several European Union Directives: Large Combustion Plants, Industrial Emissions, National Emission Ceilings Solvent Directive, as well as the UNECE Gothenburg Protocol (UNECE, 1999;Reis et al., 2012). Finally, the results of consultations with national experts, carried within the work on the review of the National Emission Ceiling Directive (Amann et al., 2012) were considered. This emission dataset was completed in April 2014 and is referred to as ECLIPSE_V5; it is part of a global emission set established during the EU funded FP7 project ECLIPSE. More detailed description of the data and applied emission calculation methodology is given in (Amann et al., 2012) and (Klimont et al., 2016b;Klimont et al., 2016a). The respective scenario is available in the freely accessible on-line version of the GAINS model[4] where more detailed outputs and all data inputs can be found.

http://[2] ec.europe.eu

[3] http://www.fao.org/statistics/en/

[4] http://magcat.iiasa.ac.at; select 'Europe' in order to access respective data and results

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7.2 Spatial distribution of emissions

The emissions were provided by INERIS for the EDT modelling domain using the spatial regridding methodology introduced in (Terrenoire et al., 2015;Bessagnet et al., 2016) which consists of:

- Europe-wide road and shipping proxies for SNAP sectors 7 and 8 (road transport and other mobile sources and machinery);
- A proxy based on the population density for residential emissions (SNAP 2: non-industrial combustion plants), note that emissions are not linearly proportional to the population density, a fit tested with the bottom-up inventory for France is used;
- For industrial emissions (SNAP 1, 3, and 4: Combustion in energy and transformation industries; Combustion in manufacturing industry; Production processes) we use the flux and location from the EPRTR inventory (prtr.ec.europa.eu). When the total emissions exceed the flux reported in EPRTR, we used a default pattern applying the CEIP spatial distribution, available by SNAP sectors ("emissions as used in EMEP models"[5]). The only exception is for particulate matter emissions for which a spatial distribution was not available for 1990; for that year a combination of officially reported emissions was produced by order of priority: SNAP, NRF01, NFR02 and NFR09 (NFR standing for "Nomenclature for Reporting" following the 2001, 2002, or 2009 guidelines).
- Bottom-up emission inventories for all SNAP for France & United Kingdom (such information was not available elsewhere);
- TNO-MACC inventory for NH3 emissions (mostly SNAP10: agricultural emissions);
- Default CEIP spatial distribution at a 50km resolution for the other sectors (SNAP5, 6, 9: Extraction and distribution of fossil fuels and geothermal energy, Solvents and other product use, Waste treatment and disposal).

In the applied method, only the spatial distribution of industrial emissions is supposed to have changed in time over the past decades. For the residential and road sector, it was considered that the recent techniques involving consistent and high-resolution proxies over Europe provide a more realistic view of emissions than the 50km resolution emission data from the 1990s and early 2000s.

8 Boundary Conditions

Two sources of chemical boundary conditions are used by the regional CTMs: a climatology of observational data, and global model results. Both have pros and cons. Global models carry biases but include a wider array of chemical species. The trend in observations matches in-situ data by nature, but only at one point over the domain. For the EDT experiment it was decided to rely on observation-based boundary conditions for most experiments (Tier 1A, 1B, 2A, 2C, 3A, 3B) but also include a sensitivity study based on modelled boundary conditions (Tier 2B).

Note that a possible impact of changing chemistry composition on large scale circulation was integrated in the forcing meteorological fields through the data assimilation of the ERA-Interim reanalysis. This factor was not considered important to isolate for the 2-decade timescale of the experiment.

[5] http://www.ceip.at/

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8.1 Observations-based boundary conditions

The boundary conditions (BCs) are a simplified version of those used in the standard EMEP/MSC-W model (Simpson et al., 2012). The values are based upon climatological data (except from those for natural particles). The most important gaseous boundary condition compounds are O_3, CO and CH_4. For ozone, the 3D climatology based on observational vertical profiles constructed by (Logan, 1998) are used in conjunction with a temporal (monthly) variation over the past 20 years. These climatological values are modified each month to ensure that their variability matches the observed variability of concentrations in the clean westerly Atlantic air masses as measured at Mace Head on the coast of Ireland. The 'Mace Head correction' has been derived for each year from ozone data from Mace Head, sorted using sector-analysis (based on trajectories obtained from MSC-W[6]). Monthly mean values of the ozone associated with easterly sectors have been calculated for respective years/months, as described in (Simpson et al., 2012).

For methane, uniform boundary conditions around the European domain are set to: 1780 ppb in 1990, 1820 ppb in 2000, and 1870 ppb in 2010 according to Mace-Head observations. For the intermediary years, an interpolation is applied.

For sulphate (SO_4^{2-}) and nitrate (NO_3^-) aerosols, the trends for 1990-2010 are derived from the trend in EPA emissions for North America of SO_2 and NOx (Hicks et al., 2002b)[7]. For ammonium (NH_4^+), the trends are derived as $2/3*SO_4^{2-} + 1/3*NOx$. The rationale for SO_2 lies in the demonstration of the close correspondence between national emissions and concentration trend in (Hicks et al., 2002a).

Monthly (3-dimensional) boundary conditions for sea salt and windblown mineral dust are constructed based on a global run performed with the EMEP/MSC-W model for 2012.

8.2 Global model based boundary conditions

A global model simulation from the Climate-Chemistry Model Initiative (CCMI) is also used in EDT. CCMI undertakes a global atmospheric chemistry reanalysis over the 1960-2010 time period (Eyring, 2014) based on the MACCity emissions (Granier et al., 2011). The CAM4-chem (Tilmes et al., 2016) member of the CCMI ensemble was made available at monthly temporal resolution for use in EDT.

Evaluation of this global reanalysis is ongoing, but the preliminary results are encouraging as illustrated in Figure 3 which shows the modelled and observed ozone trend at the Mace Head station.

9 Output format and database status

The model simulations were delivered in a common NetCDF format, so that each of the files contains gridded fields of one pollutant for a whole year. The air concentrations from only the lowest model level (or corrected to 3m height for EMEP, LOTOS-EUROS and MATCH) are delivered to the project database, but the participants are encouraged to store 3D data if their storage capacities allow such an archiving.

The requested variables are:

[6] http://www.emep.int

[7] https://www.epa.gov/air-trends

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- Hourly concentrations of O_3 and NO_2;
- Daily concentrations of aerosols: nitrate (NO_3^-), sulphate (SO_4^{2-}) and ammonia (NH_4^+), sea-salt, dust, total primary PM, anthropogenic and biogenic secondary organic aerosols, and total PM, both for the fraction below 2.5µm ($PM_{2.5}$), and the fraction below 10µm (PM_{10});
- Daily concentrations of reactive gases: NH_3, SO_2, an indicator of alpha-pinene that shall depend on the chemical mechanism of each model, isoprene, HNO_3, H_2O_2, HCHO, PAN, total VOC, biogenic VOC;
- Daily emission rate of biogenic species: isoprene, and an indicator of alpha-pinene that shall depend on the chemical mechanism of each model;
- Monthly dry and wet deposition of total oxidized sulphur (SOx), oxidised nitrogen (NOx) and reduced nitrogen (NHx);
- Hourly meteorological fields: temperature at 2m, wind speed, PBL, rain.

Additional diagnostics were subsequently computed on the common database:

- O3_DL: daily ozone computed on the basis O3_HL as the mean value for each day between 00:00 and 23:00 UTC.
- O3Aot40_DL: accumulated ozone over 40ppbv computed on the basis of O3_HL, for each day (from 1 May until 31 July) as the sum of all the daylight hourly O3_HL values exceeding the value of 40 ppb (80 ug/m3). Note that hourly values in the models correspond to instantaneous values: e.g. O3_HL(0) is for 0:00, O3_HL(23) is for 23:00). The accumulation of AOT between 8hr and 20hr was taken as the sum of O3_HL between 8:00 and 19:00, included. O3Aot40_DL is a daily quantity that must be cumulated over a given period of the year, e.g. May-June-July in the European Air Quality Directive (EC, 2008). Units are: (µg/m^3)*hours.
- O3Aot60_DL: same as before, with threshold 60 ppb (120 µg/m^3) and to be accumulated over the period 1 April - 30 September. Units are: (µg/m^3)*hours.
- O3hr8_HL: the 8-hr running mean hourly ozone computed from O3_HL. To each hour ih in O3hr8_HL the running mean is that of the 8 past values of O3_HL is assigned: O3hr8_HL(ih) = [O3_HL(ih) + ... O3_HL(ih-7)] /8.
- O3hr8Somo35_DL: Sum of ozone means over 35ppbv computed from O3hr8_HL for each day of the year as the exceedance of the daily max O3hr8_HL with respect to 35 ppb (70 µg/m^3). The accumulated value used in the Air Quality Directive is the sum over all days of the year. Units are: (µg/m^3)*days.
- O3hr8Max_DL: Maximum daily value of O3hr8_HL, sometimes also referred to as MDA8 as Ozone Maximum Daily Average.
- O3hr8Exc60_DL: computed from O3hr8_HL. For each day of the year a value of 1 is assigned if the maximum daily value of O3hr8_HL exceeds 60 ppb (120 µg/m^3), otherwise equal to zero. The value mentioned in the Directive is the sum over all days of the year. Units are: days.
- O3hr8H4th_DL: Computed from O3hr8_DL, the 4th maximum daily value over the year is calculated and assigned to all days. This corresponds to the 99 percentile of the daily max O3hr8 values over the year.
- NO2_DL: Computed from NO2_HL, same as O3_DL.
- NO2hr1Max_DL: Computed from NO2_HL, Maximum daily value of NO2_HL.
- NO2hr1Exc200_DL: Computed from NO2_HL, for each day of the year a value of 1 is assigned if the maximum daily value of NO2_HL exceeds 200 ppb, otherwise equal to zero. The value mentioned in the Directive value is the sum over all days of the year. Units are: days.
- NOx-ppb: DL and HL: Sum of NO and NO2 in ppb, i.e. NO(µg/m^3)*22.4/30 + NO_2(µg/m^3)*22.4/46.

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- PM10Exc50_DL: Computed from PM10_DL, for each day of the year a value of 1 is assigned if the (daily) value of PM10_DL exceeds 50 ug/m3, otherwise equal to zero. The value in the Directive is the sum over all days of the year. Units are: days.
- TNO3-N: Sum of NO3-10 and HNO3 in µgN/m^3, i.e. NO3-10(µg/m^3)*14/62 + HNO3(µg/m^3)*14/63.
- TNH4-N: Sum of NH4-10 and NH3 in µgN/m^3: i.e. NH4-10(µg/m^3)*14/18 + NH3(µg/m^3)*14/17.
- TSO4-S: Sum of SO4-10 and SO2 in µgS/m^3: SO4-10(µg/m^3)*32/96 + SO2(µg/m^3)*32/64.
- NOz: Sum of HNO3, and PAN in ppb. Conversion factors from µg/m^3 to ppb: [24/63,24/53].
- NOy: Sum of NO2, NO, HNO3, and PAN in ppb. Conversion factors from µg/m^3 to ppb: [24/46,24/30,24/63,24/53]

The status of models' delivery of results for each of the experiment tiers at the time of submission of the present article is summarized in Table 2. The access to the database is open for research use through the AeroCom server (see also the section on data availability)[8] .

10 Conclusions

The Eurodelta-Trend modelling experiment (EDT) will allow a better understanding of the evolution of regional scale air quality over Europe over the 1990-2010 period. This is facilitated by the thoroughly designed modelling plan. Eight modelling teams have participated in the EDT experiment, though with a variable degree of involvement. The base runs of Tier 1A, completed with eight participating models, offer a great opportunity to assess the capability of these state-of-the-art chemistry-transport models to reproduce the observed changes in the concentrations of the main pollutants, including ozone, particulate matter and its individual components, as well as in precipitation chemistry. This analysis will then be complemented by an assessment of the capability in reproducing the actual trends over the 21yr in the 1990-2010 period for the models participating in the more demanding tier 3A experiment. If this evaluation phase concludes that the skill of these models in capturing air quality evolution is satisfactory, we would then rely on the results of the trend (or decadal changes) calculations and the sensitivity experiments and recommend that they can be used when addressing science and policy questions underlying the evolution of air quality in Europe over the past couple of decades.

The critical policy question lies in the attribution of air quality trends to emission changes, to influx at the boundaries of the European domain, and to interannual meteorological variability (and natural sources of trace species) and will be addressed in a series of upcoming papers. Furthermore, thanks to the multi-model design of the experiment, other scientific questions with regard to the role of specific chemical and physical processes will be investigated in forthcoming studies based on the Eurodelta-Trends results.

The model results will also be publicly distributed in order to serve for in depth analyses to scientific communities working on the impacts of air pollution on health, ecosystems or aerosol radiative forcing.

Data availability

[8] https://wiki.met.no/aerocom/user-server

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The Eurodelta-Trends model results are made available for public use on the AeroCom server[9] under the following terms:

- Data provided on this server may be used solely for research and education purposes;
- EURODELTA-TRENDS partners cannot guarantee that the data are correct in all circumstances. Neither do they accept any liability whatsoever for any error or omission in the data, or for any loss or damage arising from its use;
- Data must not be supplied as a whole or in part to any third party without authorization;
- Articles, papers, or written scientific works of any form, based in whole or in part on data, images or other products supplied by EURODELTA-TRENDS will contain an acknowledgment concerning the supplied data reading:
    - "Modelling data used in the present analysis were produced in the framework of the EuroDelta-Trends Project initiated by the Task Force on Measurement and Modelling of the Convention on Long Range Transboundary Air Pollution. EuroDelta-Trends is coordinated by INERIS and involves modelling teams of BSC, CEREA, CIEMAT, ENEA, IASS, JRC, MET Norway, TNO, SMHI. The views expressed in this study are those of the authors and do not necessarily represent the views of Eurodelta-Trends modelling teams."
- Users of these data must offer co-authorship to the modelling teams for any study submitted for publication until June 2018. The list of modellers is: CHIMERE (A. Colette, F. Couvidat, B. Bessagnet), CMAQ (M.T. Pay), EMEP (S. Tsyro, H Fagerli, P. Wind), ex-JRC (C. Cuvelier), LOTOS-EUROS (A. Manders), MATCH (A. Andersson, R. Bergström), MINNI (M. Mircea, G. Briganti, A. Cappelletti, M. Adani, M. D'Isidoro), POLR (V. Raffort), WRF-Chem (K.A. Mar, N. Otero, N. Ojha). After this date, users must inform the Eurodelta-Trends coordinator (augustin.colette@ineris.fr) about the expected use of the data. The coordinator will, in turn, inform a representative from each modelling team.

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- The GAINS emission trends where produced as part of the FP7 European Research Project ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants); grant no 282688.
- The Chimere simulations where performed were made using the TGCC super computers under the GENCI time allocation gen7485. Also with support from the French Ministry in Charge of Ecology.
- CMAQB simulations were performed on the MareNostrum Supercomputer hosted by the Barcelona Supercomputing Center. The work developed M. T. Pay and its related expenses had been funded by the post-doctoral grant Beatriu de Pinós Program (2011 BP-A2 00015), the CICYT project CGL2013-46736-R and the Severo Ochoa Program awarded by the Spanish Government (SEV-2011-00067).
- Peter Simmonds and Gerry Spain are acknowledged for Mace Head O_3 data.

[9] https://wiki.met.no/aerocom/user-server

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- The CamChem data were produced as part of the CCMI and PEGASOS projects.
- The WRF-Chem simulations have been performed on the supercomputer HYDRA (http://www.rzg.mpg.de/).).
- The computing resources and the related technical support used for MINNI simulations have been provided by CRESCO/ENEAGRID High Performance Computing infrastructure and its staff. The infrastructure is funded by ENEA, the Italian National Agency for New Technologies, Energy and Sustainable Economic Development and by Italian and European research programmes (http://www.cresco.enea.it/english).
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- RACMO2 simulations at KNMI to provide meteorological forcings for LOTOS-EUROS were supported by the Dutch Ministry of Infrastructure and the Environment.

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Table 1 : Summary of model experiments (including label), corresponding key scientific questions. The simulations are labelled MyyByyEyy where M indicates meteorology, B indicates observation-based boundary conditions, C indicates modelled-based boundary condition, E indicates emission, and yy is the 2-digits reference of the corresponding year.

Tier	Experiment	Key question (Q) / Action (A)	Label
1A	Meteorology,boundary conditions and emissions of 1990, 2000 and 2010.	Q: What is the uncertainty within the seven CTMs ensemble in 1990, 2000, and 2010? A: Comparison 1A vs. Observations for 1990, 2000 and 2010	M10B10E10
			M00B00E00
			M90B90E90
1B	Meteorology and boundary conditions of 2010. Emissions of 1990 and 2000.	Q: What if no emission change occurred in Europe? A: Comparison 1A vs. 1B	M10B10E00
			M10B10E90
2A	Meteorology of 2010. Emissions and boundary conditions of 1990 and 2000.	Q: What if no emission changed beyond Europe? A: Comparison 2A vs. 1B	M10B00E00
			M10B90E90
2B	Meteorology and emissions of 2010. Modelled boundary conditions of 1990, 2000, 2010	Q: What is the uncertainty related to boundary conditions? A: Comparison 2A vs. 2B	M10C10E10
			M00C00E00
			M90C90E90
2C	Meteorology of 2000, emissions of 1990 and boundary conditions of 2000 and 1990.	Additional simulations for decomposition of factors in the 1990s and 2000s	M00B90E90
			M00B00E90
3A	21-years reference trend from 1990 to 2010	Q: How do the models capture the trend in observations?	MyyByyEyy
		A: Comparison 3A vs. observations	
3B	21-years trend with 2010 emissions	Q: Does meteorological variability contribute to the AQ trend over the past 20 years?	MyyByyE10
		A: Comparison 3A vs. 3B	

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1 Table 2: Synthesis of models having delivered (D) data or planning to (P) to the project database for each of the
2 experiments.

Tier	Label	CHIMERE	CMAQB	EMEP	LOTOS-EUROS	MATCH	MINNI	Polyphemus	WRF-Chem
1A	M10B10E10	D	D	D	D	D	D	D	D
	M00B00E00	D	D	D	D	D	D	D	D
	M90B90E90	D	D	D	D	D	D	D	D
1B	M10B10E00	D	D	D	D	P	D	D	D
	M10B10E90	D	D	D	D	P	D	D	D
2A	M10B00E00	D	D	D	D	P	D	D	D
	M10B90E90	D	D	D	D	P	D	D	D
2B	M10C10E10	D		D			D		D
	M00C00E00	D		D			D		D
	M90C90E90	D		D			D		D
2C	M00B90E90	D	D	D	D	P	D	D	D
	M00B00E90	D	D	D	D	P	D	D	D
3A	MyyByyEyy	D		D	D	P	D		P
3B	MyyByyE10	D		D	D	P	P		

3

4

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Table 3: Meteorological fields used in the EDT project. WRF-0.44 corresponds an optimized and nudged version of the WRF-IPSL-INERIS Eurocordex member at 0.44 degrees from EuroCordex climate downscaling programme (Jacob et al., 2013) used by most CTMs in EDT. WRF-25 corresponds to the WRF run in the same condition as WRF-0.44 in a Lambert Conformal Conic projection used to drive CMAQB. WRF-Chem indicates the configuration of WRF used within the WRF-Chem online CTM. RACMO2 is the meteorological model used by LOTOS-EUROS.

Model configuration	WRF-0.44	WRF-25	WRF-Chem	HIRLAM EURO4M	RACMO2
Model version	WRF v3.3.1	WRF v3.3.1	WRF v3.5.1	HIRLAM 3DVAR upper air analysis and OI surface analysis (for details and evaluation see (Dahlgren et al., 2016)	RACMO2.3 (Meijgaard et al., 2012)
Initial and boundary conditions	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)	WRF-0.44 simulation used by other EDT models	ERA-Interim global reanalysis (resolution ~80km) (Dee et al., 2011)	ERA-Interim global reanalysis (resolution ~80 km) (Dee et al., 2011)
Coordinate system	Rotated latitude and longitude	Lambert Conformal	Latitude and longitude	Rotated latitude and longitude	Rotated latitude and longitude with a South Pole at 47S and 10E.
Horizontal setting / number of zonal and meridional grid cells	0.44º x 0.44º (120-117)	25 km x 25 km (176-197)	Approx. 25 km x 25 km (144-154)	Approx. 22km x 22km (326-341)	0.22x0.22 (306x220)
Vertical setting	31 layers	31 layers	34 layers	60 layers eta coordinates	40 layers hybrid coordinates
Microphysics	Morrison DM (Morrison et al., 2009)	Morrison DM (Morrison et al., 2009)	Morrison DM (Morrison et al., 2009)	Large-scale condensation with Rasch-Kristjansson scheme (Rasch and Kristjánsson, 1998)	Prognostic cloud scheme (Tiedtke, 1993), Large-scale condensation (Tompkins et al., 2007), boundary-layer clouds (Neggers, 2009)
LW,RW radiation	RRTMG - (Iacono et al., 2008)	RRTMG - (Iacono et al., 2008)	RRTMG - (Iacono et al., 2008)	(Savijärvi, 1990)	Short wave radiation (Clough et al., 2005;Morcrette et al., 2008) Long wave radiation (Mlawer et al., 1997;Morcrette et al., 2001)

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Cumulus scheme	Tiedtke - (Tiedtke, 1989;Zhang et al., 2011)	Tiedtke - (Tiedtke, 1989;Zhang et al., 2011)	Grell 3D scheme[10] (Grell and Dévényi, 2002)	Convective processes Kain-Fritsch scheme (Kain, 2004)	Mass flux scheme (Tiedtke, 1989;Nordeng, 1994;Neggers et al., 2009;Siebesma et al., 2007)
Boundary & Surface layer	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	MYNN-ETA (Janjic, 2002;Nakanishi and Niino, 2006;Nakanishi and Niino, 2009)	Turbulence CBR scheme (Cuxart J. et al., 2000); adaptions for moist CBR (Tijm and Lenderink, 2003)	Eddy-Diffusivity Mass Flux Scheme with TKE prognostic variable (Lenderink and Holtslag, 2004;Siebesma et al., 2007)
Soil	NOAH (Tewari et al., 2004)	NOAH (Tewari et al., 2004)	NOAH (Tewari et al., 2004)	Further developed ISBA scheme (Noilhan and Planton, 1989;Noilhan J. and J.-F., 1996;Gollvik and Samuelsson, 2010)	TESSEL (Van den Hurk et al., 2000), HTESSEL (Balsamo et al., 2009)

1

[10] A different scheme was chosen for compatibility with chemistry, in particular so that there would be subgrid convective transport of chemical species.

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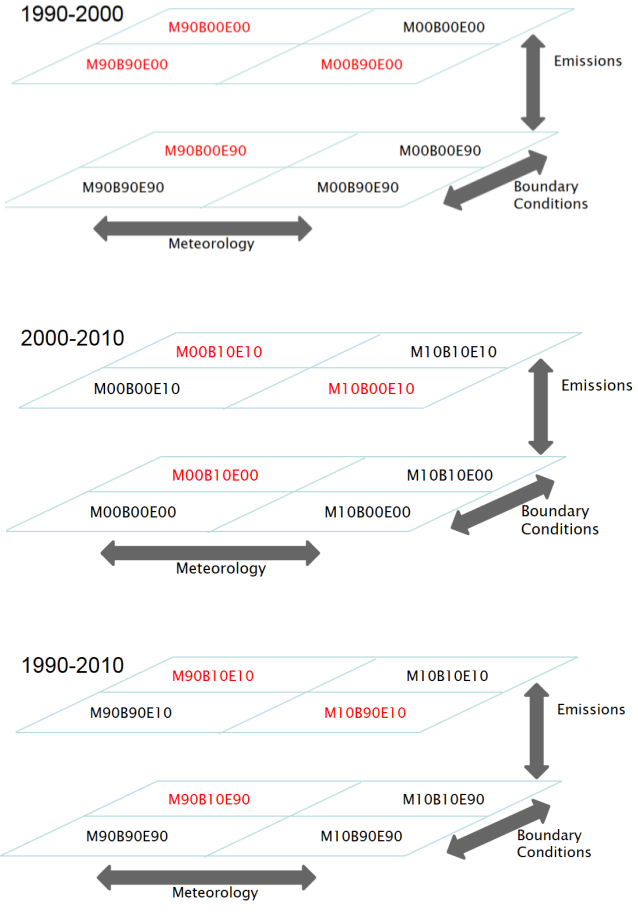
1



2

3 Figure 1 : Combination of sensitivity simulations required to perform the analysis of the contribution of (i)
4 meteorology, (ii) boundary conditions, and (iii) emission changes for the 1990-2000, 2000-2010, and 1990-2010
5 years from the top to the bottom. The key to EDT model simulations provides the 2-digit modelled year for
6 meteorology (M), boundary conditions (B) and emissions (E). Black labels are for the simulations included in the
7 experiment, and red labels are the combinations not produced in any of the tiers of experiments.

8

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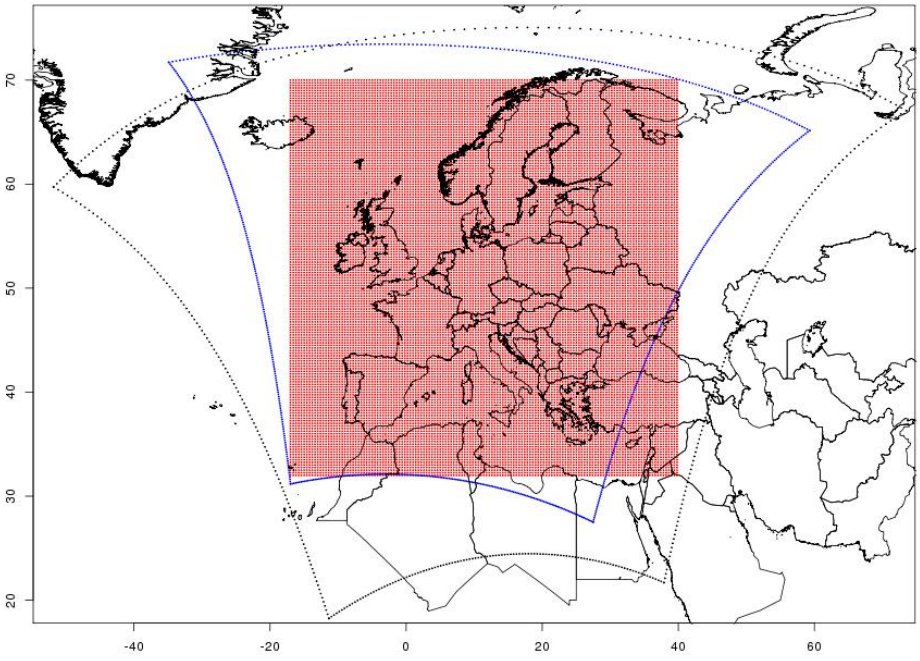

Figure 2: Modelling grid used by all the chemistry transport models involved in Eurodelta-Trends (red dots) with the exception of CMAQB that could not implement a regular latitude/longitude grid (outer grid cell of the modelling domain displayed with blue dots). The outer grid cells of the meteorological forcing data on the EuroCordex grid is also displayed (black dots).

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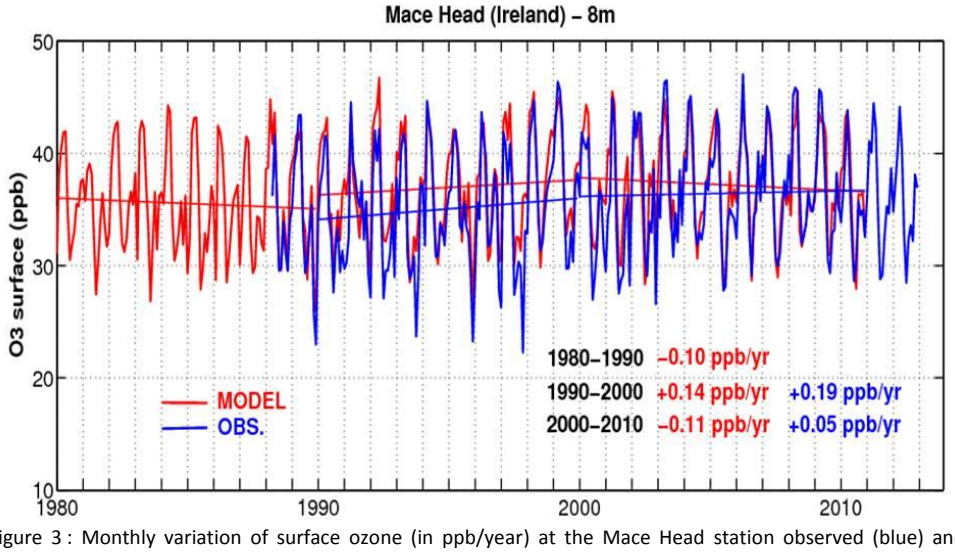
1



2
3 Figure 3 : Monthly variation of surface ozone (in ppb/year) at the Mace Head station observed (blue) and
4 modelled (red) in the CamChem member of the Climate-Chemistry Model Initiative (CCMI)

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