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Modelling Liquid Water Content of Atmospheric Aerosols

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About the author

Mizuo Kajino graduated from Kyoto University, Faculty of Science, in 2000. He received his M.Sc. and began his Ph.D course in 2002 and since then is affiliated with the Severe Storm Research Section, Disaster Prevention Research Institute, Kyoto University. His major interest is in the field of atmospheric sciences, with a focus of his current research on modelling of atmospheric aerosols and their effects on environmental acidification as well as climate change.

As a participant in IIASA's 2003 Young Scientist Summer Programme, Mizuo Kajino was a member of the Transboundary Air Pollution (TAP) Project.

Abstract

The contribution of liquid water to measured aerosol mass was assessed using a numerical approach for European measurement data. Aerosol mass is usually determined after conditioning of the filter under dry condition for a predefined period according to a standard procedure. Consequently, water in aerosols is assumed to be at very low concentrations. However, we find indications that the equilibrium of aerosol is not fully established and water is not completely evaporated after the conditioning.

In order to estimate the aerosol humidity, a mass transfer coefficient was derived for evaporating from the filters. Data from actual measurements of aerosol humidity were applied with a numerical kinetic approach, using the thermodynamic equilibrium as a boundary condition. The calculated coefficient ranges from 10^{-5} to 10^{-4} with an average value of $10^{-4.49}$.

Using this average value the expected water content of aerosol was assessed for a site in Austria. In a data set covering one year with daily samples, water contributed to the total mass between 0.00 percent and 35.9 percent with an average value of 4.1 percent. Most of the samples contained between one and two percent water, according to the calculations. In winter the air is humid and the concentration of inorganic hygroscopic compounds is also high, and so the amount of water uptake from the air is high. Under such conditions more than a half of unidentified components is considered to be water. In summer the amount of hygroscopic components as well as relative humidity is low, water then yields only 8.8 percent of unidentified components. According to the model calculations, it takes about 30 hours on annual average until 95 percent of the initial amount of water uptake is evaporated on the filter and even longer in winter.

The average water content at 49 sites of EMEP in 1999 after conditioning for 48 hours ranges from 0.54 $\mu g\ m^{-3}$ to 3.8 $\mu g\ m^{-3}$, that is 2.6 percent to 18.2 percent to total aerosol mass, while the mass transfer coefficient varies from 10^{-4} to 10^{-5}.

Modelling Liquid Water Content of Atmospheric Aerosols

Mizuo Kajino

1 Introduction

The concentration of aerosols in the atmosphere (mass of atmospheric particles) has been identified in epidemiological studies (Pope et al., 1995 Dockery et al., 1993) as a factor of adverse health effects. Both science and environmental legislation increased their attention to aerosols since, specifically to particles smaller than 10 or 2.5 μm in diameter (PM10 or PM2.5, respectively). In Europe, EU-directive 1999/30/EG prescribes limits to atmospheric concentrations. Scientific measurement programs started and modelling efforts commenced.

Many of the recent atmospheric dispersion models of aerosol formation and transport show considerable discrepancies between model results and observed concentrations of total aerosol mass in Europe. The liquid water content of aerosols may constitute a possible reason for this discrepancy. When relative humidity is very high, hygroscopic components in aerosols take up water from the atmosphere so that particles may grow to droplets that are several times their dry weight. Although aerosol weight is usually determined after storing the filter samples under defined dry conditions, there is the possibility that measured aerosols still contains a certain amount of water. While substantial research has been performed on aerosol water content under the conditions of the real atmosphere (Winkler, 1973; Meng *et al.*, 1995; Ueda *et al.*, 2000), only a few measurements are found about the water content after conditioning (Ohta *et al.*, 1998; Puxbaum *et al.* 2000). Unfortunately, these few data on aerosol humidity are not directly transferable to other experimental designs, as the conditions of conditioning such as relative humidity, temperature and conditioning period are different for each of the aerosol measurements. In this study, we therefore derive the evaporation rates of water in sampled aerosols according to the actual aerosol humidity measurement, then apply them to observations in Europe, and estimate the water content within the values of the measured aerosol mass.

2 Data and methodology

2.1 The European Monitoring and Evaluation Programme (EMEP)

The European Monitoring and Evaluation Programme (EMEP) co-ordinates monitoring from more than 200 air quality stations from different national networks all over Europe. There are about 50 stations where aerosols weight or components are measured. Daily, monthly and annual observation and calculation data of air pollutants from 1977 up to recent years are provided on the web (www.emep.int/index_pollutants.html). The measurement and analytical methods are common in all of their sites. The aerosol mass and chemical components are measured after conditioning under the standard condition of 20ºC and 50 percent relative humidity for 48 hours, which was determined according to European standard, EN 12341 (CEN, 1998).

To calculate water uptake by aerosols, temperature and relative humidity are necessary inputs. These data are not provided by EMEP but kept by national participating organizations. While national organizations were contacted for these data, no useful results were obtained within the timeframe of this study. Due to inaccessibility of measured meteorological information, we use model results, specifically the archived data of NCEP (National Centre for Environmental Prediction) final analysis data (www.arl.noaa.gov/ss/transport/archives.html) for the calculation of water in the real atmosphere just before conditioning.

2.2 AUPHEP

AUPHEP (the Austrian Project on Health Effects of Particles) conducted observations of air pollutants for more than one year at four sites in Austria with continuous monitoring of gases, aerosol parameters as well as daily aerosol sampling with consequent chemical characterization of the aerosols. The samples were weighed after conditioning for 24 hours at 20ºC and 50 percent (Hauck *et al.*, 2003a; 2003b).

2.3 The SCAPE module and mass transfer approach

For calculating aerosol water content at constant conditions, the SCAPE module (Simulating Compositions of Atmospheric Particles at Equilibrium) (Kim *et al.*, 1993a,b; Kim and Seinfeld, 1995, Meng *et al.*, 1995a) was applied. The SCAPE model is a multi-component gas-aerosol equilibrium model that forecasts concentrations of ionic compounds as well as water content at certain temperature and relative humidity. Thus the aerosol water content calculated by the model is determined by the concentrations of the inorganic ionic species in aerosols, the temperature and the relative humidity of the environment. The SCAPE model is based on some not fully realistic assumptions such as internal mixture of the aerosol composition. Still, the model has been applied to several observation data and verified its accuracy (Meng *et al.*, 1995b; Meng *et al.*, 1998, Ueda *et al.*, 2000, Kajino *et al.*, 2003).

Since the SCAPE model cannot calculate the time scale to achieve thermodynamic equilibrium, a mass transfer approach is necessary to simulate the process of aerosol drying during conditioning. In this study, we use a bulk particulate equilibrium concept (Song and Carmichael, 2001) and do not consider any mass transfer limitation. In this assumption the mass transfer coefficient is constant in each sample on the filters and water evaporation proceeds exponentially. The equation is

$$dQ_w \;/\; dt = k_w \left(Q_w^{\mathrm{eq}} - Q_w \right), \tag{1}$$

where Q_w is the water content in aerosols, k_w is the mass transport coefficient of water on the filters and Q_w^{eq} is the equilibrium concentration of water at the standard conditions.

In order to calculate k_w on the filter using measured value of aerosol humidity after conditioning, Equation 1 is turned into

$$k_w = 1 \;/\; t_c \log \left[(Q_{real} - Q_{sta}) \;/\; (Q_{obs} - Q_{sta}) \right], \tag{2}$$

where t_c is the conditioning time and Q_{real} is the simulated water content in the real atmospheric condition of T_{real} and RH_{real}. Q_{sta} is the simulated water content at the standard condition of T_{sta} and RH_{sta}. Q_{obs} is the observed water content after conditioning.

On the other hand in order to calculate aerosol humidity using known values of k_w, Q_{obs} can be introduced by

$$Q_{obs} = (Q_{real} - Q_{sta}) \exp (-k_w \; t_c) + Q_{sta}. \tag{3}$$

3 Results and discussion

3.1 Thermodynamic equilibrium on the filters

Thermodynamic equilibrium of aerosols on the filters measured for EMEP and AUPHEP is supposed to be completely established after several hours in the standard condition at 20ºC and 50 percent relative humidity. This assumes that any water still present in the aerosol is at very low concentrations. Aerosol mass then should be independent of the original relative humidity, with the exception of certain ionic compounds that are highly water-soluble, so that their presence in aerosol depends on the original water content before conditioning. Actually the concentration of hygroscopic inorganic compounds such as SO_4^{2-} and NH_4^+ have sufficiently strong correlation with relative humidity (R=0.50). As information on NH_4^+ and SO_4^{2-} concentrations is available, their contribution can be subtracted and the unidentified (=rest) of the aerosol then should not depend on the original relative humidity during sampling. However, as will be shown below, there are some indications that the water is not completely evaporated and the equilibrium not completely established.

Figure 1 shows a scatter diagram of relative humidity and unidentified components of aerosols measured at AU1, AU2 and AU3 in AUPHEP sites. The unidentified components are defined to be the difference between $PM_{2.5}$ and the sum of inorganic components (IO) and total organic compounds (TC). The concentrations of the unidentified components, which possibly include water, have a much wider scatter and include higher levels as relative humidity become high in the figure.

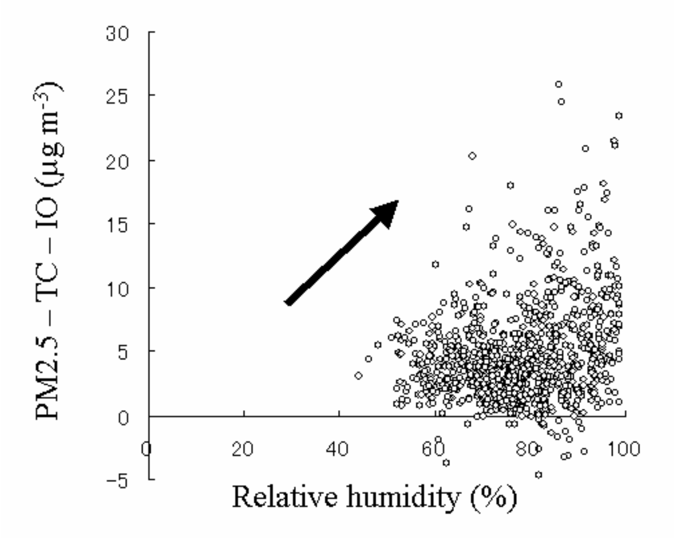


Figure 1. Scatter diagram between relative humidity and $PM_{2.5}$-TC-IO at AU1, AU2 and AU3.

The annual average water uptake calculated by the SCAPE model at the original condition (natural atmosphere) for the urban Vienna site AU1 is 14.0 $\mu g\ m^{-3}$ for a measured total aerosol mass of 36.6 $\mu g\ m^{-3}$. On the other hand the water content at the standard condition is 0.34 $\mu g\ m^{-3}$. The water content with respect to total aerosol mass at the site should be expected within these extremes, the atmospheric conditions (38.3 percent) and the equilibrium after conditioning (0.9 percent).

In order to calculate the water content at the sites in EMEP, NCEP archived data are introduced in Section 2.1. These data are available every 6 hours, 4 times a day, in polar stereographic grids, which reference interval is 190.5km (at 60ºN, 80ºW). One-day averaged temperature and relative humidity at two meter elevation with $1/R^2$ -weighted interpolation are used for the calculation. Figure 2 shows the comparison between measured meteorological data in AUPHEP and the data derived from NCEP. NCEP final analysis data have good agreement with measurement in temperature but not in relative humidity. Since aerosol water content depends critically on humidity, the model analytical data have to be taken with care for this calculation.

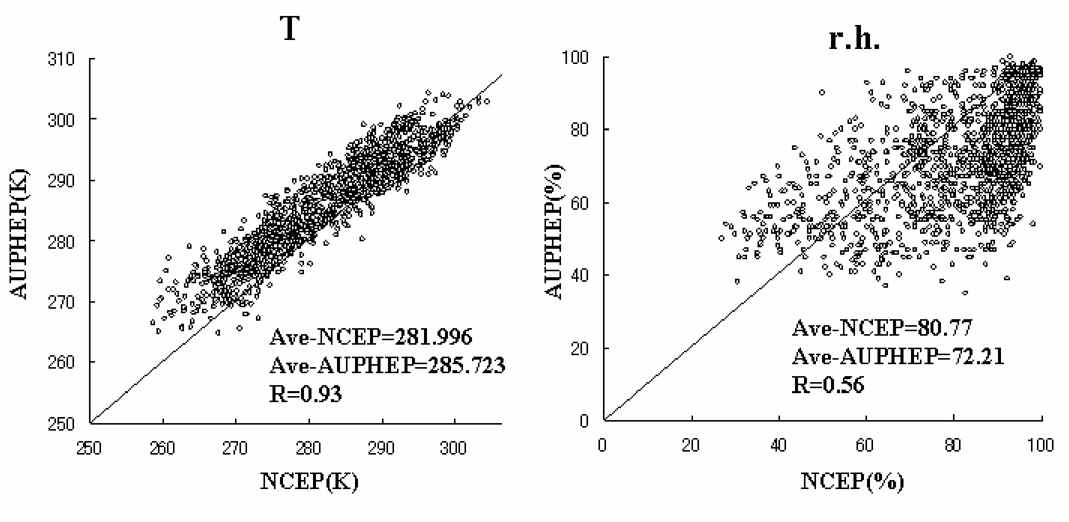


Figure 2. Scatter diagrams between measured and NCEP temperature and relative humidity at AU1, AU2 and AU3.

In spite of the discrepancy about relative humidity, gas-aerosol partitions of volatile compounds measured at the EMEP sites in the Czech Republic in 1999 (CZ01, CZ03) are well simulated by the SCAPE model. The concentrations of both gas and particulate phase ammonium and nitrate were measured every six days at the two sites in this period. The correlation coefficients R between the observed and the calculated 109 values of gaseous NH_3 and HNO_3 and particulate NH_4 and NO_3 concentrations are 0.79, 0.63, 0.75 and 0.72, respectively. This result indicates that the NCEP modelled data are possibly applicable to the simulation of the SCAPE model.

The average value of SPM concentration derived from 11677 samples at 49 sites of EMEP in 1999 is 20.9 µg m^{-3}, while the calculated water content in the NCEP condition (considered to be the real atmosphere) is 18.7 µg m^{-3}. The water content at equilibrium state under standard condition is 0.54 µg m^{-3}. The averaged concentration of the sum of measured sulphate, nitrate and ammonium is 3.2 µg m^{-3} (as much as 15.3 percent of the SPM concentration), so the difference to the total is less than the aerosol water content calculated for ambient conditions. After sampling preparation (conditioning), water content is expected to be clearly lower than 18.7 µg m^{-3}, but for the sake of consistency the water content ratio in the SPM concentration is given here as a range between 2.6 percent and 89.5 percent.

3.2 Mass transfer coefficients derived from the aerosol water content measurements

Ohta *et al.* (1998) and Puxbaum *et al.* (2000) measured the water content (humidity) of samples after conditioning under certain conditions. In their studies, humidity was determined as the weight loss of the filter in the temperature range from room temperature to 100ºC. As no evolution of organic compounds was detected in this temperature range, all of the weight loss is considered to be water.

Ohta *et al.* (1998) performed semi-monthly aerosol sampling on the 11mmφ Teflon filter using low volume sampler at a flow rate 20 l min^{-1} with 2 μm of 50 percent cut-off diameter in Sapporo (northern part of Japan) for one year from November 1991 to October 1992. Samples were collected for 14 days continually and stored at less than 30 percent relative humidity for more than a day. To calculate the mass transfer coefficient of water on the filters k_w according to Equation 2, we use the monthly averaged temperature and relative humidity in Sapporo for T_{real} and RH_{real}, 20 ºC and 30 percent for T_{sta} and RH_{sta} , 24 hours for the conditioning time t_c. The calculated mass transfer coefficients are shown in Figure 3. The horizontal axis shows the sampling periods, the left vertical axis denotes the mass-transfer coefficient k_w (note the logarithmic scale) and the right vertical axis presents the total mass of a sample (in mg). There is negative correlation between the transfer coefficient and the aerosol mass (as the aerosol mass becomes larger, the evaporation rate decreases). This implies the possibility of coagulation on the filter because the bulk coefficient should depend on the size distribution of aerosols. The average value of k_w is $10^{-4.49}$ and it ranges from $10^{-4.66}$ to $10^{-4.17}$. As the k_w values are dependent on the conditioning time, the averaged k_w would be $10^{-4.8}$ when t_c is 48 hours. However, none of the samples is stored more than 48 hours (Sachio Ohta, personal communication). The annual average values of the water content calculated by the SCAPE model are 6.2 μg m^{-3} when they were sampled and 0.00 μg m^{-3} when they would reach at the equilibrium in the standard condition while 0.42 μg m^{-3} of water were actually measured after the conditioning.

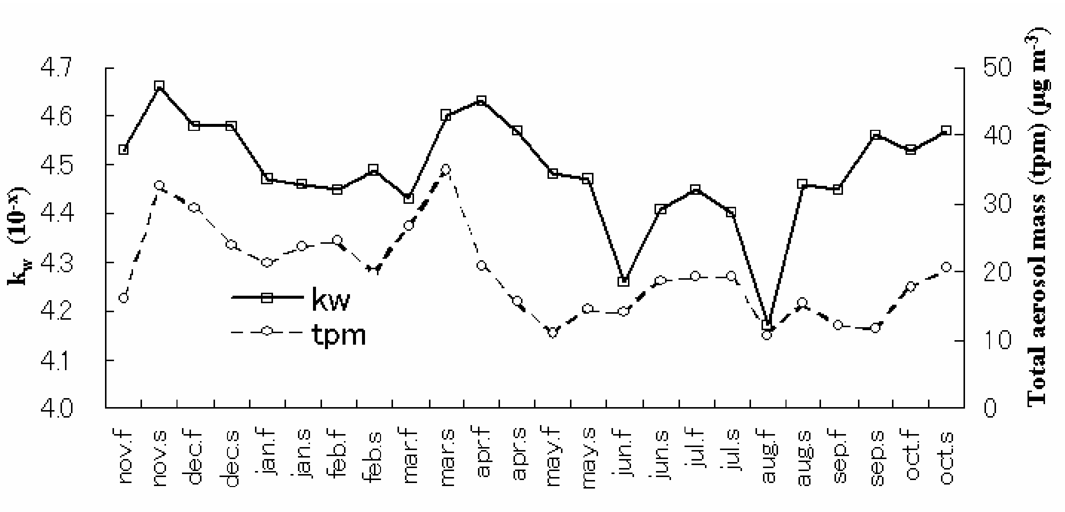


Figure 3. Mass transfer coefficient of water on the filters calculated from the measurement of Ohta *et al.* (1998)

Puxbaum *et al.* (2000) performed semi-diurnal aerosol sampling on 47mmφ quartz fibre filters using the air sampler at a rate of 2.6 m^3 h^{-1} with 30μm of upper cut-off mass median diameter at Nylsvley Natural Reserve on sub tropical savannah in the north-eastern South Africa Republic for the week from 4 May 1997 to 10 May 1997. After sampling, samples are carried by the airplane to Vienna and then there stored under the condition of 22 ºC and 50 percent for 24 hours. Since the conditions in the airplane are completely unknown, we define T_{real} as 22 ºC and RH_{real} as 60 percent or 70 percent in constant. The averaged values of water content calculated by the SCAPE model are 15.7 μg m^{-3} at 70 percent, 8.7μg m^{-3} at 60 percent and 0.68μg m^{-3} at 50 percent where measured value is 2.85μg m^{-3}. The averaged value of k_w is $10^{-4.65}$ and its range is from $10^{-4.98}$ to $10^{-4.45}$ when RH_{real} is set at 70 percent. On the other hand when RH_{real} is set at 60 percent, the averaged value is $10^{-4.82}$ and the range is from $10^{-5.25}$ to $10^{-4.53}$. There are no correlation between the coefficient and the aerosol mass.

3.3 Aerosol water contents in Europe

The water content in the aerosol mass measured by AUPHEP is estimated with the mass transfer coefficients of water on the filter discussed in Section 3.2. The coefficients are supposed to be dependent on factors like the total aerosol mass on the filter, the cut-off diameter of the sampler and the diameter of the filter. As the 50 percent cut-off diameter of the sampler in AUPHEP is 2.5μm, the coefficients are expected to be close to those of Ohta *et al.* (1998). However, within AUPHEP hi-volume samplers were used at a rate of as much as 500 *l* min^{-1}, so that the total aerosol amount on the filter is 15.3 mg sample^{-1} on average in spite of their short-period samplings (7.9 mg sample^{-1} for 14 days in Ohta *et al.*, 1998). It should also be noted that they use 150mmφ quartz fibre filter (11mmφ Teflon filter in Ohta *et al.*, 1998). This is why the water content should be estimated with a range of the coefficients. However, afterward the coefficients are assumed constant and not variable on the aerosol mass, independent of all factors and conditions because the relationships between them are uncertain at all.

Figure 4 shows the water content in the measured aerosol mass at AU1, where k_w varies between 10^{-5} and 10^{-4}. The vertical axis is the water content in μg m^{-3} and the horizontal is the mass transfer coefficient. The value to the left is the water content at equilibrium state in the standard condition. The average water contents are 14.0 μg m^{-3} in the real atmosphere and 0.34μg m^{-3} at equilibrium in the condition of 20 ºC and 50 percent. When the coefficient is 10^{-5}, half of the initial amount of water remains on the filter after 24 hours. 24 hours are enough to establish equilibrium in case that the coefficient is $10^{-4.2}$. When the coefficient is $10^{-4.6}$, the water is almost evaporated after 48 hours, but still about one third of water remains after 24 hours. If the coefficient is less than $10^{-4.8}$, more than 48 hours are necessary for complete evaporation.

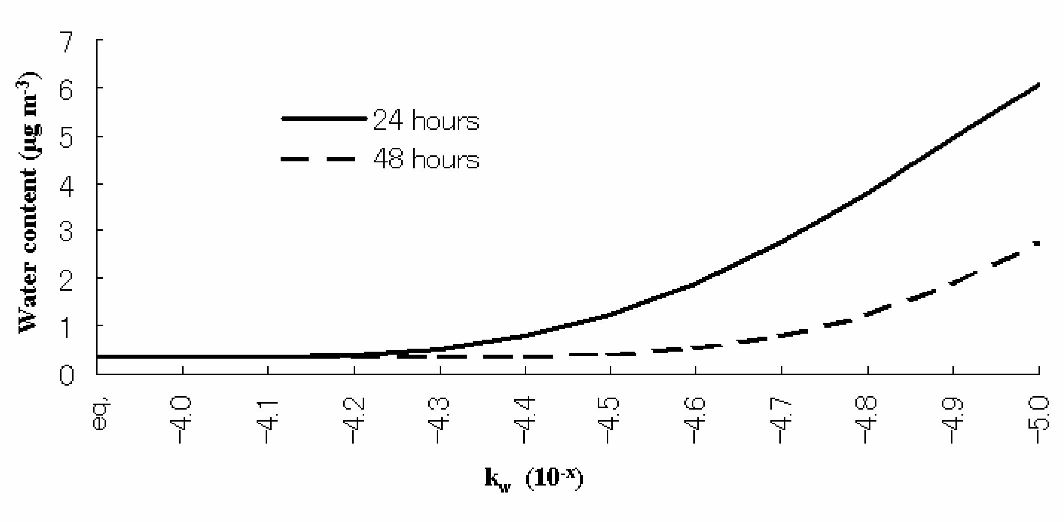


Figure 4. The water content in the measured aerosol mass at AU1 while k_w is variable between 10^{-5} and 10^{-4}.

Figure 5 shows the ratio of water content to measured $PM_{2.5}$ mass at AU1 for k_w of $10^{-4.5}$ as the average value derived from Ohta *et al.* (1998). The horizontal axis depicts the classes of ratios and the vertical axis is the number of samples (days) out of 343 samples in a year. The most frequent range is from one to two percent, annual average is 4.1 percent, the maximum 35.9 percent and the standard deviation is 20.0. The seasonal variations should be discussed because it is usually more humid in winter than summer in Europe and the aerosol concentrations get higher in winter season as well.

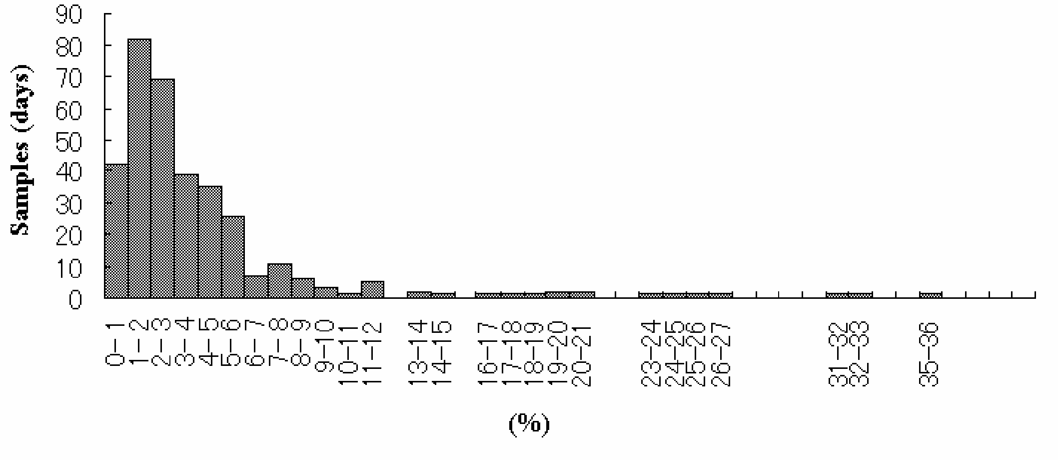
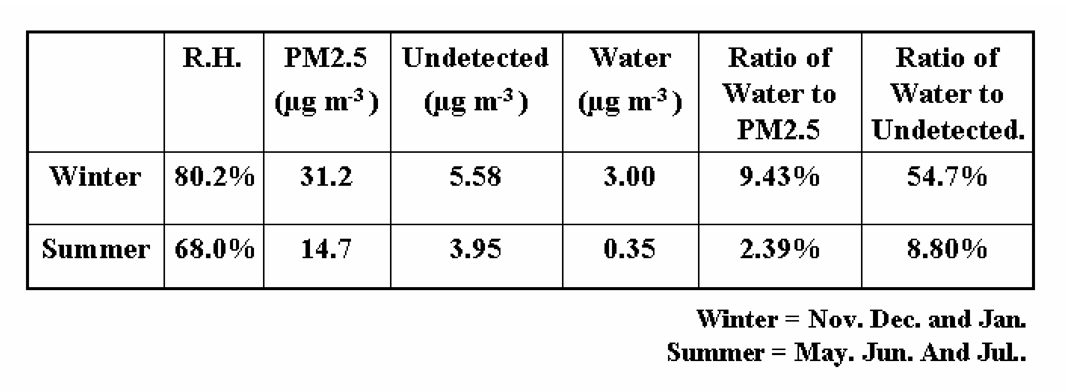


Figure 5. The ratio of water content to measured $PM_{2.5}$ mass at AU1 when k_w is $10^{-4.5}$.

Table 1 shows the difference of the water content at AU1 between the winter and summer seasons. We select the most humid months, November, December and January for the winter season and May, June and July for the summer. The averaged relative humidity is 80.2 percent in winter and 68.0 percent in the summer. The measured aerosol concentration in winter is twice as high as in the summer. Unidentified components, i.e., the difference between $PM_{2.5}$ and the sum of IO and TC, is in winter 50 percent higher than in the summer. The water content after conditioning ($k_w = 10^{-4.5}$, 24 hours) is 3.0 µg m^{-3} in the winter and 0.35 µg m^{-3} in the summer. During the winter season, 10 percent of $PM_{2.5}$, i.e., more than half of the unidentified components are supposed to be water. Figure 6 shows pie charts of the components in total suspended particles (TSP) at AU1. The left chart shows annual average, the centre is average in the summer season (same as Figure 5) and the winter to the right. The values under each chart are TSP concentrations. The unidentified components here exclude calculated water. There are more small particles and less coarse particles in winter than in summer. As a result of higher humidity and more inorganic hygroscopic particles, which cause higher water uptake and longer time to achieve equilibrium, the water content ratio is much higher in winter.

Table 1. The seasonal difference of the water content at AU1.



	R.H.	PM2.5 (µg m^{-3})	Undetected (µg m^{-3})	Water (µg m^{-3})	Ratio of Water to PM2.5	Ratio of Water to Undetected.
Winter	80.2%	31.2	5.58	3.00	9.43%	54.7%
Summer	68.0%	14.7	3.95	0.35	2.39%	8.80%

Winter = Nov. Dec. and Jan.
Summer = May. Jun. And Jul..

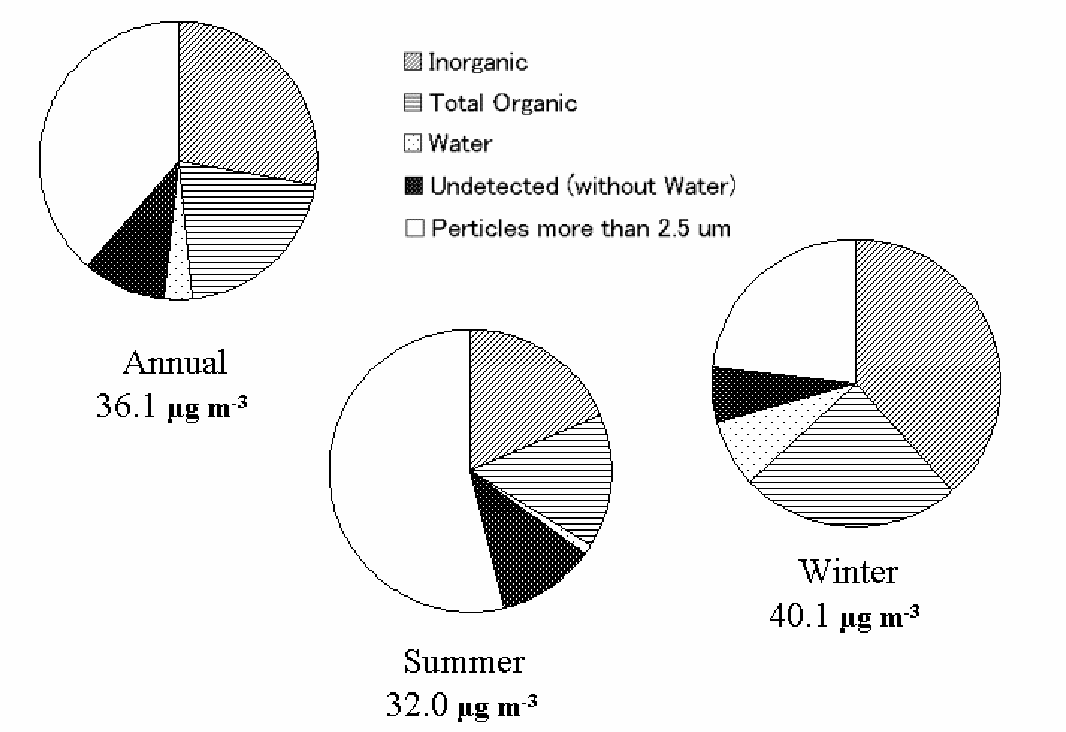


Figure 6. The pie charts of the components in total suspended particles (TSP) at AU1. The left is annual average, the centre is average in summer and the right is in winter.

Figure 7 shows the time to achieve equilibrium using different mass transfer coefficients at AU1. The vertical axis is the annual average of the water content and the horizontal axis is the conditioning time after sampling. The average water content just after sampling is 14.0 µg m^{-3} and the amount when the equilibrium is completely established is 0.34 µg m^{-3}. The four lines on the figure are drawn until the time that 95 percent of the initial amount of water is evaporated (the same time that the water content is decreased to 1.02 µg m^{-3}). With a coefficient of $10^{-4.5}$, the equilibrium of water on the filter is almost established after about 30 hours, but it takes more than two days for $10^{-4.8}$ and more than three days for $10^{-5.0}$. Moreover, it is important to note that these values are annual mean so that longer periods are necessary especially for winter season.

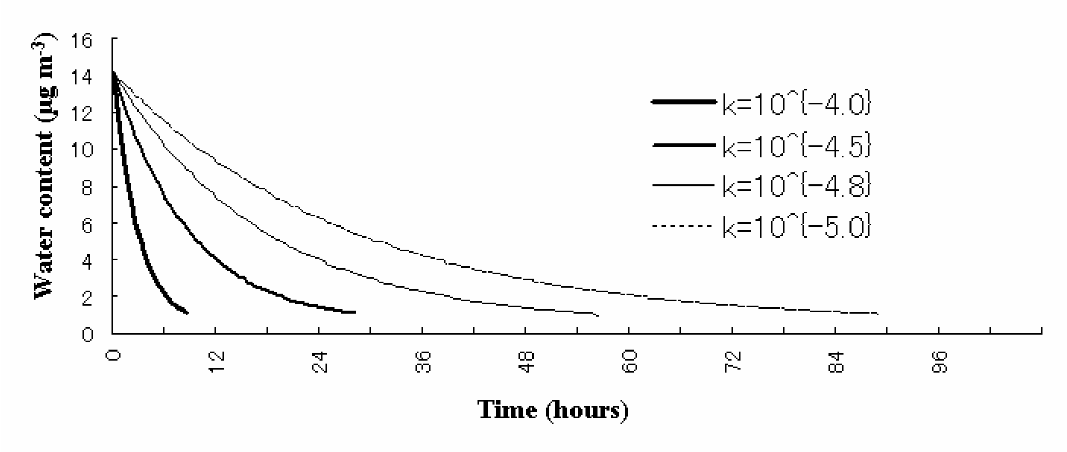


Figure 7. Annual means of time variations of water evaporation to achieve equilibrium state using different mass transfer coefficients at AU1. The lines are plotted until 95 percent of the initial amount of water is evaporated.

The average water content on the filter at 49 sites of EMEP after conditioning in 20 ºC and 50 percent for 48 hours are 0.54µg m^{-3}, 0.61µg m^{-3}, 1.7µg m^{-3} and 3.8µg m^{-3} for k_w of $10^{-4.0}$, $10^{-4.5}$, $10^{-4.8}$ and $10^{-5.0}$ respectively. These values are 2.6 percent, 2.9 percent, 8.1 percent and 18.2 percent of SPM concentration, respectively.

4 Conclusions

The thermodynamic equilibrium of aerosols on the filters will not be established after conditioning under dry conditions for several hours and a certain amount of water still remains in the measured aerosol mass.

The mass transfer coefficient of water on the filters, derived from the measurement of aerosol humidity after conditioning, ranges between 10^{-5} and 10^{-4} (the average is $10^{-4.49}$) and shows negative correlation with the total aerosol mass. As the aerosol concentration is higher, evaporation rate of water becomes lower. This implies the possibility of coagulation of aerosols on the filter.

Several examinations using k_w derived from the measurements of aerosol humidity focus on measurement of the AU1 station. For a k_w of $10^{-4.5}$, the water ratio in the total mass ranges from 0.0 percent to 35.9 percent with an average of 4.1 percent and the most frequent range between one and two percent. In winter, with humid air (average RH is 80.2 percent) and a high amount of inorganic hygroscopic components (about one third of the total mass), water uptake from the air is abundant and the aerosol humidity ratio in the unidentified components is calculated at 54.7 percent. In summer, when the average RH is 68.0 percent, the ratio of 8.8 percent is calculated. For average conditions, it typically takes about 30 hours until 95 percent of the initial amount of water will be evaporated. The time scale to achieve complete evaporation in winter takes longer than in summer.

The average water content at the 49 sites of EMEP in 1999 after conditioning for 48 hours ranges from 0.54 $\mu g\ m^{-3}$ to 3.8 $\mu g\ m^{-3}$, that is 2.6 percent to 18.2 percent to total aerosol mass, while the mass transfer coefficient varies from 10^{-4} to 10^{-5}.

While the water content in measured aerosol mass is not negligible (important especially in the winter), it does not explain the full difference between many model results and observations. Also the methodology developed in this paper is associated with many uncertainties. Since the water content after conditioning and the time to achieve equilibrium range vary widely according to the k_w value and the k_w value, further experiments are necessary to determine the k_w value and to predict the water content in aerosols more accurately.

In terms of the gap between aerosol models and measurements, the results of this paper indicate a potential to improve both: even after a longer conditioning of filters, which will decrease the water content to the thermodynamic equilibrium and decrease the measured mass, models will still not be able to fully predict observed concentrations. Furthermore, it becomes evident that the actual aerosol mass in the atmosphere is clearly higher than the measured weights indicate. It is however not clear at this time whether this water content may increase or potentially even mitigate health effects induced by atmospheric aerosols.

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