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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Lessons learnt from the first EMEP intensive measurement periods" *by* W. Aas et al.

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We would like to thank both reviewers for constructive comments to the manuscript.

We have repeated the reviewers' text and responded after each point. In addition to these comments, we have discovered a mistake in Figure 2 where the winter ammonium in PM1 gravimetric accidentally have got too high number (2.03 instead of 1.11). We will update the figure and text accordantly.

ANONYMOUS REFEREE #1

REVIEWER: "The manuscript presents a honest status report on state-of-the-art aerosol measurements and modeling for Europe. It includes a comprehensive list of potential problems, difficulties and uncertainties regarding all aspects of aerosol sam-

pling, measurements and modeling. There are basic problems in aerosol mass determination, a variety of sampling artifacts, lack of unified analytical methodologies, problems with size-cuts and instrumental capabilities, incomplete emission data, limitations of the modeling approach, to name just a few. Despite the inhomogeneity of the data the positive outcome is the relatively good match between measurements and modeling results (whatever it means).The manuscript is organized to imply that a perfect match would be desirable (using the verbs underestimate and overestimate), though previously it is admitted that sampling and measurements are themselves loaded with high biases and uncertainties due to a variety of fundamental problems. Thus, the lack of perfect agreement may not necessarily mean that the model results are 'under' or 'over' any true value set by the measurements. On the contrary, it follows that there is no 'true value' at all, a better expression would be that model and measurement do not agree. Because of the large temporal and spatial variability of atmospheric aerosols, their size distributions, chemical and physical properties, chemical formations and transformations, interactions with water vapor and droplets, nucleation, volatilization and a host of other factors, one should never expect to capture aerosol properties measured at a few sites in a short campaign with an aerosol model of 50x50 km resolution. This fundamental constraint should have been better stressed in the manuscript."

RESPONSE: We fully agree, and it is an important point. There is no true value in either of the approaches (measurements or model). The under- and overestimated terminology is nevertheless a common way of describing comparison between these two estimates. However we do agree that this may be misleading and suggest to reword all the over- and underestimation terminology with: "model gives XXX% less (or more) than observations". In this way we don't imply what the truth may be. We also suggest adding a paragraph describing the limitations of the comparison as Referee 1 suggest. I.e. by using similar wording as given by him/her: "The reader should notice that there are fundamental limitations in how good the model and measurements can resemble the same thing, due to the large temporal and spatial variability of atmospheric aerosols, their size distributions, chemical and physical properties, chemical

ACPD

12, C3530–C3536, 2012

formations and transformations, etc. “

REVIEWER: Page 3738 Line 19 There is no unbiased gas/particle separation. Using denuders the gas-to-particle equilibrium is disturbed and some volatilization of particle-phase ammonium-nitrate can be expected.

RESPONSE: OK, we agree to some extent though one would expect the bias to be marginal. We suggest rewrite the sentence to: “The only exception is IT01, which used the reference denuder/ filter method where one would expect only little if any bias in the gas/particle separation.”

REVIEWER: Page 3744 Line 4 check grammar RESPONSE:Yes, this was wrong. Suggest changing the sentence to: “IT04 and CH02 also enhanced ammonium nitrate was observed in January 2007.

REVIEWER: Page 3747 Line 12 The best example for the limitation of models with respect to point measurements is the disagreement between measured and modeled sulphate concentrations. Sulphur-dioxide has the best emission inventory, well-established chemistry and size-distribution, by far the longest history of modeling experience, and particlephase sulphate is free of sampling and measurements artifacts. Yet the fit between modeled and measured values is not at all better than that of any other aerosol component.

RESPONSE:The EMEP model generally represents sulphate better than e.g. the nitrogen species when looking at a larger dataset than what is the case for the limited numbers in this study. (ref e.g. Supplementary material to EMEP Status Report 1/2011, www.emep.int). However, we agree that the difference in performance between the different aerosol components (at least SO4,NO3, NH4) is rather small. This can probably at least partly be attributed to uncertainties in modelling of dry and wet depositions of the aerosols, which is difficult for all of the species. Furthermore, although emission inventories of SO2 are well known, information of the temporal distribution (e.g. the summer to winter ratio) is not so well known. We suggest to add a paragraph similar to

ACPD

12, C3530–C3536, 2012

this in the manuscript.

REVIEWER: Page 3748 Ammonium-nitrate deposited on filter samples may be prone to losses or gains due to changing equilibrium conditions during or after sampling. Such changes cannot be captured in models which treats instantaneous ammonium-nitrate equilibria

RESPONSE:Yes agree, we add a sentence on this.

REVIEWER: Page 3756 Line 25 Check grammar RESPONSE:Change has to have

ANONYMOUS REFEREE #1

REVIEWER: This manuscript presents atmospheric measurements of aerosols during the first EMEP intensive measurement periods. The focus of the measurements was to characterize the chemical composition of the aerosols. The experimental data was compared using the EMEP model. The work presented in this manuscript is very important from the practical point of view. There is a need for more accurate model in order to be able to fully characterise various aspects of the particulate pollution within Europe. In general, the work is carefully done and the manuscript shows results that are novel and should be published in ACP. There are some issues that need attention before potential publication, but these can be considered minor in nature.

Main comment This manuscript presents experimental data and model results that differ significantly in various parts. It would be valuable if the authors focus in discussing the uncertainty estimates of their results. This discussion can be found in parts within the manuscript, but it would deserve a separate section, especially as the difference is sometimes rather large. In general, it would be interesting to see if the observed differences can be explained by known uncertainties either in measurements or in model runs.

RESPONSE:Yes, this is an important point and probably needs a separate chapter or paragraph. We suggest adding a paragraph in the conclusion about this. It is however

ACPD

12, C3530–C3536, 2012

difficult to give one exact number for the model uncertainty for PMx - though various tests have been performed to investigate the model sensitivity to different inputs and processes. The measurements uncertainty is also difficult to quantify since there are few parallel measurements to really address this. And as pointed out by referee 1 neither model nor measurements necessarily tell the truth, adding more difficulties to actually address the uncertainties in a quantitative way and it's somewhat beyond the scope of this paper. We suggest adding the following paragraph at page 3759 line 28:

There are relatively large uncertainties in both measured and modeled estimates, but to quantify this is very difficult. The main reasons for model uncertainty are uncertainties in input data (e.g. emissions, meteorology, landuse, etc.) and uncertainties in processes descriptions in the model. The accuracy of model calculations varies a lot for different PM components, with SIA aerosols being better understood (though still far from being perfectly represented by the model), while SOA and windblown dust being rather uncertain. In the measurements the uncertainties are both related to measurement method itself and the performance of the analysis. For further details on the uncertainty in assessment of PM in Europe the reader is referred to the EMEP PM status report in 2011 (EMEP Report 4/2011).

REVIEWER: P 3737, L 20: Please report the temperature within TEOM. This is important piece of knowledge in discussing the role of volatile compounds within the instrument.

RESPONSE:These TEOMs have temperature of 50 degrees. We'll add one sentence with this information.

REVIEWER: P 3742, L 25: Give arguments for splitting the coarse nitrate evenly between PM2.5 and PM10-2.5. I have some difficulties understanding the meaning for coarse, if half of it is below 2.5. micrometers.

RESPONSE:In the model, coarse nitrate represents nitrate aerosol formed on sea salt and mineral dust. When comparing calculated PM2.5 with observations, we account in

ACPD

12, C3530–C3536, 2012

a crude way that a portion of the nitrate associated with sea salt and dust resides on aerosols with diameters smaller than 2.5 um, and thus contributes to PM2.5 mass.

In this model version, we assume the Mass Median Diameter (MMD) of coarse nitrate being 2.5 um (whereas fine nitrate has MMD of 0.33 um). That means that about a half of the coarse nitrate mass is associated with particles smaller than 2.5 um The assumption is based on a number of observations in Europe (e.g. Pakkanen, 1996; Pakkanen et al., 1996; Mehlmann and Warneck, 1995; Ottley and Harrison, 1992), showing that the MMD of coarse nitrate mostly ranging between 2.15 and 2.8 um (or being within 2-4 um size range measured with a cascade impactor). Also results from Kerminen et al. (2000) suggest a MMD for nitrate formed on sea salt lying between 1.5 and 2.5 um. Some other measurements in the US and China showed MMD for coarse nitrate somewhat larger than the aforementioned European observations (Lee et al., 2008; Zhuang, 1999; Tanner et al., 2001). On the other hand, coarse nitrate formed on dust particles may have MMD larger compared to sea salt associated nitrate (e.g. 3.8 ug as in data by Pakkanen et al., 1996). Thus, in the areas of large influence of mineral dust, the EMEP model would probably overestimate nitrate in PM2.5. The way coarse nitrate is split between PM2.5 and PM2.5-10 is rather uncertain, and currently work is in progress to implement an explicit formation of nitrate on sea salt and dust aerosols, which is going be a more sound process description. This is now better explained the updated manuscript.

It can be mentioned that in the most recent version of EMEP model, the fraction of coarse NO3 contributing to PM2.5 is reduced to 28%.

REVIEWER: P 3743, L 1: Explain in more detail how the aerosol water is calculated from ambient RH and T. What does “PM chemical” stand for?

RESPONSE:It s a mistake, it should be: “PM chemical composition”. This will be corrected. PM water is calculated with the MARS equilibrium model (see p. 3742, L. 13-16).

ACPD

12, C3530–C3536, 2012

REVIEWER: P 3743, L 8-9: Is the gravitational settling the only deposition mechanism? For fine fraction, this is not relevant for deposition.

RESPONSE:This is correct, we suggest to rewrite to: "Dry deposition velocities for aerosols are calculated following Venkatram and Pleim (1999), accounting for aerodynamic and laminar sub-layer resistances and also for gravitational settling of larger particles."

REVIEWER: P 3744, L 29: What does the authors refer to by argumenting that the central European sites are relatively more influenced by anthropogenic sources? More than south European sites or north European sites or east European sites?

RESPONSE:Yes, south and north of Europe are more influenced by natural sources, relative to the PM mass. South Europe is influenced by mineral dust while primary biological particles is (relative) important in north contribution relatively more to the coarse fraction than in the middle of Europe. We don't have much chemical composition measurements in East of Europe. To clarify we suggest rewriting the sentence the following:

"the highest PM2.5/PM10 is commonly seen for central European sites, which are relatively more influenced by anthropogenic sources., i.e. mineral dust in south of Europe and PBAB in northern Europe are contribution relatively more to the coarse fraction of PM10.

REVIEWER: P 3774, Fig 3: In the figure legend, what does PM2.5(-PM1) stand for? Why is it different compared with PM10-PM2.5?

RESPONSE:PM2.5(PM1) is the difference between PM2.5 and PM1 for those sites having both these measurement, otherwise it is representing the PM25 fraction. We add a sentence like that in the figure caption to clarify this better.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 3731, 2012.