**Atmospheric
Chemistry
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Discussions**

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Comment

Interactive comment on "Illustrating the benefit of using hourly monitoring data on secondary inorganic aerosol and its precursors for model evaluation" *by* M. Schaap et al.

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General comments

The paper by Schaap et al. provides a first comparison of the inorganic compounds predicted by a European regional chemical transport model against an annual dataset of hourly measurement data obtained with an online system capable of providing a good separation between gas and aerosol phase. A more robust capability of modelling European nitrate is important to improve our predictive capability in assessing the impact of emission reduction strategies on climate and exceedance of air quality standards. The work highlights some important discrepancies between model and

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Interactive Discussion

Discussion Paper

measurements of generic importance that should be of interest to the wider scientific community. I do, however, suggest that the authors discus some of the limitations of the model / measurement intercomparisons more clearly, and put the results into better context of other intercomparison studies and additional Dutch data (including further data from the MARGA) prior to final acceptance for ACP. Based on this, the authors should attempt to draw firmer conclusions as to where the deficiencies lie.

The manuscript is mainly well written, with some minor suggestions to improve the English provided below.

Major Scientific Comments

Contribution from coarse aerosol The main problem of the intercomparison appears to me that this study compares model results for the inorganic fractions that are expected to be contained in PM1 with measurements of PM10. The latter contain coarse nitrate (balanced by sodium and, possibly, calcium) as well as sea salt sulphate. Although this is mentioned in the paper, the magnitude of the problem and the consequences are insufficiently explored and discussed throughout. The authors should investigate the charge balance in the measurements to investigate the importance of $NaNO_3$. For example, in Dec the model appears to get the NH_4^+ concentration about right, while nitrate and sulphate are underpredicted. This implies a difference in the charge balance between model and measurements. The authors cite a report by Weijers et al. (2010), which incidentally I failed to find at www.pbl.nl, for the importance of coarse nitrate at this site, but I am aware that the MARGA measured, at least for some time, aerosol composition of PM2.5 and PM10 (as pointed out by Referee 1) and additional campaign-based measurements are available from a MARGA sizer and a MARGA / Aerosol Mass Spectrometer comparison. Thus, the authors should be able to quantify the importance of the coarse components at this site.

Similarly, the authors need to discuss the potential effect of internal vs. external mixing on the equilibrium. For example, the higher NO3a measured during daytime in July

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(Fig. 7) may be due to the contribution of non-volatile NaNO3.

It is unclear to me, why the model does not treat coarse aerosol in its current form as these authors appear to have used the LE model before, e.g. to simulate Na in the NL (Manders et al., 2010).

As pointed out by Referee 1, the MARGA provides measurements of other compounds, such as Na, SO2 and HCl (as mentioned on P 12346, but then not further used in the text). These time-series should further help elucidate the reasons for model / measurement discrepancies. For example, much of the HCl is thought to be derived from reaction of HNO3 with seasalt. Because HNO3 loss to sea salt is not treated in the model, the modelled HNO3 concentration may agree better with the sum of measured HNO3 and HCl? Performance for SO2 may shed light on the reasons why sulphate may be underestimated? The Na concentration should enable coarse sea salt sulphate to be quantified?

layer height. How good is the description of the boundary layer height in the model? If the model gets the baseline concentrations about right, but underpredicts the high episodes, this may be because the vertical resolution in the model is insufficient or the boundary layer height in the model is overestimated. The tall Cabauw tower and profiler measurements during the intensive campaigns at this site should provide information that could be used to investigate this influence. At the moment, the authors do not draw conclusions on weather model / measurement discrepancies are most likely related to the coarse mode, prediction of the boundary layer height, absolute and relative temporal pattern in emissions, the thermodynamic equilibrium model or kinetic constraints on evaporation / condensation. By taking into account further information on the importance of the coarse mode (see above), other compounds and the boundary layer height, the authors may be able to constrain the options further.

Local ammonia sources. If the site is affected by local ammonia sources (P12354, L4), this surely would affect the other concentrations also, if the equilibrium is as fast

as is assumed in the model. What would be the consequences? By the way, NH3 concentration can be very variable in urban environments, due to kerb site increases related to emissions from catalytic converters.

Link to other EMEP work. I agree with Referee 1 that a further reference is needed to the EMEP work. However, the proposed reference to the Aas et al. (2010, in prep.) paper should not be dropped, because it integrates the work presented here with other work in the Special Issue. In fact, the authors may want to add links to some further papers in preparation for the EMEP Special Issue, such as Nemitz et al. (2010a, b), Twigg et al. (2010) as well as Mensah et al. (2009) and her paper in preparation for the APCD EUCAARI Special Issue. Is the long-term concentration time-series from the MARGA at Cabauw being written up separately? It would be useful to provide details on this.

Inconsistency with earlier Dutch model validation exercises. The authors fail to explain why the results from this study are inconsistent with the earlier work of Manders et al. (2009). Is this due to the characteristics to the Cabauw site, the measurement periods or instrumental issues? As pointed out by Referee 1, sulphate should not be lost from the filters.

Minor Scientific Comments

Section 4.3. As pointed out later in Discussion, this approach (and the LE model) assume instantaneous equilibrium, while in reality there is a kinetic constraint on evaporation, while vertical exchange (including deposition) and subgrid variability linked to local sources helps to counteract the equilibrium.

Table 2. The correlation coefficient is insufficient for quantifying model skill and other metrices should be included (e.g. RMSE, bias).

P12347, L16. A reference to the CBM-IV gas-phase module would be helpful.

P12348, L1. Was the standard version of the LE model used for this study or were any

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Interactive Discussion

Discussion Paper

parameters already adjusted in light of the intercomparison results?

Section 5.1. I agree with Referee 1 that the authors should not only make recommendations for Dutch monitoring, but also for the EMEP Measurement Strategy. In this context they may want to mention that long-term monitoring by MARGA has commenced at other European sites, such as Melpitz (EMEP/TFMM presentation), Helsinki, Auchencorth (Twigg et al., 2010) and Harwell, UK.

Technical Comments

P12343, L11: Erroneous use of apostrophe.

Introduce all acronyms on first use, e.g. 'EMEP' (P12343, L16), 'PM' (P12345, L1), 'DELTA' and 'MARGA' (P12344, L15), 'LE' (P12348, L22). It would also be good to introduce EMEP and CLRTAP in a sentence or two.

Add charges to all chemical ion formulae (e.g. P12345, L21; P12346, L15/16; P12348, L27/28; Caption to Tables 1 2).

P12345, L21. I suggest you mention that this refers to $PM10$ here, as this allows readers to interpret the values of Table 1 more directly. Otherwise this information only comes on the next page.

P12345, L27. I suggest rephrasing to: "This distinguishes the MARGA from the GRAEGOR instrument, which measures NH_4^+ as the only cation, by means of a selective diffusion membrane."

P12346, L23. 'offsets' rather than 'off sets'.

P12347, L4. Better: 'model aimed at simulating air pollution'

P12347, L6. Better: 'studies directed at PM'

P12348, L14. Better: 'albeit the levels being underestimated.'

P12349, L9 and L11: Better: 'correlated with'

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Interactive Discussion

Discussion Paper

P12349, L15. Better: 'which is explainable with the role'

P12350, L24. Better: 'albeit the absolute concentrations being too low'

P12353, L10. 'results indicate that it is necessary'

References

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