

Interactive comment on “Aerosol modelling in Europe with a focus on Switzerland during summer and winter episodes” by S. Aksoyoglu et al.

Anonymous Referee #1

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General comments

The authors present a modeling study of aerosols in Europe including a comparison with experimental data obtained at two sites in Switzerland. They used an existing air quality model (CAMx) in combination with a meteorological model (MM5). The experimental data are rather detailed and provide a useful basis to diagnose some important aspects of aerosol characteristics in Switzerland. The PM chemical composition is reproduced satisfactorily by the model. However, model performance is uneven, with better performance for carbonaceous particulate species than for inorganic PM components. In summary, this is an interesting study, which deserves publication following

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revisions to complement and/or clarify a few points, as discussed below.

Specific comments

P. 10801, lines 21-24: It is not clear why the authors only mention the volatility basis-set (VBS) approach as a way to minimize the underestimation of organic PM by air quality models. The reason the VBS approach led to some improvements in the comparison between models and measurements is mainly due to the fact that it took into account the gas-phase fraction of semi-volatile POA and simulated the oxidation of semi-volatile POA. This important addition to aerosol modeling does not necessarily require the VBS approach and it is now taken into account in other modeling approaches. Furthermore, the reduction of the discrepancy between models and measurements has also been accomplished by taking into account other processes such as the oligomerization of some SOA (Morris et al., 2006; Carlton et al., 2010), the formation of SOA via aqueous chemistry (Chen et al., 2007; Carlton et al., 2008), the addition of new precursors (Morris et al., 2006; Zhang et al., 2007; Carlton et al., 2010), the treatment of SOA hygroscopicity (Pun, 2008) and the treatment of SOA formation under low-NO_x conditions (Carlton et al., 2010). These other aspects should be mentioned explicitly in the introduction with the appropriate references.

P. 10803, line 16-18: It should be noted that the treatment of oligomerization in CAMx is rather simplistic and entirely empirical. It should be noted that oligomerization may be pH-dependent (e.g., Pun and Seigneur, 2007) and that further work is needed to better understand and correctly simulate this process.

P. 10803, line 18: It should be mentioned that aqueous-chemistry of SOA is not taken into account.

P. 10804, lines 20-21: A reference is needed for the EC/OA split.

P. 10808, last paragraph: Reducing the wind speed by a factor of two is a rather brutal way to improve model performance. Although it is justified by the comparison of the

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MM5 wind speeds with the observed wind speeds for selected periods, it should be noted that the meteorological fields used in CAMx are no longer internally consistent. The authors mention that the recalculation of the vertical wind speed in CAMx leads to a mass-conserving wind field; however, this diagnostic approach to wind modeling only pertains to the wind field. The Navier-Stokes equations relate those wind speeds to other variables (temperature, pressure, relative humidity) and those equations are no longer verified when the wind speeds are independently modified. Furthermore, the authors need to explain how they treat the transition of the wind fields from a period when there is no modification of the wind field (moderate wind speeds) to one with modified wind speeds (low wind conditions). It is misleading to state in the abstract that the MM5/CAMx system was used as it suggests that a fully prognostic approach was used for meteorological modeling. In reality, a diagnostic approach was used for some periods for the wind field using MM5 to develop an initial wind field and observations to calibrate the modeled wind field. This point should be mentioned in the abstract should be modified to avoid any confusion for the reader.

P. 10810, line 5: In figure 14, it seems that some PM1 concentrations occasionally (e.g., January 24 in Zürich) exceed PM10 concentrations; some explanation is in order.

P. 10810, line 16: Performance statistics should be provided for PM10 and PM1/PM2.5.

P. 10811, line 7: The definition of the MFB performance criterion is incorrect; it should be $-60\% < \text{MFB} < +60\%$.

P. 10811, lines 6-9: The authors should elaborate. For example, it should be noted that the performance goals (error less than 50% and bias within - & + 30%) are obtained only for OA in Zürich and for EC in Payerne. Performance criteria are not met for sulfate in Zürich and nitrate and ammonium in Payerne in June.

P. 10812, lines 27-28: The authors mention that particulate nitrate is sensitive to the temperature, which is to be expected considering that the ammonium nitrate equilibrium constant is temperature- and relative humidity-dependent. One would expect the

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semi-volatile SOA to be also sensitive to temperature; see for instance Pun et al. (2008) who investigated this issue via modeling and measurements. The authors should discuss the sensitivity or lack of sensitivity of SOA to temperature.

P. 10813, first full paragraph: Did the authors include dry deposition for the SOA gas-phase fraction? Bessagnet et al. (2010) showed that neglecting their dry deposition may have a significant effect on SOA concentrations (leading to greater concentrations when it is neglected). This point should be discussed.

P. 10813, line 15: The use of a 50% change in the emissions is high. Emission control strategies typically are in the 10 to 20% range. For example, a 15% change is typically used by EMEP for emission scenario studies. Since the system is non-linear, it is not clear whether the map obtained for a 50% change would be similar to that with a 15% change. It would be appropriate to confirm this result with smaller emission changes.

Editorial comments

P. 10801, line 20; Replace "several factors" by "a large amount".

In several places, aerosol phase is used instead of particulate phase (e.g., p. 10803, line 20; p. 10812, line 17). Since the aerosol includes both the particles and the surrounding gas, it is better to refer to the particulate phase.

P. 10808, line 12: Model layers are horizontal; delete "vertical" or refer to "vertical levels".

P. 10810, line 12: Use subscript for PM1.

References

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