

Interactive comment on “Towards an online-coupled chemistry-climate model: evaluation of COSMO-ART” by C. Knote et al.

C. Knote et al.

christoph.knote@empa.ch

Received and published: 4 November 2011

S. Aksoyoglu

sebnem.aksoyoglu@psi.ch

Received and published: 23 September 2011

SC.0) This paper describes the evaluation of online-coupled model COSMO-ART. Since we have some experience for some of the modeling periods discussed in this manuscript, I'd like to give my comments and suggestions below.

SC.1) Although a lot of measurements were used for model evaluation, I find the model performance is overstated in the manuscript since important processes such as aque-

C992

ous chemistry and wet deposition are not yet implemented in COSMO-ART. In order to make it clear for the reader at the beginning, I suggest that authors mention the missing processes in the abstract.

Wet deposition of aerosols is included in the model, it is only missing for gases. We have included a sentence in the abstract mentioning that these areas need to be improved: (penultimate sentence of abstract) “Suggestions for further improvement of the modeling system consist of the inclusion of a revised secondary organic aerosols scheme, aqueous-phase chemistry and improved aerosol boundary conditions.”

We have already changed the text to more accurately reflect the model's ability due to requests from the reviewers, so we think this will be covered there.

SC.2) There is one more process -not mentioned in the manuscript - which could cause an overestimation of nitrate. Lack of wet deposition might lead to overestimation of nitric acid in the gas phase and consequently overestimation of particulate nitrate.

We thank S. Aksoyoglu for mentioning this and we have added it to the discussions of nitrate overestimation. Section 4.3 now reads:

“4.3 Nitrate

The most substantial bias found in our simulations is an overestimation of nitrate aerosol components. This is not a new phenomenon and seen also in other model evaluations (e.g. Stern et al., 2008). Accurately modeling this species is challenging (Dentener and Crutzen, 1994), as it represents the result of a dynamic, coupled system between gas- and aerosol-phase, depending on the amount of gas-phase precursors, temperature, relative humidity and aerosol composition (cf. Chapter 9 in Seinfeld and Pandis, 2006). We have tested several hypotheses to understand this deficiency in our model. We could exclude an erroneous nighttime or daytime chemistry (e.g. providing too much HNO₃) and emission sources (too high levels of NO_x). Evaluation against nitrate totals (daily averages of gas-phase HNO₃ + particulate NO₃⁻ from impregnated

C993

filter packs at the station Payerne (CH)) showed some high bias, but the overestimation is much smaller than for nitrate alone. Three hypotheses seem likely: the lack of sulfate, missing wet deposition of HNO₃ and inaccuracies in the model's ability to reproduce relative humidity and temperature well enough. In experiments, available ammonia is first neutralized by sulfuric acid and only if no more sulfuric acid is available, nitric acid serves as a replacement to form NH₄NO₃ aerosols (Seinfeld and Pandis, 2006). Suppose now that ammonia is limited, then the mass of nitrate found in the aerosol depends also on the amount of sulfate substantially. As our simulations currently underestimate sulfate, the higher amount of available NH₃ will combine to "excess" NH₄NO₃. As mentioned earlier, we could not assess the accuracy of the modeling system regarding NH₃ concentrations due to the lack of measurements. If ammonia is overestimated this will strongly influence this system as well. Secondly, due to the missing wet scavenging of gases, HNO₃ is not removed from the atmosphere as effectively as in reality. As HNO₃ is a very hydrophilic substance it will easily transfer to the aqueous-phase and is therefore efficiently scavenged. This could lead to the observed too high levels of total nitrate, which are then subject to gas-aerosol partitioning. Thirdly, the gas-aerosol partitioning for nitrates has a strong temperature dependence. If our model cannot represent daytime temperature maxima and minima to a high degree of accuracy, this will lead to errors in the partitioning. It is also known that the phase-state solid/liquid is a strongly non-linear function of aerosol chemical composition and relative humidity (see Figs. 2, 5 and 7 in Nenes et al., 1998), consequently changing the partitioning behaviour between gas and aerosol phase. As the lifetime of gas-phase HNO₃ is much shorter than for particulate NH₄NO₃, too strong partitioning to the aerosol phase results in too strong survival of total HNO₃ + nitrate in the atmosphere, which contributes to overpredictions of nitrate at later times. In summary: firstly, if the model underestimates sulfate more ammonia is available to form nitrate aerosols, which leads to an overestimation. Secondly, if not enough HNO₃ is removed more total nitrate is available for partitioning. Thirdly, even small differences in modelled temperature and relative humidity compared the situation at the instrument could change the nitrate

C994

gas/aerosol partitioning. Finally, if NH₃ concentrations are overestimated themselves, even more NH₃ is available to neutralize NO₃, and even more ammonium-nitrate is formed. We think those processes together explain a large part of our overestimation of nitrate aerosols. Other models are better able to simulate mean nitrate concentrations, for example the Comprehensive Air quality Model with extensions (CAMx) / particulate matter CAMx (PMCAMx) (e.g. Aksoyoglu et al., 2011; Andreani-Aksoyoglu et al., 2007) or WRF/Chem (e.g. Li et al., 2010), but once the diurnal cycle of gas/aerosol partitioning of nitrate is looked at, also these modeling systems exhibit problems. We have seen that there are substantial model deficiencies to accurately describe nitrate aerosols and that this is a general problem also for other, comparable model systems. One straightforward way to improve the situation will be to increase the horizontal resolution of the simulations to better represent the variability in temperature and relative humidity. A better knowledge on NH₃ emissions and concentrations is needed. The impact of the implementation of a comprehensive wet phase chemistry scheme will be investigated in a future study."

SC.3) There is not enough information about the pathways for SOA, what precursors are treated, how the yields are, whether SOA module is still based on the original SORGAM by Schell et al., (2001).

Our paper is an evaluation paper that should give an overview of model performance. It can therefore not exhaustively treat all the subprocesses in the model. It is clearly stated in the methods section that we parameterize SOA with SORGAM (Schell, 2001). As we use it in its original formulation this should be the correct way to describe the module. This also provides the necessary information for the reader where pathways and yields can be found. The manuscript is already very long so we think it is acceptable not to include more information here.

SC.4) Page 1821, first paragraph: the statement "very good agreement" (also in the conclusions) is based on mean values over all stations (Figure 2, Table 2). In line 4, it is written: "Sometimes the diurnal variability is underestimated by simulations (not

C995

shown)". It would be more useful to show time series, diurnal variations especially for wind speed to justify that agreement is "very good".

Time series at single (possibly non-representative) points are equally if not even more restrained in their informational content than are means over a large number of stations averaged over time. As stated in a response to reviewer 1 we did not focus on meteorology as our meteorological core is extensively evaluated and continuously verified by several European weather services which use it as their operational modeling system. Our setup closely follows the operational setup of the German Weather Service (DWD) - except data assimilation - and we consequently do not expect large discrepancies in quality. Evaluation of means (and time evolution) showed that there are no big differences, in fact they are very small (differences in wind direction typically below 10° , temperature ± 3 K). Figure 1 shows a timeline of meteorological parameters at Payerne (CH) for the summer 2006 period to underline our claim.

SC.5) Figure 21, top panel: Is there any reason for not using AMS data after 23rd June?

The reason we did not use AMS data at Payerne after the 23rd was a technical one (additional BC data has been available up to the 23rd and we did only plot data where both AMS & BC data was available. We have added data after the 23rd now and the new plots are shown in Fig. 2.

SC.6) Figure 21, lowest panel: It seems that there is no OA for Harwell measurement data (only inorganic aerosols are seen in the lower panel). But somehow the total seems to be similar to PM_{2.5} data at Harwell. I think this needs to be checked.

We thank S. Aksoyoglu for checking the AMS data for the 2006 period. After rechecking the data we decided to remove this plot, as the data we collected does not compare with other representations. We cannot determine the source of the error but can exclude our plotting methods as culprit. See Figure 1 for the new version.

C996

SC.7) Page 1834, line 11-14: "in the model it is almost exclusively POA". The description of SOA formation pathways in COSMO-ART would help to understand why it is like that.

We agree with S. Aksoyoglu that there is a lot of more information to be found by closer examining the data. However, we do think that this is a too detailed assessment of a single component of the modeling system for such an evaluation. The pathways can be looked up in Schell, 2001 for those looking for further information.

SC.8) Page 1838, Sulfate: We modelled the same period June 2006 with CAMx model which includes aqueous chemistry and wet deposition (Aksoyoglu et al., ACP, 2011). Comparison with our findings for the same period would give an idea about the contribution of cloud chemistry to sulfate.

We do not think that we can easily derive the contribution of cloud chemistry to sulfate by comparing these two quite different modeling systems. The setup in Aksoyoglu et al., ACP, 2011 uses CAMx, an offline-driven model with a different gas-phase chemistry, completely different aerosol formulation (2 buckets CF scheme for coarse / fine aerosols vs. 2-moment size resolved scheme), different emissions datasets, different spatial resolution. We have however added sentences to inform the reader that another modeling system has been used to model this period: "Aksoyoglu et al. (2011) simulated the summer 2006 period with another modeling system including in-cloud oxidation of SO₂ and found better agreement. Whether this can be attributed to in-cloud sulfur oxidation is unclear."

SC.9) Page 1839, line 15: Authors state that their SOA parameterization has too low yields. It would be very useful to see the yields used in this study.

The yields used are the same as in Schell (2001). See our response to a similar comment above (SC 3).

SC.10) In the section of "Organics" biogenic SOA is not mentioned at all although SOA

C997

from biogenic emissions can be very important especially in summer.

We have added a sentence: "Emissions of biogenic SOA precursors, and the effectiveness of the conversion pathways, are still in discussion and will also contribute to the discrepancies found."

SC.11) Page 1841, lines 15-20: "We think those three processes together explain a large part of our overestimation of nitrate aerosols. Other models experience similar problems in simulating aerosol nitrate, for example CAMx (PMCAMx) (e.g. Aksoyoglu et al., 2011, Andreani-Aksoyoglu et al., 2007) or WRF/Chem (e.g. Li et al. 2010) once the diurnal cycle of aerosol nitrate contributions is looked, and not only the mean". These sentences give the impression that nitrate overestimation is a general problem in models. However, CAMx version used in our studies includes aqueous chemistry and wet deposition, therefore sulfate and nitrate cannot be compared to those described in this manuscript. Our experience suggest that over-and under estimation can happen due to input parameters (e.g. boundary conditions, emissions) but not due to the CAMx model itself.

It is unclear to us what is meant with the last sentence. If the CAMx has no influence on the concentrations of nitrate / sulfate does this mean you do not have to run it at all? Table 3 in Aksoyoglu et al. 2011 e.g. states mean NO₃- measured 0.99 ug/m³, 0.97 ug/m³ modelled - which is really good. However, mean fraction bias of -91% means the variability is wrong. For nitrate this is mostly dominated by the diurnal cycle - which is what I intended to point at with our sentence. We have revised this paragraph and made it clear that other models have no problem in simulating nitrate in general: "Other models are better able to simulate mean nitrate concentrations, for example the Comprehensive Air quality Model with extensions (CAMx) / particulate matter CAMx (PM-CAMx) (e.g. Aksoyoglu et al., 2011; Andreani-Aksoyoglu et al., 2007) or WRF/Chem (e.g. Li et al., 2010), but once the diurnal cycle of gas-/aerosol partitioning of nitrate is looked at, also these modeling systems exhibit problems."

C998

References:

Aksoyoglu, S., Keller, J., Barmpadimos, I., Oderbolz, D., Lanz, V. A., Prevot, A. S. H., and Baltensperger, U.: Aerosol modelling in Europe with a focus on Switzerland during summer and winter episodes, *Atmospheric Chemistry and Physics*, 11, 7355–7373, doi:10.5194/acp-11-7355-2011, 2011.

Andreani-Aksoyoglu, S., Keller, J., and Prevot, A.: Aerosol Modelling with CAMX4 and PMCAMX: A Comparison Study, *Air Pollution Modeling and Its Application XVII*, pp. 247–256, 2007.

Schell, B., Ackermann, I., Hass, H., Binkowski, F., and Ebel, A.: Modeling the formation of secondary organic aerosol within a comprehensive air quality model system, *Journal of Geophysical research*, 106, 28 275, 2001.

Interactive comment on Geosci. Model Dev. Discuss., 4, 1809, 2011.

C999

PAYERNE (MIL/AUT) (SYNOP)

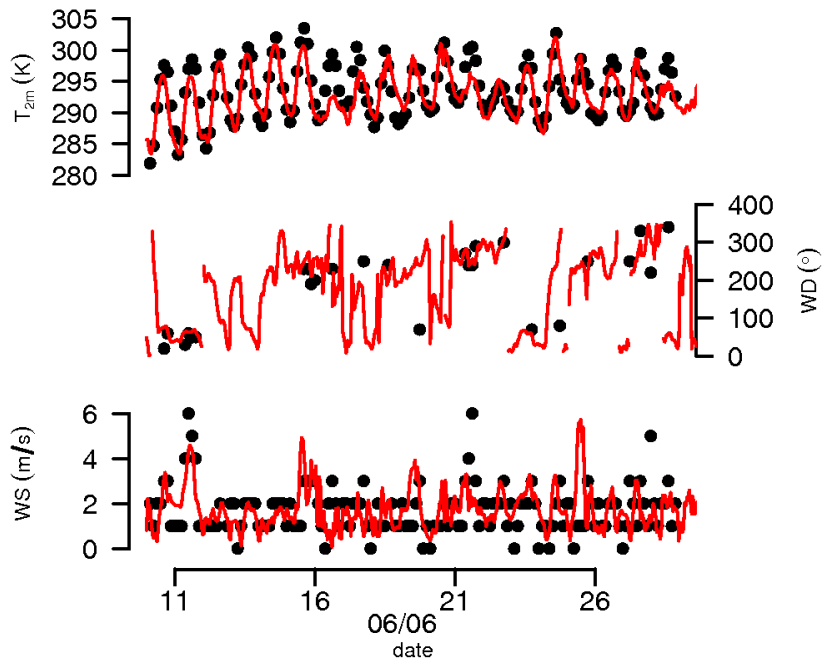


Fig. 1. Timeline of meteorological parameters as simulated by COSMO-ART (red lines) and measured by station of the SYNOP network (black circles) for the summer 2006 period.

C1000

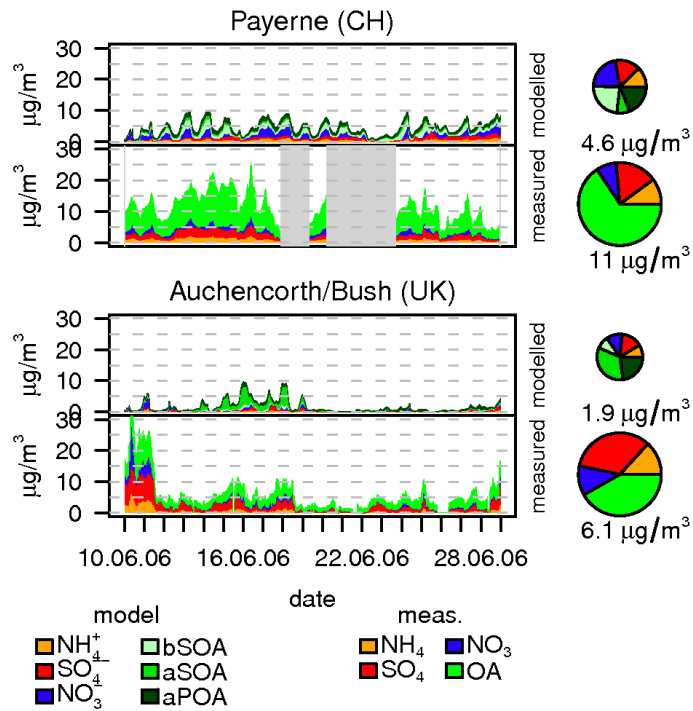


Fig. 2. revised version of Figure 21.

C1001