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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Secondary inorganic aerosols in Europe: sources and the significant influence of biogenic VOC emissions especially on ammonium nitrate" *by* Sebnem Aksoyoglu et al.

Anonymous Referee #1

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Review of Aksoyoglu et al., "Secondary inorganic aerosols in Europe: Sources and the significant influence of biogenic VOC emissions especially on ammonium nitrate"

In this article, the authors apply CAMx to two intensive monitoring periods, one in the cooler season one in the warmer season. CAMx is applied using its particulate source apportionment technology (PSAT). They double the biogenic emissions inventory to test how that impacts the formation of inorganic aerosol. They find that doubling the biogenics reduces inorganic particulate nitrate. This is tied to the reaction of nitrate radicals with terpenes. They also found that sulfate was mainly of foreign origin.

First, such a paper should have a performance evaluation in the main body of the paper. Simply saying that the chemical components are well captured by the model is not

sufficient. Actual metrics should be provided. They can show this very economically using soccer or bugle plots, along with some traditional performance metrics. The evaluation should consider the available monitors across the modeling domain. In the supplement, they provide only time series pictures, which can be very deceiving depending upon the scales chosen. They should look at all of the work done as part of the AQMEII and follow that lead. Numerical results for the performance on ozone, aerosol nitrate, sulfate and ammonium, and gaseous precursors should be in the text. (I will note, when I look at the time series, it would appear that the model is not performing well, but showing the numerical evaluation would either confirm or negate that view. . . the numerical performance measures should be given either graphically or in a tabular fashion. I think the bugle or soccer plots are best as they can show what is considered reasonable compared to past applications.)

Second, while it is good to also consider periods where intensive measurements are available, it would be good, here, to use annual simulations to limit the bias in interpretation that may be derived from using such short periods. If they were using the detailed measurements to make some process changes in the model, that would be different. Here, the measurements are used in a rather limited fashion. It is also a problem when they only show detailed results for one day (in this case, 14 June). How does this compare to other days. Provide a longer time series or provide a summer and winter average. The limited time period is also of concern when suggesting so much sulfate is coming from the boundaries. This brings up a real concern: is this article meant to support policy decisions or for science (this should be answered in the response to review, not the article). If it is to support policy-making, definitely a longer set of simulations are required. If it is for science, deeper investigation is required (in addition to a longer simulation to show how the period used for more intense investigation represents a typical period). If the period is atypical, that is fine. It just needs to be known.

They used CAMx with PSAT. It should be made clear that PSAT shows where the

species (Nitrate, ammonium, sulfate) originates, but it is not a source impact. If all of the reduced N is removed, most of the oxidized N will also go away. Given the non-linearities in the system, they should also run a series of zero-out simulations, where they zero out the major source categories of interest. These can be compared. This should be computationally quite reasonable.

How do their estimates of nitric acid formation from N2O5 s. OH compare with other historical estimates?

When I go on line, I do not see the "Rest" on their map in the supplement.

Given the huge uncertainties in the NO3-organic and NO2-organic radical and sulfate-BVOC reactions, the finding that doubling the biogenics reduces SIA should be accompanied, quite prominently, this uncertainty. How well does the model reproduce BSOA (biogenic SOA) formation, particularly from terpenes and via the IEPOX pathways? How was this assessed or addressed?

The discussion of NO3 nighttime dynamics lacks context and references, e.g., work done by Seinfeld and co-workers as well as a variety of articles by Platt and coworkers starting, in the early 1980s. They should detail what is new here. This section could also benefit from tracing the HNO3 formed by each reaction. Specifically, while nitric acid is efficiently deposited, the average deposition rate is about 1 cm/s, leading to a lifetime of about a day. It appears more HNO3 is formed during the day, so there is plenty still around at night formed during the day. Keep in mind, the HNO3 formed in the afternoon has little time to deposit. Note how quickly the NO3 raises when the air gets cool enough? The HNo3 is only being formed at a rate of o.o4 ppb/hr, which is likely not fast enough to supply the nitrate shown to be formed. Isn't much of this left over from during the day?

In summary, at present there are a number of items that need to be conducted and/or addressed before the paper should be accepted for publication. First, the model evaluation should be brought forward and discussed, and should include numerical overall

performance measures, potentially shown as soccer and/or bugle charts in the text and a more detailed set of statistics (not just some time series plots) in the supplemental. In particular, the ammonium and nitrate simulations across the domain should be evaluated and considered closely, and the ability of the model to capture BSOA should be brought out. The model should be run to examine how levels respond to removing a few major sources to show how those results compare with the PSAT results. There can be major nonlinearities that are not found when just using PSAT. It would also be advisable to run full year simulations. They need to put their results in context with past studies, e.g., look at the review by Platt and Heinz (1994) and the early work by Seinfeld and co-workers (as well as the recent work, e.g, by Nga et al. 2015). If these items are done in the revision, it would be acceptable for publication.

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