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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The simulations of sulfuric acid concentration and new particle formation in an urban atmosphere in China" *by* Z. B. Wang et al.

Anonymous Referee #2

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The authors present a a manuscript in which simulations of the sulphuric acid concentrations and particle formation are presented for an urban site in China. The study compares simulated sulphuric acid concentrations with measured ones, likewise with the concentrations of small particles. The simulation results agree at least qualitatively, and in many cases also partly on a quantitative level. However, the new knowledge emerging from the study seems slightly thin. Models should generally be seen as tools to elucidate the most important gaps in our knowledge of physical an chemical processes, and I think that the authors might try to go into more details on the key uncertainties of their assumptions they have made in their simulations. At the moment,

the study mainly presents results of tuning the model to fit measurements; this is not a problem as such, but in my opinion the tuning parameters and their implications should be discussed in more detail.

In the following, I will present some questions and comments that may be of interest when improving the manuscript:

1. Sulphuric acid modelling

As the authors state, the MATLE model clearly underestimates the H2SO4 concentration in all the cases presented. However, it seems to me that the problem lies mainly with the time period of the rising concentrations before noon: there seems to be a clear time delay of ca 2 hours between the rise of the measured and simulated concentrations, while the decay of the concentration in the afternoon is predicted quite well. If this is due to the underestimation of OH production from HONO photolysis, this would then have also effects on the estimate of condensing and nucleating vapours produced in the model. I think this should be discussed more extensively when evaluating the simulation results and their goodness.

2. Aerosol formation modelling

First and foremost, I am interested in whether simulations of aerosol formation utilised simulated or measured sulphuric acid concentrations. The simulated nucleation particle concentrations seem to follow measured concentrations very closely (Fig 3 and 5); this would to me give strong support to the role of sulphuric acid if measured SA was used, but on the other hand, I would be surprised if simulated SA (which has a clear time offset) would explain the particle concentrations this well. Can the authors clarify this? As one of the main take-away points of the manuscript is that on- and offset of particle formation is well-predicted, I think this is a key issue.

My second question concerns the performance of the simulation for so-called non-event days. To me it seems that the model clearly overpredicts particle formation on

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days when no particles are formed. This is quite interesting: this is clearly not due to overprediction of sulphuric acid, but some other mechanism is at work. The fact that particle formation is predicted even on non-event days contradicts the statement in the abstract stating that on- and offset can be predicted: for me, also time periods of no formation should be predicted for this to be true.

Regarding the modelling of [H2SO4][Org] - type particle formation (J_het): The temporal behaviour of this type of nucleation seems to be several hours off. If I have understood correctly, the parameters going into J_het are

i) measured sulphuric acid (this I assume from visually comparing figures 1d and 5)

ii) a product that results from the reaction of simulated OH and measured (or simulated?) alpha- and beta-pinene.

iii) a nucleation coefficient K_het, which is higher than previously

From this, to me one possible explanation for the time offset would be the underprediction of OH, especially before noon (as was seen for the sulphuric acid). This situation, with both measured SO2 and measured sulphuric acid and the condensation sink, would call for a rough estimate of the OH concentration from those data and comparing that to the modelled OH concentration, to shed some light on both the sulphuric acid time offset and J_het time offset. Additionally, I would suspect that using such estimated OH would reduce the need to tune K_het to a higher value.

A question of interest in studies of aerosol formation is the relative contributions of different vapours to particle growth. The simulation's performance regrading growth has been discussed very lightly apart from the statement that a condensing organic vapour yield of 0.5% was used. This seems to me to be a very low value. A more extensive description of the possibly condensing vapours, an overview of the condensing vapour concentration, and a comparison of the simulated and measured aerosol volume/mass would give a clearer picture on the accuracy of the simulation. Also, as the properties

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of the condensing vapour (molecular weight, diffusion coefficient) affect the growth; what are the assumptions made regarding these properties?

It would also be of interest to see the specific contributions that the different oxidation mechanisms (OH, Ozone, NO3) give to the added volume of aerosol, as this has been a topic of research lately.

Summarizing, I consider the manuscript as relevant and interesting, but would suggest that the authors spend some more effort in analysing the performance of the model and engage in a more detailed discussion on the specific aspects in which the simulated concentrations and aerosol parameters differ from the measurements.

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