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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sulphuric acid closure and contribution to nucleation mode particle growth" *by* M. Boy et al.

Anonymous Referee #1

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General comments:

In this manuscript, a closure study for gaseous sulfuric acid is presented, in addition to which the contribution of this vapour to nuclei growth has been estimated. The analysis relies mainly three weeks of measured data gather during a very intensive field program. In general, I consider this manuscript original, well-structured and scientifically sound. The authors should, however, to do a little bit better job in discussing some of their results. After addressing the critical comment given below, and making the necessary corrections into the text, I welcome this manuscript as part of Atmospheric Chemistry and Physics..

Specific comments:

It is kind of disturbing that equations (1), (4) and (5) include the "particle radius", while

throughout the discussion in the text the “particle diameter” is used. I understand that these equation have originally represented as a function of particle radius; however, for consistency I recommend that the equations will be rewritten as a function of particle diameter.

The analysed air masses have been divided into “less polluted” and “higher polluted”. Trajectory-based classification of air masses is always subjective, and sometimes one may end up with situations where seemingly (based on trajectories) clean air masses have picked up significant amounts of pollution, or seemingly polluted air masses are relatively clean (because of errors in trajectories). The authors should briefly comment on how “easy” or “consistent” their air mass classification was for the chosen cases.

In Table 4, the agreement between measured and calculated sulfuric acid concentrations is measured with the ratio “measured/calculated concentration”. However, the discussion in Section 5.2 has been made in such a way that following it would be much easier if the ratio in Table 4 would be other way around, that is “calculated/measured concentration”. This would also be scientifically more appealing: comparing something that is predicted (calculated in this case) with something that is supposed to be correct (measured in this case) is more straightforward when using the ratio predicted/measured.

The authors mention in Section 5.2 that their NMHC measurements include only monoterpenes (page 6353, lines 23-24). In order to avoid confusion among readers, this should be clearly brought up much earlier, preferably in section 2.4. At the very least, they should state that measured monoterpenes act as a surrogate for all NMHC’s and then justify this assumption later (as they have done in the sensitivity analysis).

I think that in the general, the authors have made a pretty good job in their sensitivity analysis. However, I wonder why they have not include a sensitivity case with [RO2]= 0.5 times [HO2] or [RO2]= 0.25 times [HO2] in Table 2. The latter, for example, would correspond to the [HO2]/[HO2]+[RO2] ratio of 0.8 measured by Cantrell et al. (1996,

ACPD

4, S2010–S2013, 2004

1997).

The overall agreement between measured and modelled sulfuric acid concentrations is quite good and, within measurement uncertainties, the closure has been achieved. This should be mentioned explicitly in the text. However, when looking at the time series in Figure 8, the are quite a few periods during which the agreement is much worse. In the end of section 5.2 something is said about this, yet the authors have not mentioned the fact that in many days (March 19, 20, 23, 24, 25, 28), the calculated peak sulfuric acid concentrations seem to exceed the measured ones by a factor of 2-4. This should be brought up in the paper and some potential explanation should be given.

In the first paragraph of Section 5.4 (page 6357) it is stated that O3, CO and NMHC's concentrations are approximately in the same level between the two air mass types. This is definitely not correct in case of NMHC's, since their concentration is about 3 times higher in polluted air compared with clean air. This is of the same order as the corresponding difference between SO2, NO2 and NO concentrations.

In the end of Section 5.4 it is stated that "sulphuric acid always participate in the aerosol formation process with a percentage with a percentage fraction between 3 and 17%" (page 6358, lines 18-20). This statement need to be totally rewritten. First, "always" is a very strong statement as this study concerns only a short period in one location. Second, one cannot estimate to percentage by which sulfuric acid participates in the aerosol formation process based on this data. Clearly, sulfuric acid explains 3 to 17% of the nucleation mode growth but the data does not reveal its contribution to nucleation (which is probably larger) or other processes associated with the very initial steps of particle formation.

The authors emphasis the important role of sulfuric acid on nuclei growth (9% contribution) (pages 6359-6360, lines $>$25 and $<$4). I would rather emphasize that sulfuric acid contribution only 9% and something else (like organic vapours) are needed.

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4, S2010–S2013, 2004

The authors further state that sulfuric acid may not be the key parameter in nucleation process itself (page 6360, line 19). What is this statement based on and what are then the key parameters?

Finally, the authors bring up the recent work by Zhang et al. (2004) in explaining nucleation due to sulfuric acid and organic vapours (last paragraph in page 6360). I would be more careful here, emphasizing a single work in the end of the manuscript may give the reader a wrong impression, especially as we do not really now what is the actual atmospheric nucleation atmospheric or whether we have plenty of those in different atmospheric environments.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 6341, 2004.