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

M. Boy et al.

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First of all we want to thank both referees for their constructive comments on this manuscript. We will include most of the advices in the final version and discuss the single points below:

Statement to the comments of referee Nr. 1:

The particle radius will be removed from the MS and only particle diameter will be used for clarity.

The trajectory based analyses were done in a way to select only days with significant pattern of polluted or unpolluted contents. For this reason we only used 16 out of 21 days for this analysis. The high discrepancy in mostly anthropogenic emitted gas con-

centrations like SO2, NO, NO2 and the condensational sink confirm that our selection is representative for “less” and “higher” polluted air masses.

We will use in the new version of the MS always the ratio predicted/measured values.

We agree with the referee that already in section 2.4 a statement concerning the NMHC should appear and will add in the new version in the end of this section one paragraph describing what organics are used in the calculations of OH and HO2.

The sensitive studies (Table 2) will be expanded with the cases [RO2] = 0.5 times [HO2] and [RO2] = 0.25 times [HO2].

The statement that NMHC are in the same order in ‘clean’ and ‘polluted’ air masses like CO and O3 are wrong and will be corrected in the new version of this MS.

We agree that the sentence page 6358, line 18-20: ‘These results indicate that sulphuric acid always participates in the aerosol formation processes with a percentage fraction between 3 to 17 %.’ should be rewritten. The authors want and will express that during the time of the campaign the concentration of sulphuric acid is high enough to explain a fraction between 3 to 17 % of the condensational growth of the particles. However, we agree with the referee that the results of this work are not able to give any statement about the participation of H2SO4 in the nucleation process or processes by itself. We will write this point more explicit in the final version.

The statement that sulphuric acid may not be the key parameter in the nucleation process itself (page 6360, line 19) will be changed in Ĕ. sulphuric acid may not be the key parameter in the particle formation process itself. This statement is based on the fact that the concentration of sulphuric acid is during some non-event days higher than on event days. If we would assume that H2SO4 is the key parameter for the particle formation process then we could expect to find direct correlation with this specie and the appearances of new formed particles. The authors will not exclude the possibility that sulphuric acid may be more important for the nucleation process itself,

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however with the results of this work and the fact that particle formation include both the nucleation process and the condensational growth of the particles the new version of this statement is justifiable in the opinions of the authors.

Concerning the link of the authors to a new article from Zhang et al. (2004) at the end of the MS, the authors will rewrite this paragraph as a possible theory for particle formation processes.

Statement to comments of referees Nr. 1 and Nr. 2:

As an answer to both referees we will include in the new version of the MS a detailed discussion on Fig. 8 and especially point out possible explanations for the higher discrepancies between predicted and measured sulphuric acid concentrations on some days.

Statement to referee Nr. 2 comments:

The accommodation coefficient used in this work was one for all particles and conditions and we will include this statement it in the new version.

The use of scenario 8 Table 4 in Fig. 8 is from the author side acceptable because it is clearly explained in the text why the concentration of NO2 is considered as too high and why the NMHC concentrations are doubled. For this reason we believe that it is not only tuning of some parameters to reach the best fit but also plausible why we used the correction of NO2 and NMHC. In this point the authors do not agree with the referee that using the closure in the title is misleading.

Concerning the statement on page 6343, line 10-12: 'Although the precursors for sulphuric acid, as well as the condensation sink, have been measured in several places, the closure between measured and calculated sulphuric acid concentration has not been investigated." The authors were reading the mentioned articles from Weber et al., (1997) and Eisele et al. (1993) and will rewrite this sentence into 'Ĕ has been investigated rarely (Weber et al., (1997) and Eisele et al. (1993))'.

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Interactive Comment

Where NMHC and HCHO particularly high when sulphuric acid made up the smallest fraction of the particle growth? No, there is no significant trend in high or low NMHC or HCHO concentrations and the fraction of H2SO4 participating in the particle growth rate.

The low contribution (3 or 4 %) of sulphuric acid on some days to the aerosol mass is surprisingly, however we did not include chemical analyses of aerosols in this work. On a hypothetic base we would suggest that on clear event days with particle growth rates between 2 and 4 nm/h other species like for example organics play the major role and during days with low growth rates (unclear event days, $<$ 2 nm/h) the contribution of sulphuric acid is much higher. However, it is obviously not strict forward to compare this growth rates with the concentrations of monoterpenes as was pointed out already in different papers (e.g. Boy et. al. ACP, Vol. 4, pp 657-678, 27-4-2004). The growth of particles may be more influenced by the reaction products of monoterpenes which automatically include the solar radiation, humidity and different gas concentrations as important parameters, too.

Concerning the discussion about the uncertainties the author's opinion differ because of the high amount of parameters included in the calculation. An estimated uncertainty would be meaningless. We agree that for example in the closure studies from Eisele et al. or Weber et al. it would give a useful estimation because in this work only measurements from sulphur dioxide, hydroxyl radicals and condensation sink values are included. However, in our case we would have to take into account also the production of OH with many parameters involved. We agree with the referee that an uncertainty between predicted and measured sulphuric acid concentrations is probably a factor of 2 but we believe also that an uncertainty study in these calculations would include too many parameters to be reliable.

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