

Interactive comment on “Changes in the production rate of secondary aerosol particles in central Europe in view of decreasing SO₂ emissions between 1996 and 2006” by A. Hamed et al.

Anonymous Referee #2

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This manuscript deals with a potentially important feedback between air quality and climate: it investigates how reduction in SO₂ level might have affected new particle formation (NPF) and consequently cloud condensation nuclei (CCN) numbers in Central Europe over the time period of 1996–2006. The topic of the study is interesting and well within the scope of ACP. For the most part, the approach seems reasonable as well. However, I do think that there are a few issues the authors will need to carefully address before the manuscript can be published in ACP.

General comments:

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1) As the authors also point out themselves, the "primary CCN" numbers they estimate from aerosol mass emissions are highly uncertain. As pointed out by Jeff Pierce in his report too, it is, for instance, likely that the emission sources and thus typical number-mass size distributions have changed over the years (the improvement of air quality being in the core of this study too!). Taking this into account, I would be quite sceptical about drawing any conclusions about the change in primary vs. secondary CCN numbers over the investigated period - particularly since the change in the secondary source is not, according to the authors' estimates extremely clear either (and has high uncertainties). In general, I would not emphasize the possible implications on CCN numbers too much in the paper, since these results do not seem scientifically extremely sound.

2) Interestingly, there seems to be a reduction in the 100–750 and 200–750 nm particle numbers, but such a clear effect is not seen in the smaller 50–750 nm size bin. The authors speculate that the increased growth rates might be a reason for this. I do not fully understand this logic. If the differences were caused by nucleation events, should not all the nucleation-originated particles in the > 100 nm size classes have grown through the sub-100 nm sizes and thus contribute to the concentration?

3) Taken into account the high uncertainties in the predicted CCN numbers (which the authors acknowledge themselves too), it would be very helpful for the reader if some - even rough - sensitivity analysis would be provided. Now, since almost all the parameters used in the CCN calculations are more or less uncertain, it is quite difficult to form an opinion about how conclusive the results are.

4) The authors report that nucleation event frequencies and particle formation rates have decreased over the investigated decade. However, simultaneously the condensation (and coagulation) sink and particle growth rate values have increased. The authors also acknowledge that there is a possible bias to this - following from the effect that the sink and growth rates have on the survival probability of the freshly-nucleated particles: the increasing sink values are likely to contribute to the decrease in the NPF

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event frequencies. Also, is it possible that the same reason is in fact causing the observed increase in the particle growth rates? That is, since the authors are only looking at days that have been classified as particle formation event days according to the criteria by Hamed et al. (2007) (and thus need to show signs of both nucleation AND growth), is it possible that due to the increase in the sink, only days with high enough vapor concentrations (and thus high enough growth rates and survival probabilities) are classifiable as NPF events in 2003-2006 as compared with 1996-1997?

5) Related to the last comment, it would be helpful if the authors could give an estimate on how much the increased sink is likely to affect the results. In light of this, I think it would make sense to also compare nucleation mode particle numbers (maybe with the influence of traffic somehow filtered away) directly to the reduced SO₂ instead of only particle formation frequencies and formation rates determined for days when also growth was observed?

6) The manuscript seems to have been written in a rush. I would recommend going the manuscript text and figures through once again, paying special attention to using consistent units and labeling them consistently, removing all unnecessary repetition from the text and checking spelling one more time.

Specific comments:

7) On p. 15089, line 12 the authors say "To quantify condensation processes during new particle formation, we calculated the condensation sink by using the method described by Pirjola et al. (1998) and Kulmala et al. . . (2001)". Besides looking at the sink for sulfuric acid, the CS acts as a measure of the capability of the particle size distribution to remove small particles. I think this point should be made clear.

8) What do the authors mean by visually estimating the growth rate? Why not use a more quantitative method? I think this is important since the growth rate plays such a crucial role in the survival probability of the freshly-formed particles. At least, the authors should give a more detailed explanation on how the growth rates were determined

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exactly.

9) Is the use of a residence time of 4 days in the SS calculation justified? For instance, how likely is it that the particles in the investigated size ranges have all the same life time? How does this assumption affect the results that the authors obtain for the CCN production rates (see also my comment about the sensitivity analysis)?

10) I think the authors should give the statistics of the NPF events and good quality data in the paper, i.e. how many days in total were analysed, how many event, non-event and equivocal days were observed each year (even monthly resolution would be good), and how large fraction of the data was considered to have a good enough quality. I think this could be done in an additional table. This would help assessing the results presented in Figs. 2b and 5, when the reader could immediately tell how many events the results are actually based on.

11) p. 15092, line 2: There is a typo in the CMD - it says it was 5 m.

12) p. 15098, line 26: The authors state "Note, however, that with decreased nucleation, the condensable vapour will be divided among fewer particles, and average growth rates could increase even if the condensable vapour levels stay constant." Is this really relevant, i.e. is the reduction in the condensational sink due to decrease in nucleation mode number concentration enough to potentially have any effect on the growth rates of the nucleation mode? I would imagine most of the vapor sink is anyway due to the larger particles.

13) p. 15100, line 5. The sentence "Based on this assumption we used the growth rate (GR) from 1nm to 3nm for each nucleation day and therefore we estimated the delay time as 2nm divided by GR." is confusing: it gives the impression that the authors estimated the time delay from the GR of 1-3 nm particles when in fact, I assume, it was vice versa (if anything - I assume 1-3 nm GRs were not even used in the study)?

14) Figure 1a is difficult to read, and the data sets difficult to compare. It might be

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a good idea to plot the y-axis in a log scale? Please also label the units in the axes consistently.

15) Figure 7. Please make all the panels of the figure equal in size.

16) In Table 1 "wind speed" should probably be capitalized.

17) Table 2 and its caption: Please use consistent symbols for the diameters. Now in the caption they are capitalized but in the actual table not.

18) Table 3 is confusing. Why does it, for instance, say in the last column "No difference" for the SO₂ concentrations, although (if I understand correctly) it has been significantly lower in the latter period as compared with the earlier period? I suspect there the rows in the last column have been mixed up at some point?

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