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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Changes in the production rate of secondary aerosol particles in central Europe in view of decreasing SO_2 emissions between 1996 and 2006" *by* A. Hamed et al.

J. Pierce (Referee)

jeffrey.robert.pierce@gmail.com

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Review of "Changes in the production rate of secondary aerosol particles in central Europe in view of decreasing SO2 emissions between 1996 and 2006" by Hamed et al.

This article explores the important topic of how aerosol microphysics (nucleation in particular) has changed due to the reduction of SO2 emissions in Europe in the past decades. The centerpiece of this paper is long-term SO2 and aerosol size-distribution

data in the mid-90s and again in the mid-00s from Melpitz, Germany. The data showed a significant decrease from the 1990s to the 2000s for SO2 concentration, nucleation event frequency and nucleation rate. The authors explore the connection between the change in SO2 and the change in nucleation by also looking at changes in the aerosol condensation sink and solar radiation, and they conclude that there is a likely a strong connection between the SO2 changes and the nucleation changes. The change in nucleation appears to have little effect on CCN concentration except for the smallest CCN sizes explored (50 nm).

The topic is very important and well within the scope of ACP. The approach is generally very good. The manuscript, however, should have been revised more thoroughly prior to its submission to ACPD (e.g. inconsistencies between figures and discussion in text and general grammar mistakes). The authors need to carefully proofread the paper for consistency and english before it can be published in ACP. Regardless, I recommend this paper to be published once the proofreading has been done and the following concerns have been addressed.

General comments:

1. During the exploration of the impact of SO2 and nucleation changes on CCN, a major assumption in both the calculation of CCN generation during the nucleation events as well as the calculation of the contribution of primary emissions to CCN is that growth of ultrafine primary particles to CCN sizes is ignored. To the extent that ultrafine primary particles are contributing to CCN in Melpitz (this may be small, but I'm not sure), the contribution of nucleation to CCN will be overestimated. This is not thoroughly addressed until the end of the paper (page 15103, lines 8-13). It would be good to also discuss this in Sections 2.3 and 2.4.

   Uncertainties in the size distribution of primary particles are also important here. More importantly, the size distribution of primary particles may have changed

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between the time periods as the source of SO2 has changed, thus changing the number of primary CCN even without a change of mass emissions. This is a significant source of uncertainty in the CCN estimates and should be mentioned in Sections 2.4 and 3.3.

Specific comments:

1. Page 15090, line 14: Is the 4-day lifetime the lifetime of CCN number or CCN mass? These lifetimes may be different if the timescale of coagulation of the smaller CCN with other CCN is ∼4 days or less. The timescale of CCN number is the correct one to use here.

2. Page 15090, line 25: Please define EMEP.

3. Page 15091: The EMEP sector numbers are in some places preceded by a capital "S", and in other places they are not. This should be changed so that it is consistent.

4. Page 15091, lines 23 and 27: I do not understand the meaning of “(22f)” and “(22g)”

5. Page 15091, line 20- and Table 2: There are many differences between the text and table 2. 420 nm vs. 430 nm. 1 mode vs. 2 modes for “other” particles. Sectors 2-6 vs. 2-8 for “Manufacture” particles. There may be others too, so please check this carefully.

6. Table 2 caption: Dp1 should be dp1. Mention if number mean or mass mean diameters. Define m% and EMEP sectors. EMEP sector 8 is listed as belonging to both the "Manufacture" and "Other" categories.

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7. Page 15092, line 2: 5 m particles?

8. Page 15092, line 12-14: Is this decrease of 50-70% the decrease for Europe as a whole? EU? Western Europe? Please be specific.

9. Page 15093, line 6: What does “ca.” mean here? It is an abbreviation for circa, but that doesn’t seem like the right word for that sentence. Also, you’ve listed 3 locations, given two values for the locations and used the word “respectively”. This is confusing because I don’t know if Halle gets grouped with Leipzig or Cottbus? Please clarify.

10. Figures 1b, 2, 3, 4, 5: Please switch the order so that the 1990s time period comes first, both in the legends and in the order in which the bars appear in the bar chart. It increases the chance of confusion when you put the later time period first. This is particularly problematic in the bar graphs.

11. Figure 3b: For the 2000s period, the CS drops by about a factor of 2 during the morning for every season. This is much less apparent in the 1990s (in the winter and autumn it doesn’t appear to happen at all!). Is there different boundary-layer growth during the two decades? Is the decrease in CS in the 1990s dampened by the faster nucleation rates that can replenish the CS? This is very interesting and deserves to be discussed in the text. Furthermore, in the text you mention that there is no remarkable difference between the two time periods during winter and autumn; there is a big difference before noon (with the 2000s having higher CS values), and only after noon are the CS values about the same.

12. Page 15095, lines 18-20: Don’t forget that water vapor is generally involved in nucleation too.

13. Figure 5a: Make sure the corrected figure makes it into the text.

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14. Table and Section 3.1.4: It is very good that you've done the statistical significance tests; however, I find Table 3 very hard to read, and the last column "Higher in" has many mistakes in it. At minimum fix this last column or get rid of it. However, if it makes sense to you, I suggest you get rid of the table and add the p-values to the plots themselves. This would require you moving the background of the stats test to earlier in the paper, but this way you can more freely discuss the statistical significance throughout the discussions of each parameter.

15. Page 15101, lines 9-12: I disagree that you should call this estimate "conservative". While you do make assumptions that may cause an underestimate in the number of CCN from nucleated particles, you also make a large assumption that would cause an overestimate of the number of CCN from nucleated particles (ultrafine primary particles may also be growing to CCN sizes with your nucleated particles). Since it would be very hard to quantify all of these errors, I suggest that you do not refer to the estimate as conservative.

16. Page 15102, lines 1-3: Also, growth of ultrafine primary emissions to CCN.

17. Page 15102, lines 21-24: Probably good to mention here that primary emissions size distributions may have changed.

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