

Interactive comment on “No statistically significant effect of a short-term decrease in the nucleation rate on atmospheric aerosols” by E. M. Dunne et al.

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We thank Anonymous Reviewer #3 for their helpful feedback and comments. We have addressed their comments below. Text in italics is quoted from the referee's comments, while bold text has been added to the manuscript.

1. The authors decreased nucleation rates by 15% for 10 days for each month. The aerosol changes that have been analyzed are monthly mean or annually mean values. On an annual mean base, the effective change of nucleation rates between perturbed and controlled cases is 15%/3=5%. It is necessary to point this out in the text and

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abstract.

Monthly or annual means are not analysed in the paper. The aerosol changes that have been analysed are daily mean values averaged between the twelve monthly periods. On ten of the thirty days, the nucleation rate has changed by 15 %. Each of the 30 days is then analysed separately, having been averaged between the twelve monthly periods. The effective change in the nucleation rate is zero for the first and last ten days of each 30 day period, and 15 % for the middle ten days.

2. Figure 8b. Based on the shown values, I assume that Fig 8b gives the percentage difference. The authors reduced the nucleation rates by 15% for every grid box. Fig. 8b shows that the changes in CN10 are negative in some locations while positive in other locations. The patterns appear to be random. Why the systematic reduction in nucleation rates didn't lead to a systematic change in CN10? Are the fluctuations in CN10 change random for all months and layers? To help the reader to better understand your modeling results, please provide figures (probably as supplementary materials) showing spatial variations (both horizontal distribution for surface and zonal average figures) of sulfuric acid vapor concentration, nucleation rate, CN10 and CCN for unperturbed case, along with the differences between perturbed and unperturbed cases.

The caption in Figure 8 has been clarified, and now reads: “**A map showing regional values of (a) the number of model levels (from a total of 31) where $F > F_{\alpha}$, and (b) the difference between unperturbed and perturbed data in number of model levels where $F > F_{\alpha}$ for the case of CN_{10} .**” Figure 8 (a) shows the number of the 31 model levels in which the variability over the 30 days in each month is greater than the variability in the full data set would predict. Approximately 5 % of the grid boxes should experience greater variability, although the number is closer to 12 %, which I

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would attribute to the underlying distribution of the data, especially across the full 31 model levels. Figure 8 (b) shows the difference between unperturbed and perturbed data in the number of model levels for which the variability is greater than would be predicted, and the point of the figure is to show the changes in variability are random and not systematic. The change in actual CN concentrations is generally negative, although there are some regions where CN concentrations increase due to feedbacks. However, the internal variability of the perturbed data sets is not systematically greater than that of the unperturbed data. In light of this explanation of the randomness of Figure 8 (b), I do not believe that spatial distributions of model tracers are necessary.

3. Page 15381. You assumed that 2.5% of Sulfur emitted as primary sulfate. The primary sulfate emission has been widely used to parameterize the sub-grid plume scale sulfate particle formation and growth. If cosmic ray ionization affects nucleation, it should also affect the sub-grid sulfate particle formation. Have you tested how the primary sulfate emission assumption may affect your results?

The analysis in this paper examines the changes in variability between two sets of model runs with identical SO₂ emissions, which is the parameter used to estimate primary sulphate emissions. Recent papers have found that uncertainty in the primary sulphate parametrisation is an important contributing factor to uncertainty in aerosol models (Stevens *et al.*, 2012). However, the majority of primary sulphate formation occurs in the boundary layer, where both ion concentrations and changes in ion concentrations will be smallest, and it is therefore unlikely to be strongly affected by changes in ionisation. While the primary sulphate parametrisation may have an effect on the ultimate uncertainty in CCN concentrations, the analysis carried out in this paper should therefore not be particularly sensitive to primary sulphate emissions.

4. Section 2.1. You used BHN of Vehkamäki *et al* (2002) for free troposphere
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*nucleation and empirical formula of Sihto *et al* (2006) for BL nucleation. Was the empirical scheme applied to global BL or limited to continental BL? How much the value of prefactor (you used 2E-6 s-1), which has large uncertainty, might affect your results and conclusions? In addition, laboratory studies (Hanson and Lovejoy, J. Phys. Chem. A, 110, 9525–9528, 2006) have shown that the BHN of Vehkamäki *et al.* (2002) overpredicted the BHN rates by about three orders of magnitude under the typical conditions of free troposphere. How sensitive are your results to the uncertainties in nucleation schemes?*

The aerosol system may be more sensitive to changes in the nucleation rate at lower nucleation rates. In the event that the nucleation parametrisation overpredicts the nucleation rate, this may reduce the sensitivity of the system to changes in the nucleation rate. Since the sensitivity of the atmosphere to nucleation is expected to saturate, but the nucleation rate at which it saturates is unknown, there may be a particular set of atmospheric conditions under which the system would be more sensitive to a 15 % change in the nucleation rate. However, under conditions where nucleation may be overpredicted, the change in ion-induced nucleation is expected to be much smaller than 15 %, and so this sensitivity study would still be looking at a larger relative change in the nucleation rate. All nucleation parametrisations currently in use in global models have a high degree of uncertainty, but the parametrisation of Vehkamäki *et al.* (2002) is an improvement on that of Kulmala *et al.* (1998), which has been shown to generate aerosol concentrations in good agreement with atmospheric observations (Spracklen *et al.*, 2010), and was therefore used in this study.

A sentence has been added to the manuscript: **The sensitivity of the aerosol system could change with the assumed nucleation rate, but the nucleation schemes chosen in this paper are consistent with observed CN concentrations (Spracklen *et al.*, 2010).**

5. *Abstract.* You mentioned twice about “observed correlation between short-term...properties”. However, in the Introduction and Discussion you questioned the existence of the correlation. If you think the correlation does not exist, you may want to modify the abstract to reflect this.

While the significance of the correlations has been questioned in comments on a recent ACPD publication, the purpose of this paper is not to question whether or not they exist, but rather to determine whether the proposed mechanism of changes in the ion-induced nucleation rate would be sufficient to generate the correlations between GCRs and cloud and aerosol properties, or whether another mechanism would be required to explain a connection. For this reason, we have kept the abstract as it is.

6. *End of Section 1.* It would be helpful to insert a short paragraph to describe the main objectives of this work and key differences to previous studies.

A sentence has now been included at the end of Section 1: “**The aim of this manuscript is to test the hypothesis that changes in the nucleation rate can account for observed correlations between GCRs and cloud and aerosol properties, and to evaluate the response in aerosol in a way that is transferable to atmospheric observations, taking into account the detectability of the response above global aerosol variability.**”

7. *Page 15393, line 14.* This statement is not necessarily true. Because of the lower sulfuric acid vapor concentration and higher ionization rate in the free troposphere, the effect of ionization rate change on particle concentration could be weak or opposite in sign.

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The sentence does not refer to a change in the ion-induced nucleation rate, but rather to the overall contribution of ion-induced nucleation to aerosol, which is likely to be higher in the free troposphere than in the boundary layer, where the coagulation sink for small particles is so much larger.

8. *Section 3.2.2.* Figure 4 should be Figure 8. Also (a) and (b) were not marked in the figure.

These corrections have been made.

References

- Kulmala, M., A. Laaksonen, and L. Pirjola, Parameterizations for sulfuric acid/water nucleation rates, *J. Geophys. Res.*, *103*, 10.1029/97JD03718, 1998.
- Spracklen, D. V., K. S. Carslaw, J. Merikanto, G. W. Mann, C. L. Reddington, S. Pickering, J. A. Ogren, E. Andrews, U. Baltensperger, E. Weingartner, M. Boy, M. Kulmala, L. Laakso, H. Lihavainen, N. Kivekäs, M. Komppula, N. Mihalopoulos, G. Kouvarakis, S. G. Jennings, C. O'Dowd, W. Birmili, A. Wiedensohler, R. Weller, J. Gras, P. Laj, K. Sellegri, B. Bonn, R. Krejci, A. Laaksonen, A. Hamed, A. Minikin, R. M. Harrison, R. Talbot, and J. Sun, Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation, *Atmos. Chem. Phys.*, *10*(10), 4775–4793, 2010.
- Stevens, R. G., J. R. Pierce, C. A. Brock, M. K. Reed, J. H. Crawford, J. S. Holloway, T. B. Ryerson, L. G. Huey, and J. B. Nowak, Nucleation and growth of sulfate aerosol in coal-fired power plant plumes: sensitivity to background aerosol and meteorology, *Atmos. Chem. Phys.*, *12*(1), 189–206, 10.5194/acp-12-189-2012, 2012.
- Vehkamäki, H., M. Kulmala, I. Napari, K. E. J. Lehtinen, C. Timmreck, M. Noppel, and A. Laaksonen, An improved parameterization for sulfuric acid–water nucleation rates for tropo-

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spheric and stratospheric conditions, *Journal of Geophysical Research*, 107(D22), 4622, 10.1029/2002JD002184, 2002.

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