

## ***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.***

**E. M. Dunne et al.**

eeemd@leeds.ac.uk

Received and published: 20 November 2012

We thank Jeffrey Pierce for his helpful feedback and comments. We have addressed his comments below. Text in italics is quoted from the referee's comments, while bold text has been added to the manuscript.

*P15375 L8: Regarding the small change in the total solar irradiance being too small to change clouds: there is a feedback mechanism in the Hadley cell that is proposed to increase subtropical clouds for a small reduction in solar irradiance.*

C9578

This mechanism is now referenced: **A feedback mechanism which increases subtropical clouds by strengthening the climatological precipitation maxima in the tropical Pacific during northern hemisphere winter has also been proposed by Meehl et al. (2008).**

*P15376 L19-21: An additional significant reason why the fractional change in nucleation is always greater than the fractional change in CCN is that the likelihood of a nucleated particle surviving to become a CCN (survival probability) decreases with increasing nucleation rates. This decrease in survival probability is because of slower growth (more competition for condensible material) and more coagulation at faster nucleation rates. This is discussed in Pierce and Adams (2009b).*

The following sentence has been added to P15376 L18: **When the nucleation rate increases, the relative probability of survival decreases as a result of slower growth due to increased competition for condensible vapours and increased self-coagulation of nucleated particles (Pierce and Adams, 2009).**

*P15378 L1-3: Laken et al. (2009) did claim that the delay was too long, but I've never agreed with this. Mean growth rates in the free troposphere are often  $\sim 1$  nm/hr or less. Therefore, it could take at least 4 days for CCN to be formed in the free troposphere, so if the FT is where the action is, this delay is expected.*

The sentence now reads: **Laken et al. (2009) concluded that the observed time delay in the liquid water cloud fraction response was longer than expected if it were due to changes in the ion-induced nucleation rate, although Svensmark et al. (2009) cite two models which suggest growth rates on the order of days (Russell et al., 1994; Arnold, 2007).**

C9579

*P15378 L9-12: Can you add a quick statement as to how Laken et al. (2010) was able to rule out ion-induced nucleation through the statistical analysis?*

The suggestion that ion-induced nucleation had been ruled out was an error. Laken et al. (2010) concluded that any effect would need to be a second-order effect, and therefore require conducive conditions in the atmosphere. This does not rule out ion-induced nucleation, and we have removed the suggestion that it would.

*P15382 L23 through end-of-paragraph: It would be useful to add a statement here about how much ion concentrations (or ion-formation rates) change during a FD. The way the paragraph is currently written might lead a novice reader to assuming that your choice of 15% reduction might have something to do with the 15% of nucleation being calculated to be ion induced (as cited earlier); however, you are actually assuming that 100% of nucleation is ion induced and that the ion concentrations decrease by 15% during a FD.*

*Also in this paragraph, you should mention that these estimates are for a forested continental boundary-layer site and that this may not be representative for the fraction of nucleation that is ion induced in other regions of the atmosphere (I believe you now have estimates of this in your modelling work that you presented in the CLOUD workshop).*

We agree with these comments, and the paragraph now reads: **The relative proportion of nucleated particles derived from charged and neutral processes is very uncertain. Analysis of observations suggests that the contribution of ion-induced nucleation to new particle formation events is between 6 % and 15**

C9580

**% in the continental boundary layer (Laakso et al., 2007; Boy et al., 2008; Gagné et al., 2008; Manninen et al., 2009), while one model suggests that as much as 80 % of nucleation events could be ion-mediated (Yu and Turco, 2008, 2011). Boundary layer nucleation is likely to have a lower ion-induced fraction than free tropospheric nucleation, due to the expected dependence of high-temperature nucleation on ternary vapours which will reduce the need for stabilising ions (Kirkby et al., 2011).**

**In this paper, we assume that all nucleation is reduced by 15 % for 10 days during a Forbush decrease. This implicitly assumes that 100 % of nucleation is ion-induced, and that ion concentrations will decrease by 15 % during a Forbush decrease when the ionisation rate decreases by 15 %. Both of these implicit assumptions are known to be inaccurate (Kirkby et al., 2011; Usoskin et al., 2010). Neutral nucleation also occurs and is likely to dominate in some parts of the atmosphere. Ion concentrations do not depend linearly on ionisation rates, and indeed the ionisation rate does not change by a uniform percentage throughout the atmosphere during a Forbush decrease. Depending on the relative proportions of neutral and ion-induced nucleation, the actual decrease in the nucleation rate resulting from a Forbush decrease could be much smaller than we simulate here.**

*Equation 1: The formula for the AE is flipped (the AOD stuff should be on top) and needs a negative sign. As the equation stands now, Rayleigh-scattering particles would have an AE of -0.25 rather than 4.*

*Equation 2: Technically the right-hand side does not exactly equal the left-hand side, or else  $X_{ij}$  in Eqn. 3 would be 0. It would be more clear to change  $Y_{ij}$  in Eqn. 2 to  $Y_{ij,fit}$ .*

C9581

*Table 2: What are S1 and S2 in the table? I assume that they should be  $S_c$  and  $S_R$ . What are the primes? I assume that the not-primed data is unperturbed and the primed is perturbed, but there is no discussion of this.*

These changes have been made. The not-primed data are unperturbed, and the primed data are perturbed. This is now mentioned in the caption.

*Figure 4b: Maybe my eyes are playing tricks on me, but it REALLY looks like the 30 days of the 12 months are correlated with each other, even in the unperturbed data. It looks like the 12 points move up and down in unison as you move across the day-of-month. I bet if you calculated the correlation coefficient between the lowest point for each day-of-month with the highest point for each day-of-month, you get an  $r$ -value much greater than 0. Why should there be any correlation between the upper values with the lower values? Is this something that is a relic of the de-trending?*

We calculated the Pearson correlation coefficient  $R$  between the highest and lowest values on each of the 30 days and find that they are poorly correlated ( $R = 0.284$ ,  $R^2 = 0.080$ ).

*P15392 L14: "Perturbed data points are no more likely to fall outside the confidence interval than unperturbed." Are you applying the confidence interval of the unperturbed data to the perturbed data? This seems like the right test to do (since the perturbed data will have more variance and thus a larger confidence interval). However, it might not matter which confidence interval you use since the difference between the perturbed and unperturbed is so small.*

C9582

Yes, we are using the unperturbed confidence interval for both sets of data. The text has been changed to reflect this.

*P15393 L8: "Responses of CN and CCN are compared in Figs. 4 and 5." CCN are in 4 and CN in 5 so maybe say "responses of CCN and CN."*

This correction has been made.

*P15393 L11: "The response in CN10 is strongest at the surface because the nucleation rate in the boundary layer  $J_{BLN} = A [H_2SO_4]$  is higher than in the free troposphere, so the 15 % decrease in the nucleation rate has a larger effect." True, but I would guess that nucleation contributes to a higher fraction of the  $CN_{10}$  in the FT than it contributes to the fraction  $CN_{10}$  than in the boundary layer (since primary emissions are emitted directly into the BL). I guess the nucleation-rate difference wins out overall though?*

Nucleation would generate almost all CN in the FT, but in terms of absolute rather than percentage contribution, nucleation creates more CN in the boundary layer. The absolute change in new particle formation at 10 nm caused by a 15% decrease in FT nucleation is not sufficiently large to generate a significant change in  $CN_{10}$ , but in the boundary layer, the nucleation rate is so much larger that the change in  $CN_{10}$  is at least noticeable, if not greater than the natural variability of the system. However, even if all nucleation were ion-induced, boundary layer ion concentrations would only be expected to vary by at most 5 % according to recent model simulations carried out using GLOMAP, which have not yet been published.

*P15393 L17: "At 10-15 km the model predicts that the concentrations of CN10 and CCN70 increase slightly in response to a decrease in nucleation rate." Is this shown in*

C9583

some figure or table? I couldn't find it.

Table 4 now includes a sign on the maximum percentage change. There was also a typo; only CN10 increase, CCN70 still decrease. This is consistent with the increase in CN10 being attributed to a decrease in CCN70 which reduces the coagulation sink for small particles.

Table 3: "Table 3 shows the proportion of unperturbed and perturbed grid boxes for which the variation between days is found to be statistically significantly greater than the residual variance in that grid box." Can you split to show the % for the unperturbed and the % for the perturbed? It would be interesting to know if there is a difference.

The table has been adjusted. I think it shows clearly that there is no major difference between the percentage of unperturbed and perturbed runs where  $F > F_{\alpha}$ .

P15394 L15 and L17: Figure 4 should be Figure 8. Figure 8: How many total model levels are there again? Figure 8b: relative difference (%) or absolute difference (units)?

These changes have been made. There are 31 model levels, and Figure 8 (b) shows the difference between unperturbed and perturbed data in number of model levels where  $F > F_{\alpha}$  for the case of CN<sub>10</sub>. Units have not been included on Figure 8 (b), since the units are model levels; however, the caption 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<sub>10</sub>.**

C9584

P15395 L5-7: I think it would be better to say "all concluded that there were no significant correlations." Saying "all concluded that the observed correlations were not causally linked to Forbush decreases" sounds like they did find significant correlations found them to not be linked through physics (i.e. the cosmic-ray change did not directly cause the cloud change); however, these studies didn't look at the physics. This is a matter of being clear on the difference between correlation and causality.

We agree with the suggestion and have modified the text accordingly.

P15395 L16-19: Similar to an earlier comment: Need to state how much ions change during a FD and also state that these observations are for a continental BL.

The section now reads: **The estimated fraction of nucleated particles derived from ion-mediated processes varies between 6 % to 15 % based on observations in the continental boundary layer (Boy et al., 2008; Gagné et al., 2008; Manninen et al., 2009) to about 80 % in the model of Yu and Turco (2008). During a Forbush decrease, ions in the continental boundary layer are expected to change by at most 5 %, though at high altitudes over the poles they could change by up to 20 %.**

Use "that" before restrictive phrases and clauses (e.g. "He drove away in the car that was red." The sentence implies that there was more than one car, and he specifically left in the red one). Use ", which" (note the comma) before nonrestrictive phrases and clauses (e.g. "He drove away in the car, which was red." This implies that there was only one car, and it just happened to be red.). There are several places in the paper where these are not done correctly (either "which" where there should be a "that" or "which" where there should be a ", which").

C9585

Some of the uses of which have been changed; however, we have since determined in discussion with Jeff Pierce that the use of “which” instead of “that” is a regional difference in English, and so not every use has been changed.

## References

- Boy, M., Kazil, J., Lovejoy, E., Guenther, A. and Kulmala, M. (2008), “Relevance of ion-induced nucleation of sulfuric acid and water in the lower troposphere over the boreal forest at northern latitudes”, *Atmospheric Research*, Vol. 90, pp. 151–158.
- Gagné, S., Laakso, L., Petäjä, T., Kerminen, V. and Kulmala, M. (2008), “Analysis of one year of Ion-DMPS data from the SMEAR II station, Finland”, *Tellus B*, Vol. 0.
- Kirkby, J., Curtius, J., Almeida, J., Dunne, E., Duplissy, J., Ehrhart, S., Franchin, A., Gagné, S., Ickes, L., Kürten, A., Kupc, A., Metzger, A., Riccobono, F., Rondo, L., Schobesberger, S., Tsagkogeorgas, G., Wimmer, D., Amorim, A., Bianchi, F., Breitenlechner, M., David, A., Dommen, J., Downard, A., Ehn, M., Flagan, R. C., Haider, S., Hansel, A., Hauser, D., Jud, W., Junninen, H., Kreissl, F., Kvashin, A., Laaksonen, A., Lehtipalo, K., Lima, J., Lovejoy, E. R., Makhmutov, V., Mathot, S., Mikkilä, J., Minginette, P., Mogo, S., Nieminen, T., Onnela, A., Pereira, P., Petäjä, T., Schnitzhofer, R., Seinfeld, J. H., Sipilä, M., Stozhkov, Y., Stratmann, F., Tomé, A., Vanhanen, J., Viisanen, Y., Virtala, A., Wagner, P. E., Walther, H., Weingartner, E., Wex, H., Winkler, P. M., Carslaw, K. S., Worsnop, D. R., Baltensperger, U. and Kulmala, M. (2011), “Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation”, *Nature*, Vol. 476, pp. 429–433.
- Laakso, L., Gagné, S., Petäjä, T., Hirsikko, A., Aalto, P. P., Kulmala, M. and Kerminen, V. (2007), “Detecting charging state of ultra-fine particles: instrumental development and ambient measurements”, *Atmos. Chem. Phys.*, Vol. 7, pp. 1333–1345.
- Laken, B. A., Kniveton, D. R. and Frogley, M. R. (2010), “Cosmic rays linked to rapid mid-latitude cloud changes”, *Atmospheric Chemistry and Physics*, Vol. 10, pp. 10941–10948.
- Manninen, H. E., Nieminen, T., Riipinen, I., Yli-Juuti, T., Gagné, S., Asmi, E., Aalto, P. P., Petäjä, T., Kerminen, V. and Kulmala, M. (2009), “Charged and total particle formation and growth rates during EUCAARI 2007 campaign in hyytiääd’ääd””, *Atmos. Chem. Phys.*, Vol. 9, pp. 4077–4089.
- Meehl, G. A., Arblaster, J. M., Branstator, G. and van Loon, H. (2008), “A coupled Air-Sea response mechanism to solar forcing in the Pacific region”, *Journal of Climate*, Vol. 21, pp. 2883–2897.
- Pierce, J. R. and Adams, P. J. (2009), “Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates”, *Atmos. Chem. Phys.*, Vol. 9, pp. 1339–1356.
- Usoskin, I. G., Kovaltsov, G. A. and Mironova, I. A. (2010), “Cosmic ray induced ionization model CRAC:CRIL: an extension to the upper atmosphere”, *Journal of Geophysical Research*, Vol. 115, p. 6 PP.
- Yu, F. and Turco, R. (2008), “Case studies of particle formation events observed in boreal forests: implications for nucleation mechanisms”, *Atmos. Chem. Phys.*, Vol. 8, pp. 6085–6102.
- Yu, F. and Turco, R. P. (2011), “The size-dependent charge fraction of sub-3-nm particles as a key diagnostic of competitive nucleation mechanisms under atmospheric conditions”, *Atmos. Chem. Phys.*, Vol. 11, pp. 9451–9463.