

## ***Interactive comment on “Cosmic rays, aerosol formation and cloud-condensation nuclei: sensitivities to model uncertainties” by E. J. Snow-Kropla et al.***

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We appreciate the insightful and useful comments by both reviewers. Our responses to the comments reviewer 1 is below. The original reviewer comments are in italics.

***Anonymous Referee #1 Received and published: 19 March 2011***

*Summary of manuscript*

*The manuscript presents a model study of the response of aerosol in the troposphere to variations in atmospheric ionization, such as occurring in the course of the decadal solar cycle, or due to Forbush decreases. The role of uncertainties in primary aerosol*

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*emissions, aerosol precursor emissions, and in the description of aerosol microphysical processes for the results (the response of aerosol to changes in atmospheric ionization) are investigated. A global chemical transport model with detailed, interactive aerosol microphysics is used for this purpose, with prescribed meteorology. The authors show that larger aerosol particles, which are more likely to form cloud drops than smaller aerosol particles, show a very modest response to changes in atmospheric ionization in the course of the decadal solar cycle. This result bears negatively upon a proposed causal link between variations in galactic cosmic ray intensity and global cloud cover. Similarly, the simulations show a very modest response of the Ångström exponent (on the global scale) to variations in atmospheric ionization, in contrast with other recently published works.*

*General comments*

*The presented work and its results are well described and discussed, and provide new insights on the topic. The manuscript is written in a transparent style, with few exceptions where text passages need clarification or rewording. Some scientific matters where additional explanations or minor corrections are needed are listed in the following. The figures are nicely done. Please check for typos.*

*Specific comments*

*Page 2698, line 1: “The flux of cosmic rays to the atmosphere has been observed to correlate with cloud and aerosol properties.” “... has been observed...” implies in scientific terms a little disputed fact, which the correlation of galactic cosmic ray intensity and cloud cover is not; a host of mutually inconsistent results has been published on the topic. “... has been reported...” would reflect this better.*

Changed the word “observed” to “reported”.

*Page 2700, line 9: ... the ion fields near clouds are strengthened by cosmic rays. The stronger field affects the collection of unactivated aerosols by cloud droplets... This de-*

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*scription of the near-cloud mechanism is incomprehensible. What does "strengthening of ion fields near clouds by cosmic rays" mean?*

Yes, this was rather confusing. We have changed this to read, "In the near-cloud mechanism, the distribution of charge on aerosols near clouds is suspected to be modulated by the cosmic-ray flux. Aerosol charging affects the collection rate of unactivated aerosols by cloud droplets. The change in collection could affect the freezing of super-cooled droplets by contact freezing (during aerosol scavenging) (Tinsley and Heelis, 2009).

*Page2700,line17:...explaining the observed correlations between cosmic rays and clouds... "reported", in contrast to "observed" would better reflect the disputed status of the correlation.*

Changed the word "observed" to "reported".

*Page 2701, line 21:"The observed 5–10 day delay in the minimum of aerosol and cloud values after the minimum in the cosmic ray flux by Svensmark et al. (2009) is evidence for the ion-aerosol clear-sky mechanism." The delay is consistent with the clear-sky mechanism, but by no means evidence.*

Changed "evidence for" to "consistent with".

*Page 2703, line 22:"SOA is assumed to be non-volatile and is condensed onto the aerosol surface area." Secondary organic aerosol does not condense onto aerosol surface area.*

Recent studies show that a large (50% or larger) fraction of SOA measured in the field (in both remote and urban sites) is low enough volatility that condensed molecules do not likely cycle back to the gas phase during the lifetime of the aerosol (see Riipinen et al., "Organic condensation – a vital link connecting aerosol formation to climate forcing", ACPD, 11, 387-423, doi:10.5194/acpd-11-387-2011, 2011 and Cappa and Jimenez, "Quantitative estimates of the volatility of ambient organic aerosol", Atmos.

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Chem. Phys., 10, 5409-5424, doi:10.5194/acp-10-5409-2010, 2010). Regardless if this low volatility was caused by gas-phase or particle-phase reactions, this means that once the low-volatility SOA is in the aerosol phase, it stays there. It gets to the aerosol in the first place by condensation to the Fuchs-corrected surface area. Thus, even though the SOA is partitioning to the aerosol mass, new SOA mass is distributed across aerosol sizes proportionally to aerosol surface area for effectively non-volatile SOA (i.e.  $C^* \leq 1E-3$  ug m<sup>-3</sup>).

Of course, there is also some truly semi-volatility SOA that will quickly cycle between the aerosol and gas phase. This SOA will condense more proportionally to the aerosol mass distribution. Thus, what we use in this paper is an approximation that favors the growth of ultrafine particles. This should make CN40 and CN80 more susceptible to changes in nucleation rates. Even with this favorable assumption of SOA condensation to surface area, we find the sensitivity of CN40 and CN80 is low.

We have modified the text to read: "SOA is assumed to be non-volatile and is distributed across the aerosol size distribution proportionally to the Fuchs-corrected aerosol surface area (Riipinen et al., 2011). Since some SOA will be volatile enough to cycle between the gas and aerosol phase (which would cause the net SOA condensation to partition to the aerosol mass distribution rather than surface area), this assumption favors growth of freshly nucleated particles to CCN sizes."

*Page2708,line15: "...the modelled accumulation mode may be too large on average." Please, clarify whether the accumulation mode consists of too many or too large particles.*

We now say, "the size of the modelled accumulation mode may be too large on average."

*Page 2710, line 16:"The competition between these three phenomena [sic] lead to some regions showing increases of CCN with cosmic rays and some regions showing the opposite."*

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“Phonomena” is now “phenomena”.

- *This is a plausible explanation for the patterns seen in Figure 6, but can you provide some supporting evidence for it?*

We don't believe we can prove our proposed explanation without adding additional diagnostics and re-running the model. Considering (1) the regions largely cancel each other out in the global mean and (2) most correlations of cosmic-ray effects on clouds have been seen in low clouds, the additional information probably doesn't justify the additional effort.

We have added the word “may” to the sentence preceding the three phenomena: “These may occur because of several competing microphysical processes.”.

We also have modified the paragraph after the three phenomena based on your suggestions below.

- *The model runs cover March, June, September and December, with a prescribed meteorology. Can you exclude the possibility that the patterns seen in Figure 6 are a result of the specific meteorology in these months?*

No, we can't exclude this possibility and this is a good point. (We'll show how we changed the manuscript text to reflect this below.)

- *Is there a strong variation in the patterns seen in Figure 6 between the months?*

Yes, there is variation. (We'll show how we changed the manuscript text to reflect this below.)

- *Would the same patterns be obtained from averaging a 12-month run?*

No, not likely the same. Regions of increasing and decreasing CCN were found in the FT for changes in nucleation rates in Pierce and Adams 2009a and 2009b. The locations of increases and decreases were not in the exact same place as the current manuscript, however, a different host model was used. We suspect that the patterns

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may be governed by regions of net uplift and subsidence, which will certainly vary seasonally and may vary between host models.

The paragraph after listing the 3 phenomena had been revised to include the points the reviewer made above: “The competition between these three phenomena might lead to some regions showing increases of CCN with cosmic rays and some regions showing the opposite. It is also possible that the magnitude and location of the increase and decrease regions will change if longer time periods are analyzed. The regions of increases and decreases occur in different locations for each of the 4 months. However, these competing regions were also found for changes in cosmic rays in Pierce and Adams (2009a) and for changes in the nucleation rate in general in Pierce and Adams (2009b). Both of these previous studies used a different host model (GISS GCM II') and evaluated longer time periods (1 year). On average in Figure 7c and 7d, the areas of increases and decreases in the free troposphere largely offset each other (global FT change less than 0.05%, Figure 8) for both CN40 and CN80.”

*Page 2714, line 1-10: This passage describes in detail the content of Figure 8, but it could be shortened to its most relevant portions.*

This paragraph has been shortened in the revised version of the manuscript.

*Table 1: Simulation "CHARGE": Is the sulfuric acid condensation rate charge-enhanced for all sub-10 nm particles, or only for those particles that formed from a gas phase ion? If the former is the case, can that approach be justified?*

It is the former case. No, it is not realistic and we should have made this more clear in the text. We have added the following sentence to the description of the CHARGE simulations: “It is unrealistic that every particle contains a charge, so this provides an upper bound to this enhancement.”

*Figure 8: The change from solar maximum to minimum in CN10 in the "ALL" simulation is always smaller than in the "LoPE" simulation over land areas (Fig. 8 b). This is*

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explained on page 2713 line 19-24:

*“The change in CN10 between solar-maximum and solar-minimum is less for the ALL case than for the LoPE case for all surface locations. This is because the additional SOA and charge-enhanced growth cause a decrease in the ability of nucleated particles to grow to 10nm when primary emissions are reduced (the relative increase in the coagulation sink from the extra SOA and charge-enhanced growth is larger when primary emissions are reduced).”*

*However, over the land areas (with the exception of the tropics), the change from solar maximum to minimum in CN80 is LARGER in the "ALL" simulation compared to the "LoPE" simulation (Fig. 8 c). Is this consistent with the above explanation, and is this behavior of C10 and C80 consistent?*

Yes, it certainly can be consistent. Adding extra SOA to the reduced primary emissions case (the ALL case) causes ultrafine particles to grow more rapidly to CN80 sizes. Thus even though the LoPE cases may have a higher sensitivity of CN10 to changes in nucleation than the ALL case, it may have a lower sensitivity of CN80 to changes in nucleation than the ALL case.

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