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Atmospheric Chemistry and Physics Discussions

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 of reviewer 2 are below. The original reviewer comments are in italics.

Anonymous Referee #2 Received and published: 31 March 2011

General:

In this manuscript, the so-called ion-aerosol clear-air mechanism connecting cosmic rays and climate is investigated using global model simulations. The investigation expands over the previous studies on the subject 1) by using a global model having very

detailed description for aerosol microphysics, and 2) by considering both Forbush decreases and typical changes between solar maximum and minimum. The paper is very well written and easy to follow. I do not find any scientific mistakes. After the authors have addressed the few minor issues given below, the paper can be accepted for publication in Atmospheric Chemistry and Physics.

Scientific comments:

Section 2.1: Two minor issues: 1) Was there some specific reason for averaging the results over 8 simulated Forbush decreases as done in the paper, 2) Why were the wavelengths 340 and 440 nm chosen as the basis for calculating the Angstrom exponent? Was it because of model size resolutions? How does it correspond to respective calculations made from AERONET or satellite retrievals?

Regarding the first question, we added the sentence, "We simulated multiple events to improve the signal to noise ratio of the aerosol parameters."

Regarding the second question. These wavelengths were used entirely because these were the wavelengths used in the AERONET measurements presented in Svensmark et al. (2009), so we could have the most straightforward comparison. In Svensmark et al. (2009) this wavelength pair showed the strongest response to the Forbush decrease. We now say in the paper: "These wavelengths are the same as used by Svensmark et al. (2009) to calculate the AE from AERONET measurements."

Section 2.2: The authors should explain how the additional SOA in xSOA-experiments is distributed over the particle size spectrum. I suppose it is done similarly to other SOA. How is gas-particle portioning of organic vapors taken care of in the model? This is important since it is expected to affects greatly the relation between CN and CCN formation.

Yes, the details of SOA condensation greatly affect aerosol growth and CCN predictions. The SOA is assumed to be non-volatile and condensed onto the Fuchs-corrected aerosol surface area similar to sulfate. This was mentioned in the paper, but we have expanded upon it. It now reads: "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."

Riipinen, I., Pierce, J. R., Yli-Juuti, T., Nieminen, T., Häkkinen, S., Ehn, M., Junninen, H., Lehtipalo, K., Petäjä, T., Slowik, J., Chang, R., Shantz, N. C., Abbatt, J., Leaitch, W. R., Kerminen, V.-M., Worsnop, D. R., Pandis, S. N., Donahue, N. M., and Kulmala, M.: Organic condensation – a vital link connecting aerosol formation to climate forcing, Atmos. Chem. Phys. Discuss., 11, 387-423, doi:10.5194/acpd-11-387-2011, 2011.

Section 3.1: Can the authors pinpoint reasons for the substantially weaker prediction of Angstrom exponent as compared with total particle number concentrations (CN10), as indicated by Figure 3? Intuitively, one would expect that CN10 values are substantially more sensitive to various model assumptions than accumulation mode particle concentrations which determine the value of Angstrom exponent.

Accumulation mode particles are indeed less sensitive to model assumptions than ultrafine particles. It is hard to directly compare the model evaluation of CN10 to the model evaluation of AE. CN10 is plotted in log scale in order to see the full range of particle number concentrations in the atmosphere. Thus we calculate log-mean biases. AE cannot be plotted on log scale since AE values can be negative. Thus we plot mean biases.

Additionally, one may predict the correct number of accumulation-mode aerosols, but if the size is somewhat wrong, the AE will be incorrect.

Finally, correctly predicting AE also requires aerosol water uptake to be correctly predicted, whereas number comparisons (CN10) do not directly require this.

Section 3.1: Besides aerosol number concentrations and Angstrom exponents, it would be very interesting to know how large growth rates of nucleated particle the model predicts in the boundary layer and free troposphere, and how this compares with observed growth rates. As the authors certainly know, the growth rate is a very essential quantity in determining the relation between modeled CN and CCN concentrations.

Absolutely. We are currently doing a detailed evaluation of modelled nucleation rates, growth rates and survival probabilities (of nucleated particles growing to CCN sizes) to measurements in Hyytiälä, Po Valley, Pittsburgh, Atlanta and St. Louis. It will hopefully be submitted to ACPD this summer.

Westervelt, D.M., I. Riipinen, J.R. Pierce, W. Trivitayanurak, and P.J. Adams: Formation, Growth, and Cloud Condensation Nuclei Production of Nucleated Particles: Comparison of Observations to a Global Aerosol Microphysics Model, in preparation for ACPD, 2011.

Sections 3.2.1 and 3.2.2: The changes in CN and CCN concentrations resulting from changes in cosmic ray-induced changes in nucleation are logical and well explained in the paper. How about changes in Angstrom exponent? How and by which atmospheric processes do cosmic ray flux variations influence the value of Angstrom exponent? A brief discussion on this issue would be very helpful for the reader.

Yes, this should have been discussed. We have modified the discussion of the AE in Figure 7a: "Regarding the AE, an increase in the nucleation rate associated with an increase in cosmic rays could increase the flux of particles to the accumulation mode. This increase could shift the optical effective diameter to smaller sizes and increase the AE. However, the column-integrated AE (Figure 7a) shows only a small change between the solar-minimum and solar-maximum simulations. The absolute changes are all between -0.02 and 0.02. The globally averaged change in AE is less than 0.001 (Figure 8a). This shows that there is not a significant change in the optical effective diameter at these wavelengths. These maximum predicted changes in AE ($\pm$0.02) are an order of magnitude smaller than the average change in the AE measured by Svensmark et al. (2009) during five major Forbush-decrease events. The Forbush-decrease events in Svensmark et al. (2009) had a similar change in the cosmic-ray flux as the solar cycle."

In the discussion of Figure 8, we added the sentence: "Again this shows that the optical effective diameter does not change greatly between solar maximum and solar minimum."

Section 4, last paragraph: The authors appear to demonstrate very convincingly that the ion-aerosol clear-air mechanism is too weak to affect the connection between cosmic rays and climate. Yet they leave the door open for this mechanism by saying that they might have misinterpreted some processes. I am not very much favor in this kind of a statement. What could possibly change the conclusion about the weakness of the ion-aerosol clear-air mechanism? The authors correctly point out that we know too little about other potential mechanism, such as the near-cloud mechanism, too say anything definite for the overall connection between cosmic rays and climate.

We agree that we cannot think of anything that would change the weakness of the ion-aerosol clear-air mechanism in our model. However, we are more comfortable leaving this statement in the manuscript since currently the only other published global estimate of cosmic rays on CCN is by a related group of authors. We do have confidence in our work; however, you can never be 100% sure you aren't missing something. We do not feel that our 2 studies alone should be enough to entirely lay the clear-sky mechanism to rest (particularly considering the week-long delay in reported changes in AERONET AE in Svensmark et al. (2009)).

However, we have somewhat rewritten this paragraph. Hopefully this is more agreeable to this reviewer. "Although the simulated ion-aerosol clear-sky mechanism is shown to be very minor, this by no means disproves the connection between cosmic rays and climate. In this paper we tested the uncertain model inputs that we suspected

were most likely to affect the connection between cosmic rays and CCN. However, it is possible that a different model process that was untested here is being misrepresented and causes an underestimation of the effects of cosmic rays on CCN, though it is unclear what model processes this might be. Additionally, another mechanism may be at work such as the ion-aerosol *near-cloud* mechanism, but strategies to test the magnitude of the near-cloud mechanism have yet to be developed."

Technical issues: Page 2701, line 7: "coagulation a larger particle"; grammatical error

Fixed. Thanks.