



Can cosmic rays affect cloud condensation nuclei by altering new particle formation rates?

J. R. Pierce,^{1,2} and P. J. Adams¹

Received 3 March 2009; revised 1 April 2009; accepted 13 April 2009; published 13 May 2009.

[1] Although controversial, many observations have suggested that low-level cloud cover correlates with the cosmic ray flux. Because galactic cosmic rays have likely decreased in intensity over the last century, this hypothesis, if true, could partly explain 20th century warming, thereby upsetting the consensus view that greenhouse-gas forcing has caused most of the warming. The “ion-aerosol clear-air” hypothesis suggests that increased cosmic rays cause increases in new-particle formation, cloud condensation nuclei concentrations (CCN), and cloud cover. In this paper, we present the first calculations of the magnitude of the ion-aerosol clear-air mechanism using a general circulation model with online aerosol microphysics. In our simulations, changes in CCN from changes in cosmic rays during a solar cycle are two orders of magnitude too small to account for the observed changes in cloud properties; consequently, we conclude that the hypothesized effect is too small to play a significant role in current climate change. **Citation:** Pierce, J. R., and P. J. Adams (2009), Can cosmic rays affect cloud condensation nuclei by altering new particle formation rates?, *Geophys. Res. Lett.*, *36*, L09820, doi:10.1029/2009GL037946.

1. Introduction

[2] The effect of solar variability on the Earth’s climate remains a controversial area of climate change science. Cycles in numerous climate phenomena, including tropospheric and stratospheric temperatures, sea-surface temperatures, sea-level pressure, and low-level cloud cover have been observed to correlate with the 11-year solar cycle [Crooks and Gray, 2005; Gleisner and Thejll, 2003; Labitzke, 2005]. Although uncertainties remain, for many current estimates of climate sensitivity and response times of the climate system to external forcings, the variation in solar irradiance over the cycle of about 0.1% is insufficient by itself to explain the ~ 0.1 K variation in tropospheric temperature associated with the solar cycle [Douglass and Clader, 2002; Svensmark and Friis-Christensen, 1997]. Therefore, there has been a search for other mechanisms to explain observed climate correlations with the solar cycle, including the effect of UV variations on stratospheric chemistry as well as hypothesized effects coupling cosmic rays, aerosols, and clouds [Forster et al., 2007].

[3] The galactic charged-particle (cosmic-ray) flux to the Earth’s lower atmosphere varies by about 15% over the

11-year solar cycle, driven by changes in the solar wind with higher cosmic ray fluxes to the troposphere during the solar minimum [Carslaw et al., 2002]. A correlation between the atmospheric cosmic-ray flux and global low-level cloud cover was first reported by Svensmark and Friis-Christensen [1997] with less cloud cover during the solar maximum (when the cosmic-ray flux is at a minimum). The global cloud cover was found to vary by 2–4% through the solar cycle, corresponding to changes in radiative forcing of 1–2 W m⁻² (globally averaged, the shortwave forcing of clouds is about 47 W m⁻² [Kim and Ramanathan, 2008]). The observed correlation remains controversial, however, as other analyses have shown both strong correlations [e.g., Harrison and Stephenson, 2006] and weaker or no correlation [e.g., Sun and Bradley, 2002].

[4] Cosmic-ray proxies indicate that the cosmic-ray flux has decreased over the past century by an amount similar to the cosmic-ray change during a solar cycle [Carslaw et al., 2002]. If the strong potential connection of cosmic rays and the cloud radiation budget described above is correct, this cosmic-ray change would have caused a significant positive (warming) radiative forcing of 1–2 W m⁻² during the 20th century (for present-day aerosol conditions, similar to the observed solar cycle correlations). Such a connection would strengthen the relationship between solar variability and global temperature and greatly upset a consensus view of anthropogenic climate change; however, the physics connecting cosmic rays and clouds are still uncertain and a physical representation of the effect of cosmic rays on clouds has not yet been tested in climate models.

[5] Much of the uncertainty in the cosmic ray-cloud coupling comes from poor understanding of the physical mechanisms connecting cosmic rays to clouds. The driving factor of these physical connections is the formation of small ions when cosmic rays interact with atmospheric gases in the Earth’s lower atmosphere. Two mechanisms have been proposed that link cosmic-ray-induced ion formation to changes in cloud cover: the ion-aerosol “clear-air” mechanism and the ion-aerosol “near-cloud” mechanism [Carslaw et al., 2002]. The ion-aerosol clear-air mechanism has generally received the most attention and is the focus here. It occurs when the formation of new atmospheric particles (aerosol nucleation) is enhanced by the presence of these ions (ion-induced nucleation) [Carslaw et al., 2002]. The newly formed particles, with diameters of approximately 1 nm, may grow in the atmosphere through the condensation of gases, such as sulfuric acid and low-volatility organics, to sizes where they act as cloud condensation nuclei (CCN), generally around 80–100 nm for stratus clouds. Increases in CCN number lead to increases in the number of cloud droplets, which in turn may lead to more reflective and more persistent clouds [Forster et al., 2007]. There are,

¹Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, Pennsylvania, USA.

²Now at Department of Physics and Atmospheric Science, Dalhousie University, Halifax, Nova Scotia, Canada.

Table 1. Overview of Simulations With Global Aerosol Number Budgets^a

Simulation Name	Ion-Induced Nucleation Parameterization	Solar Activity	Nucleation Rate ($\text{cm}^{-3}\text{s}^{-1}$)	CCN(0.2%) (cm^{-3})	CCN(0.2%) Min:Max Ratio
MODGIL-MIN	<i>Modgil et al.</i> [2005]	min	8.7E-03	1.45E+02	1.00004
MODGIL-MAX	<i>Modgil et al.</i> [2005]	max	1.9E-03	1.45E+02	1.00004
IONLIMIT-MIN	All ions form new particles	min	6.3E+01	1.63E+02	1.00079
IONLIMIT-MAX	All ions form new particles	max	5.1E+01	1.63E+02	1.00079

^aDetailed descriptions of simulations in text. Values normalized by tropospheric volume at 273 K and 1 atm.

however, several factors dampening the effect of cosmic ray/ion-induced nuclei on CCN and cloud cover: 1) most nucleated particles are lost through coagulation before reaching CCN sizes [*Pierce and Adams*, 2007], 2) a significant fraction of CCN are derived from primary emissions (particles emitted directly from sources such as cars and the ocean), which should not be modulated by cosmic rays [*Pierce and Adams*, 2009], and 3) most of the ionization potential of cosmic rays is at high altitude and high latitude, so in order to modify low clouds at low latitudes (as is often seen in the cosmic ray/cloud correlations [*Carlsaw et al.*, 2002]), the CCN must be transported vast distances. Physical models exploring the potential of the ion-aerosol clear-air mechanism as a possible connection between cosmic rays and clouds must account for these dampening factors.

[6] In this paper, we determine the ability of cosmic rays to modify cloud properties through the ion-aerosol clear-air mechanism using a global model of the Earth's atmosphere with online aerosol nucleation and microphysics. Previous global evaluations have addressed the contribution of nucleation in general to CCN [e.g., *Pierce and Adams*, 2009; *Spracklen et al.*, 2008] and the global contribution of cosmic rays to new-particle formation [*Lucas and Akimoto*, 2006], but not their subsequent growth to CCN. To our knowledge, this is the first paper to evaluate ability of cosmic rays to modulate CCN concentrations globally.

2. Methods

[7] To address the coupling of cosmic rays and CCN, we calculate the number and composition of aerosol particles as a function of particle size and time throughout the lower atmosphere (troposphere and lower stratosphere). Additionally, we simulate the gas-phase DMS, SO₂, H₂SO₄ and secondary organic aerosol (SOA) precursor concentrations relevant, allowing us to calculate the aerosol nucleation rates and growth of these new particles to CCN sizes. The simulations are performed in the Goddard Institute for Space Studies General Circulation Model II-prime (GISS GCM II-prime) [*Hansen et al.*, 1983; *Rind and Lerner*, 1996] with online aerosol microphysics using the Two-Moment Aerosol Sectional (TOMAS) microphysics algorithm [*Adams and Seinfeld*, 2002]. The up-to-date details of aerosol emissions and microphysics in the model are described by *Pierce and Adams* [2009] (base-case present-day emissions). Each run is initialized for three months followed by a full year of simulation. The results in this paper are annually averaged.

3. Simulations Details

[8] Table 1 describes the four simulations presented here and provides global particle number budget data. We

simulate years with high (solar min) and low (solar max) cosmic ray fluxes approximately corresponding to years 1986 and 1990, respectively, leaving all other model inputs constant. Ion-formation rates from cosmic rays are calculated using the method of *Usoskin and Kovaltsov* [2006] with input for solar minimum and solar maximum time periods (modulation potential of 0.4 and 1 GV, respectively). Decay of ²²²Rn contributes to ion production near the Earth's continental surface and is prescribed based on *Bricard* [1965] and is constant between simulations. Throughout most of the free troposphere, the ion-pair production rate changes by 15–30% between the solar maximum and minimum, with changes less than 10% in the boundary layer. Because the mean cosmic-ray flux is thought to have decreased throughout the 20th century by a similar magnitude as the change during the solar cycle [*Carlsaw et al.*, 2002], these simulations show whether or not these cosmic ray changes may be affecting present-day climate. We simulate two different ion-induced nucleation schemes in order to address uncertainties in ion-induced nucleation. The first scheme (MODGIL) is the parameterization of *Modgil et al.* [2005], in which the aerosol nucleation rate as a function of the ion-pair formation rate, sulfuric acid concentration, temperature, relative humidity and existing aerosol surface area. We apply the scheme only when temperature is less than 260 K to prevent unrealistic behavior of the curve fit at higher temperatures [*Lucas and Akimoto*, 2006]. This limitation of the MODGIL scheme may affect its ability to predict changes in low-level cloud cover [e.g., *Svensmark and Friis-Christensen*, 1997] as nucleation will not occur in the boundary layer except for at high latitudes; however, nucleation events in the free troposphere may still affect CCN in the boundary layer. In the second scheme (IONLIMIT), we assume that every ion produced, positive or negative, will nucleate a particle provided that there is enough sulfuric acid to grow the particle to a diameter of 1 nm. While this may produce nucleation rates that are unreasonably high, it will provide an upper limit on the contribution of ion-induced nucleation to new-particle formation and CCN within our model framework. Each of the two ion-induced nucleation schemes is simulated for both high and low cosmic ray years.

4. Aerosol Nucleation Rates

[9] Figure 1 shows zonal average maps of the annually averaged nucleation rates (formation rate of stable particles with diameters ~ 1 nm) during maximum solar activity and the ratio of aerosol nucleation rates between years of minimum and maximum solar activity for both nucleation schemes. The nucleation rates in MODGIL-MAX (Figure 1a) show moderate nucleation rates ($\sim 10^{-2} \text{ cm}^{-3} \text{ s}^{-1}$ at 273 K and

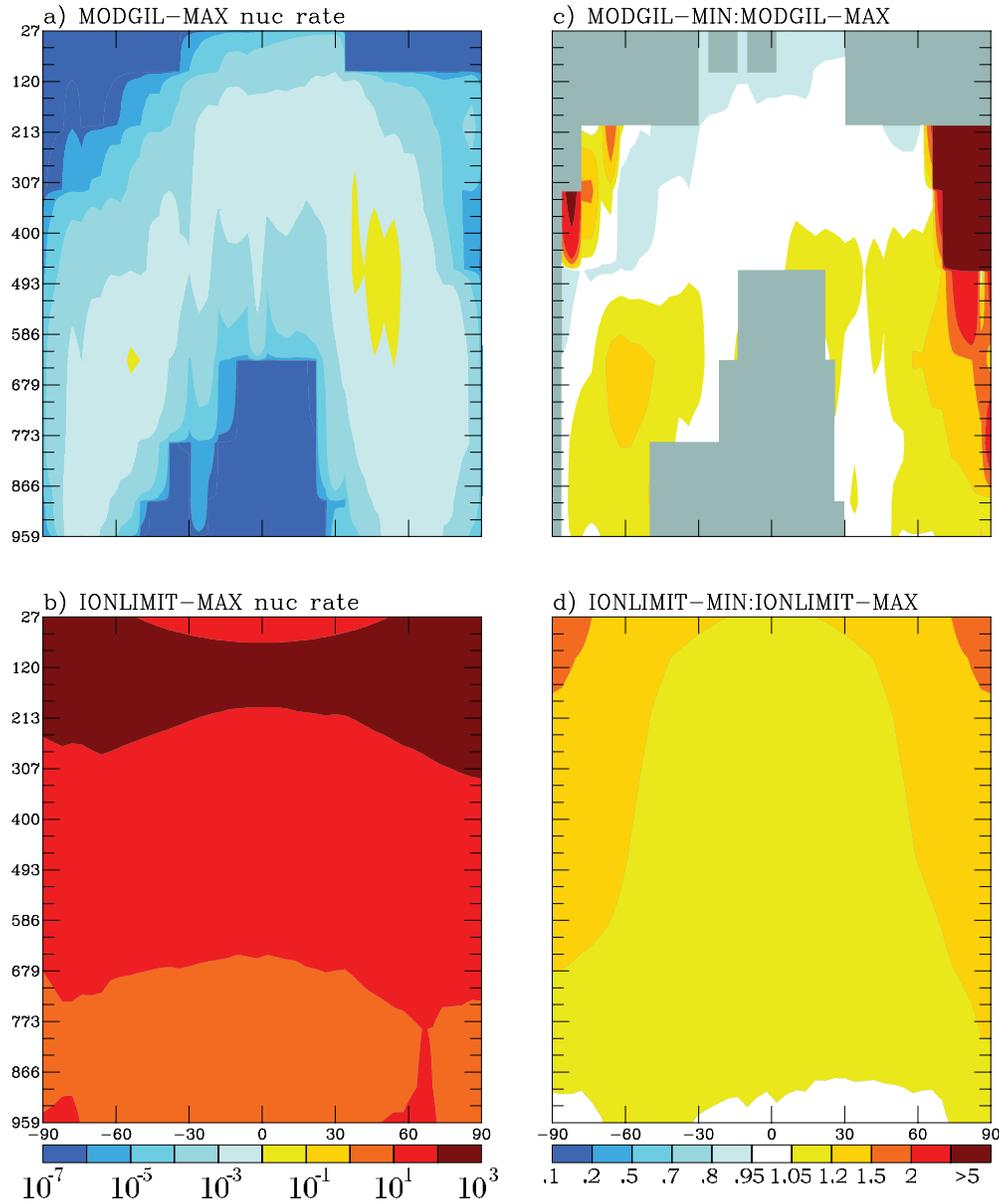


Figure 1. Annually averaged pressure (hPa) versus latitude plots of (a) nucleation rates ($\text{cm}^{-3} \text{s}^{-1}$ at 273 K and 1 atm) in the MODGIL-MAX simulation, (b) nucleation rates ($\text{cm}^{-3} \text{s}^{-1}$ at 273 K and 1 atm) in the IONLIMIT-MAX, (c) ratio of nucleation rates in MODGIL-MIN to those of MODGIL-MAX, and (d) ratio of nucleation rates in IONLIMIT-MIN to those of IONLIMIT-MAX. Gray areas in panel Figure 1c reflect areas where average nucleation rates were less than $10^{-6} \text{cm}^{-3} \text{s}^{-1}$ at 273 K and 1 atm during one of the simulations.

1 atm) in the upper troposphere and colder lower troposphere. Nearly everywhere, the rates change by less than a factor of 2; however, the nucleation rates increase greatly near the poles above the 500 hPa level (Figure 1c). Globally, the nucleation rates increased between MODGIL-MAX and MODGIL-MIN by over a factor of 4 (Table 1).

[10] Because every ion in the presence of even a tiny amount of sulfuric acid vapor nucleates a particle, the nucleation rates in IONLIMIT-MAX (Figure 1b) follow the same spatial pattern as the ion formation rates. The nucleation rates in IONLIMIT-MAX ($>10 \text{cm}^{-3} \text{s}^{-1}$ at 273 K and 1 atm) are much larger than the MODGIL-MAX simulations ($\sim 25,000$ times faster globally, Table 1). The increase in the nucleation rates between IONLIMIT-MAX and

IONLIMIT-MIN (Figure 1d) is similar to the increase in ion production rates between the solar maximum and minimum: a 24% increase in nucleation rate globally (Table 1).

[11] The changes in the global nucleation rates between the solar minimum and maximum provide an upper bound on the global change in CCN (if primary emissions did not contribute to CCN and all nucleated particles became CCN). However, the changes in CCN in the atmosphere must be lower.

5. CCN Concentrations

[12] Figure 2 shows zonal-average maps of the annually averaged CCN at 0.2% supersaturation ($\text{CCN}_{0.2\%}$), a typical

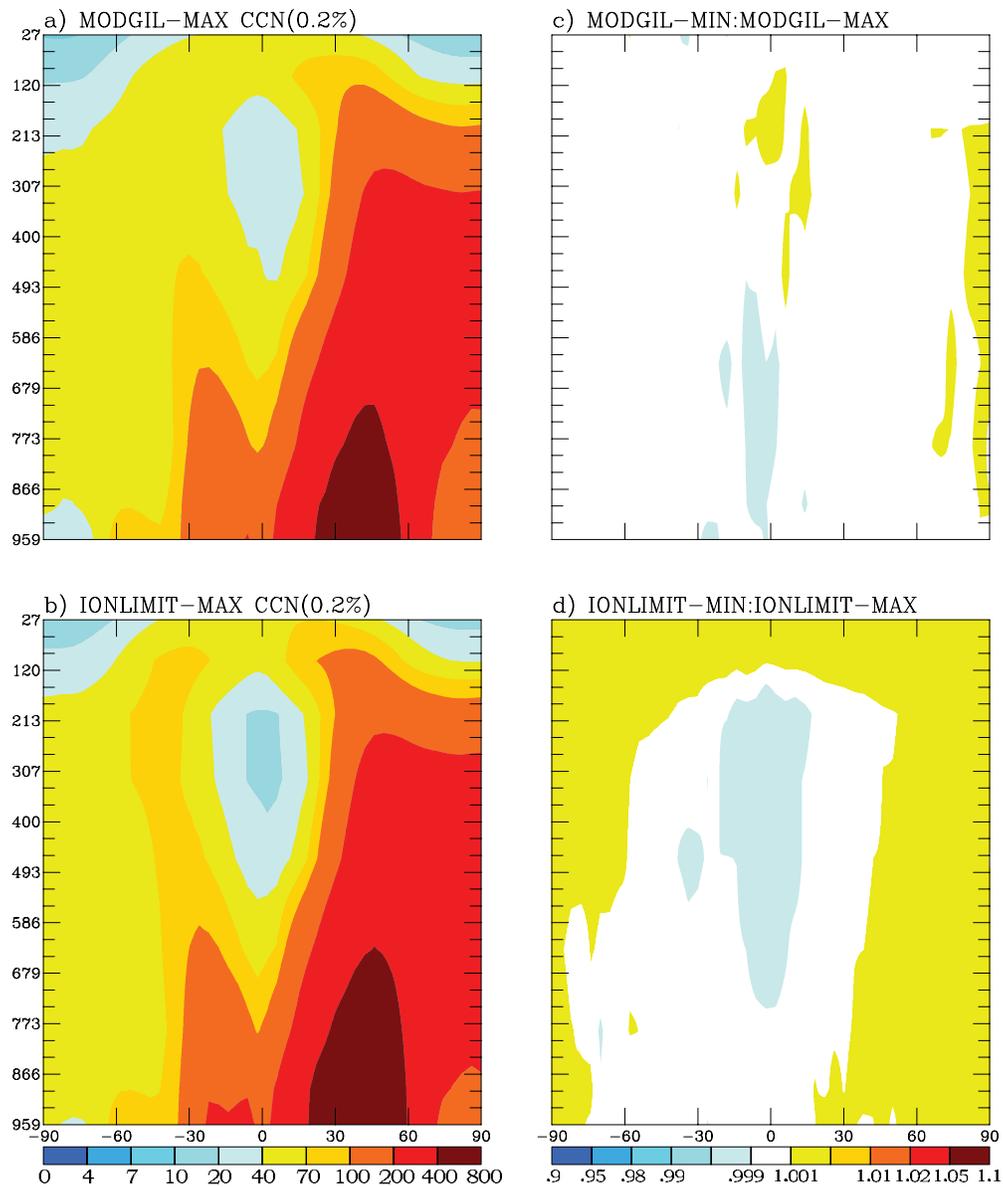


Figure 2. Annually averaged pressure (hPa) versus latitude plots of (a) $\text{CCN}_{0.2\%}$ concentrations (cm^{-3} at 273 K and 1 atm) in the MODGIL-MAX simulation, (b) $\text{CCN}_{0.2\%}$ concentrations (cm^{-3} at 273 K and 1 atm) in the IONLIMIT-MAX, (c) ratio of $\text{CCN}_{0.2\%}$ concentrations in MODGIL-MIN to those of MODGIL-MAX, and (d) ratio of $\text{CCN}_{0.2\%}$ concentrations in IONLIMIT-MIN to those of IONLIMIT-MAX.

supersaturation reached in low stratus clouds [Pruppacher and Klett, 1997]. Figures 2a and 2b show the $\text{CCN}_{0.2\%}$ concentrations during maximum solar activity. In both MODGIL-MAX (Figure 2a) and IONLIMIT-MAX (Figure 2b), the $\text{CCN}_{0.2\%}$ are highest in the northern hemisphere mid-latitude lower troposphere. The $\text{CCN}_{0.2\%}$ concentrations here are dominated by emissions of particles directly from anthropogenic sources. Globally, IONLIMIT-MAX has 12% more $\text{CCN}_{0.2\%}$ than MODGIL-MAX due to the faster nucleation rates. A simulation that turned off all nucleation while keeping all other inputs constant (NONUC of Pierce and Adams [2009], not shown here) has 17% fewer $\text{CCN}_{0.2\%}$ than MODGIL-MAX and 26% fewer $\text{CCN}_{0.2\%}$ than IONLIMIT-MAX globally. Although IONLIMIT-MAX has

several orders magnitude faster nucleation than MODGIL-MAX, there is only the relatively small increase in $\text{CCN}_{0.2\%}$. This insensitivity is due to a limited amount of material that can condense onto the particles to allow them to grow from ~ 1 nm to ~ 80 nm in diameter.

[13] Figures 2c and 2d show the ratio of the $\text{CCN}_{0.2\%}$ concentrations between the years of minimum and maximum solar activity for both ion-induced nucleation schemes. For either scheme, there is almost negligible sensitivity of $\text{CCN}_{0.2\%}$ to the changes in the nucleation rate between the solar maximum and the solar minimum. Globally for the MODGIL simulations, there is a 0.004% increase in $\text{CCN}_{0.2\%}$ even though there is a factor of 4 increase in the new particle formation rate, and for the

IONLIMIT simulations, there is a 0.08% increase in $CCN_{0.2\%}$ from a 24% increase in the nucleation rate (Table 1).

6. Discussion

[14] The CCN concentrations in these tests are quite insensitive to the changes in nucleation rates that occur between the solar minimum and maximum. The insensitivity of CCN to aerosol nucleation merits further explanation. At faster nucleation rates, more nm-sized particles are competing for condensable gases that grow to CCN sizes. Each particle thus grows more slowly, keeping the particles at the smallest sizes where they have fast coagulation scavenging timescales [Pierce and Adams, 2007]. Although more particles have formed giving the potential for more CCN to form, the probability of any particle surviving to CCN size is much lower. This strong negative feedback strongly dampens the sensitivity of CCN to changes in the new-particle formation rate (dampening effect). Finally, emissions of primary particles contribute a significant fraction of tropospheric CCN and do not vary with solar activity (primary emissions effect). These two effects are strongly apparent when comparing three simulations with very different nucleation rates. A simulation by Pierce and Adams [2009], NONUC, without nucleation but otherwise identical to these simulations, has an annual-average $CCN_{0.2\%}$ concentration of 116 cm^{-3} . This simulation can be compared with MODGIL-MIN (annual-average nucleation rate is $8.7 \times 10^{-3} \text{ cm}^{-3} \text{ s}^{-1}$ and $CCN_{0.2\%}$ is 145 cm^{-3}) and IONLIMIT-MIN (annual-average nucleation rate is $6.3 \times 10^1 \text{ cm}^{-3} \text{ s}^{-1}$ and $CCN_{0.2\%}$ is 163 cm^{-3}). The primary emissions alone in NONUC account for much of the CCN seen in the other simulations (primary emissions effect). The $CCN_{0.2\%}$ increase by 25% with the addition of the relatively low nucleation rates in the MODGIL simulations; however, the $CCN_{0.2\%}$ only increase by another 12% when the nucleation rates are increased to the IONLIMIT rates that are 4 orders of magnitude higher (dampening effect).

[15] How might these small changes in CCN affect cloud properties? Here we use a simple test to estimate the potential shortwave radiative forcing due to an increase in cloud droplets. We assume that the change in cloud radiative forcing is primarily by low-level stratus clouds with a supersaturation 0.2% and a dark surface underneath, that cloud liquid water content is constant, and that changes in cloud droplet number correspond linearly to changes in $CCN_{0.2\%}$. The following equation is used to estimate the change in the sun's energy reflected by these clouds between the solar maximum and minimum [Seinfeld and Pandis, 2006].

$$\Delta F_c = -\frac{1}{3} F_0 A_c T_a^2 R_c (1 - R_c) \Delta \ln N \quad (1)$$

We assume that: F_0 , the incoming shortwave flux, is 343 W m^{-2} ; A_c , the fractional area covered by clouds, is 0.3; R_c , the albedo of the clouds, is 0.5; T_a , the globally-averaged above-cloud transmittance, is 0.76; and N , the number of cloud droplets is equal to the globally averaged $CCN_{0.2\%}$ [Seinfeld and Pandis, 2006]. Given these values, the change in the sun's energy reflected by clouds, ΔF_c , induced by a 0.1% change in CCN (the maximum effect of cosmic rays

on $CCN_{0.2\%}$ simulated here) is 0.005 W m^{-2} (averaged over cloudy and clear areas).

[16] The test above assessed the change in cloud reflectivity due to a change in cloud-droplet number caused by cosmic rays at constant cloud cover; however, this does not include the various cloud microphysical and dynamical feedbacks, such as changes in precipitation, cloud lifetime and cloud depth, which may also occur because of changes in CCN (generally cosmic ray correlations in the literature are with cloud-cover extent [Carslaw *et al.*, 2002]). Nevertheless numerous modeling studies show that these feedbacks to changes in CCN affect the solar reflection comparably to the cloud-albedo effect [Denman *et al.*, 2007]. Therefore, the changes in cloud cover and thickness from the changes in CCN induced by clear-sky ion-induced nucleation changes during both the solar-cycle and centennial-scale changes in cosmic rays should also be of order 0.01 W m^{-2} . Even including the feedbacks, this is two orders of magnitude too small to explain the changes seen in cloud cover [Svensmark and Friis-Christensen, 1997] or to be a significant contributor to present-day climate change. Because we have only simulated present-day conditions, the role of cosmic rays on CCN prior to present-day conditions remains uncertain, although Pierce and Adams [2009] found that the sensitivity of CCN to nucleation was similar between pre-industrial and present-day simulations. Regardless, any changes in the cosmic ray flux during the 20th century have likely had little impact on present-day climate.

[17] Because models are imperfect, uncertain factors such as the condensation rate of secondary organic aerosol (SOA) and the primary emissions rate will, under more favorable conditions, increase the effect of cosmic rays on CCN. Pierce and Adams [2009] and Spracklen *et al.* [2008] showed that the sensitivity of $CCN_{0.2\%}$ to changes in aerosol nucleation rates is greater with more SOA and/or less primary aerosol emission. These known uncertainties will affect the results; however, based on the sensitivity studies of Pierce and Adams [2009] and Spracklen *et al.* [2008], even at the most favorable conditions for $CCN_{0.2\%}$ sensitivity to nucleation, the $CCN_{0.2\%}$ would only change by several tenths of a percent throughout the atmosphere because of changes in the cosmic-ray flux, still more than an order of magnitude too low to account for the reported changes in cloud properties. An additional and potentially non-trivial uncertainty exists in the growth and loss of particles smaller than 10 nm diameter within the model. This growth is parameterized and does not account for self coagulation of nucleation-mode particles (this will be more important at the faster nucleation rates of the IONLIMIT cases).

[18] Based on the results of these simulations, we conclude that – barring any strong biases due to model uncertainties – the ion-aerosol clear-air mechanism is too weak to explain putative correlations between cloud cover and the solar cycle. This does not rule out the potential legitimacy of the connection between cosmic rays and clouds by other physical mechanisms. Further work needs to be done with detailed cloud models to determine the strength of other potential mechanisms.

[19] **Acknowledgments.** This research was supported by the Environmental Protection Agency (EPA) through the Science to Achieve Results

(STAR) Graduate Fellowship (91668201-0) as well as a research grant from the National Aeronautics and Space Administration (NASA grant NNG04GE86G).

References

- Adams, P. J., and J. H. Seinfeld (2002), Predicting global aerosol size distributions in general circulation models, *J. Geophys. Res.*, *107*(D19), 4370, doi:10.1029/2001JD001010.
- Bricard, J. (1965), Action of radioactivity and of pollution upon parameters of atmospheric electricity, in *Problems of Atmospheric and Space Electricity*, edited by S. C. Coroniti, pp. 83–117, Elsevier, New York.
- Carslaw, K. S., et al. (2002), Cosmic rays, clouds, and climate, *Science*, *298*(5599), 1732–1737.
- Crooks, S. A., and L. J. Gray (2005), Characterization of the 11-year solar signal using a multiple regression analysis of the ERA-40 dataset, *J. Clim.*, *18*(7), 996–1015.
- Denman, K., et al. (2007), Couplings between changes in the climate system and biogeochemistry, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 499–587, Cambridge Univ. Press, Cambridge, U.K.
- Douglass, D. H., and B. D. Clader (2002), Climate sensitivity of the Earth to solar irradiance, *Geophys. Res. Lett.*, *29*(16), 1786, doi:10.1029/2002GL015345.
- Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 129–234, Cambridge Univ. Press, Cambridge, U.K.
- Gleisner, H., and P. Thejll (2003), Patterns of tropospheric response to solar variability, *Geophys. Res. Lett.*, *30*(13), 1711, doi:10.1029/2003GL017129.
- Hansen, J., et al. (1983), Efficient 3-dimensional global-models for climate studies—Model-I and Model-II, *Mon. Weather Rev.*, *111*(4), 609–662.
- Harrison, R. G., and D. B. Stephenson (2006), Empirical evidence for a nonlinear effect of galactic cosmic rays on clouds, *Philos. Trans. R. Soc., Ser. A*, *462*(2068), 1221–1233.
- Kim, D., and V. Ramanathan (2008), Solar radiation budget and radiative forcing due to aerosols and clouds, *J. Geophys. Res.*, *113*, D02203, doi:10.1029/2007JD008434.
- Labitzke, K. (2005), On the solar cycle-QBO relationship: A summary, *J. Atmos. Sol. Terr. Phys.*, *67*(1–2), 45–54.
- Lucas, D. D., and H. Akimoto (2006), Evaluating aerosol nucleation parameterizations in a global atmospheric model, *Geophys. Res. Lett.*, *33*, L10808, doi:10.1029/2006GL025672.
- Modgil, M. S., S. Kumar, S. N. Tripathi, and E. R. Lovejoy (2005), A parameterization of ion-induced nucleation of sulphuric acid and water for atmospheric conditions, *J. Geophys. Res.*, *110*, D19205, doi:10.1029/2004JD005475.
- Pierce, J. R., and P. J. Adams (2007), Efficiency of cloud condensation nuclei formation from ultrafine particles, *Atmos. Chem. Phys.*, *7*, 1367–1379.
- Pierce, J. R., and P. J. Adams (2009), Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, *Atmos. Chem. Phys.*, *9*, 1339–1356.
- Pruppacher, H. R., and J. D. Klett (1997), Microphysics of clouds and precipitation, in *Atmospheric and Oceanographic Sciences Library*, edited by R. D. Rosen, Kluwer Acad., Dordrecht, Netherlands.
- Rind, D., and J. Lerner (1996), Use of on-line tracers as a diagnostic tool in general circulation model development: 1. Horizontal and vertical transport in the troposphere, *J. Geophys. Res.*, *101*(D7), 12,667–12,683.
- Seinfeld, J. H., and S. N. Pandis (2006), *Atmospheric Chemistry and Physics*, 2nd. ed., John Wiley, New York.
- Spracklen, D. V., et al. (2008), Contribution of particle formation to global cloud condensation nuclei concentrations, *Geophys. Res. Lett.*, *35*, L06808, doi:10.1029/2007GL033038.
- Sun, B., and R. S. Bradley (2002), Solar influences on cosmic rays and cloud formation: A reassessment, *J. Geophys. Res.*, *107*(D14), 4211, doi:10.1029/2001JD000560.
- Svensmark, H., and E. Friis-Christensen (1997), Variation of cosmic ray flux and global cloud coverage—A missing link in solar-climate relationships, *J. Atmos. Sol. Terr. Phys.*, *59*(11), 1225–1232.
- Usoskin, I. G., and G. A. Kovaltsov (2006), Cosmic ray induced ionization in the atmosphere: Full modeling and practical applications, *J. Geophys. Res.*, *111*, D21206, doi:10.1029/2006JD007150.

P. J. Adams, Center for Atmospheric Particle Studies, Carnegie Mellon University, 5000 Forbes Avenue, Pittsburgh, PA 15213, USA.

J. R. Pierce, Department of Physics and Atmospheric Science, Dalhousie University, Halifax, NS B3H 3J5, Canada. (jeffrey.robert.pierce@gmail.com)