# COSMIC RAYS, PARTICLE FORMATION, NATURAL VARIABILITY OF GLOBAL CLOUDINESS, AND CLIMATE IMPLICATIONS

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## Abstract

Via its role in aerosol formation, cosmic ray may affect the global cloudiness and hence climate. Here we show that an increase in cosmic ray fluxes may lead to an increase in particle production in the lower troposphere but a decrease in particle production in the upper troposphere. In addition to the reported positive correlation between cosmic ray variations and low cloudiness, our analysis of satellite-based cloud cover data reveals that high cloudiness may be anti-correlated with cosmic ray variations if volcano and El Niño impacts are excluded. The observed different correlations between cosmic ray variations and low, middle and high cloud anomalies are consistent with the predicted different sensitivities of particle production to cosmic ray changes at various altitudes. The influence of the solar-modulated cosmic ray fluxes on global cloudiness, if confirmed, may provide the external forcing needed to reconcile the apparent differences between observed surface and troposphere temperature trends.

# 1. INTRODUCTION

Clouds play a key role in the energy budget of Earth's surface and lower atmosphere, and are probably the largest contributor to the uncertainty concerning the global climate change<sup>1</sup>. Small modifications of the amount, distribution, or radiative properties of clouds can have significant impacts on the predicted climate<sup>2</sup>. To detect and attribute anthropogenic influences on climate, it is crucial to quantify the natural fluctuations of cloudiness and the associated radiative forcing. In 1997, Svensmark and Friis-Christensen<sup>3</sup> reported a surprising discovery that total cloud cover over midlatitude ocean correlates closely with the galactic cosmic ray (GCR) intensity. The cloud data analyzed include the C2 data sets from the International Satellite Cloud Climatology Project (ISCCP)<sup>4</sup>. Recently it has become possible to infer global cloud properties at different altitudes from the ISCCP-D2 data, which come from an improvement of procedures leading to the C2 data<sup>5</sup>. Analyses of the ISCCP-D2 data indicate that a clear correlation can only be seen between GCR fluxes and the global average of low cloud cover<sup>6,7</sup>. Due to its potential importance and implication, the GCR-cloud-climate hypothesis<sup>3</sup> has been under close scrutiny<sup>8-10</sup>. Two of the main questions and doubts raised against the hypothesis are: (1) no convincing physical mechanism is available to explain the correlation, (2) there is no obvious correlation between solar activity and high cloudiness (where, it is argued, if GCR ionization has any impact on cloud microphysics, it would most likely be found in the upper troposphere where GCR incidence is greatest).

Here we first try to address these two raised issues by investigating the role of GCR ionization in particle formation and the potential altitude-dependent influence of GCR variations on particle production and global cloudiness. We then explore the possibility of GCR-induced global cloud changes as an external forcing that may reconcile the apparent differences in global mean temperature trends between ground and atmosphere measurements. Over the last two decades, the temperature records taken at the Earth's surface show rapid warming (globally 0.15  $\pm$  0.05 °C per decade), however the data produced by satellite and balloon studies indicate little if any warming (globally 0.05  $\pm$  0.10 °C per decade) of the low to mid-troposphere - the

atmospheric layer extending up to about 8 km from the Earth's surface<sup>11,12</sup>. Climate models generally predict that this atmospheric layer should warm faster than the surface if increased concentrations of greenhouse gases are causing the warming. Model simulations taking into account the effects of sulfate aerosols, stratospheric ozone depletion, and volcano eruptions were not able to reconcile these inconsistencies<sup>13-15</sup>. *Gaffen et al*<sup>16</sup> suggested that these inconsistencies may be associated with external forcings of climate system that result in different surface and low tropospheric temperature changes. It is of interest and importance to investigate if the GCR-induced global cloud changes can provide the kind of external forcing needed to reconcile the inconsistencies.

# 2. GCR-CN-CCN-CLOUD HYPOTHESIS



# **GCR-CN-CCN-Cloud Hypothesis**

**Figure 1**. Schematic illustrating of GCR-CN-CCN-Cloud Hypothesis that, if confirmed, might explain the correlation between variations of GCR flux and low cloud cover. The possible dominating species involved in the different phases of CN formation and growth processes are also indicated. The organics species may play an important role in growing the CN into the size of CCN.

Figure 1 shows the GCR-CN-CCN-Cloud hypothesis that, if confirmed, might offer a physicallybased link between GCR fluxes and global low-level cloud properties. Several steps are involved in this hypothesis. First, the modulation of galactic cosmic radiation by the solar cycle will cause a notable variation in aerosol production and condensation nuclei (CN) population in the lower atmosphere. Second, a systematic change in the ultrafine particle production rate will affect the population of cloud condensation nuclei (CCN). Third, a change in CCN abundance will affect the cloud properties. Clouds that form in air containing high CCN concentrations tend to have high droplet concentrations, which leads to both an increase in albedo and an increase in absorption. Increase in the CCN concentration also inhibits rainfall and therefore increases cloud lifetimes (cloud coverage). These effects – which are due to more, smaller droplets at fixed liquid water content – are particularly significant in marine air, where the CCN concentrations are generally quite low. In our proposed hypothesis, the first key process connecting GCR flux and low cloud cover is that ions generated by GCR ionization play an important role in new particle formation in the lower atmosphere which is the focus of discussion of the next section.

# 3. COSMIC RAY IONIZATION AND PARTICLE FORMATION

Ambient ions are continuously generated by galactic cosmic rays at the rate of ~2 ion-pairs cm<sup>-3</sup>s<sup>-1</sup> at ground level and up to ~20-30 ion-pairs cm<sup>-3</sup>s<sup>-1</sup> in the upper troposphere<sup>17,18</sup>. Due to enhanced growth and stability of charged clusters (as a consequence of electrostatic interactions), air ions produced from GCR ionization may play an important role in the production of new particles under typical tropospheric conditions<sup>19,20</sup>. The proposed ion-mediated nucleation (IMN) theory can physically explain the enhanced growth rate (a factor of ~ 10) of sub-nanometer clusters and the square of sulfuric vapor concentration ( $[H_2SO_4]^2$ ) dependence of nucleation rate as observed by *Weber et al*<sup>21</sup>, and seems to account consistently for ultrafine aerosol formation in jet plumes, in clean continental air and in marine boundary layer, as well as for the diurnal variation in the atmospheric mobility spectrum, as demonstrated by *Yu* and his colleagues <sup>19,20,22-24</sup>.

It is generally known that sulfuric acid vapor concentration  $([H_2SO_4])$ , temperature (T), relative humidity (RH), pressure (P), and the surface area of preexisting particles are among the list of parameters controlling the particle formation in the troposphere. The IMN theory adds another important parameter-ion concentration ([ion], or ionization rate O)-to this list. Here we focus on investigating the influence of GCR variations on particle formation and CN abundance at different altitudes. We employ an advanced particle microphysics (APM) model that simulates a size-resolved multicomponent aerosol system via a unified collisional mechanism involving both neutral and charged particles down to molecular sizes<sup>20</sup>. The size-resolved ion-ion recombination coefficients, ion-neutral collision kernels, and neutral-neutral interaction coefficients calculated in the model are physically consistent and naturally altitude (temperature, pressure, and relative humidity) dependent<sup>20</sup>. For the simulations presented below, the ion concentration is initialized as  $\sqrt{Q/\alpha}$  where  $\alpha$  is ion-ion recombination coefficient. The pre-existing particles are initialized as two log-normal modes with total number densities of 19.5/cm<sup>3</sup> and 0.6/cm<sup>3</sup>, median dry diameters of 0.09 µm and 0.3 µm, and standard deviations of 1.6 and 1.5, respectively. This gives an initial wet surface area of ~ 4.5  $\mu$ m<sup>2</sup>/cm<sup>3</sup> at 90% relative humidity, corresponding to a cloud-processed clean air mass where typical significant aerosol nucleation has been observed.

Figure 2 shows the total condensation nuclei bigger than 3nm  $(N_{d>3 nm})$  after three hours of simulations as a function of ionization rates at three different altitudes (0, 5, 8 km). The values of  $[H_2SO_4]$ , T, P, and RH for each altitude (as specified in the figure legend) are fixed during the three-hour simulations. The shaded areas in Figure 1 are the ranges of Q values corresponding to low (>680 mb), middle (440-680 mb), and high (<440 mb) cloud regions as defined in ISCCP cloud data according to the cloud top pressures. It is clear from Figure 2 that significant number of ultrafine particles have formed under all the considered conditions. Most of these newly formed particles began as electrically charged clusters that have the advantage of enhanced growth and stability due to electrostatic effects. The neutral sub-critical clusters, on the other hand, grow too slowly to exceed the critical size under the prevailing conditions. The production rate of ultrafine particles is most sensitive to  $[H_2SO_4]$  and [ion] (or ionization rate).  $[H_2SO_4]$  controls the growth rate of ion clusters, while *[ion]* determines the lifetime of charged clusters and the availability of ions. The neutralization by ion-ion recombination will make the growing charged clusters lose their growth advantage and the resulting neutral clusters may dissociate if smaller than the critical size. At typical  $[H_2SO_4]$  where nucleation has been observed, for very low Q most of the ion clusters have sufficient time to reach the larger stable sizes prior to recombination and the nucleation rate is limited by Q. As Q increases, ion concentration increases but the lifetime of ions decreases and hence the fraction of ions having sufficient time to grow to the larger stable sizes decreases. As a result, the total number of particles nucleated first increases but later on decreases as Q increases. Figure 2 demonstrates that, as Q increases,  $N_{d>3 nm}$  increases rapidly in the low cloud region but decreases in the high cloud region. The Q value at turning point (i.e., dN/dQ=0) is sensitive to  $[H_2SO_4]$  and is most likely located in middle cloud region.



Figure 2. Simulated concentrations of total condensation nuclei larger than 3nm ( $N_{d>3 nm}$ ) after three hour of simulations for various ionization rates (Q) at three altitudes (0, 5, and 8 km). The shaded areas are the ranges of Q corresponding to low (>680 mb), middle (440-680 mb), and high (<440 mb) cloud regions as defined in ISCCP cloud data.  $N_{d>3 nm}$  increases rapidly in the low cloud region but decreases rapidly in the high cloud region as Q increases.

During a solar cycle, the values of Q vary by ~20-25% in the upper troposphere and ~5-10% in the lower troposphere. To study the effect of such systematic change of ionization rates on particle production at different altitudes, we increase the baseline ionization rate at each chosen altitude by 20% and compare the CN abundance after three hours of simulations. The altitude-dependent values of  $[H_2SO_4]$ , Q, T, RH, P, and the surface area of preexisting particles are specified and some of them are shown in Figure 3. The baseline values of Q at different altitudes are from observations<sup>17,18</sup>, and the temperature and pressure are according to the US standard atmosphere. The  $[H_2SO_4]$  and RH are parameterized in a way so that they are constant in the lowest 2 km of atmosphere (2x10<sup>7</sup>/cm<sup>3</sup> and 90%, respectively) and gradually decrease with altitude above 2 km. These parameterizations are reasonable and are within the range of the observed values in various field campaigns<sup>25,26</sup>.

Figure 4 shows the total condensation nuclei bigger than  $3nm (N_{d>3 nm})$  after three hours of simulations at different altitudes. The black line (with opaque circles) is for the baseline Q values while the green line (with filled circles) is for Q values 20% over the corresponding baseline values. The shaded areas in Figure 4 are low, middle, and high cloud regions as defined in ISCCP cloud data. [ $H_2SO_4$ ], Q, T, and RH at each altitude (see Figure 3) are fixed during the three-hour simulations. It is clear from Figure 3 that an increase in GCR ionization rate associated with solar activity leads to an increase in the ultrafine production rate (i.e., dN/dQ>0) in the lower

troposphere (as indicated by the red arrows) but a decrease in the ultrafine production rate (i.e., dN/dQ<0) in the upper troposphere (as indicated by the blue arrows). In the middle troposphere, dN/dQ changes sign and the average value of dN/dQ is small compared to that of lower and upper troposphere. It is interesting to note that the optimum particle formation layer is located in the middle troposphere (3-5 km altitude, likely in cloud outflows or top of low clouds), which is consistent with the measurements obtained in recent field campaigns such as ACE-1<sup>26</sup>.



**Figure 3.** The vertical profiles of  $[H_2SO_4]$ , baseline Q, T, and RH used in the model to study the effect of a systematic change in ionization rates on particle production at different altitudes.



**Figure 4.** Simulated concentrations of total condensation nuclei larger than 3nm after three hours of simulations at different altitudes. The black line is for the baseline Q values while the green line is for Q values 20% over corresponding baseline values. The arrows indicate the changes in  $N_{d>3 nm}$  as ionization rates increase by 20%. The shaded areas are ISCCP low, middle, and high cloud regions.

#### 4. NATURAL VARIABILITY OF GLOBAL CLOUDINESS

It is well known that the abundance of cloud condensation nuclei (CCN) affects cloud properties<sup>27-</sup> <sup>29</sup>. Clouds that form in air containing high CCN concentrations tend to have high droplet concentrations, which lead to an increase in both cloud albedo and absorption. Increases in the CCN concentration also inhibit rainfall and therefore increase cloud lifetimes (cloud coverage). Since the dominating number of CCN is evolved from newly formed ultrafine particles, a systematic change in the ultrafine particle production rate will affect the population of CCN. It is physically plausible that an increase in ultrafine production rate will increase the CCN abundance and cloudiness. During a solar cycle, the values of Q vary by  $\sim 20-25\%$  in the upper troposphere and ~5-10% in the lower troposphere. Therefore, based on the influence of GCR ionization change on particle formation rate at different altitudes as shown in Figure 4, we can expect that if GCR variations have any impact on cloudiness, they should correlate positively with low cloud amount and negatively with high cloud amount. For middle clouds, such a correlation (if any) is likely to be weak. With these insights, we analyze the ISCCP–D2 cloud data sets to study if the expected different correlations between GCR fluxes and low, middle, and high cloudiness exist. ISCCP-D2 data sets are considered as the most reliable measure of global cloud cover<sup>30</sup> and are widely used for diagnostic studies of the climate system<sup>31</sup> as well as verification of climate model simulations<sup>32</sup>. We look into the infra-red (IR) cloud data because they provide spatially and temporal unrestricted measurements that include clouds over the entire globe during both day and night<sup>6,7</sup>.

Figure 5 shows the global average monthly mean anomalies of (a) high, (b) middle, and (c) low IR cloud cover during last solar cycle. To smooth out the seasonal variations, the monthly anomalies are calculated by subtracting the climatic monthly average from each month on an equal area grid before averaging over the globe<sup>6,7</sup>. The variations of GCR fluxes as measured from CLIMAX (normalized to May, 1965) are also indicated in each panel (dot-dashed lines). It is very clear that the low cloud anomalies highly co-vary with the change of GCR fluxes as has been reported by *Marsh and Svensmark*<sup>6,7</sup>. During one solar cycle, the absolute amount of low cloudiness changes by ~1.5-2%. The fluctuation of middle cloud anomalies is small compared to that of low cloud, and no obvious correlation exists between middle cloudiness and GCR variations.

For the high cloud anomalies, there is no obvious correlation for the whole solar cycle. There may have several explanations for this. First, it takes much longer time for new particles to grow to the size of CCN or ice nuclei (IN) in high altitude than in low altitude due to much lower precursor vapor concentrations. As a result, the initial difference in CN production rate may not lead to obvious difference in CCN/IN abundance as a result of coagulation, scavenging, and mixing. Second, the properties of high cloud are determined by ice nuclei abundance which may be insensitive to CN production rate. The processes controlling IN abundance in high altitude are currently not well known. Third, there may exit a negative correlation but it does not appear in the ISCCP-D2 data of last solar cycle because of the influence of other processes such as volcano eruptions and El Niño events.

We note that there were two major volcano eruptions during the period (El Chichón in April 1982 and Mt Pinatubo in June 1991). Volcano eruptions can inject large amount of  $SO_2$  into the stratosphere which leads to the formation of sulfate aerosols. On one hand, the cooling of upper troposphere as a result of volcano eruption may enhance the high cloud formation. On the other hand, the volcano aerosols descending from the stratosphere to the upper troposphere are likely to increase the frequency and lifetime of cirrus clouds<sup>33-35</sup> and hence high cloudiness. The timescale to disperse the volcanic stratospheric aerosols around the whole globe through meridional circulations is 1-2 years<sup>36-38</sup>. Therefore, the effect of volcanic eruptions on global high cloudiness may become obvious 1-2 years after the eruptions. This is consistent with the observed increase in

high cloudiness in 1984 and 1993 (i.e., 1-2 years after the El Chichón and Mt Pinatubo eruptions). A detailed analysis of Stratospheric Aerosol and Gas Experiment (SAGE) I and II aerosol extinction data for the upper troposphere<sup>39,40</sup> indicates that a substantial enhancement of aerosols down to 2-3 km below the tropopause persisted until 1986 for the El Chichón eruption (i.e., ~4 years after the eruptions). The high cloudiness in 1987 may have been affected by the El Niño event during that year<sup>41</sup>.



Figure 5. The global average monthly mean anomalies of (a) high, (b) middle, and (c) low IR cloud cover during last solar cycle. The variations of galactic cosmic ray (GCR) fluxes as measured from CLIMAX (normalized to May, 1965) are also indicated in each panel (dot-dashed lines). The shaded areas in Figure 2(a) corresponding to the years that global high cloudiness might have been affected by volcano eruptions and El Niño event.

The shaded areas in Figure 5(a) corresponding to the years that global high cloudiness might have been affected by volcano eruptions and El Niño event. From 1988 to 1993, the impact of volcano eruptions and El Niño on global high clouds is likely negligible, and it is during this period that we find a significant anti-correlation between GCR fluxes and high cloud anomalies. The increase of high cloudiness during 1988-1989 and the decrease of high cloudiness during

1991-1992 can be readily explained by the potential role of GCR in aerosol formation and CCN abundance. Furthermore, if we look at the whole period from 1984 to 1994, we do not see obvious enhancements of high cloudiness over average values due to volcano eruptions. If we consider what the volcano eruptions might have superimposed on the natural variable high cloudiness, it is clear that volcano eruptions might have enhanced the global high cloudiness by up to ~ 1.5% (without volcano effect, the high cloudiness during the solar minimum 1986-1987 is expected to be 1.5-2% less the value during solar maximum 1990-1992).

In summary, the predicted different sensitivities of the particle production to cosmic ray changes at different altitudes seem to be consistent with the observed different correlations between cosmic ray variations and low, middle and high cloud anomalies. However, due to the limit of cloud cover data available and uncertainties in the volcano and El Niño impacts, our conclusions, especially with regard to the existence of anti-correlation between high cloudiness and cosmic ray variations, are not definitive. More research is obviously needed.

#### 5. CLIMATE IMPLICATIONS

While the first key step of GCR-CN-CCN-Cloud hypothesis seems to be consistent with the observed spatially-dependent correlations of GCR variations and global cloudiness, much more work is needed to clearly establish the GCR-Cloud connection. Nevertheless, it is meaningful to discuss the climate implications associated with the possible GCR-induced cloud changes. We assume that the anomalies of high cloud cover correlate negatively while that of low cloud cover correlate positively with GCR variations, and the magnitudes of the fluctuations are similar (1.5-2% absolute change). As a result of opposite systematic variations of low and high clouds associated with solar activity, the total global cloud cover may show no obvious correlation with GCR variations. However, the radiative effects are unlikely to cancel each other.

First, the net radiative forcing of clouds depends on their altitude and optical thickness. High optically thin clouds tend to warm while optically thick high and low clouds tend to  $cool^2$ . Since cloud plays an import role in the Earth radiation budget, a systematic absolute increase of high cloud amount by ~1.5-2% and a decrease of low cloud amount by ~1.5-2% from solar minimum to solar maximum, if confirmed, may represent an important mechanism to amplify the effect of solar variability on Earth's climate.

Second, the opposite change in high and low clouds may change the atmosphere heating profile and the distribution of energy between the atmosphere and the surface, and hence may have far-reaching dynamical and climatic consequences. A systematic increase in high cloud may either warm or cool the atmosphere and the earth's surface below, depending on the types of high clouds and the underlying atmospheric properties. For example, it has been shown that the presence of a cirrostratus (with a base height of 16 km and thickness of 1.5 km) in an otherwise clear tropical atmosphere has a net cooling effect for the atmosphere below ~6 km but has a net heating effect for the atmosphere above ~ 6 km when solar zenith angles are small (< ~  $60^{\circ}$ )<sup>42</sup>. A systematic decrease in low cloud is likely to warm the surface by allowing more sunlight to reach the earth surface, while the same decrease will cool the lower troposphere by reducing the visible absorption in the cloud layer and infra-red absorption in the cloud layer and the atmosphere below<sup>42</sup>.

The long-term trend of global low and high cloud cover as a result of GCR variations may become an important external forcing of Earth climate system. Based on observations, *Lockwood et al*<sup>43</sup> have shown that from 1964 to 1996 the strength of the solar magnetic flux, shielding the Earth from GCR, has increased by ~ 41% while GCR has decreased by ~3.7%<sup>43</sup>. The ion chamber measurements<sup>44</sup> also indicate that the sea level GCR intensity has decreased by ~2% from 1979 to 1994. The GCR intensity decrease is expected to be larger at higher altitudes in the troposphere. From the data available, we estimate that the decrease in GCR fluxes during the past two decades

(1979-1999) is about 1/3 to 1/2 of the maximum variations during the last solar cycle. Thus, if the connection between low and high cloudiness exists, the global mean low cloud amount might have been decreasing (0.25-0.5%) per decade) and high cloud amount increasing (0.25-0.5%) per decade) during the past two decades. The net impact of GCR variations during the past two decades are likely to have warmed the earth surface but cooled the lower troposphere. Note that the potential GCR-induced change in cloud albedo and absorption may enhance such an impact (a decrease in cloud droplet concentration due to fewer CCN as a result of reduced GCR fluxes may also imply a low cloud albedo and absorption). While the exact amount of net radiative forcing associated with GCR-induced low and high cloud changes remains to be investigated, it is physically plausible that the decrease in GCR fluxes during the past two decades has led to a net warming of  $\sim 0.05$  °C per decade at the surface while a net cooling of  $\sim 0.05$  °C per decade in the lowest 8 km of atmosphere. In other words, the GCR-induced low and high cloud changes may explain why the Earth's surface has warmed much more rapidly (~0.1 °C per decade) than the lowest 8 km of atmosphere during the last two decades. A piece of suggestive evidence to support such a claim is that the regions of large differences in surface and low atmospheric temperature trends remarkably coincide with the regions of high correlation between cosmic ray and low cloud top temperature as shown in Figures 6 and 7.

The spatial correlation map in Figure 6 shows how ISCCP-D2 low cloud top temperatures co-vary with GCR flux. The correlation coefficients, r, are calculated from the 12-month running mean at each grid point (following Marsh and Svensmark <sup>6.7</sup>). Figure 6 reveals a band of significantly high correlation centered around the tropics where stratocumulus and marine stratus clouds are dominant<sup>6.7</sup>. The fraction of Earth with r > 0.6 is ~ 30% which is significant at 99.9%.

Figure 7 shows the global distribution of temperature trends in the lowest five miles of the atmosphere derived from MSU t2lt data for the period 1979-2000. If we compare MSU t2lt trends with similar temperature trends derived from surface observations (not shown), we see warming over the northern third of the globe both at the surface and in the five-mile-deep layer of air above. The largest differences in the trends of temperature between surface and low atmosphere measurements are over the tropical regions where the surface data show a significant warming while MSU t2lt data show a slightly cooling. The tropical radiosonde temperature data show the same patterns of surface warming and tropospheric cooling since 1979 as the independent surface and low atmospheric temperature trends remarkably coincide with the regions of high correlation between cosmic ray and low cloud top temperature (Figure 6). Such a nice coincidence may suggest that the differences in surface and low atmospheric temperature trends over tropics are associated with the solar indirect forcing via GCR-Cloud link.

It may become necessary to include the solar indirect forcing via GCR-induced cloud change in the future climate model, as current models still cannot account fully for the apparent difference between observed surface and troposphere temperature trends since 1979<sup>13-15</sup>. Unlike homogeneous greenhouse gases which warm both the surface and low troposphere, the potential influence of GCR variations on clouds are different at different regions of the atmosphere and the associated radiative forcing are spatial and temporal inhomogeneous. The observed rapid warming during the past two decades over the northern third of the globe both at the surface and in the air above is likely due to the greenhouse effect.



Figure 6. Global correlation map of GCR with anomalies of IR low cloud top temperature. White pixels indicate regions with either no data or an incomplete monthly time series.



Figure 7. Low-to-middle atmosphere decadal linear temperature trend ( °C/decade) for period 1979-2000 derived from MSU t2lt data.

#### 6. SUMMARY AND DISCUSSION

The dependence of ultrafine production rate on galactic cosmic ray ionization rate at different altitudes has been investigated. Our primary studies indicate that an increase in GCR ionization rate leads to an increase in CN production in the lower troposphere (>680 mb), but a decrease in CN production in the upper troposphere (<440 mb). In the lower troposphere, the ionization rate is low and the  $H_2SO_4$  concentration is relatively high, the particle formation is limited by ionization and an increase in ionization rate leads to an increase in nucleation. In the upper troposphere, the ionization rate is very high and the  $H_2SO_4$  concentration is relatively low, the particle formation is limited by  $H_2SO_4$  concentration and an increase in ionization rate inhibit the nucleation by reducing the lifetime of ion clusters. The average change of CN production as the ionization rate increases is small in the middle troposphere (440-680 mb).

Since an increase in ultrafine production rate is likely to increase the CCN abundance and cloudiness, we can expect that the correlation between GCR changes and global cloud cover (if any) should be positive for low cloud, negative for high cloud, and weak for the middle cloud. In addition to the reported positive correlation between GCR variations and low cloudiness, our analyses of ISCCP D2 IR cloud data further reveal that high cloudiness may be anti-correlated with GCR variations if volcano and El Niño impacts are excluded. During a solar cycle, the absolute change of high and low cloud amounts is opposite in sign but similar in magnitude (~1.5-2%). The fluctuations of middle cloud anomalies are small compared to that of low clouds, and no obvious correlation between GCR variations and low, middle and high cloud anomalies seem to be consistent with the predicted dependence of CN production on GCR variations at different altitudes. Such a consistency suggests that solar activity might affect global cloudiness by modulating GCR fluxes. However, due to the limit of cloud cover data available and uncertainties in the volcano and El Niño impacts, our conclusions, especially with regard to the existence of anti-correlation between high cloudiness and cosmic ray variations, are not definitive.

The climate implications associated with the possible GCR-induced cloud changes are discussed. Since cloud is critical to Earth radiation budget, opposite systematic variations of low and high clouds associated with solar activity, if confirmed, may represent an important mechanism to amplify the effect of solar variability on Earth's climate. The decrease in GCR intensity during the last two decades might have led to a decrease in global mean low cloud amount and an increase in high cloud amount, which might have warmed the Earth's surface and cooled the low troposphere. The potential GCR-induced change in cloud albedo and absorption may enhance such an impact (a decrease in cloud droplet concentration due to fewer CCN as a result of reduced GCR fluxes may also imply a low cloud albedo and absorption). We suggest that, the GCR-induced natural variability of global cloudiness, together with the greenhouse gases which warm both the surface and low troposphere, may reconcile the apparent differences in global mean temperature trends at Earth's surface (rapidly warming, as recorded by thermometers) and in the lowest 8 km of atmosphere (little if any warming, as monitored by satellites and balloons).

While this study provides additional evidence for the inferred correlation between variations in global cloud properties and the solar-modulated GCR fluxes, much more work is needed to understand how and how much the GCR variations will affect the global cloud properties. The first key process (i.e, influence of GCR variations on nucleation and CN abundance) in our proposed GCR-CN-CCN-Cloud hypothesis seems to be consistent the spatially dependent influence of GCR variations on cloud properties. However, we currently do not know how much the natural GCR variations will affect the CCN abundance and cloud properties. Laboratory and field measurements, as well as theoretical studies are needed to validate the predicted dependentbehaviors of nucleation on ionization rates at different altitudes, to investigate the effect of GCR variations. The current analyses of GCR-cloud correlations are limited by the uncertainties associated with the cloud data and short periods of cloud data available. Improved cloud cover data covering longer time periods will be very useful in studying GCR-cloud connections.

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