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**Cosmic rays induced
formation of NAT
particles**

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Formation of large NAT particles and denitrification in polar stratosphere: possible role of cosmic rays and effect of solar activity

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Abstract

The formation of large nitric acid trihydrate (NAT) particles has important implications for denitrification and ozone depletion. Existing theories can't explain the recent observations of large NAT particles over wide Arctic regions at temperature above ice frost point. Our analyses reveal that high-energy cosmic rays may induce the freezing of supercooled $\text{HNO}_3\text{-H}_2\text{O-H}_2\text{SO}_4$ droplets when they penetrate these thermodynamically unstable droplets. The cosmic ray-induced freezing (CRIF) is consistent with the observed highly selective formation of NAT particles. We suggest that the physics behind the CRIF mechanism is the reorientation of polar solution molecules into the crystalline configuration in the strong electrical fields of moving secondary ions generated by passing cosmic rays. Our simulations indicate that strong solar proton events (SPEs) may significantly enhance the formation of large NAT particles and denitrification. The CRIF mechanism can explain the high correlations between the thin nitrate-rich layers in polar ice cores and major SPEs. The observed enhancement in aerosol backscattering ratio at PSC layers shortly after an SPE and the significant precipitation velocity of the enhanced PSC layers also provide strong support for the CRIF mechanism.

1. Introduction

Polar stratospheric clouds (PSCs) are crucial to the ozone depletion because of their well-recognized role both in activating chlorine (through heterogeneous reactions on the surface of PSC particles) and denitrifying the lower stratosphere (through gravitational settling of large HNO_3 -containing PSC particles) (e.g. Solomon, 1999; Word Meteorology Organization, 1999). During the recent SAGE III Ozone Loss and Validation Experiment (SOLVE), large nitric acid trihydrate (NAT) particles ("NAT rocks") at low number densities has been observed in a synoptic scale at temperature above frost point (Fahey et al., 2001). As a result of their significant fall speeds, these large

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NAT particles can lead to an irreversible removal of nitric acid which may prolong the lifetime of reactive chlorine and thereby enhance the ozone depletion. Climate models predict that the increased concentrations of greenhouse gases and depletion of the ozone layer may lead to lower stratospheric temperatures and more widespread PSC formation and denitrification in the Northern Hemisphere (Shindell et al., 1998; Waibel et al., 1999). Modeling simulations show that widespread denitrification could enhance future Arctic ozone loss by up to 30% (Tabazadel et al., 2000). To evaluate properly the consequences of future lower stratospheric temperatures on the arctic ozone layer, details about the formation of large NAT particles must be known.

The large NAT particles observed in a synoptic scale at temperature above frost point (Fahey et al., 2001) are believed to be formed through the freezing of supercooled ternary solutions (STS) composed of H_2SO_4 , HNO_3 and H_2O . Since the observations indicate that only $\sim 0.002\%$ – 0.01% of these STS droplets froze, the freezing mechanism must be highly selective (Fahey et al., 2001; Tolbert and Toon, 2001). Currently we have no satisfying theories to explain this “mysterious” highly selective freezing process observed during SOLVE (e.g. Fahey et al., 2001; Tolbert and Toon, 2001; Carslaw et al., 2002a; Jensen et al., 2002; Drdla et al., 2003). Laboratory studies indicate that homogeneous freezing of STS particles are negligible at the temperature above ice frost point under polar stratospheric conditions (Koop et al., 1995; Knopf et al., 2002). Homogeneous freezing on particle surface may enhance the nucleation rate by a factor of ~ 100 (Tabazadeh et al., 2002), however this enhancement is still too small to account for observed production rate of large NAT particles. Since the large NAT particles were observed within a synoptic scale pool of air, the role of mountain waves in generating these particles (Carslaw et al., 1998) can also be excluded (Carslaw et al., 2002a). While laboratory experiments by Bogdan and Kulmala (1999) and Bogdan et al. (2003) suggested that silicas could trigger heterogeneous nucleation of nitric hydrates at temperature above the ice frost point, these experiments were carried out for binary HNO_3 - H_2O solutions which are different from the ternary H_2SO_4 - HNO_3 - H_2O solutions in the polar stratosphere. It is known to be much more difficult to initiate freezing

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in ternary $\text{H}_2\text{SO}_4\text{-HNO}_3\text{-H}_2\text{O}$ solutions than in binary $\text{HNO}_3\text{-H}_2\text{O}$ solutions. Biermann et al. (1996) tested the efficiency of meteoritic dust grains and a variety of other solid materials as nuclei in ternary $\text{H}_2\text{SO}_4\text{-HNO}_3\text{-H}_2\text{O}$ solutions at temperatures as low as 188 K and detected negligible effects.

5 In this study, we explore the freezing of the supercooled ternary solutions (STS) as a result of the interactions between the thermodynamically unstable STS droplets and high-energy cosmic rays. The possible physics behind the cosmic ray-induced freezing (CRIF) is described. The CRIF mechanism is formulated and used in a 1-D polar stratospheric cloud (PSC) model to simulate the formation of PSC particles.
10 The effect of solar activity on PSC properties and denitrification, and the observations supporting the CRIF mechanism are discussed.

2. Cosmic ray-induced freezing (CRIF)

Cosmic rays (CRs) entering Earth's atmosphere are known to be able to initiate phase changes in supercooled vapor (e.g. Wilson, 1927) and superheated liquid (e.g. Glaser, 1960). Role of cosmic ray ionization in the formation of ultrafine particles in the atmospheric has received increasing interest in recent years (e.g. Yu and Turco, 2000; Yu, 2002; Carslaw et al., 2002b; Harrison and Carslaw, 2003; Lee et al., 2003). The possible enhancement in the freezing of supercooled water as a result of electrical interactions (i.e. electrofreezing) has been suggested in various studies (see textbook by Pruppacher and Klett, 1997, and review articles by Harrison and Carslaw, 2003 and Tinsley and Yu, 2003). The electrofreezing of supercooled water could be induced by
15 (1) the direct passage of energetic particles, (2) strong electric fields, and (3) contact with ions.

Electrofreezing processes may also play a role in the formation of NAT particles in the polar stratosphere. It has been suggested that the freezing of STS particles in the polar stratosphere may be initiated by the contact of the supercooled droplets with small ions or charged $\text{HNO}_3/\text{H}_2\text{O}$ clusters (Hamil and Turco, 2000; Carslaw, 2001; D'Auria
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and Turco, 2001). In this study we propose that the freezing may be initiated by the passage of high-energy cosmic rays directly through the thermodynamically unstable STS droplets. All the molecules in the STS droplets (HNO_3 , H_2O , H_2SO_4) have high dipole moments. For freezing, the dipoles need to be oriented and arranged into the crystalline configuration. Our proposed mechanism of CR-induced freezing (CRIF) involves the reorientation of polar solution molecules into crystalline configuration in the strong electrical fields of moving secondary ions generated by passing CRs.

When a primary CR particle (proton) hits a solution atom or molecule, secondary electrons are ejected in various directions with a spectrum of energies and strong electric fields exist between the positive parent ion and ejected electrons. The strength of electric fields (E) between the positive ion and the ejected electrons separated by l nm can be approximated by

$$E(x) = \frac{e}{4\pi\epsilon_0\epsilon_r} \left[\frac{q_1}{x^2} + \frac{q_2}{(l-x)^2} \right], 0 < x < l \quad (1)$$

where x is the position from positive ion in the line connecting two ions. ϵ_0 is the vacuum permittivity, ϵ_r is the relative permittivity of liquid solution. q_1 is the number of elementary charges (e) carried by the parent positive ion. $q_1=q_2=1$ for single ionization. For multiple ionization, $q_1>1$ and $q_2=1$ if the contribution of other electrons is ignored. q_2 could be >1 when electrons are ejected in same directions. Figure 1 shows the values of $E(x)$ corresponding to $q_1=1, 4, 8$ ($q_2=1$) and (a) $l=2$ nm, (b) $l=3$ nm. As an approximation, the relative permittivity for water ($\epsilon_r=80$) is used since the value of ϵ_r for STS is not available.

The electric fields between positive ion and electrons are sensitive to l and q_1 (number of electrons ejected upon the collision of proton with atoms). In order to initiate freezing through the reorientation of solution molecules in the electrical fields of CR-generated positive ion and electrons, at least two conditions have to be met.

First, the maximum separation between the parent ion and the ejected electrons has to be larger than the critical size of crystalline embryo. The ejected energetic electrons will make inelastic collision with liquid molecules and transfer its kinetic energy to

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these molecules. The electrons will be neutralized by positive ion or absorbed by liquid molecules after certain time. How far an electron can travel before being neutralized or absorbed depends on the initial velocity of the electron and hence the velocity or energy of incoming CR particles. The size of critical embryo depends on the degree of supercooling and solution properties.

Second, the minimum strength of electric field between the positive ion and electrons separated by a distance of critical embryo size should reach above a certain threshold value. Laboratory studies indicate that the electric fields have to be very large to cause the freezing of supercooled water drop (e.g. Abbas and Latham, 1969; Braslavsky and Lipson, 1998). Analysis of both positive and negative experimental results relevant to the question of whether nucleation of ice is enhanced by the presence of electric field indicate that electric fields on the order of 10^8 V/m might be essential in orienting polar molecules in a microscopic volume to initiate freezing (Tinsley and Deen, 1991). For $l=2-3$ nm (typical sizes of critical crystalline embryo), Fig. 1 indicates that multiple ionization is needed to reach electric fields of 10^8 V/m. It is noteworthy that simultaneous electric fields and laminar flow have been found to be important in orienting crystalline embryos to initiate freezing (Pruppacher, 1963; Abbas and Latham, 1969; Doolittle and Vali, 1975; Tinsley and Deen, 1991; Pruppacher and Klett, 1997). Since the electrons are moving away from the positive parent ion at a certain speed, the polar molecules between the positive ion and the electron(s) experience simultaneous electric fields and laminar flow (relative to ions).

There is not much experimental study on the possible role of cosmic ray penetration in inducing the freezing of supercooled droplets. Varshneya (1969) observed tracks of ice crystal when CRs passed through the supercooled triple distilled, suspension free, pure water. Later, he developed a classical ion induced freezing theory which considers the contribution of electrostatic energy to the ice nucleation activation energy when the ice-like clusters are formed around ions (Varshneya, 1971). Tinsley and Deen (1991) showed clear correlations of winter cyclone intensity with day-to-day changes in the cosmic ray flux and suggested ice nucleation by passage of energetic CR particles as

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one of possible mechanisms. Detwiler and Vonnegut (1980) and Seeley et al. (2001) tested the hypothesis about the ionization-induced freezing by irradiating supercooled water droplets with 5-Mev alpha particles and found that alpha particles ionization has negligible effect on water droplet freezing. Based on Varshneya's theory (1971), all ions generated by different sources should have similar ability in initiating freezing. In this regard, Detwiler (1993) questioned the applicability of both Varshneya's experimental results and theory to ice nucleation in droplets in upper tropospheric cloud. Nevertheless, Seeley et al. (2001) themselves pointed out that their measurement does not exclude the possibility that another type of or energy range of cosmic rays induces ice nucleation.

In contrast to the ion-induced freezing as a result of the contribution of electrostatic energy to ice nucleation activation energy (as proposed by Varshneya, 1971), the physics behind our proposed CRIF is the reorientation of solution molecules into crystalline configuration in the strong electrical fields of moving secondary ions generated by passing cosmic rays. In order to initiate freezing, the energy of cosmic rays has to be above certain level in order to cause certain multiple ionizations and generate electrons that meet the two conditions listed above (i.e. electrons energetic enough to travel beyond the critical size and the electric fields between positive ion and electrons above certain threshold value). For a critical crystalline embryo size of around 2–3 nm, Fig. 1 indicates that q_1 may have to be around 8 (a complete ionization of oxygen atom) to have electric field above 10^8 V/m. It is possible that the energy of alpha particles Detwiler and Vonnegut (1980) and Seeley et al. (2001) used in their study is not high enough to initiate freezing under the conditions they investigated. Most of CRs entering polar stratosphere have energy larger than 100 Mev with significant fraction above 500 Mev. It is conceivable that the direct passage of these high-energy cosmic rays through the highly supercooled liquid may initiate freezing. Laboratory measurement using cosmic ray particles in the energy range common in the atmosphere is needed to test the CRIF theory proposed above. While such measurement is currently unavailable, we demonstrate below that there exists strong observational evidence supporting

the CRIF theory.

The CRIF production rate of solid NAT particles ($J_{\text{NAT}}^{\text{CRIF}}$, in $\text{cm}^{-3} \text{sec}^{-1}$) from STS particles with number density n (cm^{-3}) and radius r can be estimated as,

$$J_{\text{NAT}}^{\text{CRIF}} = n\pi r^2 F_{\text{CR}} P \quad (2)$$

5 where F_{CR} (in $\text{cm}^{-2} \text{sec}^{-1}$) is the omni-directional flux of high energy CRs (protons). P is the probability of freezing when a CR particle hits a supercooled STS droplet. P can be expressed as,

$$P = P_1 * P_2 \quad (3)$$

10 where P_1 represents the average chance that a CR particle going through the droplet has a collision with a solution atom, and P_2 represents the probability that freezing is initiated when the CR particle collides and ionizes the atom. Following the approach of Loeb (1939) in deriving the collision probability of an ion entering a sphere with an air molecule, we derive P_1 as,

$$P_1 = 1 - \frac{L^2}{2r^2} \left[1 - \left(\frac{2r}{L} + 1 \right) e^{-\frac{2r}{L}} \right] \quad (4)$$

15 where L is the mean free path of CR particles (protons) in liquid solution and r is the radius of droplet. L can be estimated as,

$$L = \frac{1}{\sigma N} \quad (5)$$

20 where σ is the proton ionization cross section and N is the number of molecules per unit volume of liquid. σ is a function of incident proton energy. We don't have information about the ionization cross section for protons in ternary solution but we can use that of protons in liquid water as an approximation. Table 1 shows the values of σ and L for protons of different energies in liquid water and the corresponding values of P_1 with

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droplet radius of $r=0.1\ \mu\text{m}$, $0.2\ \mu\text{m}$, and $0.3\ \mu\text{m}$. For STS droplets of typical sizes in polar stratosphere ($\sim 0.2\ \mu\text{m}$), P_1 is likely to be above ~ 0.5 .

P_2 depends on the detailed interaction of CRs (protons) with solution molecules/atoms (for example the degree of ionization) and the critical size of crystalline embryo. Thus P_2 is likely to be a function of many parameters including the energy of the incoming CRs, temperature, NAT supersaturation ratios (S_{NAT}), composition and size of STS particles, and interfacial tension between the liquid and solid phases. Just like the values of the activation energy used in the classical homogeneous freezing theory (Tabazadeh et al., 2000; Knopf et al., 2002), the values of P_2 may also have to be decided through laboratory studies and/or observations. Below we estimate the range of $P(=P_1 \cdot P_2)$ by comparing the calculated values of $J_{\text{NAT}}^{\text{CRIF}}$ with the observed NAT formation rates.

Figure 2 shows the dependence of $J_{\text{NAT}}^{\text{CRIF}}$ on the radius of STS particles at $P=0.01$, 0.1 , 1 . Typical CR flux in the polar stratosphere of $2.5\ \text{cm}^{-2}\ \text{s}^{-1}$ and STS particle number concentration (n) of $8\ \text{cm}^{-3}$ are assumed. The observed NAT production rates shown in the figure are inferred from large mode (number density $\sim 2.3 \times 10^{-4}\ \text{cm}^{-3}$, mean diameter $\sim 14.5\ \mu\text{m}$) and small mode (number density $\sim 2 \times 10^{-3}\ \text{cm}^{-3}$, mean diameter $\sim 3.5\ \mu\text{m}$) NAT particles observed by Fahey et al. (2001), assuming that these particles were accumulated in a $3(\pm 2)$ -day period. The background sulfate aerosols generally have mean radius of $\sim 0.07\ \mu\text{m}$ but can grow to a mean radius of $\sim 0.25\ \mu\text{m}$ by taking up HNO_3 when $S_{\text{NAT}} > 1$. Since the maximum radius of STS particles in typical stratospheric conditions is around $0.25\ \mu\text{m}$, we put the observed NAT production rates at the position of x-axis corresponding to a radius of $0.25\ \mu\text{m}$. In the real situations, freezing could happen on all sizes of STS particles as they grow from $\sim 0.07\ \mu\text{m}$ to $0.25\ \mu\text{m}$ by taking up HNO_3 . Figure 2 indicates clearly that the CRIF can explain the formation rate of the observed NAT particles. Most importantly, CRIF rate has an upper limit of around $10^{-4}\ \text{cm}^{-3}\ \text{h}^{-1}$ ($P=1$) which is controlled by the chance that a CR particle hits a STS droplet. Thus CRIF is highly selective which meets the critique of nucleation mechanism inferred from observations (Fahey et al., 2001; Tolbert and

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Toon, 2001). Figure 2 indicates that the freezing probability is around 0.1 with a range of 0.01–1 under the conditions where the observations (Fahey et al., 2001) were made. The less than unity of the freezing probability can be easily understood as some CR particles may penetrate the STS droplets without inducing freezing because of various reasons including (but not limited to): (1) low energy of the CRs, (2) low supersaturation of droplets, and (3) short CR path in the droplet (especially when CR penetrates the edge of the droplets).

3. Simulations

To study the effect of CRIF on PSC formation and denitrification, we use a one-dimensional (1-D) PSC model from Danish Meteorological Institute (DMI) (Larsen, 2000; Larsen et al., 2002). The DMI PSC model calculates the time dependent PSC particle size distributions and chemical compositions together with changes in gas phase mixing ratios of water vapor (H_2O) and nitric acid vapor (HNO_3). The formation (nucleation), growth, evaporation, and sedimentation of various types of PSC particles are simulated in the model with up-to-date thermodynamic data.

The initial profile and sinusoidal oscillation of temperature and the initial nitric acid mixing ratio profile follow those of Jensen et al. (2002). The temperature oscillation is an idealized representation of the temperature variations experienced by air mass circulating around the pole during the winter of 2000 where SOLVE measurements were made. The initial altitude (h) dependent temperature ($T_0(h)$) has a minimum of 197 K at $h=20$ km, and increases linearly to 202 K at 30 km and to 217 K at 10 km. The temperatures at all altitude follow a sinusoidal evolution as $T(h)=T_0(h) + 4.5 \times \{\sin[2\pi(1/4 + t/\tau)]-1\}$ where τ is the oscillation period. The temperature profile and oscillation amplitude were based on the early to mid-January Arctic conditions (Newmann and Harries, 2002). We follow Jensen et al. (2002), using $\tau=6$ days as the baseline case and include $\tau=12$ days in the sensitivity study. The HNO_3 gas mixing ratio has a maximum value of 9 ppbv at 20 km, with mixing ratio decreasing

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below this altitude down to about 2 ppbv in the lowermost stratosphere, which is typical of the polar stratosphere in early winter (Gille et al., 1996). A water mixing ratio of 5 ppmv is assumed. The initial background sulfate aerosol is from Deshler et al. (2003). At the 20 km, the total number concentration, median radius, standard deviation for mode 1 and mode 2 are 7.67 cm^{-3} , $0.069 \mu\text{m}$, 1.63 and 0.006 cm^{-3} , $0.42 \mu\text{m}$, 1.11 , respectively. Same mixing ratio of sulfate is assumed at other altitudes. The vertical space resolution is 200 m. The cosmic flux (F_{CR}) profile corresponding to the 2000 winter polar stratosphere is interpolated based on the balloon measurements of cosmic ray fluxes in February 1987 (maximum fluxes) and November 1989 (minimum fluxes) in polar latitudes (Bazilevskaya and Svirzhevskaya, 1998) and cosmic ray intensities measured with Climax Neutron Monitor.

As discussed earlier, P depends on S_{NAT} and other parameters and the dependence relation remains to be investigated. As a first approximation, we assume $P=0.1$ and that the freezing becomes possible only when S_{NAT} is larger than 10 (Jensen et al., 2002) in this study. While the details of simulated PSC properties will vary when different values of P are used, the main features of the results and conclusion will not change.

Figure 3 presents the model results plotted versus time and altitude: (a) temperature, (b) S_{NAT} , (c) HNO_3 gas mixing ratio, (d) CR-induced freezing rate (J_{NAT}), (e) total number concentration of NAT particles, (f) median diameter of NAT particles. The temperature reaches the lowest value at $t=3$ days and S_{NAT} can reach up to 30. The classical homogenous freezing theory predicts negligible ($<10^{-8} \text{ cm}^{-3} \text{ h}^{-1}$) nucleation under the condition if the activation energy at the similar S_{NAT} derived from recent laboratory data (Knopf et al., 2002) is used. With $P=0.1$, the CR induced freezing rate can reach up to $3.5 \times 10^{-5} \text{ cm}^{-3} \text{ h}^{-1}$ (Fig. 3d). The concentrations of NAT particles reach above 10^{-4} cm^{-3} between 17 km and 21 km with a maximum concentration of $\sim 10^{-3} \text{ cm}^{-3}$ at 20 km (Fig. 3e). The median diameters of NAT particles are in the range of 8–13 μm in the area where significant NAT particles are present (concentration $>10^{-4} \text{ cm}^{-3}$) (Fig. 3f). The simulated size and concentration of NAT particles are qualitatively con-

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sistent with the measurements of Fahey et al. (2001), with the understanding that the temperature history and vertical structures of the air mass where the observations were made are more complex than those assumed in our simulation. There are some denitrification at altitudes between 19.5 and 22 km and some renitrification between 16 km and 19.5 km (Fig. 3c). The depth of NAT saturated layer limits the sedimentation of NAT particles as NAT particles sublime when $S_{\text{NAT}} < 1$. A longer PSC duration time will increase the sizes of NAT particles and enhance the denitrification (Jensen et al., 2002, also see next section).

The main properties of PSC shown in Fig. 3 are similar to that presented in Jensen et al. (2002). This is not surprising since the initial profile and sinusoidal oscillation of temperature and the initial nitric acid mixing ratio profile used in this study are same as those of Jensen et al. (2002). The major difference between our study and Jensen et al's study is that we use equation (2) to calculate the cosmic ray-induced freezing while Jensen et al used a constant freezing rate to represent an unidentified freezing mechanism. With CRIF mechanism included in the model, we can investigate how solar activity may affect PSC properties and denitrification.

4. Effect of solar activity on PSC properties and denitrification

It is known that solar activities affect F_{CR} in polar stratosphere significantly. F_{CR} at altitudes between 15–25 km in the polar stratosphere almost doubles from solar maximum to solar minimum during regular 11-year solar cycle, and strong solar proton events (SPEs) can increase the CR fluxes in polar stratosphere by a factor of ~ 10 –30. Figure 4 shows the balloon-based measurements of F_{CR} as a function of altitude in northern polar regions (Murmansk, cut-off rigidity=600 MeV) under different conditions of solar activities. The data for F_{CR} during solar minimum (February 1987, F_{CR} maximum), during solar maximum (November 1989, F_{CR} minimum), and during the strongest solar proton event in October 1989 are from Bazilevskaya and Svirzhevskaya (1998). The F_{CR} profile corresponding to the 2000 winter polar stratosphere (used for calculations

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presented in Fig. 3) is interpolated based on F_{CR} maximum and minimum profiles and the CR intensities measured with Climax Neutron Monitor. It is noteworthy that CRs have the maximum flux in the polar stratosphere where PSCs form.

CRIF is very selective (less than 0.1% of particles with radius of $0.2 \mu\text{m}$ will be hit by CRs during a 3-day period) and the freezing rate is proportional to F_{CR} (Eq. 2). Thus, we should expect to see the enhanced production rate of large NAT particles and hence denitrification when CR fluxes increase. Using representative F_{CR} profiles during four different solar activities (maximum, minimum, and average solar activities, and during strong SPEs; Fig. 4) and with temperature oscillation periods (τ) of 6 days and 12 days, we compare the corresponding HNO_3 mixing ratio profiles at $t = \tau$ (Fig. 5). Significant denitrification and renitrification at different altitudes are obvious and the extended life of PSC significantly enhances the denitrification. From the solar maximum (F_{CR} minimum) to solar minimum (F_{CR} maximum), the denitrification at altitudes between 20–22 km increases by up to 0.6 ppbv (7%) for $\tau=6$ days (3.5-day cloud lifetime) and up to 1.3 ppbv (15%) for $\tau=12$ days (7-day cloud lifetime).

The strong SPEs significantly enhance the denitrification, especially in the top layer of PSCs. The renitrification layer during SPEs also shifts to lower altitudes. Thus, compared to non-SPE periods, SPEs lead to extra denitrification in the cloud top layer of around 2 km depth and move the renitrification layer by 1 km. For the temperature assumed in this study, NAT particles are not able to reach the surface due to sublimation. However, significant amount of nitric acid has been moved downward by up to ~ 5 km for $\tau=6$ days and by up to ~ 8 km for $\tau=12$ days. This downward-moved nitric acid can precipitate to the ground in subsequent cloud and precipitation processes. As a result, the enhanced denitrification during the SPEs might have a signal in the nitric acid deposited in polar ice cores.

Actually, the thin nitrate-rich layers in both Arctic and Antarctic firn and ice cores have been found to be highly positively correlated with periods of major solar proton events (the probability of chance correlation less than 10^{-9}) (McCracken et al., 2001). Most importantly, the impulsive events in the nitrate record are all short lived (many

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have a time constant of 2 weeks, none has one greater than 6 weeks) (McCracken et al., 2001). Solar proton events are known to increase NO_x concentration in the mesosphere and upper stratosphere (e.g. Jackman et al., 2001). However, it has been estimated (Dunkerton, 1978) that the downward transport of HNO_3 in the gaseous phase from mesosphere and upper stratosphere to the troposphere would take at least 3–6 months. McCracken et al. (2001) concluded that the timescale of the nitrate events is too short to be understood in terms of transport mechanisms in the gaseous phase, and suggested that PSC formation processes and the nitrate precipitation might be enhanced during and after major SPEs (which is consistent with CRIF mechanism). Thus, the high correlations between the nitrate-rich layers in polar ice cores and major solar proton events (McCracken et al., 2001) may support the CRIF mechanism.

Shumilov et al. (1996) reported a considerable increase in aerosol backscatter ratio after a strong SPE. Analysis of meteorological data indicates that the increase was not associated with temperature change and seems to be caused by the SPE (Shumilov et al., 1996). The mechanism Shumilov et al. (1996) offered to explain the aerosol backscatter ratio enhancement is the increase in aerosol concentration as a result of SPE-induced enhancement in the ion nucleation rate of ultrafine particles. This explanation is questionable because the concentration of sulfuric acid gas in winter polar stratosphere is extremely low ($< \sim 10^5 \text{ cm}^{-3}$) and the increase in ionization rate may not lead to increase in the formation of fresh ultrafine particles (Yu, 2002). It also takes a long time to grow the fresh nucleated sulfuric acid particles to a size that can be detected by lidar (radius $> 0.69 \mu\text{m}$). Furthermore, the profiles of aerosol backscattering ratio indicates that enhanced particle layer formed precipitated quickly (1–2 km/day). Only PSC particles of around $10 \mu\text{m}$ can have this precipitation velocity. To form PSC particles of around $10 \mu\text{m}$, the freezing process must be very selective (Fahey et al., 2001). The considerable increase in aerosol backscatter ratio after a strong SPE and the fast precipitation velocity of the enhanced particle layer are actually consistent with the CRIF mechanism.

Obviously, the CRIF process provides a direct and convincing explanation for the

high correlation between the thin nitrate-rich layers in polar ice cores and major SPEs (McCracken et al., 2001) and the observed enhancement in aerosol backscattering ratio at PSC layers shortly after the SPE (Shumilov et al., 1996). This in turn provides strong indirect support for the CRIF mechanism.

5. Conclusions

The existing theories on nitric acid trihydrate (NAT) particle formation can't explain the recent observations of large NAT particles over wide regions in Arctic vortex. We propose that high energy cosmic ray particles may induce the freezing of the supercooled $\text{HNO}_3\text{-H}_2\text{O-H}_2\text{SO}_4$ droplets when they hit and penetrate these droplets in polar stratosphere. The mechanism of CR-induced freezing (CRIF) we suggest involves the reorientation of polar solution molecules into crystalline configuration in the strong electrical fields of moving secondary ions generated by passing CRs, which is different from the ion-induced freezing theory considering the contribution of electrostatic energy to ice nucleation activation energy. In order to initiate freezing, the energy of cosmic rays has to be above certain level in order to induce certain multiple ionizations and generate electrons that are energetic enough to travel beyond the critical size and the electric fields between positive ion and electrons above certain threshold value. The CRIF mechanism is highly selective which can naturally explain the formation of a few large nitric acid trihydrate (NAT) particles ("NAT rocks") observed during SOLVE (Fahey et al., 2001).

The CRIF mechanism is formulated and included in a 1-D polar stratospheric cloud (PSC) model to simulate the formation and properties of type 1 PSCs and the associated denitrification. Under the temperature profile and oscillation amplitude of the early to mid-January Arctic conditions during SOLVE, our simulated size and concentration of NAT particles are qualitatively consistent with the measurements of Fahey et al. (2001) while the classical homogenous freezing theory predicts negligible nucleation. Our simulations indicate that strong solar proton events (SPEs) may significantly

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enhance the formation of large NAT particles and denitrification. The CRIF process provides a convincing explanation for the observed enhancement in aerosol backscattering ratio at PSC layers shortly after the SPE and the high correlations between the thin nitrate-rich layers in polar ice cores and major SPEs. This in turn provides strong indirect support for the CRIF mechanism.

Our proposed CRIF mechanism, which can naturally explain the highly selective formation of NAT particles in the polar stratosphere, may represent an important improvement in our understanding of NAT particle formation and properties. Such an understanding is crucial to evaluate the consequences of future lower stratospheric temperatures on the arctic ozone depletion. The CRIF process may shed new light on the effect of solar activities on the NAT particle formation, denitrification, and ozone depletion, and may provide a theoretical foundation to use nitrate events in polar ice cores to infer past SPEs and solar activities. The CRIF could also contribute to the highly selective freezing of supercooled water droplets in upper troposphere which may have important climatic implications. Laboratory study using cosmic ray particles in the energy range common in the atmosphere is needed to test the CRIF theory. Observations of PSC particle changes during and after strong SPEs may provide useful information on CRIF. Theoretical investigation of how protons interaction with solution molecules/atoms and how polar molecules in solution react to the strong electric fields are needed to advance the CRIF theory.

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Table 1. Total ionization cross section σ (from Dingfelder et al., 2000), mean free path L , probability of ionization for proton of different energies going through liquid water droplet of different radius r .

Incident proton energy	σ (m ²)	L (μ m)	P_1 ($r=0.1 \mu$ m)	P_1 ($r=0.2 \mu$ m)	P_1 ($r=0.3 \mu$ m)
5 Mev	3.4E-21	0.009	1.0	1.0	1.0
50 Mev	5.0E-22	0.061	0.85	0.96	0.98
200 Mev	1.8E-22	0.168	0.53	0.76	0.86
500 Mev	1.1E-22	0.275	0.37	0.60	0.73
1 Gev	8.8E-23	0.344	0.31	0.52	0.66

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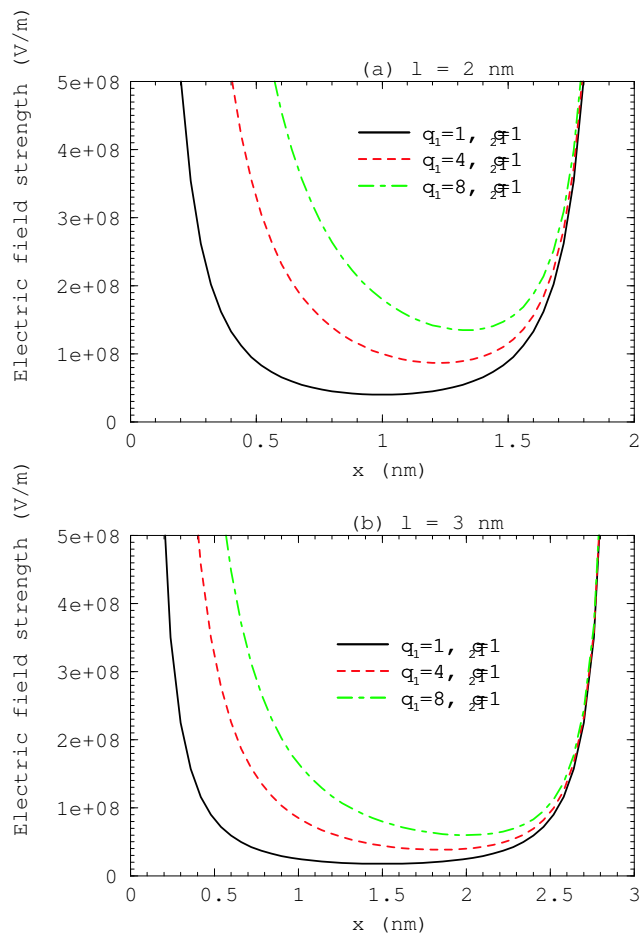


Fig. 1. The strength of electric fields at different positions in the line connecting positive parent ion and electrons. **(a)** $l=2$ nm, **(b)** $l=3$ nm.

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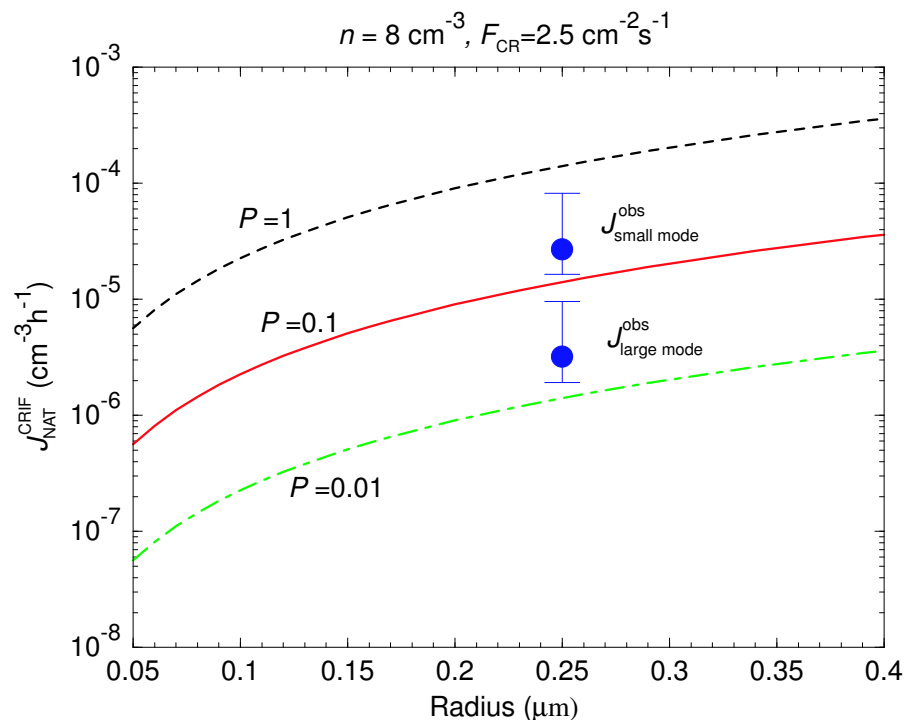


Fig. 2. Cosmic ray-induced freezing (CRIF) production rate of solid NAT particles ($J_{\text{NAT}}^{\text{CRIF}}$) as a function of the radius of STS droplets at three different values of P (0.01, 0.1, 1). P is the probability of freezing when a cosmic ray particle hits a supercooled STS droplet, which is likely to be a function of many parameters (see text for details). The shown observed NAT production rates are inferred from large mode and small mode NAT particles measured by Fahey et al. (2001), assuming that these particles were accumulated in a 3(\pm 2)-day period. The error bars indicate the possible ranges of freezing rate if the observed NAT particles are assumed to be accumulated in a period of 1–5 days.

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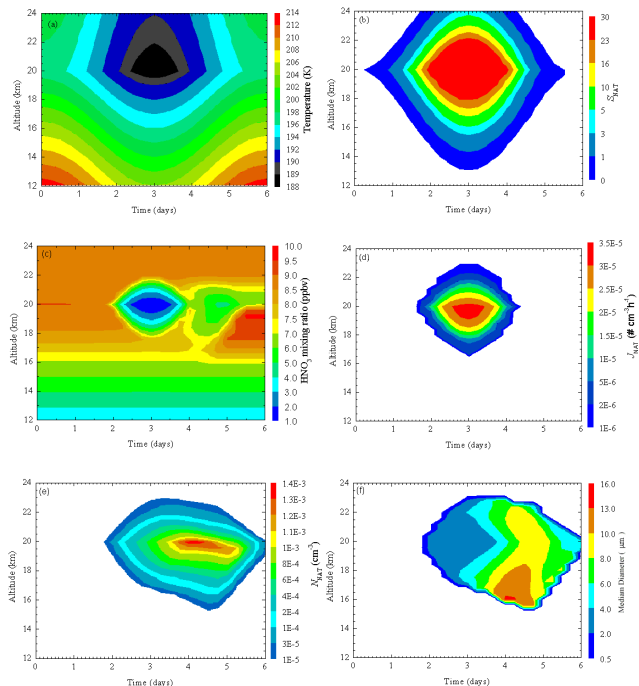


Fig. 3. PSC simulation using a 1-D PSC model with the cosmic ray-induced freezing mechanism included. Fields are plotted versus time and altitude: **(a)** temperature (K), **(b)** S_{NAT} , **(c)** HNO_3 gas mixing ratio (ppbv), **(d)** NAT particle production rate ($\text{cm}^{-3} \text{h}^{-1}$), **(e)** total number concentration of NAT particles (cm^{-3}), **(f)** median diameter of NAT particles (μm). The initial profile and sinusoidal oscillation of temperature follow those of Jensen et al. (2002) which is an idealized representation of the temperature variations experienced by air mass circulating around the pole during the winter of 2000 when SOLVE measurements were made. The lowest temperature is above ice frost point and classical homogenous freezing theory predicts negligible nucleation. The cosmic ray-induced freezing can qualitatively explain the highly selective formation of NAT particles observed during SOLVE (Fahey et al., 2001).

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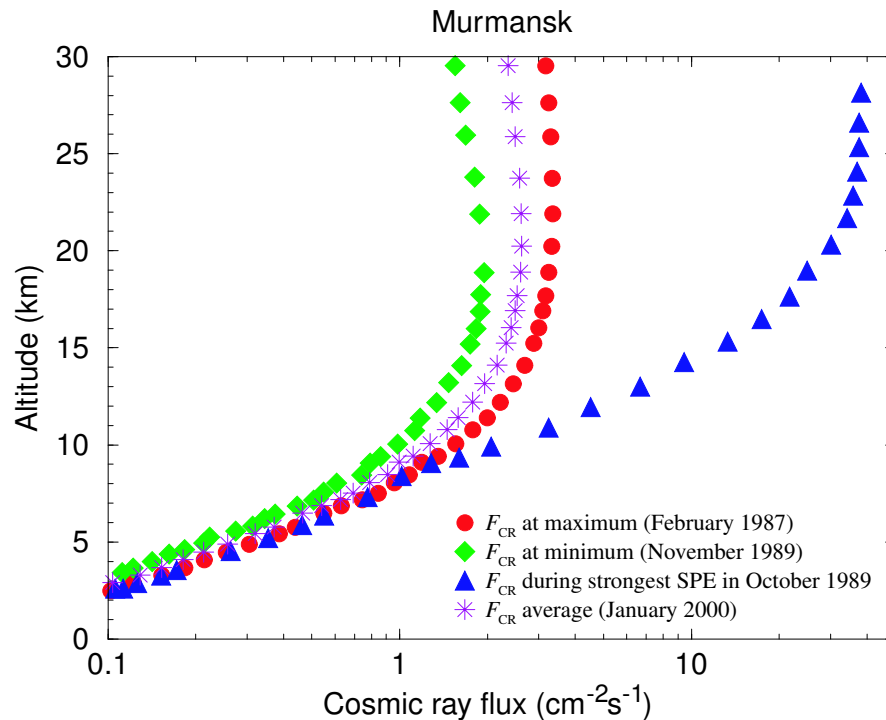


Fig. 4. Balloon-based measurements of F_{CR} as a function of altitude in northern polar regions (Murmansk, cut-off rigidity = 600 MeV) under different conditions of solar activities. The data for F_{CR} during solar minimum (February 1987, F_{CR} maximum), during solar maximum (November 1989, F_{CR} minimum), and during the strongest solar proton event in October 1989 (F_{CR} SPE) are from Bazilevskaya and Svirzhetskaya (1998). The F_{CR} profile corresponding to the 2000 winter polar stratosphere (F_{CR} average; used for calculations presented in Fig. 3) is interpolated based on F_{CR} maximum and minimum profiles and the cosmic ray intensities measured with Climax Neutron Monitor.

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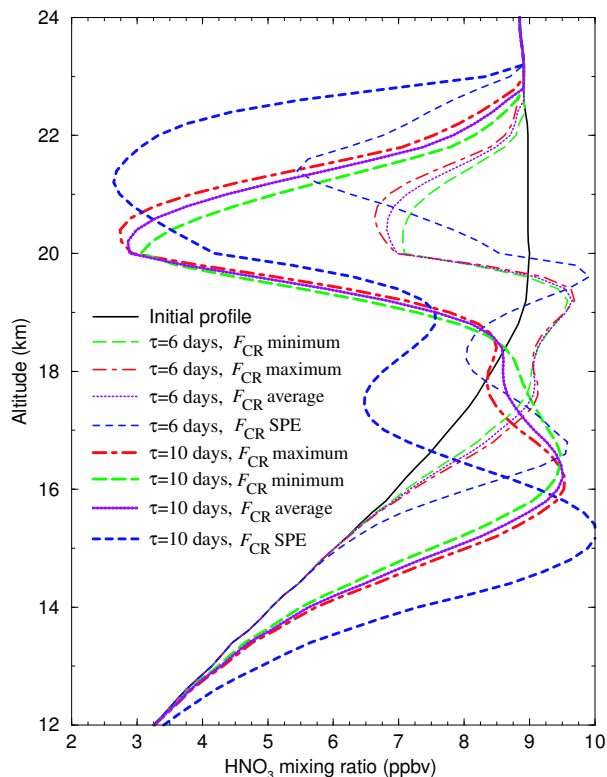


Fig. 5. Simulated HNO_3 mixing ratio profiles at $t = \tau$ using representative F_{CR} profiles corresponding to four different solar activities (maximum, minimum, and average solar activities, and during strong SPEs; Fig. 4) and with temperature oscillation periods (τ) of 6 days and 12 days. The initial HNO_3 profile is also shown for comparison. The strong SPEs significantly enhance the denitrification, especially in the top layer of PSCs. The renitrification layer during SPEs also shifts to lower altitudes. The cosmic ray-induced freezing may explain the high correlation between the impulsive nitrate events in polar ice cores with major PSCs and the existence of such correlations provides strong indirect support for the CRIF mechanism.

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