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Interactive Comment

Interactive comment on "Formation of large NAT particles and denitrification in polar stratosphere: possible role of cosmic rays and effect of solar activity" *by* F. Yu

F. Yu

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The author thanks the referee for taking time to review the paper. It appears that this referee missed some major points of the paper. Our point-to-point reply to the comments is given below.

(1) This paper presents: (I) cosmic rays-induced freezing (CRIF) mechanism which may explain the observed highly selective NAT particle formation; (II) the possible physics behind the CRIF; (III) a simple formula to include CRIF in PSC model; (IV) investigation on the effect of the solar activities on polar stratosphere denitrification; and (V) analyses of the observed signals in polar regions associated with solar proton events from a new perspective. All these are original in a sense that no previous similar work has been reported (see below for elaboration).

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(2) The referee may not realize that the CRIF mechanism proposed in this paper is different from the suggestion made by K. Carslaw at the workshop in April 2001.

As can be seen from Carslaw's presentation overhead posted on CERN website (http://cloudws.web.cern.ch/cloudws/documents_talks/Ken_Carslaw/P18.html), Carslaw suggested that the freezing may be initiated by the contact of the STS droplets with cosmic ray-generated ions or charged HNO3/H2O clusters (i.e., contact freezing). Similar suggestion has been made by Hamil and Turco (2000), and Turco and Hamil (2000) at AGU Spring meeting in June 2000 (see abstracts posted on http://www.agu.org/meetings/sm00top.html) and by D'Auria and Turco (2001) in their GRL paper. The concentration of small ions in polar stratosphere is around 10,000 cm-3 and a typical STS droplet will collide with a small ion in about every 10 seconds. Thus, the ion contact freezing can't explain the observed high selective freezing unless it is true that only certain very big charged clusters can initiate freezing (D'Auria and Turco, 2001). Carslaw and co-authors acknowledged in their papers (Fahey, Gao, Carslaw et al., 2001; Carslaw et al., 2002) that there is no established mechanism to explain the observed highly selective formation of large NAT particles. Obviously Carslaw and co-authors didn't consider the role of CR ionization in freezing as a possible mechanism since this was not even mentioned when the possible NAT formation mechanisms were suggested in these papers.

Our CRIF mechanism differs from ion contact freezing in that the CRIF mechanism involves the direct collision of high-energy CR particles with atoms inside the STS droplets while the ion contact freezing involves the collision of STS droplets with CR-generated ions or charged clusters in the air. As stated clearly in this paper, we propose that the CRIF is a result of the reorientation of polar solution molecules into crystalline configuration in the strong electrical fields of parent positive ions (may carry multiple charges) and ejected electrons generated when CR particles collide with an atom in the droplet. We further suggest that, in order to initiate freezing, at least two conditions (i.e., electrons energetic enough to travel beyond the critical size and the electric fields

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between positive ion and electrons above certain threshold value) have to be met. As far as I understand, when ion contact freezing was suggested, possible physics behind the mechanism was not given.

The CRIF suggestion is original because it is different from the existing suggestion of ion contact freezing and we demonstrate in the paper that the CRIF is physically plausible and consistent with a couple of the observed phenomena. I would like to emphasize again that the possible influence of CRs on STS freezing (either through CRIF or ion contact freezing) has not been considered as a possible mechanism in the recent papers discussing the observed highly selective formation of NAT particles.

(3) The electrical alignment of molecules in the electrical field to provoke freezing (which, I agree, is a fairly elementary view) is only one piece of the physics behind the CRIF. Another piece of the elementary physics is that the electric field strength has to be above certain level to induce freezing. The original and critical parts of CRIF include: (I) When a high-energy CR particle penetrates a STS droplet, it may collide with an atom which leads to the ejection of electrons. It is the strong electric field between the parent positive ions and ejected electrons that may initiate the freezing of STS droplets. (II) In order to initiate freezing, the energy of CRs should be high enough to induce certain multiple ionizations so that the electrons can travel beyond the critical crystalline embryo size and the minimum electric field strength between positive ion and electrons (when separated by a distance of the critical size) is above certain threshold value.

The point (II) is especially important as it may explain why Detwiler and Vonnegut (1980) and Seeley et al. (2001) didn't find the enhancement in freezing when the supercooled water droplets were irradiated with 5-Mev alpha particles (see discussion in page 1043 of this paper). As I understand, the result of the laboratory study by Detwiler and Vonnegut (1980) is one of the reasons leading Tinsley (1993) to agree with Detwiler (1993) that "the passage of energetic particles through droplets is not likely to be important for ice nucleation in the atmosphere" and to focus his attention (in

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his later studies) on contact ice nucleation (or electroscavenging) as being a candidate for electrofreezing. If our proposed theory turns out to be correct, we can infer that CRs may actually be able to induce the freezing of supercooled water droplet in upper troposphere which might have significant climatic implication. Based on the physics of our proposed CRIF, when a STS droplet collides with an air ion, the electric field near the single ion is not able to induce freezing (see Figure 1 and discussion in the paper).

(4) With the clarifications given above, I hope that this referee can agree that the statement "the related theory outlined offers little details and no new insight" is a misinterpretation of this paper. We also want to point out that our investigation on the effect of solar activities on denitrification is the first of its kind. Our re-interpretations of the observed enhancement in aerosol backscattering ratio at PSC layers shortly after an SPE and the high correlations between the thin nitrate-rich layers in polar ice cores and major SPEs clearly offer new insights into the issues.

With regard to P2, we agree that this parameter is not well constrained theoretically. As we pointed out in the paper, P2 is likely to be a function of many parameters including the energy of the incoming CRs, temperature, NAT supersaturation ratios, composition and size of STS particles, and interfacial tension between the liquid and solid phases. At this stage, it is almost impossible to derive theoretically the values of P2 though further theoretical study can advance the theory. One should not dismiss a new theory or hypothesis because it contains an unconstrained parameter. Even for the widely used classical homogenous freezing theory, the key parameter activation energy is not well constrained theoretically and has to be decided through laboratory studies (Knopf et al., 2002).

As we have already pointed out in the paper, laboratory study using CR particles in the energy range common in the atmosphere is needed to test the CRIF theory and the values of P2 may have to be decided through laboratory studies and/or observations. Since CR particles in the energy range common in the atmosphere can't be generated in a regular laboratory, the practical approach is to observe if freezing occurs when

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natural CR particles penetrate STS droplets. Techniques to detect the incoming CRs need to be developed to confirm the penetration of STS droplets by CRs. It is interesting to note that, in their laboratory measurements studying homogeneous freezing of supercooled solutions, Koop et al. (1995) and Beyer et al. (1994) found that, for the same conditions, usually a subgroup of the sample froze in a very short time, while the rest took very long or did not freeze at all. While Koop at al. (1995) have attributed such freezing to the formation of ice frost or NAT on the containment walls from the vapor phase, such spontaneous freezing could also be due to the cosmic ray-induced freezing. Note that the flux of CRs in a laboratory room is likely to be very small ($\lesssim$ 0.005 cm-2s-1), thus only a very limited number of droplets are likely to be hit by CRs in the laboratory at a given time period.

In summary, we do not agree with this referee about the original contribution of this paper. We have shown above that the major points of the paper were misinterpreted. We will make our points clearer in the revised version of the paper.

The technical points will be considered in the revised paper.

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Other papers cited in this reply can be found in the reference list of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 1037, 2004.

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