Atmospheric Chemistry and Physics Discussions

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. Our reply to the comments is given below.

1. With regard to Figure 3. As we pointed out in the paper, the similarity of Figure 3 to earlier study is expected since the same initial profiles and sinusoidal oscillation of temperature were used and the freezing rates used in both cases are constrained by the observation. The major difference is that we used equation (2) to calculate the cosmic ray-induced freezing while others used a constant freezing rate. As can be seen in Figure 3(d), the freezing rates in our model changes with time (due to change in the radius of STS droplets). The major purpose of figure 3 is to demonstrate that equation (2) can be used in the PSC model to predict PSC formation. Since equation

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(2) contains several variables (cosmic ray flux, particle concentration, particle radius), a PSC model with CRIF mechanism included can physically study the dependence of PSC properties on solar activity (as shown in the paper) and other parameters (such as background aerosol concentrations, HNO3 mixing ratio, etc.). Figure 3 also provides a base for the following discussion with regard to the effect of solar activity on denitrification.

2. The real question. we agree with the referee that the real question is whether CRIF is a viable mechanism.

While the possible effect of cosmic ray on the freezing of supercooled droplets has been suggested for more than 30 years (Varshneya, 1969), we propose here for the first time 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. As we point out in the paper, the mechanism we proposed is totally different from the mechanism presented by Varshneya (1971) and may explain why Detwiler and Seeley observed negligible effect of alpha particle radiation on freezing. The equation (2), while simple and straightforward, is the first of its kind to link the freezing rate with several important parameters (cosmic ray flux, particle concentration, particle radius). In the past, the parameterization of the classical homogeneous freezing rate or a fixed constant freezing rate has been used in PSC models to simulate NAT formation. Equation (2) offers another quite different mechanism linking PSC properties with several important parameters.

Now again the question is whether the CRIF represented by equation (2) is a viable mechanism. This referee raised several doubts which we address below.

3. What is the value of P_2? As we pointed out in the paper, P_2 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. We may be able to derive the possible range of P_2 by looking into the multi-ionization cross section of cosmic rays of different energies and study the behavior of polar molecules in the strong electric fields near the ions (which are under investigation). However, the final values of P_2 have to be decided through laboratory studies and/or observations. 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).

In this paper, we infer that P_2 should be around 0.1 from Fahey et al.'s observations. While we are not able to give a physical reason why P_2 is 0.1 at this point, I don't see why P_2 should not be 0.1 either. This referee argued that P_2 should be much lower for those droplets which remain supercooled for a long time. This is not true because the main reason that lots of droplets remain supercooled for a long time is that the chance that these droplets hit by a cosmic ray particle is very small. Based on equation (2), less than 1% of particles with radius of 0.2 μm (0.1 μm) will be hit by a cosmic ray in 30 days (120 days). This is exactly why CRIF is highly selective.

4. Shumilov et al.'s observations. I am not sure how significant the subside of air may enhance the sulfuric acid gas concentration ($[H_2SO_4]$) in polar stratosphere (any measurements?). Even if $[H_2SO_4]$ is high enough to induce formation of new ultrafine particles, I don't think that the further increase in the ionization rate (as a result of SPE) will increase the nucleation rate because the nucleation should be limited by $[H_2SO_4]$. Otherwise, we should see very high concentration of new particles as the ionization rate in the polar stratosphere is very high. On the other hand, it takes a long time to grow the fresh nucleated sulfuric acid particles to a size that can be detected by lidar (radius > 0.69 μm). As pointed in the text, the profiles of aerosol backscattering ratio indicates that enhanced particle layer formed precipitated quickly (1-2 km/day). Only PSC particles of around 10 μm can have this precipitation velocity. To form PSC

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particles of around 10 μm, the freezing process must be very selective. I doubt that the possible nucleation of NAT or NAD on the large number of ions has this highly selective property and I don't see how the increase in ionization during SPE will enhance the nucleation (again the nucleation on ions is unlikely to be limited by ion concentration).

5. The correlation between NO_3^- core with SPE. It is well know that the sedimentation of large NAT particles denitrificate the polar stratosphere (where HNO_3 mixing ratio is high). The NAT particles will not make it to the surface in a single PSC cycle. However, the sedimentation of NAT particles moves the HNO_3 to a lower layer which will then be precipitated to surface in the subsequent cloud processes. This may explain why the nitrate events in ice core after the SPEs have duration of a few weeks.

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