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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

K. Carslaw (Editor)

CARSLAW@ENV.LEEDS.AC.UK

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Both referees are correct in pointing out that (a) the suggestion that cosmic rays may be responsible for the formation of low number concentrations of nitric acid hydrate particles in the polar stratosphere is not new and (b) that this paper does not quantify the rate of hydrate formation and that therefore we are no closer to including this mechanism in any meaningful way in atmospheric models.

I concur with the referees on both points. However, despite the uncertainties that remain, Yu has provided us with a useful phenomenology for cosmic ray-induced freezing that identifies some key parameters that need to be constrained by experiments. Yu also offers a physical explanation for why the freezing nucleation rate might depend on the energy of the ionising radiation (due to the strength of the electric field induced in the droplet by ionisation). It would be wrong to suggest that this paper represents only a modest extension of earlier work. In particular, my own contribution (Carslaw et al. 2001) can only be considered at the level of a suggestion rather than an explanation, and the work of d'Auria and Turco (2001) considers a different mechanism.

The observations of enhanced aerosol backscatter after a solar proton event in the 1984 Arctic polar vortex (Shumilov et al., 1996) are interesting and deserve further investigation. The observations were made in air that was most likely cold enough to form nitric acid trihydrate (NAT) PSCs, although Shumilov et al. attribute the enhanced backscatter to sulphuric acid aerosol formation. The correlation between the enhanced backscatter and the SPE; the occurrence of temperatures below those required for NAT; and the subsequent apparent sedimentation of the particle layer are suggestive of a SPE-induced NAT event. However, there are many factors that complicate such an interpretation, including the temperature histories of the air parcels that were probed and the possibility of NAT formation elsewhere in the Arctic vortex followed by advection over the observation site. However, the suggestion of Yu to investigate other similar events is a useful one.

The observations of enhanced nitrate in Antarctic and Arctic firn cores correlated with SPEs are an open issue and it is conceivable that denitrification of the stratosphere plays a role. However, as with the Shumilov et al. observations, the links remain to be established quantitatively. Firstly, denitrification of the Antarctic stratosphere tends to be essentially complete every winter, so it needs to be explained how a small modulation of the NAT nucleation rate could affect denitrification (in contrast, the Arctic is much less denitrified). Secondly, it needs to be shown that the amount by which nitrate is enhanced corresponds to the amount by which denitrification of the stratosphere might be modulated. I suggest these as topics for further investigation by other, but Yu needs to acknowledge the uncertainties.

One of the referees argues that the denitrification simulations presented in section 3

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are not very illuminating as they essentially reproduce the results of earlier studies. Here I agree, although in Yu's defence it should be pointed out that readers of this paper from outside the stratospheric community may find these simulations helpful. This section serves only to establish the value of P2 required to explain observed particle concentrations and magnitude of denitrification. Nevertheless, it does need to be stressed that these simulations are as uncertain as earlier simulations that assumed a constant NAT nucleation rate. The uncertainty in the value of P2 used in the model and its unknown dependence on several physical parameters means that any dependence of denitrification on particle radius (in eq. 2) is probably irrelevant. It will not be possible to confirm a radius dependence of nucleation rate from observations so long as the functional form of P2 remains unknown.

Some effort needs to be made to explain laboratory freezing experiments of Koop et al. (1995). They measured upper limits to the freezing rate for liquid volumes of approximately 1 cm^3 or less under stratospheric conditions. Is it possible to use the results to determine an upper limit to the value of P2?

Overall, this paper does a good job of assembling concepts from a range of atmospheric sciences. It provides a useful conceptual picture of what might be involved in cosmic ray-induced freezing of stratospheric droplets and those involved in stratospheric aerosol processes will find something useful in the paper. If presented less as a quantitative 'explanation' of NAT formation but more as a framework from which further studies can develop, then it is acceptable for publication. The paper needs to stress that cosmic rays *can* explain NAT nucleation rates and identify how the processes might be better quantified so that it can be established whether this mechanism really does explain the observations.. Both referees raise some important issues that need to be addressed in any revised version.

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