

## ***Interactive comment on “Nitrogen and oxygen isotopic constraints on the origin of atmospheric nitrate in coastal Antarctica” by J. Savarino et al.***

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The authors are to be congratulated on new and interesting work. Some comments that might be relevant to the interpretation are as follows: [1] Over Antarctica, the temperatures and water vapour partial pressures from mid-winter into spring have vertical profiles that will allow an ice crystal (or NAT ‘rock’) to continue to grow, both by vapour deposition and by coagulation with underlying crystals, as it falls under gravity. Once into the lower stratosphere and upper troposphere, this process can be expected to accelerate into a runaway condition all the way down to the surface, because most PSCs are forced by synoptic systems from below, with ice cloud reaching from the surface to the top of the type II PSCs, as encountered by the ER-2 19870817 and 19870818. Further out in the vortex, often at locations near DDU, ER-2 observations in 1994 sug-

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gest that such runaway does not always occur, with enriched NO<sub>y</sub> content sometimes occurring as the result of the kind of evaporation seen over the Arctic by the DC-8. There is a discussion of these mechanisms in section 4 of Q. J. R. Meteorol. Soc., 123, 1-69, (1997). There are also maps of the frequency of occurrence of PSCs as a function of geographical position, showing a maximum over west Antarctica for both 1987 and for several years' climatology from SAM II and SAGE, published in J. Geophys. Res., augmented later by emission limb scanners on UARS. [2] If sedimentation can be modulated by evaporation and followed by later re-condensation, a route for isotopic exchange may exist via what happens in the aqueous surface layer and interior of the particles: the water vapour that goes on to and into the particle may ionize, giving opportunities for both N and O isotopic exchange as well as H and D. Because of the mobility via tunneling of protons and deuterons in liquid water and ice, the H and D which leave the particle when a water or nitric acid molecule leaves may not be the ones that were attached to the O atom when it entered the particle as part of a water or nitric acid vapour molecule. [3] Overtone pumping can effect photodissociation of molecules like HNO<sub>3</sub>, HNO<sub>4</sub> and H<sub>2</sub>SO<sub>4</sub> by near infrared and visible light (Geophys. Rev. Lett., 24, 2651-2654, 1997; Geophys. Res. Lett., 26, 1373-1376, 1999; Chem. Revs., 103(12), 4717-4729, 2003; Science, 299, 1566-1569, 2003). Isotope exchange seems probable, given the recent theoretical work of Y. Miller and R. B. Gerber in J. Phys. Chem., earlier this summer.

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