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- Recent Final Revised Papers
- Volumes and Issues
- Special Issues
- Library Search
- Title and Author Search

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General Information
Submission
Review
Production
Subscription
Comment on a Paper

Volumes and Issues | Contents of Issue 2

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Factors controlling Arctic denitrification in cold winters of the 1990s

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Abstract. Denitrification of the Arctic winter stratosphere has been calculated using a 3-D microphysical model for the winters 1994/95, 1995/96, 1996/97 and 1999/2000. Denitrification is assumed to occur through the sedimentation of low number concentrations of large nitric acid trihydrate (NAT) particles (as inferred by e.g. Fahey et al., 2001). We examine whether the meteorological conditions that allowed particles to grow to the very large sizes observed in 1999/2000 also occurred in the other cold winters. The results show that winter 1999/2000 had conditions that were optimum for denitrification by large NAT particles, which are a deep concentric NAT area and vortex. Under these conditions, NAT particles can circulate in the NAT-supersaturated air for several days, reaching several micrometres in radius and leading to a high downward flux of nitric acid. The other winters had shorter periods with optimum conditions for denitrification. However, we find that NAT particles could have grown to large sizes in all of these winters and could have caused significant denitrification. We define the quantity "closed-flow area'' (the fraction of the NAT area in which air parcel trajectories can form closed loops) and show that it is a very useful indicator of possible denitrification. We find that even with a constant NAT nucleation rate throughout the NAT area, the average NAT number concentration and size can vary by up to a factor of 10 in response to this meteorological quantity. These changes in particle properties account for a high degree of variability in denitrification between the different winters. This large meteorologically induced variability in denitrification rate needs to be compared with that which could arise from a variable nucleation rate of NAT particles, which remains an uncertain quantity in models. Sensitivity studies show that although denitrification was likely approaching asymptotic minimum values throughout much of the 1999/2000 vortex, decreases in the volume-averaged nucleation rate would have substantially reduced the denitrification.

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