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3-D microphysical model studies of Arctic denitrification: comparison with observations

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Abstract. Simulations of Arctic denitrification using a 3-D chemistry-microphysics transport model are compared with observations for the winters 1994/95, 1996/97 and 1999/2000. The model of Denitrification by Lagrangian Particle Sedimentation (DLAPSE) couples the full chemical scheme of the 3-D chemical transport model, SLIMCAT, with a nitric acid trihydrate (NAT) growth and sedimentation scheme. We use observations from the Microwave Limb Sounder (MLS) and Improved Limb Atmospheric Sounder (ILAS) satellite instruments, the balloon-borne Michelsen Interferometer for Passive Atmospheric Sounding (MIPAS-B), and the in situ NO_y instrument on-board the ER-2. As well as directly comparing model results with observations, we also assess the extent to which these observations are able to validate the modelling approach taken. For instance, in 1999/2000 the model captures the temporal development of denitrification observed by the ER-2 from late January into March. However, in this winter the vortex was already highly denitrified by late January so the observations do not provide a strong constraint on the modelled rate of denitrification. The model also reproduces the MLS observations of denitrification in early February 2000. In 1996/97 the model captures the timing and magnitude of denitrification as observed by ILAS, although the lack of observations north of ~67° N in the beginning of February make it difficult to constrain the actual timing of onset. The comparison for this winter does not support previous conclusions that denitrification must be caused by an ice-mediated process. In 1994/95 the model notably underestimates the magnitude of denitrification observed during a single balloon flight of the MIPAS-B instrument. Agreement between model and MLS HNO_3 at 68 hPa in mid-February 1995 is significantly better. Sensitivity tests show that a 1.5 K overall decrease in vortex temperatures, or a factor 4 increase in assumed NAT nucleation rates, produce the best statistical fit to MLS observations. Both adjustments would be required to bring the model into agreement with the MIPAS-B observations. The agreement between the model and observations suggests that a NAT-only denitrification scheme (without ice), which was discounted by previous studies, must now be considered as one mechanism for the observed Arctic

denitrification. The timing of onset and the rate of denitrification remain poorly constrained by the available observations.

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