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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Homogeneous nucleation of NAD and NAT in liquid stratospheric aerosols: insufficient to explain denitrification" *by* D. A. Knopf et al.

D. A. Knopf et al.

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Question 1 + 2:
In the Experimental section, the geometry of the cell which houses the droplets should be described in more detail. In particular, how are the droplets sealed from the atmosphere?
What evidence do the authors have that the droplets do not change their composition from their initial values?
Reply:
The following text will be appended:
p. 673, l. 27: ...flow clean bench. Either a Teflon plate or an o-ring, each covered by a thin layer of high-vacuum-grease, served as a spacer for a second quartz plate which sealed the droplets against ambient air. The inner diameter of the spacer depended

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on the investigated droplet volume and varied between 0.3–0.6 cm and the spacer thickness ranged between 0.125–0.175 cm. The total volume of the cell was about 8.8 10^{-3}–5 10^{-2} cm^3. The volume of the droplets varied between 10^{-3}–10^{-2} cm^3. Therefore, even at room temperature the number of water and HNO_3 molecules in the gas phase of the cell is negligible when compared to the number of condensed water and HNO_3 molecules in the droplets. Hence, the composition of the droplets stays constant during a freezing experiment. The preparation of the droplet cell took about 15 s. The number of molecules which may evaporate during that time is negligible to the total number of molecules in the condensed phase. This was confirmed by checking the melting points of the droplets after freezing which were found to be in agreement with the phase diagram.

Question 4:
On page 675, the meaning of n* should be elaborated upon.
Reply:
The following sentence will be appended:
P. 675, l. 7: ...(Koop et al., 1997) – i.e., if the experiments were repeated an infinite number of times the observed number of nucleation events will be smaller than n^* in 99.9 % of the cases.

Question 3+5:
How many droplets were used?
What is the reproducibility of the measurements? How many times was the experiment conducted on droplets of the same composition?
Reply:
The total number of experiments and the total number of individual droplets will be included in table 1.
p. 674, l. 17: Table 1 shows the composition, volume, total number of different per-

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formed experiments, and total number of individual droplets.

Table 1: Composition, volume, total number of performed experiments, and total number of individual droplets. The symbols refer to the ones in Fig. 5

Solution	HNO_3 [wt%]	H_2SO_4 [wt%]	H_2O [wt%]	Volume [$10^{-3}cm^{-3}$]	Symbol	# Exp.	# Drop.
1	63.6	0	36.4	5–10	$\times$	16	16
2	53.8	0	46.2	1–10	$+$	28	28
3	32.2	13.8	54.0	10	$*$	22	5
4	38.3	7.6	54.1	10	$\bullet$	16	4

All droplets of solution 1 nucleated within a temperature range of about 205–208 K. A large fraction of the droplets of solution 2 froze below about 192 K, however, some droplets froze above 200 K, most likely due to heterogeneous nucleation. This might indicate that heterogeneous NAT nucleation occurs more readily than heterogeneous NAD nucleation. None of the droplets of solutions 3 and 4 did nucleate during the observation time.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 669, 2002.