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Atmospheric Chemistry and Physics Discussions

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

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As I mentioned in my previous comment, the possibility of pseudo-heterogeneous nucleation occurring at the surface of liquid droplets do not not affect the conclusions drawn in the paper by Knopf et al. 2002. Nevertheless, I take the opportunity to discuss here some of the features of pseudo-heterogeneous nucleation.

I agree that the idea of pseudo-heterogeneous is very interesting and should be pursued further in experiments and theoretically. Also, the experiments presented in

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Tabazadeh et al. 2002 seem to support the suggestion although a rigorous proof is still missing. However, I do not agree with the statement made in one of the comments by A. Tabazadeh that the presented formulation for pseudo-heterogeneous nucleation is in agreement with all experimental data published to date:

1. It seems a little to easy to discount the bulk sample data (which do not fit the parameterization) because of possible impurities that are "*present in every laboratory*", and then go ahead and use all other laboratory data without much discussion about a possible surface contamination of those. In fact, a possible surface contamination of the Salcedo et al. data is discussed, but then these data are used anyway to show that they fit the parameterization.

2. An aerosol study by Bertram et al. JGR 2000 has been ignored. Only data from binary HNO_3/H_2O droplets (including data from previous measurements by Bertram et al.) with HNO_3:H_2O mole ratios of 1:2 and 1:3 have been used to derive a linear concentration dependence of the Gibbs activation energy and, hence, the nucleation rate coefficient. This is then linearly extrapolated to stratospheric HNO_3 concentrations. However, in their 2000 JGR paper Bertram et al. present data for non-stoichiometric mole ratios over a much broader concentration range. These data clearly show that a linear concentration dependence is incorrect. Hence the current parameterization for pseudo-heterogeneous nucleation presented in Tabazadeh et al. 2002, when linearly extrapolated to stratospheric conditions, overestimates the pseudo-heterogeneous nucleation rates at stratospheric conditions. I want to stress once more that I am not saying that pseudo-heterogeneous nucleation is not possible in the lab or in the stratosphere. However, the current parameterizations is not in agreement with available aerosol data and, therefore, must not be used at stratospheric conditions.

Another point in the same direction is that the current parameterization is based exclusively on data of binary HNO_3/H_2O samples. As A. Tabazadeh points out several times in the discussion, minor constituents may have a very strong effect on the surface composition and, therefore, on the pseudo-heterogeneous surface nucleation rate coefficient. So the rate coefficients in ternary droplets in the stratosphere might be very

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much different from those in binary droplets in the lab. Even if HNO_3 is enriched at the surface of ternary droplets, the surface will not be free from sulfate or bisulfate ions. These effects definitely need to be taken into account before any applications to the stratosphere can be made.

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