Atmos. Chem. Phys. Discuss., 14, C1935–C1939, 2014
www.atmos-chem-phys-discuss.net/14/C1935/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Denitrification by large NAT particles: the impact of reduced settling velocities and hints on particle characteristics" *by* W. Woiwode et al.

Anonymous Referee #2

Received and published: 29 April 2014

This paper presents a limited study of denitrification occurring in the 2009/2010 Arctic stratosphere, combining in situ particle measurements (FSSP-100 on the Geophysica), airborne FTIR (MIPAS-STR) and a 3-D simulation of chemistry with Lagrangian transport and sedimentation (CLaMS driven by ERA-Interim winds and temperatures.)

Particles detected by FSSP-100 have diameters up to 30 microns. It is claimed that the NAT particles cannot grow to these large sizes assuming the standard NAT mass density and compact near-spherical shapes within the actual time available for growth under super-saturated conditions (i.e. amount of time following nucleation the particles remain below TNAT). An attempt to model the particle growth with CLaMS shows that

the sedimentation velocities may have to be reduced significantly to match the HNO3 gas phase vertical distributions. The reduced settling velocities are attributed to the formation of compact columnar particles with large aspect ratios ∼8.

The main concern I have is that far too little has been presented to rule out more mundane explanations of the discrepancy between the observed FSSP particle sizes / MIPSA-STR HNO3 vertical distributions and the CLaMS modeled results. More detailed comments are given in the section below. Presumably more simulations runs would be required address these concerns, but these are essential to strengthen the conclusions of the paper and are clearly within the scope of this study.

/xxx/ ==> delete xxx

[xxx] ==> add xxx

P5895-L26: "composed of NAT" What about ICE coated NAT?

P5898-L22: Mie theory accounts for scattering from spherical particles only. Please give a reference for how scattering from "slightly aspheric" particles with random orientation can be approximated adequately by Mie theory.

P5898-L21-26: It's not clear if you used the advanced methods cited or how large the difference is in particle sizing between the MIE and more advanced methods. This seems important given that your conclusions are that the NAT particles have an aspect ratio of about 8 and are therefore not at all "slightly aspheric".

P5899-L20: contributions [from aerosols] are typical

P5899-L25: high dynamic/s/[al] range

P5899-L29 and P5900-L3: I find the term "stray light" somewhat misleading. It would normally used to describe an instrument artefact such as unwanted reflections which could maybe have been reduced by a better optical design. Here you are talking about atmospheric radiation upwelling from the surface and troposphere and being scattered

by clouds into the instrument line of sight i.e. an external contamination of the requisite limb signal. In principle, this effect could be modeled. Since the unwanted tropospheric signature is not the fault of instrument engineering the effect should not be labeled as a stray light problem.

First you state that tropospheric CO2 and H2O signatures were not identified, but then state that as H2O was not retrieved that "stray light cannot be ruled out for this species". As shown in Hopfner (2004) this is viewed directly by examining the radiance spectrum for evidence of a tropospheric signature of the H2O lines (i.e. an absorption feature) providing particle sizes are in the range 1 to 7 microns. Overall, I think you need to make a better job of explaining how you extract information on aerosols, temperature and gases from the MIPAS-STR measurements. Additionally, no indication is given at all on the uncertainties in the retrieved HNO3 and temperature from MIPAS-STR.

P5900-L22: /probable/ [potential]

P5900-L23: /sizes in/ diameter[s]

P5900-L27: The text apparently gives the differential number densities at the peaks of the two modes (i.e. the peak bin values corresponding to Fig 1). This is misleading because normally the total number density in each mode would be reported (e.g. Fahey et al (Fig 4 caption) give the integral of their large NAT mode as 2.3E-4 cm-3). You state that your FSSP large NAT mode is a factor of 5 larger than Fahey et al, so we have 5*2.3E-4 cm-3 => 0.00115 cm-3. You need to give the FSSP integrated mode densities so these can be compared directly. Also, try working out what would be the HNO3 content of both NAT modes. This is useful information.

P5901-L2: mode is /by/ about [a] factor [of] 5

P5901-L19: "yielding compact spherical particles". The particle shape and form is not being simulated and is therefore not "yielded". Suggest changing this to "resulting in growth of the simulated spherical particles to maximum diameters..."

P5903-L1: A small temperature bias can have a considerable effect on particle growth. What would be the effect of a reasonable uncertainty in the CLaMS ERA-Interim temperatures on the eventual particle sizes? What about temperature fluctuations experienced along the trajectory? Errors in ambient HNO3/H2O? Could these act to reduce the discrepancy in the modeled and FSSP particle sizes? These effects should be ruled out before looking for more complex explanations. Did you compare the MIPAS-STR and ERA-Interim temperatures? There is also the question of the nucleation mechanism. Have alternatives been considered? What about the NAT nucleation rate? These issues must be addressed adequately.

P5903-L6-7: /ppbv/ [ppmv]

P5903-L18-25:The measurements from MIPAS-STR should be explained in more detail, such as the uncertainties on the retrieved quantitities. Without this information the reader is unsure what significance to attribute to "excess HNO3" etc. Please quote some HNO3 values for the maxima and excess rather than leaving it to the reader to work these out from the plots. It will also help understand the comment below about P5904-L7.

P5903-L13: Here you are invoking effects of retrieval uncertainties and horizontal gradients without giving any indication of their magnitudes.

P5903-L13: Uncertanties in HNO3 also affect the calculated TNAT.

P5904-L6-7: the [continiuum] retrieval: You need to explain why the continuum is not sensitive to large NAT/low number density. Although I think your interpretation is incorrect if the amount of HNO3 in the NAT particles is significant (say around 1 ppbv or more?). I would expect this amount of HNO3 uptake into large NAT particles to generate detectable mid-ir emission.

P5904-L8-10: "large NAT particles falling ... dense PSC cloud ... increased opaqueness". The wording here would appear to contradict your earlier assertion that the

mid-ir is insensitive to large NAT/low number density particles. To what do you attribute the cause of the increased opaqueness? Are you talking about the large NAT falling out of a mixed phase STS/NAT cloud or from a population of large/small NAT? Is it cold enought to form STS? It is not clear from your description.

P5906-L16-18: Essentially you are saying that trying to match up individual vertical profile comparisons is not possible, but never mind because over the vortex the ensemble profile is more reliable?

P5910-L7: give the dimensions [of] potential

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 5893, 2014.