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Atmospheric Chemistry and Physics Discussions

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2, S32–S38, 2002

Interactive Comment

Interactive comment on "NAT-rock formation by mother clouds: a microphysical model study" *by* S. Fueglistaler et al.

S. Fueglistaler et al.

Received and published: 20 March 2002

Reply to interactive comments by Reviewer 1 and 2.

General Comments:

We agree with reviewer 2 that the model description is short and contains only the minimum of what is needed to repeat the calculations. The new manuscript will describe the model and choice of parameters in more detail.

Both reviewer 1 and 2 express their concern about the impression given in the paper that the mother cloud/NAT-rock mechanism might be the only explanation for the NAT-rock observations.

Our model study is supposed to show that sedimentation of NAT particles out of a mother cloud produces a NAT population a few kilometers below the mother cloud with

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Interactive Discussion

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the characteristics (particle size, number density) of the recent NAT-rock observations. The study does not investigate what other processes may lead to NAT-rocks, and we do NOT claim based on this study that it is the only mechanism to lead to NAT-rocks. However, in contrast to the remark of reviewer 2 that it is "not really surprising" (last specific comment) for NAT particles falling into supersaturated air to become NAT-rocks, we maintain that it is a major finding of this paper that the calculated particle number densities are consistent with the observations.

We can show that it is not preferrential sedimentation of initially larger particles but two other effects that control particle number density of the sedimenting particle ensemble: particle number density reduction due to particle acceleration and additional particle number density reduction due to gas phase depletion. It is shown in this study, that these two effects lead to $n \approx 10^{-4} cm^{-3}$ a few kilometers below the mother cloud for a wide range of particle number densities in the mother cloud. This finding could not be anticipated a priori - it could have been that the particle number densities decrease too much or not enough to be consistent with measurements. It is only this qualitative understanding of the process and the quantitative analysis that allows to conclude that NAT-rocks may form from mother clouds. To better explain this point we intend to add in the revised manuscript a new figure and a short paragraph comparing the steady-state solution (without gas phase depletion) with the results of the full model.

We agree that the first sentence of "Conclusions" is too strong and suggest the following: "This study shows that type 1a and type 1a-enh PSCs with $n = 0.01 - 1.0 cm^{-3}$ can be a source of NAT-rocks, which efficiently denitrify the polar vortex."

Reply to specific comments of anonymous reviewer 1

1. The authors seem to say "QED". ...

Reply: See reply to 'General comments' and response to last point of reviewer 2.

p.30, l. 17: Reference to Gao et al.

Reply: Will be added

p.31, l.8-10: 'observed characteristics'

Reply: Will be specified to be particle number density and size in the revised manuscript.

—: Discussion of the Flux Reduction Layer (FRL)

Reply: We consider this discussion to be an essential part of the work. Of course the flux reduction occurs in the entire column, but its relative importance is most pronounced in the layer just below the mother cloud. As stated in the paper, the FRL depends also on the number density of the mother cloud. For a dense mother cloud ($n \approx 1cm^{-3}$) it is a distinctive region of a few 100 m thickness which remains throughout the lifetime of the mother cloud. Conversely, a less dense mother cloud ($n \approx 0.01cm^{-3}$) does not show a pronounced FRL at any time. It was not intended to introduce additional 'jargon' to the PSC field, rather it was our intention to concisely describe the mechanism. The revised version will explain this part in more detail (see also 'General comments').

p.32, l. 23 - p.33, l.10: Profiles of HNO3 and temperature

Reply: The gas phase HNO3 in the modelled mother cloud is 0.8 ppb (given S-NAT=10) and 8 ppb everywhere else (partitioning into STS at S-NAT = 10 is negligible), the temperature is in the entire column about 3 K below T-NAT. At p = 40 hPa this corresponds to approximately 191.5 K. The revised manuscript will give these numbers, but does not emphasize it, as what is important is S-NAT.

p.33, l.17: Units (ppb x km / day)

Reply: We agree that this unit is unusual. However the SI unit molec/m2/s is much less intuitive.

p.34, l.11: STS existence temperature

ACPD

2, S32–S38, 2002

Interactive Comment

Reply: The term is indeed imprecise and will be removed.

p.35, l.12: no -> not

Reply: Will be changed.

Reference to Dhaniyala et al.

Reply: Will be given.

Proposed sentence: p.35, l. 16: In addition, wind shear causes particles to sediment through different air masses, which increases the volume of air denitrified by the sedimenting particles [Dhaniyala et al., 2002].

p.35, l.18: First sentence in Conclusions, 'reveal'

Reply: See discussion in 'General comments' and (1)

Reply to specific comments of anonymous reviewer 2

p.30, l.12: atmospheric dilution

Reply: Effects of wind shear and turbulence - since we do not discuss these effects in detail in the paper we will omit this in the revised version.

p.31, l.19:

Reply: Apparently we were not able to explain sufficiently well the steady-state solution and why it is important. The paragraph will be rewritten, and a new figure will be added where the importance of the steady-state solution to the understanding of the particle number density reduction is discussed.

p.32, l.14/18: Eulerian/Lagrangian

Reply: We do not consider these terms as 'jargon'.

p32., l.21: Radius of particles in mother cloud

ACPD
2, S32–S38, 2002

Full Screen / Esc
Print Version
Interactive Discussion
Original Paper

Reply: We will add the radius in the revised version.

p.33, l.4: Thickness of mother cloud, STS particles below

Reply: All calculations were done for an "infinite" thick cloud above 22 km. We chose this setup to be independent of the cloud thickness. This point will be discussed in the section "Model limitations and sensitivity tests" of the revised version.

Below the mother cloud only in the 'STS' case liquid droplets play a role, at S-NAT = 10 the partitioning into STS is neglectable which will be explicitely stated in the section "Description of the microphysical model".

p.33, l.6: STS, Profiles

Reply: STS -> see above

Profiles: As stated, we want the calculations to be unobstructed by effects caused from temperature/HNO3 profiles. This is why we set HNO3-concentration = 8ppb and S-NAT = 10 in the first place. Temperature is then calculated from these two parameters (about 3 K below the NAT existence temperature, i.e. about 191.5 K at p = 40 hPa, see also response to reviewer 1).

p.32, l.18: Treatment of liquid particles

Reply: We do not calculate growth/evaporation of the liquid particles but consider them in thermodynamic equilibrium and hence calculate the available HNO3 at each time step. This is justified since equilibration time of STS is fast for the small gas phase changes due to HNO3 uptake from the sedimenting NAT particles.

p.33, l.15: Properties of falling particles

Reply: Yes, will be added.

p.33, l.16: FRL

Reply: As stated above we will discuss this point better in the revised version. The

ACPD

2, S32–S38, 2002

Interactive Comment

increase in number density observed by the reviewer at a fixed altitude is due to the (slow) sedimentation of the mother cloud itself. It is intended to show in the revised version profiles of particle number density at fixed times and with a height axis relative to the base of the mother cloud.

p.34, l.8: Dependence on n and radius

Reply: We assume the mother cloud to be in thermodynamic equilibrium, hence the particle radius is fixed when SNAT, number-density and HNO3 mixing ratio are given. These equilibrium radii will be given in the revised version.

p.35, l.18: Conclusions

Reply: The study 'reveals' that sedimentation from mother clouds can lead to NAT-rocks, i.e. that the presence of mother clouds (and S-NAT > 1 below the cloud) is a sufficient condition for NAT-rocks. The slightly changed first sentence (see 'General Comments') should make this clear. As already pointed out in the 'General Comments' we do not agree that what is basically shown is that sedimenting particles can grow to NAT-rock sizes. Rather we show what mechanisms control the sedimentation and that the particle number density turns out to be $\approx 10^{-4} cm^{-3}$ independently of particle number density and size distribution in the mother cloud.

Whether a theoretical model is not able to prove anything is a debate of its own. However, we think that the finding that the mechanism leads to NAT-rocks with $n \approx 10^{-4} cm^{-3}$ independent of:

- particle number densities in the mother cloud

- particle size distribution in the mother cloud

- particle shape

- temperature and gas phase profiles (as long as S-NAT > 1 below mother cloud)

- Gas phase and eddy diffusion

ACPD

2, S32–S38, 2002

Interactive Comment

is a very strong argument that the process also takes place in the 'real' stratosphere.

Technical corrections:

p.35, l.12: Change 'no' to 'not'

Reply: Will be done.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 29, 2002.

ACPD

2, S32–S38, 2002

Interactive Comment