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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Brightening of the global cloud field by nitric acid and the associated radiative forcing" *by* R. Makkonen et al.

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We thank reviewer 1 for the useful comments and suggestions. Our detailed responses are as follows (reviewer comments in bold):

As the forcing estimate of nitrate acid gas is quite large here (0.32 W/m2 for cloud albedo effect, and -0.46 W/m2 for cloud lifetime effects), it is important to put these numbers in context. Here are three things that need further clarification in the manuscript:

i). How do nitrate acid gas concentrations used in this study compare with observations and other models? Off-line monthly nitrate acid gas concentrations are taken from ECHAM5-MOZ, as documented in Rast et al. (2012). Unfortunately,

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Rast et al. (2012) is still in preparation. So it is important to provide the information regarding these offline nitrate acid gas dataset, such as global burden, how they are compared with other model studies, how do surface concentrations compared with observations.

Rast et al. (2012) will be made available soon. The chemistry scheme of the simulations used to obtain the HNO_3 fields is the same as it was used by Horowitz et al. (2003), which includes a comparison of HNO_3 concentrations to observations. Over 15 (14) measurement campaigns we get at altitudes of 500hPa (900hPa) arithmetic mean biases of 75% (124%) respectively. These measurement campaigns are (PEM-West-A, ABLE-3B etc.) are giving representative values for the respective regions. Since there is no nitrate aerosol formation in this simulation, HNO_3 is just removed by wet and dry deposition and chemical reactions (photolysis and reaction with OH). Wet deposition may be on the low side, making HNO_3 concentrations particularly high. Nevertheless, the sources of HNO_3 are spatially highly unevenly distributed and the full coupling of reactions and transport influences the HNO_3 burden. Although the comparison of NO_2 column density with satellite data shows a generally good agreement, NO_2 column density is overestimated in some industrialized regions of Asia (but underestimated over the China sea e.g.) and Europe but underestimated in some regions of the US.

Furthermore, we can say that the profiles of the measurement campaigns are generally well matched by the model in shape although too high values at around 800hPa are present almost everywhere. The applied nitric acid fields are quantitatively similar to those in Xu and Penner (2012), with global average surface concentrations of 165 pptv and 174 pptv in this study and Xu and Penner (2012), respectively. Surface HNO_3 concentrations over remote oceans are slightly higher in ECHAM5-MOZ, while Xu and Penner (2012) show generally higher concentrations over continents. We will add a sentence about the bias in HNO_3 concentrations to the revised manuscript, and a detailed comparison to several campaing observations. We added the following reference

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under which the report about the RETRO run will appear from which the HNO_3 fields are taken.

ii) Uncertainty in the parameterizaiton of Romakkaniemi et al. (2005). The nitrate effect on droplet activation is based on Romakkaniemi et al. (2005). What is the uncertainty in this parameterization? Is there any other parameterizations to treat the effects of nitrate acid gas on droplet activation?

It is difficult to estimate the absolute accuracy of the HNO_3 parameterization. The parameterization is based on an extensive set of cloud parcel model simulations that should cover the range of aerosol size distributions and nitric acid concentrations present in the global simulation with the limitations that only two modes can be included (soluble Aitken and accumulation modes in our case). The parameterization is fitted into the parcel model data so that the the relative error between simulation results and parameterization is minimized and thus, on average, it gives the correct result. In Romakkaniemi et al. (2005) it is shown that the agreement between parameterization and cloud parcel model simulations is really good in the conditions it is validated. Xu and Penner (2012) used a simple method to redistribute the gas-phase nitric acid to fine-mode aerosols and calculated aerosol activation after this redistribution.

iii) The anthropogenic aerosol indirect forcing other than nitrate acid gas in ECHAM5-HAM2. Here I wonder how the indirect forcing of nitrate acid gas compared with the other anthropogenic aerosol indirect forcing from the same model (present-day aerosols – preindustrial aerosols, without nitrate in ECHAM5-HAM2). A table to compare the indirect forcing of nitrate acid gas with that of anthropogenic aerosol indirect forcing as calculated by the ECHAM5-HAM2 will be desirable.

In the revised manuscript, we have included the anthropogenic indirect forcing calculated with ECHAM5.5-HAM2.

p. 5229, line 26: how about coarse mode? The effect can be small, but these may

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affect how much nitrate acid available for the finer mode.

The parameterization of Romakkaniemi et al. (2005) can only be applied using two modes of the aerosol size distribution. Over continents, the number concentration of coarse mode particles is relatively small. In marine conditions, coarse mode sea-salt particles decrease the effect of HNO_3 on CDNC. We have mentioned this in the revised manuscript.

p. 5230, line 9: Rast et al. (2012) is still in preparation. So it is important to include the basic information about nitrate acid here, such as burden, distributions, and how nitrate acid concentrations and burdens in this study compared with other published results?

Please see the first answer.

p. 5230, lines 19-27: cloud albedo forcing. Are CDNC used here directly diagnosed from the scheme of Abdul-Razzak and Ghan? If this is the case, please clarify this here.

First, the CDNC is calculated with the Abdul-Razzak and Ghan parameterisation. Second, the parameterisation of Romakkaniemi et al. (2005) is used to calculate an increase in CDNC due to HNO_3. Finally, the radiation code is ran two times (with these two CDNC values) to obtain the cloud albedo forcing. We have clarified the method in the revised manuscript.

p. 5231, line 10-16: I understand the argument here, but I still think it is better to provide the anthropogenic indirect effect of nitrate acid gas, but not just the total indirect effects of nitrate acid gas, as the anthropogenic indirect effect of nitrate acid gas will have broader impact.

We agree that it would be beneficial to extract the anthropogenic contribution the the indirect forcing, however, the RETRO-runs applied in this study do not have pre-industrial HNO_3 fields available. In this respect, the paper by Xu and Penner (2012) gives some

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indication of the anthropogenic fraction of the effect: their present-day gas+aerosol nitrate total effect is -0.23 W m-2 and the anthropogenic forcing is -0.9 W m-2. If our result would scale similarly, the anthropogenic forcing from nitric acid condensation would amount to -0.11 W m-2.

p. 5231, line 25-27: why does the enhancement in activation fraction increase with height? Also, are data shown in Figure 1 from the annual-mean value at each GCM grid?

Several factors influence the vertical distribution of the nitric acid effect. The soluble mass addition from nitric acid increases with height, relative to sulfate mass. At lower temperatures, nitric acid has more time to condense on particles, leading to increased effect on CDNC. As indicated in the figure caption, the data in Fig. 1 are 5-yr annual averages at each grid point.

Section 3.2: how about the anthropogenic aerosol indirect forcing from the preindustrial time to the present-day? This can help to put the number you get here into context. As the magnitude of aerosol indirect forcing can vary a lot from different models, the relative magnitude will be more meaningful.

In the revised manuscript, we have included a value of the aerosol indirect forcing from the same model setup to help putting the nitric acid effect into perspective. Using the same model setup, the anthropogenic aerosol forcing in the year 2000 amounts to -1.6 W m-2.

p. 5234, lines 18-19: I am not sure I understand this first part of this sentence ('even though : : : in rather polluted areas'. Specifically, I am not sure the logic between the first part of the sentence and the second part of the sentence ('the results indicate : : :').

We have rewritten the sentence in the revised manuscript.

p. 5228, line 22: remove 'other'.

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p. 5229, line 12: surface to the 10hPa

p. 5232, line 4: 'more that' ! 'more than'.

We have corrected these in the revised manuscript.

References:

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