

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

Anonymous Referee #2

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This paper aims at quantifying the indirect aerosol effect caused by nitric acid and corresponding nitrate using a global climate model with explicit aerosol treatment and a two moment cloud microphysics taking the aerosol–cloud interactions into account. The incorporation of the effects of nitrate is performed via a parameterization derived from a cloud parcel model.

This is the main weakness of the paper. It is explicitly stated that nitrate aerosols are not treated at all. The modifications of the activated cloud droplet number only depends on the parameterization, which mainly considers the change in maximum supersaturation mimicking modifications of the hygroscopicity and aerosol mass due to condensation of gaseous nitric acid during droplet formation.

However, an underlying change in the aerosol distribution is not considered at all. Fur-

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thermore, the nitric acid concentrations are taken from a monthly climatology (without any evidence of how realistic the used HNO_3 distributions are), since the corresponding paper describing the nitric acid distribution is not (yet) available.

It has been shown before, that aerosol nitrate has a considerable influence on the direct aerosol effect (e.g. Bauer et al, ACP, 2007). Since the radiative properties of nitrate are not so different from those of sulphate, the resulting differences cannot be only an effect of additional uptake of nitric acid, but a substantial modification of the aerosol size distribution due to the additional aerosol mass.

Aerosol nitrate is known to be semivolatile; consequently using the total nitric acid as a proxy does not resemble realistically the aerosol fraction of nitrate. Due the diurnal temperature cycle the aerosol fraction in especially warm climate regions is lowest during the day and consequently the effect on clouds is lower during the day than during the night. Since the short wave spectrum is dominating the cloud forcing, the effect of nitrate is relatively low under those conditions.

On the other hand, the gaseous HNO_3 concentrations mostly increase during the day due to the photochemical production, which cannot be resembled by monthly mean concentrations. Consequently the monthly mean concentrations applied in this study are not a realistic representation of the effects of nitrate.

Additionally, the nitrate aerosol fraction is known to substantially depend on the chemical composition of the aerosol, e.g. in marine environment NaNO_3 can be formed, forcing a release of HCl into the gas phase as an acid replacement process. However, since NaNO_3 has a lower hygroscopicity than NaCl the activation potential of the aerosol should decrease, which cannot be reproduced by the applied parameterisation. On the other hand, if no replacement reactions take place additional mass (mostly in form mostly of NH_4NO_3) is added to the aerosol distribution. This aerosol mass (due to condensation on existing particles) alters the size distribution, affecting coagulation and other microphysical aerosol properties similarly (such as conversion of hydropho-

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bic to hydrophilic material via coating) as the aerosol hygroscopicity (and subsequent water uptake) and therefore the ambient aerosol size. Both of these effects can play a significant role in the activation process of the total aerosol. However, the amount of nitrate formed does not only depend on the available nitric acid, but also on other neutralising compounds, mainly NH_3 . Consequently, a scheme not taking the ammonia concentration into account is not able to reproduce a realistic nitric acid effect.

Even though the parameterization tries to mimick some of the effects it is not evident, if all of those processes are properly taken into account or if e.g. the temperature is only used as a measure for the change of HNO_3 solubility. Since neither in this paper nor the parameterization derivation by Romakkaniemi et al. (ACP, 2005) it is shown how well the parameterization reproduces "real conditions" from either chamber or field measurements, the quality of the numerically derived theoretical considerations is ambiguous.

The parameterization is able to reproduce the fact that HNO_3 is highly soluble and will be taken up by the activated cloud droplets; this dissolution will occur mainly after the activation process and more during the droplet growth due to diffusional limitations, such that even though a reduction in the critical supersaturation is achieved, this mainly occurs after the droplets have reached the critical radius and will anyhow continue to grow.

After obtaining the modified cloud droplet numbers, this paper is a consistent study using state of the art techniques to analyse the indirect effects. The obtained numbers of modifications of the indirect effect are mainly within the order of uncertainty associated with the total indirect effect (based on the multi-model spread from model intercomparison studies).

The fact that the importance of aerosol nitrate will potentially increase in future scenarios is realistic, equally the assumption that it will replace some of the ammonium-sulphate. However, the quantification of this effect is again difficult without taking the

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aerosol interactions of nitrate into account. Furthermore, to apply for the future conditions based on an emission scenario the HNO_3 field should be consistent (modified by the different NO_x emissions and the altered oxidation capacity of the atmosphere). The reduction in the NO_x emissions in the scenario will lead to a smaller aerosol load; the increase in the activated fraction is unlikely to compensate the reduced aerosol in such a way that the indirect aerosol effect by nitrate is even to increase. The only reason for this can be a more efficient activation of partly organic aerosol particles compared to a case without any activation modification by nitrate. This underlines the potential importance of aerosol nitrate, but does not increase the indirect effect; only the disturbance by nitric acid becomes more significant.

Consequently, even though the study is performed well with respect to simulation setup and scientific question investigated, I cannot recommend publication due the severe weaknesses in the methodology. In my opinion the scientific answers obtained from this study are flawed by the strong limitations of effects being included with the presented approach. Since this study is one of the first actually providing quantifying numbers of the indirect aerosol effect of nitrate, wrong assumptions and strong limitations will substantially influence follow-up studies. The presented approach leaves out too many effects to actually quantify the indirect aerosol effect of nitric acid properly. Therefore, the answers provided by this study are a quantification of the indirect aerosol with heavy restrictions, such that the obtained values are not representing the realistic state of the atmosphere. As the authors state themselves in their conclusions such a study should be performed with a coupled aerosol chemistry climate model, treating aerosol nitrate explicitly.

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