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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Summertime nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer monsoon region" *by* Y. Gu and H. Liao

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This study investigates the spatial distribution of nitrate aerosols in the UTLS over the Tibetan Plateau and the South Asian summer monsoon (TP/SASM) region using the GEOS-Chem model. With GEOS-Chem, the authors simulate elevated concentrations of summertime aerosols in the UTLS over the TP/SASM region, confirming the findings of previous observation and model studies. In contrast to previous model studies, which generally emphases the importance of sulfate, they argue that nitrate aerosol is the most dominant aerosol species in the UTLS over the studied region. According to their model result, nitrate contributes more than half of the aerosol mass concentration

(PM2.5) at 100 hPa height. The issue addressed by this study is interesting and is within the scope of ACP. The manuscript is well written and organized, but the scientific quality of the current manuscript is not sufficiently high for publication. The authors may consider performing additional model analyses to provide stronger evidence to support their arguments.

The study is focused on aerosol formation in the UTLS, a region influenced by both tropospheric and stratospheric processes. In contrast to complete tropospheric chemistry, the simplification of stratospheric chemistry in GEOS-Chem and possible influence on HNO3 and nitrate aerosol formation in the UTLS is not well introduced in the manuscript. It is stated that the chemical reaction rates in the stratosphere are taken from other model than GEOS-Chem (Line 190-195). The authors performed a 10-year spin-up run to generate the initial conditions (Line 239-241). How are the long-lived species like CFCs and N2O are treated in the model? Could the year 2005 stratospheric chemistry be simulated?

HNO3 concentrations from MLS are used for comparison with model results in this study. As shown in Fig. 4, however, no MLS data are available at 200 hPa and 100 hPa over most of the TP/SASM region. Surface concentrations of aerosols in the SASM region are also used for model evaluation, with normalized mean biases of +51.5% for NO3- and 74.9% for NH4+ in summertime. What are the causes of such large biases? Do such large biases have great impact on the simulation of nitrate in the UTLS, as upward transport from the lower troposphere is suggested to be a mechanism for high nitrate concentrations in the UTLS? Specifically, might the concentrations of nitrate aerosol in the UTLS be greatly overestimated as well?

With regards to the mechanisms for high nitrate concentrations in the UTLS, the investigation appears to be skin-deep without in-depth analysis of physical-chemical processes. For instance, what are the different chemical mechanisms for NO3- and SO4= formation that contribute to the differences in their vertical distributions (Line 509-513)? The possibility of the nitric acid trihydrate (NAT) formation in the investigated region is

discussed in this study (Line 555-566). However, while only the range of temperatures is referred, other chemical and physical conditions that are also important for the NAT formation (see Kirner et al., 2011) are not addressed. Could GEOS-Chem be used for the simulation of NAT?

In addition to the reaction of nitric acid with ammonia, as stated in the manuscript (Line 244-245), nitrate aerosol can also form by multi-phase chemistry including mineral and sea salt aerosols, even in the free troposphere over TP (Ma et al., 2003). Previous studies showed that mineral dust could make a considerable contribution to elevated summertime aerosol concentrations in the UTLS over the TP/SASM region (Fadnavis et al., 2013). While it is stated that mineral dust aerosols are included in GEOS-Chem (Line 177-181), they are neither presented nor discussed in the manuscript.

In this study simulated O3 profiles are compared with balloon-borne sonde measurements at Kunming in August 2009 and at Lhasa in August 2010 (337-350). How frequent were O3 profiles measured at these two sites? Are there sufficient measured O3 profiles for calculating the monthly mean values for comparison?

Also for model evaluation, the observed PM10 concentrations were converted to PM2.5, following the suggestions of Zhang et al. (2002) (Line 398-399). Is that work of Zhang et al. (2002) related to PM2.5 and PM10 in the TP/SASM region? The authors are suggested to consider referring to measurement work in the investigated region.

With respect to the hygroscopic growth of aerosols (432-434), are the growth factors calculated on-line for internal-mixed aerosols in GEOS-Chem? It is stated that the region with relatively high aerosols extinction coefficients (Fig. 9) corresponds to that with high PM2.5 concentrations at 100 hPa (Fig.7) (Line 435-438). This statement might not be true if one would compare the two plots carefully.

Line175: 34 layers in the troposphere?

Line 279: Fig. 4(a) and Fig. 4(b)?

Line 529: Fig. 12(c)?

There are several literatures listed in the References but not cited in the formal text.

References

Fadnavis, S., Semeniuk, K., Pozzoli, L., Schultz, M. G., Ghude, S. D., Das, S., and Kakatkar, R.: Transport of aerosols into the UTLS and their impact on the Asian monsoon region as seen in a global model simulation, Atmos. Chem. Phys., 13, 8771-8786, 10.5194/acp-13-8771-2013, 2013.

Kirner, O., Ruhnke, R., Buchholz-Dietsch, J., Jöckel, P., Brühl, C., and Steil, B.: Simulation of polar stratospheric clouds in the chemistry-climate-model EMAC via the submodel PSC, Geosci. Model Dev., 4, 169-182, 10.5194/gmd-4-169-2011, 2011.

Ma, J., Tang, J., Li, S.-M., and Jacobson, M. Z.: Size distributions of ionic aerosols measured at Waliguan Observatory: Implication for nitrate gas-to-particle transfer processes in the free troposphere, J. Geophys. Res., 108, 4541, 10.1029/2002jd003356, 2003.

Zhang, X., Cao, J., Li, L., Arimoto, R., Cheng, Y., Huebert, B., and Wang, D.: Characterization of atmospheric aerosol over Xian in the south margin of the Loess Plateau, China, Atmos.Environ., 36, 4189–4199, 2002.

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