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Responses to Reviewer #2

General Comments: 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 ($PM_{2.5}$) 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.

Responses to general comments:

We have performed additional sensitivity studies to examine the uncertainties associated with simulated nitrate in the UTLS. Anthropogenic emissions of NO_x, NH_3, and SO_2 are changed according to the NMBs of the simulated concentrations of sulfate and nitrate. These sensitivity studies all confirm that nitrate aerosol is a dominant aerosol species in the UTLS, which are presented in our new Section 7 of “Impacts of uncertainties in surface-layer aerosol concentrations on simulated nitrate in the UTLS”.

Our point-to-point responses to the reviewer’s comments are listed below.

Specific Comments:

1. 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 HNO_3 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 N_2O are treated in the model? Could the year 2005 stratospheric chemistry be simulated?

Response:

(1) Both CFCs and N_2O are considered in the linearized stratospheric chemistry in the GEOS-Chem model, and their concentrations are updated at

the end of each chemistry time step by using the archived three-dimensional monthly mean production rates and loss frequencies from the NASA Global Modeling Initiative (GMI) Combo simulations. More details are described at http://wiki.seas.harvard.edu/geos-chem/index.php/Stratospheric_chemistry and in Murray et al. (2012).

We have revised the description on stratospheric chemistry in Section 2.1:" The monthly mean production rates and loss frequencies of other stratospheric species (including long-lived species such as CFCs and N_2O) use those from NASA Global Modeling Initiative (GMI) Combo simulations (Duncan et al., 2007; Considine et al., 2008; Murray et al., 2012)."

(2) The archived three-dimensional monthly mean production rates and loss frequencies in the stratosphere are the averages over years of 2004–2010 (http://wiki.seas.harvard.edu/geos-chem/index.php/Stratospheric_chemistry). Although we use the emissions and meteorological fields of year 2005 in our simulations, we would consider that the tropospheric simulation can be representative of year 2005 but stratosphere simulation should represent a multi-year average. This is now clarified in Section 2.2 where we describe our numerical experiments.

2. HNO_3 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 $\mathrm{NO_3^-}$ and 74.9% for $\mathrm{NH_4^+}$ 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?

Response:

(1) Since dense high clouds were observed to locate in the upper troposphere over the Tibetan Plateau (Li et al., 2005), most cloud-contaminated measurements were screened out for MLS v4.2 datasets as described in Lively et al. (2015). As a result, no MLS data are available at 200 hPa and 100 hPa over a large fraction of the TP/SASM region. To have more datasets for comparisons between the simulations and the observations, we have replaced MLS 4.2 datasets with MLS v3.3 datasets, which have been widely used in previous studies (Livesey et al., 2011; Miyazaki et al., 2012; Fiorucci et al., 2013; Schwartz et al., 2015; Yan et al., 2015). The MLS v3.3 measurements are available in about 60% of the grid cells at 200 hPa and in about 95% of the grid cells at 100 hPa over the TP/SASM region (70–105°E, 10–40°N). We have added the following sentence for HNO_3 in the third paragraph of Section 3.1: "Considering all the grid cells with MLS HNO_3 data available, the simulated seasonal mean HNO_3 concentrations show

normalized mean bias (NMB) of +15.9% at 100 hPa over the TP/SASM region in summer of year 2005.”

(2) As noted in Section 4.2 of the manuscript, the reasons for the large NMBs of $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ include (a) the measurements of $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ are quite limited in terms of the number of samples, and (b) the mismatch of the model year 2005 with the years of 1992–2010 with observations available.

(3) We have performed four new sensitivity studies to examine the impacts of uncertainties in surface-layer aerosol concentrations on simulated nitrate in the UTLS. In the first three cases, anthropogenic emissions of NO_x, NH_3, and SO_2 in Asia are changed by –50%, –50%, and +20%, respectively, relative to those in our standard simulation. In the last case, anthropogenic emissions of all these three species are changed simultaneously, with NO_x reduced by 50%, NH_3 reduced by 50%, and SO_2 increased by 20% in Asia relative to the standard simulation. All these sensitivity studies confirm that nitrate is the most dominant aerosol species in summertime in the UTLS over the TP/SASM region (see Table 3 below). See also our new Section 7 “Impacts of uncertainties in surface-layer aerosol concentrations on simulated nitrate in the UTLS” in the revised manuscript.

Table 3. Sensitivity simulations to examine the impacts of uncertainties in surface-layer aerosol concentrations on simulated NO_3^- in the UTLS. "Conc" and "Ctri" denote, respectively, simulated seasonal mean concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, BC and their contributions to $PM_{2.5}$ (in percentages) during summertime (June-August) of 2005. The mass concentrations are averaged over the TP/SASM region, with unit of μg m^{-3} at the surface layer and of 10^{-2} μg m^{-3} at 200 hPa and 100 hPa. Also shown are the NMBs, as the simulated surface-layer concentrations are compared with measurements described in Section 4.2.

Species	Baseline Case			SO_2 (+20%)			NO_x (–50%)			NH_3 (–50%)			All Change		
	Conc.	Ctri.	NMB	Conc.	Ctri.	NMB	Conc.	Ctri.	NMB	Conc.	Ctri.	NMB	Conc.	Ctri.	NMB
Surface															
SO_4^{2-}	1.70	35.9%	–14.7%	1.92	38.1%	–4.4%	1.58	39.5%	–18.1%	1.70	38.1%	–14.7%	1.78	43.2%	–8.3%
NO_3^-	0.94	19.8%	+51.5%	0.94	18.7%	+53.5%	0.50	12.5%	–11.7%	0.73	16.4%	+24.1%	0.39	9.5%	–27.0%
NH_4^+	0.85	18.1%	+74.9%	0.94	18.6%	+93.8%	0.68	17.1%	+44.1%	0.78	17.6%	+64.6%	0.71	17.3%	+55.4%
OC	0.94	19.8%	–57.2%	0.94	18.6%	–57.2%	0.94	23.4%	–57.2%	0.94	21.0%	–57.2%	0.94	22.7%	–57.2%
BC	0.30	6.4%	–32.2%	0.30	6.0%	–32.2%	0.30	7.5%	–32.2%	0.30	6.8%	–32.2%	0.30	7.3%	–32.2%
200 hPa															
SO_4^{2-}	3.27	20.2%		3.67	22.9%		3.31	20.6%		3.29	29.1%		3.74	33.7%	
NO_3^-	7.57	46.8%		7.05	43.9%		7.41	46.0%		3.86	34.2%		3.19	28.7%	
NH_4^+	2.67	16.5%		2.67	16.6%		2.71	16.8%		1.49	13.2%		1.50	13.5%	
OC	2.22	13.7%		2.22	13.8%		2.22	13.8%		2.22	19.7%		2.22	20.0%	
BC	0.44	2.7%		0.44	2.7%		0.44	2.7%		0.44	3.9%		0.44	4.0%	
100 hPa															
SO_4^{2-}	2.60	21.4%		2.80	23.0%		2.66	21.9%		2.60	25.2%		2.87	27.2%	
NO_3^-	6.90	56.8%		6.72	55.3%		6.81	56.1%		5.68	55.0%		5.62	53.3%	
NH_4^+	1.43	11.8%		1.43	11.7%		1.45	12.0%		0.84	8.1%		0.84	8.0%	
OC	1.05	8.6%		1.05	8.6%		1.05	8.7%		1.05	10.2%		1.05	10.0%	
BC	0.16	1.3%		0.16	1.3%		0.16	1.3%		0.16	1.6%		0.16	1.5%	

3. 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 SO42- 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?

Response:

(1) We have added in Section 6.1 the following detailed description on the chemical mechanisms for SO_4^{2-} and NO_3^- that contribute to the differences in their vertical distributions: "The chemical mechanisms for the formation of SO_4^{2-}, NO_3^-, and NH_4^+ aerosols in the GEOS-Chem model were described in R. J. Park et al. (2004), which are comprehensive and have been used extensively in previous studies to simulate these three aerosol species (R. J. Park et al., 2004; Pye et al., 2009; L. Zhang et al., 2010; Zhu et al., 2012; Jiang et al., 2013; Lou et al., 2014). Sulfate aerosol forms from gas-phase oxidation of SO_2 by OH and from in-cloud oxidation of SO_2 by O_3 and H_2O_2. Nitrate forms from the partitioning of HNO_3 between gas and aerosol phases, which is calculated by the ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes, 2007) in the GEOS-Chem model. Major reactions for the production and loss of HNO_3 were listed in Liao and Seinfeld (2005). HNO_3 is produced by the reaction of NO with OH during daytime and by hydrolysis of N_2O_5 on aerosol surfaces at night. The chemical mechanisms for SO_4^{2-} and NO_3^- have different sensitivity to meteorological conditions. During the vertical transport, temperature decreases, which reduces the gas-phase oxidation of SO_2 (Yao et al., 2002; Seinfeld and Pandis 2006; X. Y. Zhang et al., 2012) but promotes the formation of NO_3^- by shifting gas-particle equilibria (Dawson et al., 2007; Liao et al., 2009). Therefore the different chemical mechanisms for SO_4^{2-} and NO_3^- formation contribute to the differences in their vertical distributions."

(2) We have revised the description on the mechanism of NAT formation, in Section 6.2: "Previous studies have also reported that nitric acid trihydrates (NAT, $HNO_3 \bullet (H_2O)_3$) could form in the polar and tropical stratosphere at low temperatures through two mechanisms: (1) the homogeneous nucleation out of supercooled ternary solutions, and (2) the heterogeneous formation on ice particles (Hofmann et al., 1989; Carslaw et al., 1998; Voigt et al., 2000; Popp et al., 2006; Kirner et al., 2011). A typical NAT condensation temperature is approximate 193 K (Kirner et al., 2011)."

Currently the GEOS-Chem model does not have simulation of NAT. The balloon-borne measurements of depolarization ratio and backscattering ratio of aerosols at Lhasa during August-October of 1999 by Kim et al. (2003) and Tobo et al. (2007) suggested that coarse and aspherical particles such as NAT are scarce in the UTLS of the TP/SASM, so the missing of NAT mechanism in the GEOS-Chem model should not compromise the conclusion of our study. We have added a sentence to discuss NAT at the end of the conclusion section.

4. In addition to the reaction of nitric acid with ammonia, as stated in the manuscript (Line244-245), 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.

Response:

We agree with the reviewer that nitrate aerosol can also form by multi-phase chemistry on mineral and sea salt aerosols (Ma et al., 2003; Liao et al., 2004). The GEOS-Chem model uses the aerosol thermodynamic equilibrium module ISORROPIA to calculate concentrations of nitrate, which accounts for the formation of ammonium nitrate and the formation of nitrate on sea salt aerosol. However, the formation of nitrate on mineral dust aerosol is not considered in the publicly released versions of the GEOS-Chem model. Fadnavis et al. (2013) showed, by using the ECHAM5-HAMMOZ model, that mineral dust aerosol contributed to the enhanced aerosol extinction in the UTLS over the anticyclone region during summer. Their simulated seasonal mean concentrations of mineral dust were 0.010–0.029 μg m^{-3} at 110 hPa over the TP/SASM region in summer of 2003. The upper end of their simulated mineral dust concentration is close to the simulated SO_4^{2-} concentration at 100 hPa in our work. We have added a sentence at the end of the conclusion section to clarify that future studies need to account for mineral dust aerosol.

5. In this study simulated O_3 profiles are compared with balloon-borne sonde measurements at Kunming in August 2009 and at Lhasa in August 2010 (337-350). How frequent were O_3 profiles measured at these two sites? Are there sufficient measured O_3 profiles for calculating the monthly mean values for comparison?

Response:

The measurements used in the comparisons were 11 profiles of O_3 in Kunming over August 7–13, 2009, and 12 profiles of O_3 in Lhasa over August 22–28, 2010, which should be enough for calculating the monthly mean O_3 values. To help with our argument, we have added in Figure 6 the monthly mean O_3 profile obtained from daily observations in Kunming during August 12–31, 2012, and the monthly mean O_3 profile obtained from the daily observations in Lhasa during August 4–26, 2013. The updated Figure 6 is shown below.

We have revised Section 3.2 accordingly: "In addition to the comparisons against MLS products, the simulated O_3 profiles are compared with balloon-borne sonde measurements in Fig. 6. The measurements were carried out at Kunming (KM, 102.7°E, 25.0°N) in August of 2009 and 2012, and at Lhasa (LH, 91.1°E, 29.7°N) in August of 2010 and 2013. The uncertainties of the observed O_3 mixing ratios were estimated to be within 5–10% (Bian et al. 2012). The comparisons with multi-year observations show that the model can reproduce the vertical distributions of O_3 in Kunming and Lhasa. At 100 hPa, the simulated monthly mean O_3 mixing ratio in KM is 112.6 ppbv, and the observed value is 124.2 ppbv in 2009 and 113.5 ppbv in 2012. In LH, the simulated monthly O_3 mixing ratio at 100 hPa is 152.6 ppbv, and the observed O_3 mixing ratio at that altitude is 142.4 ppbv in 2010 and 167.9 ppbv in 2013. The magnitudes of O_3 mixing ratios from these balloon-borne sonde measurements support those from MLS; O_3 mixing ratios in the UTLS are less than 200 ppbv over the TP/SASM region."

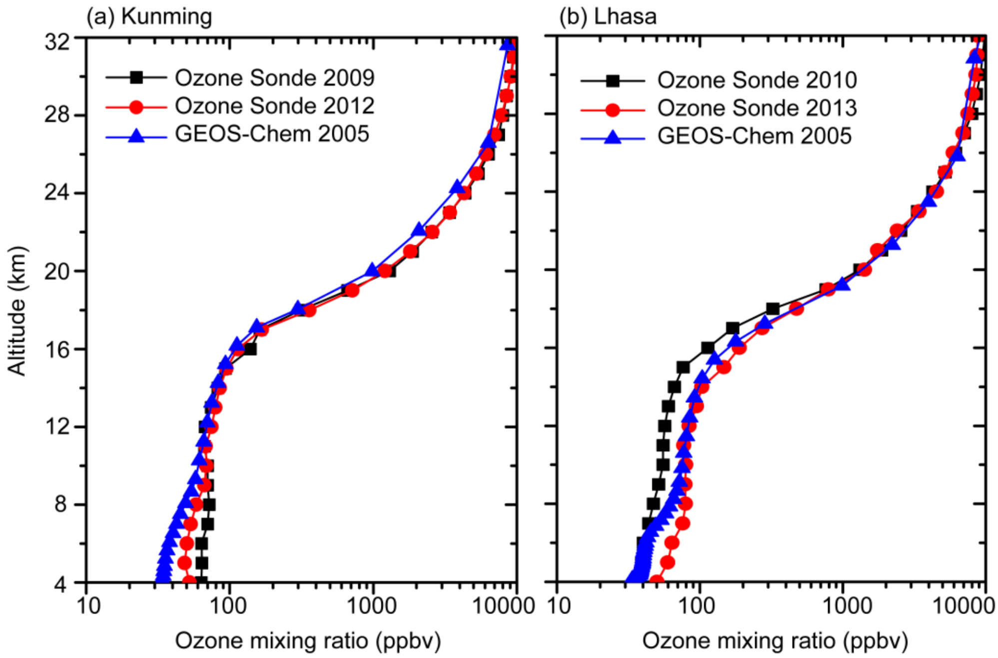

Figure 6. The simulated and observed vertical profiles of monthly mean O_3 mixing ratios at (a) Kunming and (b) Lhasa in August. The model results are from the simulation of year 2005. The observations in Kunming were conducted during August 7–13 (11 profiles of O_3 collected) in 2009 and during August 12–31 in 2012 (daily observations). The observations in Lhasa were conducted during August 22–28 in 2010 (12 profiles of O_3 collected) and during August 4–26 in 2013 (daily observations).

6. Also for model evaluation, the observed PM_{10} concentrations were converted to $PM_{2.5}$, following the suggestions of Zhang et al. (2002) (Line 398-399). Is that work of Zhang et al. (2002) related to $PM_{2.5}$ and PM_{10} in the TP/SASM region? The authors are suggested to consider referring to

measurement work in the investigated region.

Response:

The observations in Zhang et al. (2002) were conducted in Xi'an in northwestern China. Chatterjee et al. (2010) measured aerosol concentrations in Darjeeling, a station in northeastern Himalayas, during January-December of 2005, and reported that fine aerosol ($PM_{2.5}$) concentration accounted for 58.3% of PM_{10} concentration on average. Therefore it is OK to use the factor of 0.6 from Zhang et al. (2002) to convert PM_{10} to $PM_{2.5}$ for the TP/SASM region. We have revised this sentence as: "The observed PM_{10} concentrations listed in Table S1 are multiplied by 0.6 to convert to $PM_{2.5}$ for model evaluation, following the suggestions in Zhang et al. (2002) and Chatterjee et al. (2010)."

7. 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 $PM_{2.5}$ concentrations at 100 hPa (Fig.7) (Line 435-438). This statement might not be true if one would compare the two plots carefully.

Response:

(1) Yes. It is clarified in Section 4.3: "The hygroscopic growth of each aerosol species with relative humidity is accounted for, using the hygroscopic growth factors listed in Martin et al. (2003)".

(2) Thanks for pointing this out. At 100 hPa, the simulated $PM_{2.5}$ concentrations are high over the anticyclone region, with peak values near the south slop of the Himalayas. The simulated aerosol extinction coefficients are also high over the anticyclone region, but the peak values extend to the Arabian Peninsula and the nearby ocean. The reasons for the differences are as follows. While simulated $PM_{2.5}$ considers only sulfate, nitrate, ammonium, OC and BC aerosols, simulated aerosol extinction coefficient accounts for the contributions of sulfate, nitrate, ammonium, OC, sea salt, and mineral dust to compare with SAGEII measurements. Mineral dust aerosol has been shown to contribute largely to elevated summertime aerosols in the UTLS over the Asian summer monsoon region, especially over the Arabian Peninsula and the nearby ocean (Fadnavis et al., 2013). We have revised the sentence as: "At 100 hPa, the simulated aerosol extinction coefficients are relatively high over the anticyclone region, where anthropogenic aerosol species (Fig. 7) and natural aerosols such as mineral dust and sea salt contribute to aerosol extinction coefficients in summer".

8. Line175: 34 layers in the troposphere?

Response:

Yes.

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

Response:

We have changed “Fig. 3(a) and Fig. 3(b)” to “Fig. 4(a) and Fig. 4(b)”.

10. Line 529: Fig. 12(c)?

Response:

We have changed “Fig. 13(c)” to “Fig. 12(c)”.

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

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Response:

We have included these studies in our reference list.

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