Manuscript # ACP-2015-793

Responses to Referee #3

Before a publication some major points must be addressed (and modified in the manuscript accordingly):

The authors have partially responded to many concerns in their authors replies, but have not substantially modified the manuscript. Therefore, the paper still not very much improved to readers not analysing all the discussion concerns and responses.

The most prominent example is the role of chemical production to the nitrate formation. In the current manuscript version, this is still not well discussed and gas phase HNO3 production and gas aerosol partitioning are not treated separately, such that it cannot be concluded properly whether the meteorological influences on the chemical reaction system or the altered thermodynamical stability of NH4NO3 in the UT dominate the importance of nitrate over the TP. This could be elucidated by e.g. analysing the ratio of particulate nitrate to total oxidised nitrogen (N(V), i.e. gaseous HNO3, N2O5, NO3, and particulate nitrate).

Response:

(1) Following the Reviewer's comments, we have calculated the net chemical production of HNO_3 by gas-phase reactions and heterogeneous reactions (chemical production by reactions R1-R23 minus chemical loss by reactions R24-R25) (new Table 3 in our manuscript, see below). The vertical profiles of net chemical production of HNO_3 and nitrate formation from gas aerosol partitioning over the TP/SASM region during summertime of year 2005 are shown in Figure 12 (see below) of our manuscript. While the net chemical production of HNO_3 has an overall trend of decreasing with altitude, nitrate formation from gas to aerosol conversion of HNO_3 peaks between 100–300 hPa. The magnitude of gas to aerosol conversion of HNO_3 is several times higher than the net chemical production of HNO_3, indicating that the gas-aerosol partitioning plays a dominant role in the enhancement of nitrate in the UTLS. A new Sec. 6.1 has been added into the manuscript to discuss this.

Table 3: List of gas-phase reactions and heterogeneous reactions involve HNO_3 in the GEOS-Chem model (Version 9-01-03)

	Reactants		Products
	Chemical productions		
R1	$NO_2 + OH + M$	→	$HNO_3 + M$
R2	$NO_3 + CH_2O$	→	HNO_3 + prod.
R3	$ALD_2 + NO_3$	→	HNO_3 + prod.
R4	$RIO_1 + NO$	→	HNO_3 + prod.
R5	$IAO_2 + NO$	→	$0.08HNO_3$+ prod.
R6	$ISN_1 + NO$	→	$0.05HNO_3$ + prod.
R7	$VRO_2 + NO$	→	HNO_3 + prod.
R8	$MRO_2 + NO$	→	HNO_3 + prod.

R9	INO_2+NO	→	$0.85HNO_3$+ prod.
R10	$ALK_4 + NO_3$	→	HNO_3 + prod.
R11	RCHO + NO_3	→	HNO_3 + prod.
R12	MEK + NO_3	→	HNO_3 + prod.
R13	$INO_2 + MO_2$	→	$0.425HNO_3$ + prod.
R14	GLYX + NO_3	→	HNO_3 + prod.
R15	MGLY + NO_3	→	HNO_3 + prod.
R16	MACR + NO_3	→	HNO_3 + prod.
R17	$C_2H_6 + NO_3$	→	HNO_3 + prod
R18	$INO_2 + MCO_3$	→	$0.85HNO_3$ + prod.
R19	NO_2 + (aerosol)	→	$0.5HNO_3$+ prod.
R20	NO_3 + (aerosol)	→	HNO_3 + prod.
R21	N_2O_5 + (aerosol)	→	$2HNO_3$
R22	DMS + NO_3	→	HNO_3 + prod.
R23	$BrNO_3 + H_2O$(l,s)	→	HNO_3 + prod.
		Chemical loss	
R24	HNO_3 + OH	→	$H_2O + NO_3$
R25	HNO_3 + hv	→	$OH + NO_2$

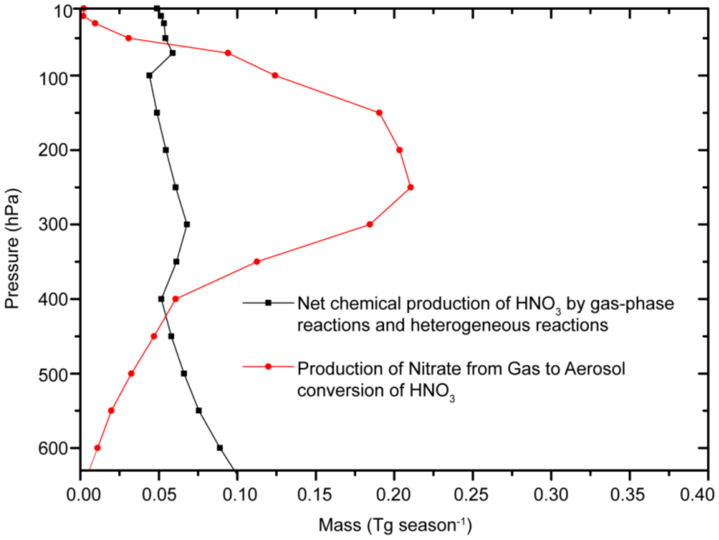

Figure 12. The net chemical production of HNO_3 by gas-phase reactions and heterogeneous reactions (black dotted line) and the production of nitrate from gas to aerosol conversion of HNO_3 (red dotted line) over the TP/SASM region during summertime of year 2005.

(2) The Reviewer suggests to analyze the ratio of particulate nitrate to total oxidised nitrogen (N(V), i.e. gaseous HNO_3, N_2O_5, NO_3, and particulate nitrate). Such ratio cannot answer the question raised by the reviewer with respect to whether the meteorological impacts on the chemical reaction system or on the thermodynamical stability of NH_4NO_3 dominate the enhancement of nitrate concentration over the TP, because the TP region isn't a black box for chemical reactions. The ratio of particulate nitrate to total oxidised nitrogen varies with grid cells and the concentrations of nitrate and the total N(V) are influenced by vertical and horizontal transport, dry and wet deposition, temperature, humidity,

pressure, light but also the concentrations of various precursors.

Furthermore, upward transport is still one of the dominant processes according to the authors. Even though they mention deep convection, they use ECMWF vertical winds for displaying ascent of air masses, which does not represent convective activity. And as deep convection is mostly associated with precipitation, HNO3 and aerosol particles would be efficiently removed. This might also contribute to the underestimated OC and BC concentrations in the simulations for the UT, i.e. too strong wet removal. Therefore, such a strong contribution to UTLS nitrate from the lower tropopause appears unlikely to the reviewer. It would be much more easy to explain the enhanced simulated nitrate by the vertical transport acting on low solubility compounds such as NOx, which are then converted in the UTLS by chemical reactions into N(V).

Response:

(1) The upward transport associated with the deep convection over the TP/SASM during the Asian summer monsoon period has been presented in a lot of previous studies, evidenced by both satellite observations (Fu et al., 2006; Luo et al., 2011) and the calculation of the outgoing long-wave radiation which is a proxy for convection over the region (Randel and Park, 2006; Park, et al., 2007; Fadnavis et al., 2013). The ECMWF vertical winds are used to show upward transport that is associated with the deep convection (Randel et al., 2010; Bian et al., 2011; Fadnavis et al., 2013, 2014; Qie et al., 2014; He et al., 2014).

(2) Deep convection over the TP/SASM region consists of both precipitating and nonprecipitating segments, as observed by CALIPSO during summertime (Luo et al., 2011). Previous studies used both phrases of “deep convection” and “vertical transport” to describe the transport of chemical species over the TP/SASM region. For example, here are some original sentences from the literature:

> In the abstract of Fadnavis et al. (2013): “The simulations show persistent maxima in black carbon, organic carbon, sulfate, and mineral dust aerosols within the anticyclone in the UTLS throughout the ASM (period from July to September), when **convective** activity over the Indian subcontinent is highest, indicating that boundary layer aerosol pollution is the source of this UTLS aerosol layer. The simulations identify **deep convection** and the associated heat-driven circulation over the southern flanks of the Himalayas as the dominant **transport** pathway of aerosols and water vapour into the tropical tropopause layer (TTL).”
>
> In the third paragraph of Sec. 3 in Fadnavis et al. (2013): “… large-scale **vertical transport** within the anticyclone is related to **deep monsoon convection** over the region 10–35°N, 60–120°E.”
>
> In the first paragraph of Sec. 5 in Fadnavis et al. (2013): “Model simulations indicate the **transport** of boundary layer aerosol pollutant by **ASM convection** to the UTLS.”
>
> In the first paragraph of Randel et al. (2010): “The anticyclone is a region of persistent enhanced pollution in the upper troposphere during boreal summer, linked to **rapid vertical transport** of surface air from

> Asia, India, and Indonesia in **deep convection**, and confinement by the strong anticyclonic circulation."

Although precipitation associated with deep convection can wash out a fraction of soluble species, enhanced concentrations of soluble compounds have been observed over the Asian monsoon region during boreal summer, as a result of the rapid vertical transport of pollutants from the surface. For example, hydrogen cyanide (HCN), a water soluble pollutant emitted from biomass and biofuel burning, was reported to have a maximum concentration in the UTLS by observations of the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) satellite instrument (Randel et al., 2010). Vernier et al. (2011) observed, by CALIPSO lidar measurements, a layer of aerosols at the tropopause, which extended horizontally from the eastern Mediterranean to western China and vertically from 13 to 18 km during the Asian monsoon season. Fadnavis et al. (2013) found that aerosols (sulfate, OC, BC and mineral dust) showed high values within the anticyclone region during the summer of year 2003.

The wet deposition of both gas-phase species and aerosols has been implemented in the GEOS-Chem model (Liu et al., 2001). It accounts for scavenging in convective updrafts, rainout and washout from convective anvils and large-scale precipitation, and allows for return of chemical species to the atmosphere following evaporation. For the scavenging of aerosols, $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, and hydrophilic OC and hydrophilic BC aerosols are assumed to be fully soluble. The model also includes soluble gases on the basis of their effective Henry's law partitioning in warm clouds, and surface coating or cocondensation of ice crystals in cold clouds (Park et al., 2004). The GEOS-Chem model has also been widely applied in simulating HNO_3 and aerosols in upper troposphere (Li et al., 2005; Murray et al., 2012; Eastham et al., 2014), which indicating that the model has the ability to investigate the issue addressed in this study.

(3) It is suggested by the Reviewer that the enhanced simulated nitrate may be explained by the vertical transport of low solubility compounds such as NO_x, which are then converted in the UTLS by chemical reactions into N(V). However, model results do not support this assumption. As shown by Fig. 12 in our response to your first comment, net chemical production does not show a maximum in the UTLS. Furthermore, as discussed in Sec. 7 of our manuscript, nitrate concentrations in the UTLS over the TP/SASM region are most sensitive to changes in NH_3 and show small sensitivity to NO_x emissions during summertime.

The treatment of aerosol species as an external mixture for the extinction calculation is not very reasonable as most aerosol particles containing nitrate will be in the form of solution doplets – therefore the contribution from nitrate cannot such easily be estimated as stated by the authors and their result can only be considered as a rough approximation.

Response:

We agree with the Reviewer that the treatment of aerosol species as an external mixture may lead to uncertainties in simulated aerosol extinction coefficients, but such treatment should not compromise conclusions from this work. The water uptake by aerosols is accounted for as described in Martin et al. (2003) and Drury et al. (2010). The ability of the GEOS-Chem model in simulating aerosol optical depth (AOD) and aerosol extinction coefficients has been evaluated by a number of observational studies (Drury et al., 2010; Pierro et al., 2011; Johnson et al., 2012; Zhang et al., 2013; Kim et al., 2015); simulated aerosol optical properties agreed fairly well with ground-based, aircraft, and space-borne observations in the United States, Europe and Asia.

The authors mention that MLS data is assumed to have uncertainties which are of the same magnitude as the actual values – therefore a comparison of HNO3 values of the magnitude of the uncertainty limits is of reduced value. The MLS data is much more trustful in the stratosphere, where higher mixing ratios are found. The questioned aspect of vertical resolution of the data (i.e. 3 to 4 km) is not discussed at all, such that especially at 100hPa stratospheric air masses can influence the observations.

Did the authors use the same averaging kernels for the model data as the MLS retrieval? If not, this alone makes it difficult to compare model results with satellite data and the authors did not mention how the model-observation comparison is done in detail.

The geographical distribution of the MLS data with simulated HNO3 does not provide a good pattern correlation, except for the main features, which are characterised by the differentation of tropospheric and stratospheric air masses, e.g., there is a simulated minimum over the Gulf of Bengal which does not show up in the data. For the 200 hPa data over the TP there is hardly any data.

Response:

(1) The uncertainty in MLS data cited in the manuscript was single profile precision obtained by comparisons with ground-based, balloon-borne, and satellite measurements, which may not be representative of the precision of profiles averaged over summertime and over the TP/SASM. For the cases that HNO_3 abundances are smaller than the single-profile precision, precision can be improved by averaging, with the precision of an average of N profiles being $1/\sqrt{N}$ times the precision of an individual profile (Livesey et al., 2011). In our manuscript, the observed HNO_3 concentrations used in the comparisons were seasonal and regional averages, which should have much smaller uncertainties than the single-profile precision.

We agree with the Reviewer that the coarse vertical resolution might limit the use of the MLS data to obtain information on examining the vertical gradients in the UTLS. Following the suggestion of the Reviewer, we have added the following discussions in Sect. 3 of the manuscript: "The MLS data HNO_3 product at 100 hPa represents an average of a 3–4 km layer reaching from below the tropopause layer into the lower stratosphere (Duncan et al., 2007).

Currently, the UTLS datasets of MLS are still considered to be valuable in evaluating model results (Ducan et al., 2007; Miyazaki et al., 2012; Brakebusch et al., 2013; Liu et al., 2013) despite the limitations of the MLS data product.

(2) We didn’t apply the averaging kernels to model results in the previous version of the manuscript, because the instrument’s vertical resolution is reasonably comparable to the model grid in the upper troposphere and the lower stratosphere (Miyazaki et al., 2012).We did linear interpolation to convert simulated HNO_3 and O_3 dataset to the MLS vertical grid as suggested in Livesey et al. (2011).

In the revised version, we have applied the averaging kernels to model results to address the concern of the Reviewer. The new Figure 4 and Figure 5 are shown below and the comparisons have been updated in Sec. 3. The full MLS averaging kernels are complicated entities which are not available to the public (Livesey et al., 2011). Since the MLS averaging kernels typically change little with latitude/season/atmospheric state, the MLS group provides only two representative kernels, one for the equator and one for polar (70°N) which could be representative of profiles over other regions (Livesey et al., 2011). According to Santee et al. (2007), the variation in the averaging kernel of MLS is sufficiently small so that they are representative for all profiles. Therefore, we choose the averaging kernels for the equator, and the simulated HNO_3 and O_3 mixing ratios are converted to $\hat{\mathrm{x}}$, following the method of Livesey et al. (2011).

$$\hat{\mathrm{x}} = \mathrm{x_a} + \mathrm{A}\,(\mathrm{x} - \mathrm{x_a})$$

where A is the averaging kernel matrix of the MLS, x is the modeled vertical profile of HNO_3 or O_3 interpolated at the vertical grid of MLS, and $\mathrm{x_a}$ is the HNO_3 or O_3 prior profile of MLS. This is now clarified in Sec.3 and in Captions of Figs. 4 and 5.

Figure B below displays the seasonal average distributions of simulated HNO_3 mixing ratios with and without the averaging kernels. The comparisons indicate that the application of the averaging kernels to model results makes little differences in the simulated distributions of HNO_3 in the UTLS.

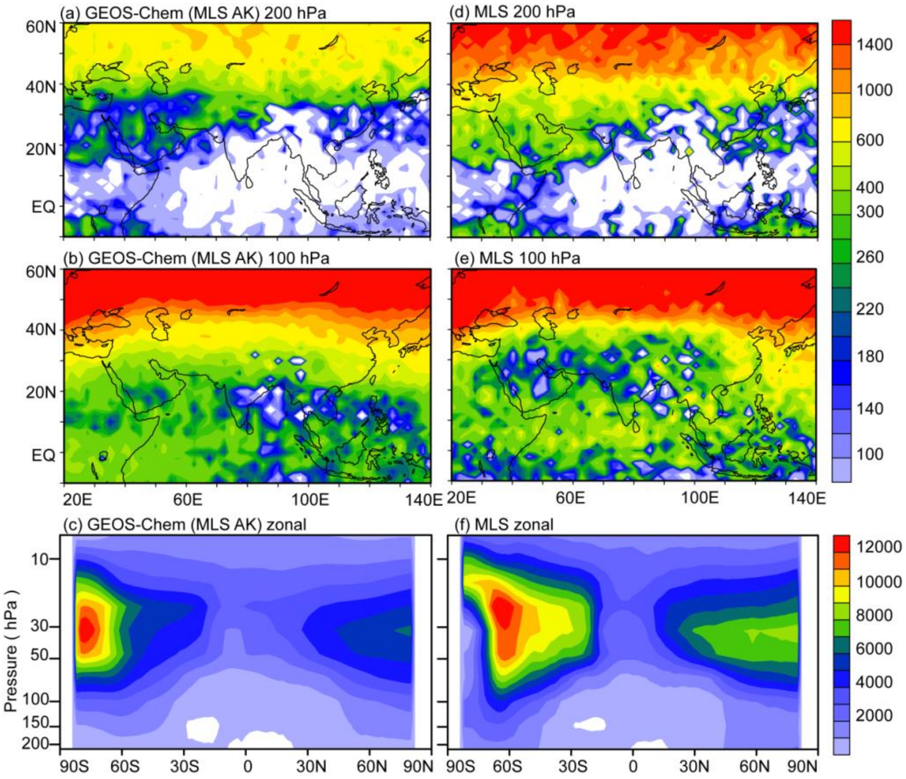

Figure 4. Comparisons of simulated HNO_3 concentrations (pptv) with observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated HNO_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem HNO_3 simulations are smoothed by the corresponding averaging kernels (AK). White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

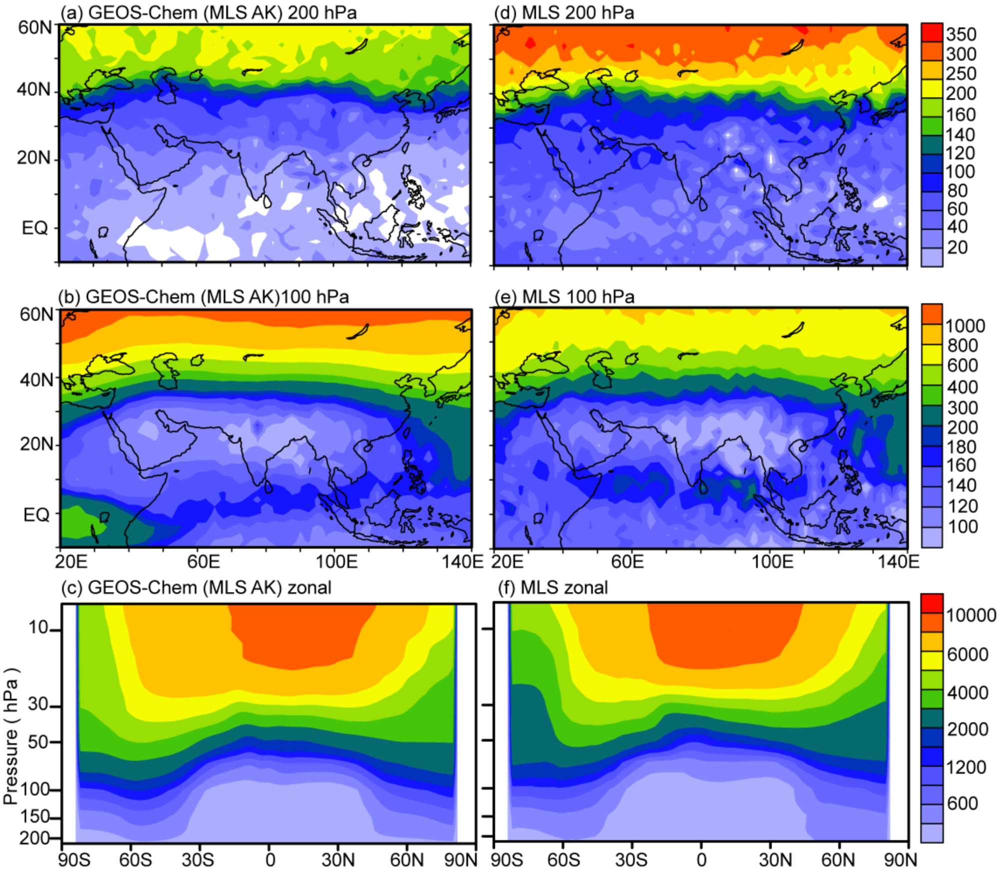

Figure. 5. Comparisons of simulated O_3 concentrations (ppbv) with observations (ppbv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated O_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem O_3 simulations are smoothed by the corresponding averaging kernels. White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

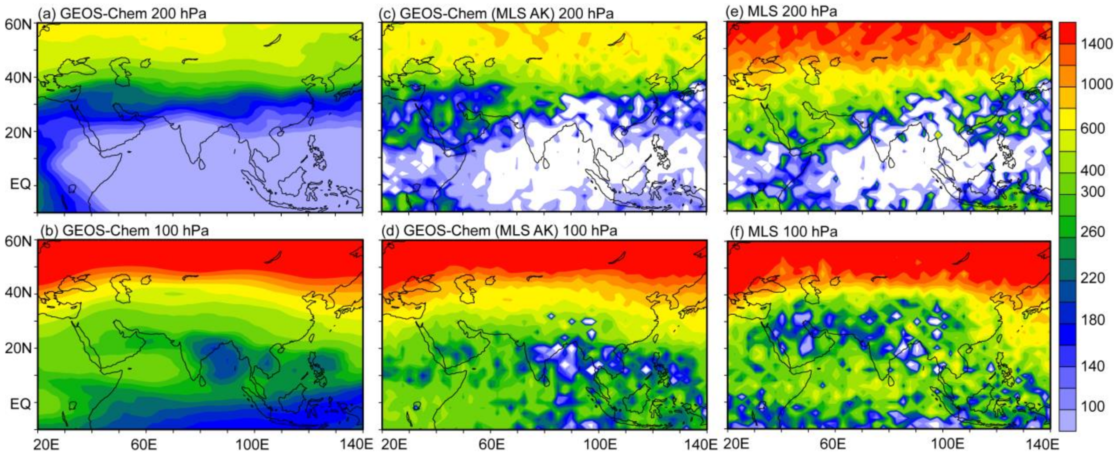

Figure B. Comparisons of simulated HNO_3 concentrations (pptv) with observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively, without the MLS averaging kernels (AK) applied. (c) and (d) are simulated concentrations at 200 hPa and 100 hPa, respectively, smoothed by the MLS AK at Equator (Livesey et al., 2011). (e) and (f) are observations from MLS. All the data are averaged over June–August of 2005.

(3) An unfortunate limitation of the MLS v3.3 HNO_3 data is that it might be affected by clouds. Thick clouds can lead to spikes in the HNO_3 mixing ratios in the UTLS. As a result of the deep convection, convective clouds develop in the troposphere over the TP/SASM region during the Asian summer monsoon region (Fu et al., 2006; Luo et al., 2011; Fadnavis et al., 2013). To avoid cloud contamination, many profiles are screened out according to the instructions of Livesey et al. (2011), resulting in limited data available in the UTLS over the studied region, especially at 200 hPa. The Gulf of Bengal, which is mentioned by the Reviewer, is where oceanic convection occurs (Luo et al., 2011). Thus, profiles that might be impacted by the convective clouds in this region are also screened out. Low values of the simulated HNO_3 can be seen with the limited observations over the Gulf of Bengal; however, the pattern might not be significant since there are limited qualified data available.

The surface data points for the various aerosol compounds (Fig. 8) contains a lot of data from Southern India which are of minor importance to the Tibetean plateau but more to the SASM region. As these are two different regions a separation of the data points would be better (even though this reduces the number of data points for a potential regression). Anyhow, this comparison does not show a very good model representation of the measurements, which the authors attribute to different years and other factors.

Response:

We have separated the data points in TP and SASM with different colors as displayed in Fig. 8(a). The impacts of uncertainties in surface-layer aerosol concentrations on simulated nitrate in the UTLS have been discussed in Sec.7. Four sensitivity simulations are performed to examine the impacts. According to the sensitivity results, the uncertainties in surface aerosol concentrations do not compromise the conclusion of this study that nitrate is the most dominant aerosol species in summertime in the UTLS over the TP/SASM region.

Concluding, I am not fully convinced that the manuscript is worth publication, as there is hardly any innovative, conclusive content in the paper. I would merely see this as (one of many) modelling studies covering Southeast Asia including a comparison with measurements, which does not show a very good model performance.

Response:

We have in abstract, introduction and conclusion sections the new information this work brings: 1) A number of previous studies have reported the persistent maxima of aerosols in the UTLS above TP/SASM during summertime (Kim et al., 2003; Li et al., 2005; Tobo et al., 2007; Vernier et al., 2009; Fadnavis et al., 2013; He et al., 2014), but no previous studies have reported and examined nitrate aerosol in the UTLS. We present such a study here. 2) We have examined the mechanisms for the accumulation of nitrate in the UTLS over the TP/SASM region. The vertical transport and the gas-to-aerosol conversion of HNO_3 to form nitrate occurs during the upward transport are found to lead to high nitrate in the UTLS. We believe our study is a novel contribution to research on the summertime chemical composition of aerosols, especially nitrate, in the UTLS over the TP/SASM region.

Minor (language and formulation points):

196: irons → ions

Response:

Changed.

699-700: in from of → in form of

Response:

Changed.

222: citation should not contain first names

Response:

Because there are two references of Park et al. (2004) in the manuscript, ‘R. J.’ was added to differentiate them by the journal’s Production Office.

244-245: NH3 emissions follow the global inventory described in Fisher et al.

Response:

Changed.

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Summertime nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer monsoon region

Yixuan Gu[a,b], Hong Liao[c,*], and Jianchun Bian[d]

[a]State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.

[b]University of Chinese Academy of Sciences, Beijing, China

[c]School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing, China

[d]Key Laboratory of Middle Atmosphere and Global Environment Observation (LAGEO), Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.

*Corresponding author address:

Prof. Hong Liao
School of Environmental Science and Engineering
Nanjing University of Information Science & Technology
Nanjing 210044, China
E-mail: hongliao@nuist.edu.cn

Abstract

We use the global three-dimensional Goddard Earth Observing System chemical transport model (GEOS-Chem) to examine the contribution of nitrate aerosol to aerosol concentrations in the upper troposphere and lower stratosphere (UTLS) over the Tibetan Plateau and the South Asian summer monsoon (TP/SASM) region during summertime of year 2005. Simulated surface-layer aerosol concentrations are compared with ground-based observations, and simulated aerosols in the UTLS are evaluated by using the Stratospheric Aerosol and Gas Experiment II satellite data. Simulations show elevated aerosol concentrations of sulfate, nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$ (particles with diameter equal or less than 2.5 μm, defined as the sum of sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols in this study) in the UTLS over the TP/SASM region throughout the summer. Nitrate aerosol is simulated to be of secondary importance near the surface but the most dominant aerosol species in the UTLS over the studied region. Averaged over summertime and over the TP/SASM region, C_{NIT} (the ratio of nitrate concentration to $PM_{2.5}$ concentration) values are 5–35% at the surface, 25–50% at 200 hPa, and could exceed 60% at 100 hPa. The mechanisms for the accumulation of nitrate in the UTLS over the TP/SASM region include vertical transport and the gas-to-aerosol conversion of HNO_3 to form nitrate. The high relative humidity and low temperature associated with the deep convection over the TP/SASM region are favorable for the gas-to-aerosol conversion of HNO_3.

1 Introduction

Aerosols in the upper troposphere and lower stratosphere (UTLS) have much longer residence times than those in the lower troposphere, influencing atmospheric chemistry and the Earth’s climate with large spatial and temporal coverage (Rasch et al., 2008). Aerosols in the UTLS influence the concentrations of chemical species via changes in photolysis rates and heterogeneous reactions (Pitari et al., 2014). For example, heterogeneous reactions on sulfate aerosol can perturb the chemical partitioning in the lower stratosphere, leading to significant O_3 depletion through enhanced chlorine, bromine, and odd-hydrogen catalytic cycle (Zhao et al., 1997; Considine et al., 2001; Talukdar et al., 2012; Tang et al., 2014; Pitari et al., 2014). Aerosols in the UTLS also influence climate by altering properties of cirrus clouds via homogeneous or heterogeneous ice nucleation (Li et al., 2005; Liu et al., 2009; Yin et al., 2012; Fadnavis et al., 2013). Injection of aerosols into the UTLS has been reported to induce complex responses in circulation, temperature, and water vapor (Liu et al., 2009; Wu et al., 2011; Su et al., 2011; Fadnavis et al., 2013).

Aerosols over the Tibetan Plateau (TP) and the Asian summer monsoon region are especially important. The TP is surrounded by countries with large anthropogenic emissions (Li et al., 2005; Lau et al., 2006). Aerosols from India, Southeast Asia, and southern China can be transported to the TP by prevailing winds in the premonsoon and monsoon seasons (Lawrence and Lelieveld, 2010; Xia et al., 2011). Observational and modeling studies have shown that persistent maxima of atmospheric constituents, such as water vapor (Gettelman et al., 2004; Randel and Park, 2006; Park et al., 2007), CO (Kar et al., 2004; Li et al., 2005; Park et al., 2007, 2008, 2009), CH_4 (M. Park et al., 2004; Xiong et al., 2009), NO_x (M. Park et al., 2004), HCN (Park et al., 2008; Randel et al., 2010), C_2H_6 and C_2H_2 (Park et al., 2008), exist in the UTLS above the TP and the South Asian summer monsoon (SASM) region because of the deep convection during boreal summer. Satellite observations suggested that the convection associated with the SASM is a vital pathway to transport air mass from the lower troposphere into the stratosphere (Chen et al., 2006; Randel and Park, 2006; Randel et al., 2010; Bian et al., 2011a). The heating associated with the persistent deep convection during summertime leads to the formation of the Tibetan anticyclone in the UTLS, which acts to isolate air within the anticyclone and traps the uplifted pollutants at that altitude (Park et al., 2007; Vernier et al., 2011; Bourgeois et al., 2012; Fadnavis et al., 2013; He et al., 2014). The stratosphere-troposphere exchange (STE) over the TP contributes largely to the global STE (Chen et al., 2006).

Previous studies have reported that aerosols exist in the UTLS over the TP/SASM region. Kim et al. (2003) carried out optical measurements with a ground-based lidar in Lhasa from August to October of 1999, and found an enhancement in aerosol concentration near the local tropopause with scattering ratio (SR, the ratio of aerosol plus molecular backscatter to molecular backscatter alone) of 1.1–1.2. Tobo et al. (2007) reported an enhancement of sub-micron aerosols (effective radius r = 0.15–0.6 μm) near the summertime tropopause (about 130 to 70 hPa), on the basis of in situ balloon measurements from an Optical Particle Counter at the same location in August of 1999. Vernier et al. (2009) examined satellite measurements from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard

Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) and reported the presence of small depolarizing particles with high SR values (about 1.20 at 532 nm) at 16–17 km altitude over South Asia in July and August of 2007 and 2008. Bourgeois et al. (2012) found that an aerosol layer existed at 16–18 km altitude over the Asian continent and Indian Ocean (20°S–30°N, 5–105°E) on the basis of the CALIOP observations. Recently, He et al. (2014) examined the vertical profiles of aerosol extinction coefficients measured with a Micro Pulse Lidar at Naqu, a meteorological station located in the central part of the TP, and also showed a maximum in aerosol extinction coefficient ($\sim 2.10^{-3}$ km^{-1}) in the UTLS (18–19 km) during the summer of 2011.

A number of previous studies have attempted to understand the chemical composition of aerosols in the UTLS. Froyd et al. (2009) measured aerosol composition with the National Oceanic and Atmospheric Administration (NOAA) single-particle mass spectrometer aboard the National Aeronautics and Space Administration (NASA) WB-57 high altitude aircraft platform, and reported that particles in the tropical tropopause layer were rich in nitrogen. Vernier et al. (2011) suggested that aerosol layer at the tropopause of Asia could be sulfur and/or organics, considering that Asian pollutants consisted of black carbon, organic carbon, SO_2, and NO_x (Park et al., 2009; Randel et al., 2010). Weigel et al. (2011) analyzed the volatility of aerosols obtained from in situ airborne measurements and reported that about 75–90 % of the particles in the tropical tropopause layer were volatile, but this study did not give any detailed analyses of chemical composition of aerosols. Bourgeois et al. (2012) showed, by using the ECHAM5.5-HAM2 model, that sulfate, water, and OC contributed, respectively, 53%, 29%, and 11% to aerosol extinction in the vicinity of the tropical tropopause layer. The ECHAM5.5-HAM2 model used by Bourgeois et al. (2012) simulated all major aerosol species in the atmosphere except for nitrate.

Few previous studies have examined nitrate aerosol in the UTLS, although nitrate is expected to be important for the following reasons. First, emissions of precursors of nitrate, such as NO_x and NH_3, are high over India, Southeast Asia, and China (Streets et al., 2003; Datta et al., 2012; Huang et al., 2012). Second, simulated nitrate concentrations are high over those regions (Liao and Seinfeld, 2005; Mu and Liao, 2014; Lou et al., 2014). Third, measured concentrations of nitrate are comparable to or larger than those of sulfate at rural and urban sites in the SASM region. Shrestha et al. (2000) carried out measurements of aerosols at Phortse, Nepal, during September 1996–November 1997, and showed that the average concentration of nitrate during the monsoon season (June-September) was 0.34 μg m^{-3}, higher than that of sulfate (0.17 μg m^{-3}). Decesari et al. (2010) reported, on the basis of measurements at the Nepal Climate Observatory-Pyramid from 2006 to 2008, that the concentrations of nitrate and sulfate were 0.37 μg m^{-3} and 0.50 μg m^{-3}, respectively, during the monsoon season. Chatterjee et al. (2010) measured aerosols at a high altitude station in northeastern Himalayas during January-December 2005. They found that the average concentrations of fine-mode nitrate and sulfate were 3.31±2.25 μg m^{-3} and 3.80±2.9 μg m^{-3}, respectively. At Lahore, an urban site in Pakistan, the observed daytime nitrate concentration of 21.8 μg m^{-3} was also higher than sulfate concentration of 12.6 μg m^{-3} (Lodhi et al., 2009), as the observations were averaged over November 2005 to March 2006. Fourth, the low temperatures in the UTLS would favor nitrate formation (Seinfeld and Pandis, 2006). Therefore, it is of interest to take nitrate aerosol into consideration when we examine aerosols in the UTLS.

In this work we simulate nitrate aerosol and its contribution to aerosol concentrations in the UTLS over the TP (70–105°E, 25–40°N) and the SASM region (70–105°E, 10–25°N) by using the global chemical transport model GEOS-Chem driven by the assimilated meteorological fields. These regions of interest are shown in Fig. 1. Simulated surface-layer aerosol concentrations are compared with ground-based observations, and simulated aerosols in the UTLS are evaluated by using the Stratospheric Aerosol and Gas Experiment II (SAGE II) satellite data. Section 2 is a brief description of the GEOS-Chem model and numerical experiment. Section 3 presents the simulation and evaluation of distributions and concentrations of HNO_3 and O_3 to show model's capability in simulating the NO_x-O_3-HNO_3 cycle over the studied regions. Section 4 shows simulated aerosols and Section 5 presents the simulated contribution of nitrate to aerosol concentrations in the UTLS over the TP and the SASM region. Section 6 discusses the mechanisms for high concentrations of nitrate in the UTLS. Section 7 discusses the impacts of uncertainties in surface-layer aerosol concentrations on simulated nitrate in the UTLS.

2 Model description and numerical experiment

2.1 GEOS-Chem model

We simulate gas-phase species and aerosols using the global chemical transport model GEOS-Chem (version 9-01-03, http://geos-chem.org) driven by the GEOS-5 assimilated meteorological fields from the Goddard Earth Observing System of the NASA Global Modeling and Assimilation Office. The version of the model used here has a horizontal resolution of 2° latitude by 2.5° longitude and 47 vertical layers extending from the surface to 0.01 hPa. Over the TP and the SASM region, the model has about 34 layers in the troposphere and 12 layers in the stratosphere.

The GEOS-Chem model has a fully coupled treatment of tropospheric NO_x-CO-hydrocarbon-aerosol chemistry and aerosols including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic carbon (OC), black carbon (BC) (R. J. Park et al., 2003; 2004; Pye et al., 2009), mineral dust (Fairlie et al., 2007), and sea salt (Alexander et al., 2005; Jaeglé et al., 2011). Anthropogenic aerosols are treated as bulk mass concentrations (particles of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC are not size-resolved). Sea Salt mass is simulated for two size bins (0.1–0.5 and 0.5–8 μm) and mineral dust is simulated for four size bins (0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm). Both BC and OC consist of hydrophilic and hydrophobic fractions in the model. It is assumed that 80% of BC and 50% of OC emitted from all primary sources are hydrophobic (Cooke et al., 1999; Chin et al., 2002; Chung and Seinfeld, 2002), which become hydrophilic with an e-folding time of 1.2 days following Cooke et al. (1999) and Chin et al. (2002). All secondary OC is assumed to be hydrophilic. Hydrophilic fractions of both BC and OC aerosols are assumed to be fully soluble.

The gas-aerosol partitioning of nitric acid and ammonium is calculated using the ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes, 2007). In the version of the GEOS-Chem model used in this work, ions considered in ISOROPIA II include $H^+/Na^+/NH_4^+/Cl^-/SO_4^{2-}/HSO_4^-/NO_3^-/OH^-$. The two-way coupling between aerosols and gas phase chemistry provides consistent chemical fields for aerosol simulation and aerosol mass for heterogeneous processes and calculations of gas-phase photolysis rates. Heterogeneous reactions include hydrolysis of N_2O_5 (Evans and Jacob, 2005), irreversible absorption of NO_3 and NO_2 on wet aerosols (Jacob, 2000), and the uptake of HO_2 by aerosols (Liao and Seinfeld, 2005; Thornton et al., 2008). Aerosol species are treated as an external mixture in the calculation of aerosol optical properties.

With respect to chemistry in the stratosphere, stratospheric O_3 concentrations are calculated using the linearized parameterization scheme (McLinden et al., 2000). 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).

Convective transport in GEOS-Chem mimics that in the parent GEOS general circulation model (GCM) (Hack, 1994; Zhang and McFarlane, 1995), which accounts for updraft, downdraft, and entrainment mass fluxes for deep and shallow convection (Wu et al., 2007). The aerosol wet deposition scheme in the GEOS-Chem follows that of Liu et al. (2001). For the scavenging of aerosols, $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, and hydrophilic OC and hydrophilic BC aerosols are assumed to be fully soluble. Dry deposition follows the standard resistance-in-series model of Wesely (1989).

Global emissions of aerosols and their precursors in the GEOS-Chem follow R. J. Park et al. (2003, 2004), with anthropogenic emissions of NO_x, CO, SO_2, and non-methane volatile organic compounds (NMVOC) in Asia overwritten by David Streets' 2006 emission inventory (http://mic.greenresource.cn/intex-b2006). Emissions of NH_3 in Asia are taken from Streets et al. (2003). Since NH_3 emissions in China showed large uncertainties in previous studies (Streets et al., 2003; Kim et al., 2006; Y. Zhang et al., 2010; Huang et al., 2011, 2012), we use the most recent estimate of NH_3 emissions in China by Huang et al. (2012), which is 9.8 Tg yr^{-1}, instead of 13.5 Tg yr^{-1} from Streets et al. (2003). Table 1 summarizes the annual emissions of NO_x, SO_2, NH_3, OC, and BC in Asia domain (60–155°E, 10–55°N).

Natural NO_x emissions from lightning are calculated using the scheme described by Sauvage et al. (2007) and Murray et al. (2012), and those from soil are simulated following Wang et al. (1998). Natural NH_3 emissions from soil, vegetation, and the oceans are taken from the Global Emissions Inventory Activity inventory (Bouwman et al., 1997). Biomass burning emissions are from the monthly Global Fire Emissions Database (GFED v3) driven by satellite observations of fire activity (van der Werf et al., 2010). Biogenic VOC (volatile organic compounds) emissions are calculated from the Model of Emissions of Gases and Aerosols from Nature (Guenther et al., 2006).

The monthly variations of emissions of SO_2 and NO_x follow Wang et al. (2013) and those of BC and OC follow Lou et al. (2014). The monthly scaling factors for NH_3 emissions follow the global inventory described in Fisher et al. (2011). Monthly variations of emissions (anthropogenic plus natural emissions) of NO_x SO_2, NH_3, OC, and BC over Asia are displayed in Fig. 2. The emissions of NH_3 are the highest in June as a result of the agriculture practice and high temperatures (Wang et al., 2013).

2.2 Numerical experiment

To examine the contribution of nitrate to aerosol concentrations in the UTLS over the TP/SASM region, we simulate aerosol concentrations by using the emissions of and meteorological fields of year 2005. Year 2005 is chosen so that we can use the observational datasets for this year from SAGE II and MLS, as described in Sects. 3 and 4. Following Rasch et al. (2008), we perform a 10-year spin-up run to generate the initial conditions (to allow the stratospheric species to reach quasi-steady state conditions). We would consider that the tropospheric simulation can be representative of year 2005 but stratosphere simulation should represent a multi-year average, because the 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).

3 Simulated concentrations of HNO_3 and O_3 and model evaluation

Nitrate aerosol forms when nitric acid (HNO_3) reacts with alkaline gases (for example, ammonia) in the atmosphere (Seinfeld and Pandis, 2006). HNO_3, as the important precursor of nitrate, is the major oxidation product of nitrogen oxides ($NO_x = NO+NO_2$) (Seinfeld and Pandis, 2006). To show the model's capability in simulating the NO_x-O_3-HNO_3 cycle over the studied regions, we present and evaluate the simulated HNO_3 and O_3 in this section.

Simulated mixing ratios of HNO_3 and O_3 in the UTLS are evaluated by using datasets from the limb viewing satellite instrument of Microwave Limb Sounder (MLS, version 3.3, level 2, ftp://acdisc.gsfc.nasa.gov/data/s4pa///Aura_MLS_Level2/). The MLS datasets provide valuable information on atmospheric compositions in the UTLS

(Waters et al., 2006). For HNO_3, the MLS provides scientifically useful datasets for 215 to 1.5 hPa, with a vertical resolution of 3–4 km and a horizontal resolution of 400–500 km. Since further evaluations are needed for datasets at altitudes with pressures higher than 215 hPa (Livesey et al., 2011), we use only datasets for pressures lower than that. For O_3, the MLS provides scientifically useful datasets for 261 to 0.02 hPa, with a vertical resolution of 2.5–3 km and a horizontal resolution of 300–400 km in the UTLS (Santee et al., 2007; Livesey et al., 2011). The uncertainties of the MLS HNO_3 and O_3 datasets in the UTLS are discussed in Livesey et al. (2011).The MLS data HNO_3 product at 100 hPa represents an average of a 3–4 km layer reaching from below the tropopause layer into the lower stratosphere (Duncan et al., 2007). The data screening is conducted strictly according to the instructions of Livesey et al. (2011). For comparison with the MLS observations, the simulated HNO_3 and O_3 mixing ratios are converted to $\hat{\mathrm{x}}$, following the method of Livesey et al. (2011):

$$\hat{\mathrm{x}} = \mathrm{x}_\mathrm{a} + \mathrm{A}\,(\mathrm{x} - \mathrm{x}_\mathrm{a})$$

where A is the averaging kernel matrix of the MLS, x is the modeled vertical profile of HNO_3 or O_3 interpolated at the vertical grid of MLS, and x_a is the HNO_3 or O_3 prior profile of MLS.

3.1 HNO_3

Figure 3(a) shows the simulated global distribution of HNO_3 concentrations averaged over June-August of 2005. Concentrations of HNO_3 exceed 1 ppbv over the industrialized areas such as Europe, North America, central and eastern Asia, and over biomass burning regions in the tropics, in agreement with the distributions and magnitudes reported in Liao et al. (2003). Over South

Asia, simulated HNO_3 concentrations are high (0.3–1 ppbv) in the northern Indian subcontinent, because the emissions of NO_x and NH_3 are high in this region (Streets et al., 2003; Zhang et al., 2009; Datta et al., 2012).

Figures 4(a)-4(b) show the simulated HNO_3 concentrations in the UTLS averaged over June-August of 2005. Since the tropopause is located at 70–150 hPa (12–15 km) over the TP/SASM region (Li et al., 2005; Bian et al., 2011b; Fadnavis et al., 2014), we choose the vertical layers of 200 hPa and 100 hPa to represent the UTLS. At both 200 hPa and 100 hPa, the highest HNO_3 concentrations are simulated to occur in the high latitude regions in the Northern Hemisphere (NH) (Fig. 4(a) and Fig. 4(b)). Simulated HNO_3 concentrations at 100 hPa are low over the region of 40–100°E and 10–30°N, which is part of the anticyclone region defined in Fig. 1. Figure 4(c) shows the latitude-altitude cross section of simulated seasonal mean HNO_3 mixing ratios averaged over 70–105°E. In boreal summer, the highest HNO_3 mixing ratios are simulated to occur at 30 hPa over the Polar Regions in both hemispheres. Over high latitudes, HNO_3 concentrations in the Southern Hemisphere (SH) are simulated to be higher than those in the NH.

To evaluate the simulated HNO_3, Figures 4(d)-4(f) show HNO_3 concentrations in the UTLS from MLS that are averaged over June-August of 2005. At 200 and 100 hPa altitudes, the observed HNO_3 mixing ratios are high in the high latitudes in the NH, which are captured by the GEOS-Chem model. The observed HNO_3 at 100 hPa exhibits low values of less than 400 pptv over 30–100°E and 10–30°N in the Asian monsoon anticyclone region (Fig. 4(e)). At 100 hPa, the observed HNO_3 mixing ratio averaged over the TP/SASM region (70–105°E, 10–40°N) is 335.4 pptv, which is lower than the simulated value of

372.6 pptv. Considering all the grid cells with MLS HNO_3 data available, the simulated seasonal mean HNO_3 concentrations show normalized mean bias (NMB) of +11.1 % at 100 hPa over the TP/SASM region in summer of year 2005. The observed pattern of the HNO_3 vertical distribution (Fig. 4(f)) is also captured by the GEOS-Chem model (Fig. 4(c)). The distributions of HNO_3 in the UTLS are associated with the Brewer-Dobson circulation proposed by Brewer (1949) and Dobson (1956), traveling upwards across the tropopause to the stratosphere at the equator and downwards to the troposphere near the Polar region.

3.2 O_3

Figure 3(b) shows the global distribution of simulated summertime surface-layer O_3 concentrations. Simulated O_3 concentrations are in a range of 40–70 ppbv over Europe, North America, China, and the biomass burning region of South Africa. Our model results agree closely with the simulated distributions and magnitudes reported in Mickley et al. (1999), Collins et al., (2000), Liao et al. (2003), Wu et al., (2008), Zeng et al. (2008), and Fadnavis et al. (2014). Fadnavis et al. (2014) also presented aircraft measurements over India in September of 2010 during the Cloud Aerosol Interaction and Precipitation Enhancement Experiment (CAIPEEX). Our simulated O_3 concentrations of 30–40 ppbv over India agree with the CAIPEEX measurements.

Figures 5(a)-5(b) show the simulated O_3 concentrations in the UTLS averaged over June-August of 2005. The distributions of O_3 concentrations in the UTLS are similar to those of HNO_3, with elevated values in the high latitudes of the NH. Relatively low O_3 mixing ratios of less than 200 ppbv are simulated at 100 hPa over 10–30°N, 20–110°E, within the anticyclone region defined in Fig. 1. Our simulated distributions and magnitudes of O_3 agree with those reported in Bian et al. (2011b), which examined the summertime distributions of O_3 in the UTLS during 2005–2009 by using the MLS version 2.2 level 2 products (Livesey et al., 2008). Because the background O_3 concentrations are generally high in the UTLS and the stratosphere, the low O_3 concentrations in the UTLS over the TP/SASM region are caused by the deep convection that transports O_3-poor air upward (Fu et al., 2006; Randel and Park, 2006; Park et al., 2007; Bian et al., 2011b). Figure 5(c) displays the latitude-altitude cross section of seasonal mean O_3 mixing ratios averaged over 70–105°E. As a result of the Brewer-Dobson circulation, O_3 concentrations in the UTLS are lower over the tropics than in the Polar Regions, even though the maximum O_3 concentrations are located around 10 hPa over the tropics (Brewer, 1949). Our simulated O_3 concentrations in the UTLS agree well with the measurements from MLS (Fig. 5(d)-5(f)). At 100 hPa, simulated and MLS observed O_3 mixing ratios averaged over the TP/SASM region (70–105°E, 10–40°N) are 151.7 and 146.6 ppbv, respectively. Compared to MLS observations, simulated O_3 concentrations at 100 hPa have a NMB of +3.5 % over the TP/SASM region in summer of 2005. Our simulated global STE of O_3 is 420 Tg yr^{-1}, which is within the range reported in previous studies (475±120 Tg yr^{-1} in McLinden et al. (2000), 420 Tg yr^{-1} in Škerlak et al. (2014), and 556±154 Tg yr^{-1} in Stevenson et al. (2006)).

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 above 12 km 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.

4 Simulated aerosols and model evaluation

4.1 Simulated aerosols

Figure 7 (a) shows the simulated surface-layer concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, BC, and $PM_{2.5}$ (the sum of the mass of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC aerosols) averaged over June-August of year 2005. As expected, simulated aerosol concentrations are high over polluted regions such as India and eastern China as a result of the high anthropogenic emissions of aerosol precursors and aerosols (Streets et al., 2003; Huang et al., 2012). Over the TP/SASM region (70–105°E, 10–40°N), the average concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC are 1.70, 0.94, 0.85, 0.30, and 0.94 μg m^{-3}, respectively. NO_3^- is simulated to be of secondary importance at the surface over the region of our interest. The simulated distributions and magnitudes of these aerosol species are similar to those reported in Wang et al. (2013) and

Mu and Liao (2014).

Figures 7(b) and 7(c) also show the simulated concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, BC, and $PM_{2.5}$ in the UTLS. Elevated concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, BC and $PM_{2.5}$ are simulated over the TP and Plateau south slope at 200 hPa altitude, and extend from eastern Mediterranean to western China at 100 hPa. The simulated enhanced concentrations of SO_4^{2-}, OC, and BC at 100 hPa over the anticyclone region (20–120°E, 10–40°N) agree with previous observational and modeling studies (Lelieveld et al., 2001; Li et al., 2005; Fadnavis et al., 2013). Li et al. (2005) reported elevated CO concentrations in the upper troposphere over the TP, on the basis of both MLS measurements and the GEOS-Chem simulation for September 2004. Fadnavis et al. (2013) also simulated maximum concentrations of SO_4^{2-}, OC, BC, and mineral dust aerosols in the UTLS during the Asian summer monsoon season owing to convective uplifting of the boundary layer pollutants. With NO_3^- aerosol accounted for in our simulation, NO_3^- is simulated to be the most dominant aerosol species in the UTLS over the TP/SASM region, followed by SO_4^{2-}, NH_4^+, OC, and BC. At 100 hPa, the averaged concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, and BC over the TP/SASM region (70–105°E, 10–40°N) region are 0.026, 0.069, 0.014, 0.011, and 0.002 μg m^{-3}, respectively.

4.2 Comparisons of simulated aerosol concentrations with in-situ observations

The simulated aerosol concentrations in East Asia in the GEOS-Chem model have been evaluated in previous studies (L. Zhang et al., 2010; Fu et al., 2012; Jeong and Park, 2013; Jiang et al., 2013; Wang et al., 2013; Lou et al., 2014). Here we are focused on the evaluation of aerosols in the South Asian monsoon region. For lack of publicly accessible in situ measurements of summertime aerosols in South Asia monsoon area, we compiled monthly or seasonal mean measured concentrations of each aerosol species based on measurements reported in the literature (see Table S1 in the Supplementary Material). These measurements were carried out over years of 1992–2010. The locations of sites with measurements available are shown in Fig. 8(a). Most sites are located in the upwind directions of the TP, with pollutants that can be transported to the UTLS during the South Asian summer monsoon season. 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).

Figures 8(b)–8(f) show the scatterplots of simulated versus observed seasonal mean aerosol concentrations. Compared with measurements, simulated $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, OC and BC have NMBs of –17.0%, +38.8%, +42.0%, –69.7% and –41.0%, respectively, as the concentrations of all seasons are considered. The correlations between model results and observations have *R* values of 0.49–0.85 for all aerosol species, indicating that the model is capable of capturing the spatial distributions and seasonal variations of each aerosol species in the South Asian monsoon region despite the biases in concentrations. If we consider simulated and measured concentrations for JJA alone, the simulated concentrations of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, OC and BC exhibit seasonal NMBs of –14.7%, +51.5%, +74.9%, –57.2% and –32.2%, respectively, and the values of *R* are in the range of 0.24–0.85. Note that the measurements of $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ are quite limited in terms of the number of samples, and the discrepancies between model results and measurements may also arise from the mismatch of the model year 2005 with the years of 1992–2010 with observations available.

4.3 Comparisons of simulated aerosol extinction coefficients with SAGE II datasets

Satellite datasets from the Stratospheric Aerosol and Gas Experiment II (SAGE II, https://eosweb.larc.nasa.gov/project/sage2/sage2_v620_table) are used to evaluate the simulated aerosol extinction in the UTLS. The SAGE II instrument was launched in October 1984 aboard the Earth Radiation Budget Satellite (ERBS) and terminated on 8 September 2005 (McCormick et al. 1987; Chu et al. 1989). The datasets used here are aerosol extinction coefficients at 525 nm from the version 6.20 SAGE retrievals, covering from 0.5 to 40 km with a vertical resolution of 0.5 km. Many validation studies have been conducted on the SAGE II aerosol data (Russell and McCormick, 1989; Oberbeck et al., 1989; Wang et al., 1989), which indicated that extinction coefficients have uncertainties of 20–30%. The extinction coefficients of aerosols in the GEOS-Chem model are calculated using aerosol mass concentration, extinction efficiency, effective radius, particle mass density, and the assumed aerosol size distribution (Drury et al., 2010). The hygroscopic growth of each aerosol species with relative humidity is accounted for, using the hygroscopic growth factors listed in Martin et al. (2003).

Figure 9(a) presents the simulated monthly mean distribution of aerosol extinction coefficients at 100 hPa for July of 2005. 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. Note that the contributions of sulfate, nitrate, ammonium, OC, sea salt, and mineral dust are all considered when we calculate aerosol extinction coefficients. Aerosol extinction coefficients are simulated to be 1.2–2×10^{-3} km^{-1} at 100 hPa over the Asian continent and Indian Ocean (20°S–30°N, 30°–105°E). These values agree closely with aerosol extinction coefficients measured at Naqu during August of 2011 for the same altitude, the maximum of which was 2.4×10^{-3} km^{-1} (He et al., 2014). Vernier et al. (2011) also identified this Asian aerosol layer with high SR at 100 hPa by observations of CALIPSO for JJA of 2006–2008.

Figure 9(b) displays the monthly mean vertical profiles of aerosol extinction coefficients averaged over the Asian monsoon anticyclone region (20–120°E 10–40°N) (Fig. 1) for July of 2005. The SAGE II datasets are available for July only in 2005. The profiles from SAGE II and the GEOS-Chem simulation are all shown. The vertical distributions of aerosol extinction coefficients “with nitrate” and “without nitrate” are both from the baseline run with full chemistry. The vertical distribution of aerosol extinction coefficient “with nitrate” (or “without nitrate”) indicates that the contribution of nitrate aerosol to aerosol extinction is (or is not) accounted for. Accounting for all aerosol species, the GEOS-Chem model reproduces well the aerosol extinction coefficients above 10 km, but the discrepancies are rather large in altitudes less than 10 km. Note that the uncertainties in satellite datasets increase as the altitude decreases (Vanhellemont et al., 2008；Kulkarni and Ramachandran, 2015), and the missing data in the lower troposphere along the satellite trajectories over the region of our interest also contribute to the discrepancies

Comparisons of profiles of aerosol extinction coefficients with and without nitrate aerosol indicate that the profiles show small differences in altitudes less than 6 km but large discrepancies from 6 km to the tropopause. With nitrate aerosol accounted for, the simulated aerosol extinction coefficients agree closely with SAGE II datasets in the UTLS (averaged over 14–16 km, the simulated value is 8.6×10^{-4} km^{-1} while the observed value is 8.0×10^{-4} km^{-1}). Without nitrate aerosol, the simulated aerosol extinction coefficient at 14–16 km altitude is 1.5×10^{-4} km^{-1}, which underestimates the aerosol extinction coefficient by 82.6% compared to that calculated with all the aerosol species. These comparisons of extinction coefficients with and without nitrate aerosol suggest that nitrate aerosol plays an important role in aerosol extinction in the UTLS over the region of our interest.

5 Contribution of nitrate to aerosol concentrations in the UTLS

Since nitrate aerosol is simulated to be the most abundant aerosol species in the UTLS over the TP/SASM region, we analyze the contribution of nitrate to $PM_{2.5}$ concentration (C_{NIT}=nitrate concentration/$PM_{2.5}$ concentration) in this section. Figure 10 shows the simulated seasonal mean distributions of C_{NIT} for June-August of year 2005. At the surface layer (Fig. 10(a)), simulated high C_{NIT} values are located over the areas with high nitrate concentrations (India and eastern China) as well as the oceans where $\mathrm{NO_3^-}$ also forms on sea salt and mineral dust particles (Arimoto et al., 1996; Nakamura et al., 2005; George and Nair, 2008). Over the TP/SASM region, the C_{NIT} values in JJA are 5–35% at the surface, 25–50% at 200 hPa (Fig. 10(b)), and could exceed 60% at 100 hPa (Fig. 10(c)). The latitude-altitude cross section of C_{NIT} (Fig. 10(d)) shows that C_{NIT} over 20–40°N increases with altitude and reaches maximum values around the extratropical tropopause.

Table 2 lists the mean concentrations of SO_4^{2-}, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, BC and OC, and their contributions to $PM_{2.5}$ during summertime of 2005 over the TP/SASM, TP, and SASM regions. Over the TP/SASM region, $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, BC and OC are simulated to contribute 35.9%, 19.8%, 18.1%, 6.4%, and 19.8%, respectively, to $PM_{2.5}$ mass concentration at the surface layer. The contributions increase significantly in the UTLS. The largest C_{NIT} is simulated in the SASM region at 100 hPa, where $\mathrm{NO_3^-}$ accounts for 60.5% of $PM_{2.5}$ mass concentration. The high C_{NIT} values indicate that $\mathrm{NO_3^-}$ plays an important role in the aerosol layer in the UTLS over the TP/SASM region.

Considering the large uncertainties in simulated sea salt (Jaeglé et al., 2011) and mineral dust (Fairlie et al., 2007) aerosols, we tend to be focused on anthropogenic aerosol species (SO_4^{2-}, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, BC, and OC) in this work. In our model, concentrations of sea salt (or mineral dust) are simulated to be 1.0–1.7 ng m^{-3} (or 5.0–7.0 ng m^{-3}) over the studied region in the summer of 2005, which contribute less than 1.2% (or 5.0%) to total aerosol mass at 100 hPa. Therefore the consideration of sea salt and mineral dust can slightly reduce C_{NIT} values, but C_{NIT} values at 100 hPa are still as high as 45-65% over the TP/SASM region in summer.

6 Mechanisms for high nitrate concentrations in the UTLS

6.1 Upward transport of nitrate from the lower troposphere

The intense convective transport of chemical species into the UTLS over the TP/SASM region during summertime has been widely discussed in previous studies (Randel et al., 2010; Bian et al., 2011a; Fadnavis et al., 2013, 2014;

Qie et al., 2014; He et al., 2014), evidenced by both the satellite observations (Fu et al., 2006; Luo et al., 2011) and the calculation of the outgoing long-wave radiation which is a convective proxy over the region (Randel and Park, 2006; Park, et al., 2007; Fadnavis et al., 2013). Since nitrate aerosol is simulated to be of secondary abundant aerosol species in the surface layer over the TP/SASM region (Fig. 7), the vertical mass transport through the deep convection in this region contributes to the accumulation of NO_3^- in the UTLS. Figure 11 shows the latitude-altitude cross sections of simulated concentrations of SO_4^{2-} and NO_3^- averaged over 70–105°E in June-August of 2005, together with the wind vectors obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data. Note that the assimilated GEOS-5 meteorological fields do not have vertical winds (http://wiki.seas.harvard.edu/geos-chem/index.php/List_of_GEOS-5_met_fields), so we use the ECMWF reanalysis wind fields to do the analysis here. High values of aerosol concentrations are found on the south slope of the Himalayas, where the deep convection exists. Although both SO_4^{2-} and NO_3^- are transported upward to the extratropical tropopause, the details of the vertical distributions are different. At altitudes higher than 8 km, the concentrations of NO_3^- do not decrease with altitude as quickly as those of SO_4^{2-}, and the concentrations of NO_3^- over 10–40°N are higher than those of SO_4^{2-}.

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. HNO_3 is produced by the reaction of NO_2 with OH during daytime and by hydrolysis of N_2O_5 on aerosol surfaces at night (Table 3). The chemical mechanisms for $\mathrm{SO_4^{2-}}$ and $\mathrm{NO_3^-}$ 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) but promotes the formation of $\mathrm{NO_3^-}$ by shifting gas-particle equilibria (Dawson et al., 2007; Liao et al., 2009). Dawson et al. (2007) examined the sensitivities of sulfate and nitrate concentrations to temperature by using the Particulate Matter Comprehensive Air Quality Model with extensions (PMCAMx). The sensitivity test was performed by fixing all meteorological parameters but perturbing temperature. Their sensitivity simulations showed that the increases in temperature led to increases in sulfate concentrations and decreases in nitrate concentrations. Compared to nitrate, sulfate concentrations showed smaller sensitivity to temperature changes (Dawson et al., 2007); as temperature increased, nitrate concentrations decreased by 19% K^{-1} and 17% K^{-1} in January and July respectively, while sulfate concentration increased by 0.12% K^{-1} and 1.3% K^{-1} in January and July, respectively. Therefore the different chemical mechanisms for $\mathrm{SO_4^{2-}}$ and $\mathrm{NO_3^-}$ formation contribute to the differences in their vertical distributions.

6.2 Net chemical production of HNO_3 during the vertical transport

As mentioned above, the formation of gas-phase HNO_3 and the partitioning of HNO_3 between gas and aerosol phases are the two major chemical processes that influence NO_3^- concentrations. The ability of the GEOS-Chem model to simulate gas-phase HNO_3 has been evaluated in Section 3.1 (by comparisons of our model results with MLS observations and concentrations from previous modeling studies). Major reactions for the production and loss of HNO_3 are listed in Table 3. Figure 12 shows the net chemical production of HNO_3 by gas-phase reactions and heterogeneous reactions (chemical production by reactions R1-R23 minus chemical loss by reactions R24-R25 in Table 3) summed over the TP/SASM region. The net chemical production has an overall trend of decreasing with altitude.

6.3 The gas-to-aerosol conversion of HNO_3 to form nitrate during the vertical transport

NO_3^- formation from gas-to-aerosol conversion of HNO_3 is calculated by using the ISORROPIA II thermodynamic equilibrium module (Fountoukis and Nenes, 2007). As shown in Fig. 12, nitrate formation from gas to aerosol conversion of HNO_3 peaks between 100–300 hPa. The magnitude of gas to aerosol conversion of HNO_3 is several times higher than the net chemical production of HNO_3, indicating that the gas-aerosol partitioning plays a dominant role in the enhancement of nitrate in the UTLS.

The gas-to-aerosol conversion of HNO_3 to form NO_3^- is very sensitive to relative humidity (RH) and temperature (Fountoukis and Nenes 2007; Dawson et al., 2007). Low temperature and high RH are favorable for NO_3^- formation.

Figure 13 shows the seasonal mean horizontal distributions of RH and temperature at 100 hPa and the latitude-altitude cross sections of these two parameters averaged over 70–105°E. RH exhibits high values in the TP/SASM region, which are consistent with the high H_2O mixing ratios in this area reported in Gettelman et al. (2004), M. Park et al. (2004), and Fu et al. (2006). At 100 hPa, the locations with high RH of exceeding 45% correspond well with those with high C_{NIT} values (Fig. 10(c)). The latitude-altitude cross section of RH (Fig. 13(c)) shows that RH has high values over the places with intense upward transport (Fig. 11). For temperature, as Fig. 13(b) and 13(d) show, summertime temperatures are cold (190–200 K) at 100 hPa in the TP/SASM region, consistent with the distribution and magnitude reported for August, 2011, in He et al. (2014) on the basis of the NCEP Reanalysis data. The low temperatures over the TP/SASM region are associated with the adiabatic expansion of ascending air mass of the deep convections (Yanai et al., 1992; Park et al., 2007; He et al., 2014).

Because of the favorable conditions of RH and temperature, the gas-to-aerosol conversion of HNO_3 to form nitrate can occur during the upward transport and in the UTLS. Figure 14 shows the mass budget for nitrate aerosol within the selected box of (70–105°E, 10–40°N, 8–16 km) to see the role of nitrate formation over the TP/SASM region. The horizontal mass fluxes have a net negative value of 0.10 Tg season^{-1}, reducing nitrate aerosol in the selected box. The vertical transport and the gas-to-aerosol conversion of HNO_3 increase nitrate mass in the selected box, with values of 0.09 Tg season^{-1} and 0.11 Tg season^{-1}, respectively, indicating that the gas-to-aerosol conversion plays an important role in the enhancement of nitrate in the UTLS over the TP/SASM region. Although relatively high RH exists near the tropopause of the TP/SASM region, the air near the tropopause is still dryer compared to that in the lower altitudes. Model results show that the gas-to-aerosol partition of HNO_3 decreases with altitude over 8–16 km, indicating that the gas to aerosol conversion contributes to nitrate accumulation in the UTLS mainly during the process of upward transport.

Previous studies have also reported that nitric acid trihydrates (NAT, $HNO_3 \cdot (H_2O)_3$) 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). As shown in Fig. 13, the temperatures around 100 hPa over the TP/SASM region are in the range of 190–200 K, which are low enough to produce some NAT particles. However, 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.

7 Sensitivities of simulated nitrate in the UTLS to anthropogenic NO_x, NH_3, and SO_2 emissions in Asia

Since simulated SO_4^{2-}, NO_3^- and NH_4^+ concentrations have, respectively, NMBs of –17.0%, +38.8%, and +42.0% on an annual mean basis and of –14.7%, +51.5%, and +74.9% in summer (Section 4.2), we perform four sensitivity simulations 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 case. The purpose of these sensitivity studies is to reduce NMBs of simulated surface-layer concentrations of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ and see whether $\mathrm{NO_3^-}$ is still the most dominant aerosol species in the UTLS. Model results from these sensitivity studies for summer of 2005 are presented in Table 4.

As anthropogenic emissions of SO_2 in Asia are increased by 20%, the NMB of simulated surface-layer $\mathrm{SO_4^{2-}}$ concentrations is –4.4%, which is an improvement compared to the NMB of –14.7% in the standard simulation. However, the increases in SO_2 emissions lead to larger NMBs of surface-layer $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ because of the increased formation of ammonium sulfate or ammonium bisulfate. The percentage contributions of $\mathrm{SO_4^{2-}}$ to total aerosol mass in the UTLS increase slightly by 2.7% at 200 hPa and by 1.6% at 100 hPa, and nitrate in the UTLS also shows small sensitivity to the change in SO_2 emissions.

With anthropogenic emissions of NO_x in Asia reduced by 50%, the NMB of simulated surface-layer $\mathrm{NO_3^-}$ concentrations changes from +51.5% in the standard simulation to –11.7% in this sensitivity run. The contribution of each of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ aerosols to total aerosol mass in the UTLS is not sensitive to this reduction in NO_x emissions at the surface; the percentage contribution obtained from this sensitivity run is very close to the value obtained in the standard simulation (Table 4). Similarly, in the sensitivity study with NH_3 emissions reduced by 50% in Asia, simulated surface-layer concentrations of $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ are improved in terms of the values of NMBs, but the improvement in simulated aerosol concentrations at the surface-layer does not influence our conclusion of high nitrate aerosol concentration in the UTLS.

As shown in Table 4, for the surface layer, simulated nitrate concentration over the TP/SASM region decreases by 46.8% with a 50% reduction in anthropogenic NO_x emissions in Asia, and it decreases by 22.3% when anthropogenic NH_3 emissions are reduced by the same percentage, indicating that surface-layer nitrate aerosol is more sensitive to anthropogenic emissions of NO_x than to those of NH_3. Relative to the baseline simulation, simulated nitrate concentrations at 200 hPa and 100 hPa decrease, respectively, by 49.0% and 17.7% with a 50% reduction in NH_3 emissions, whereas only by 2.1% and 1.3% with a 50% reduction in NO_x emissions. Over the studied region, the role of NH_3 in the sulfate-nitrate-ammonium aerosol system can be quantified by the gas ratio of $\mathrm{GR} = \frac{\text{free ammonia}}{\text{total nitrate}} = \frac{\mathrm{TA - 2 \times TS}}{\mathrm{TN}}$ (Ansari and Pandis, 1998), where $\mathrm{TA} = \mathrm{NH_3} + \mathrm{NH_4^+}$, $\mathrm{TS} = \mathrm{SO_4^{2-}}$, and $\mathrm{TN} = \mathrm{HNO_3} + \mathrm{NO_3^-}$. Over the TP/SASM region, GR is generally positive both at the surface and in the UTLS, especially over 20–40°N where deep convection exits (Fig. 11), indicating that S(VI) is in the form of sulfate and free ammonia is available to react with nitrate (Seinfeld and Pandis 2006). However, GR is generally less than 1.0 above 400 hPa in summer over the TP/SASM region, which indicates nitrate concentrations are most sensitive to changes in NH_3 and explains the small sensitivity of nitrate aerosol to NO_x emissions in the UTLS.

In the sensitivity study with emissions of NO_x, NH_3, and SO_2 in Asia changed simultaneously, simulated surface-layer concentrations of SO_4^{2-}, NO_3^- and NH_4^+ have NMBs of –8.3%, –27.0% and +55.4%, respectively, which are all improved compared to those in the standard simulation. Even though nitrate aerosol is now underestimated at the surface, it still accounts for 53.3% of the $PM_{2.5}$ concentration at 100 hPa over the TP/SASM region in summer.

It should be noted that the concentrations of OC and BC are also underestimated, with NMBs of –57.2% and –32.2%, respectively, in summer (Section 4.2). We have done a simple calculation with the concentrations of OC and BC in the UTLS multiplied by 2.3 and 1.5, respectively, and nitrate is still the most dominant aerosol species in summertime in the UTLS over the TP/SASM region (not shown in Table 4). Therefore the uncertainties in surface aerosol concentrations do not compromise the conclusion of this study.

8 Conclusions

In this work we simulate nitrate aerosol and its contribution to aerosol concentrations in the UTLS over the TP/SASM region (70–105°E, 10–40°N) for summertime of year 2005, using the global chemical transport model GEOS-Chem driven by the assimilated meteorological fields.

Simulated HNO_3 and O_3 are evaluated to show the model's ability to simulate the NOx-O_3-HNO_3 cycle over the studied region. In the UTLS, both the horizontal and vertical distributions of simulated HNO_3 and O_3 agree well with the MLS observations. At 100 hPa, simulated seasonal mean HNO_3 and O_3 mixing ratios show NMBs of +11.1 % and +3.5 %, respectively, over the

TP/SASM region (70–105°E, 10–40°N) in summer of year 2005, and the model biases lie within the confidence range of the MLS instruments. Both simulated and observed O_3 concentrations show relatively low values of less than 200 ppbv at 100 hPa over the TP/SASM region.

Averaged over the TP/SASM region, the surface-layer concentrations of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, BC, and OC are simulated to be 1.70, 0.94, 0.85, 0.30, and 0.94 μg m^{-3}, respectively. Nitrate aerosol is simulated to be of secondary importance near the surface over the region of our interest. Comparisons of simulated aerosol concentrations with ground-based observations show that simulated summertime concentrations of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, OC and BC have NMB of –14.7%, +51.5%, +74.9%, –57.2% and –32.2%, respectively. Note that the measurements of $\mathrm{NO_3^-}$ and $\mathrm{NH_4^+}$ are quite limited in terms of the number of samples.

Model results show elevated concentrations of $\mathrm{SO_4^{2-}}$, $\mathrm{NO_3^-}$, $\mathrm{NH_4^+}$, OC, BC and $PM_{2.5}$ in the UTLS over the TP/SASM region throughout the summer. $\mathrm{NO_3^-}$ is simulated to be the most dominant aerosol species in the UTLS of the TP/SASM region. Accounting for $\mathrm{NO_3^-}$ aerosol, the GEOS-Chem model reproduces well the magnitude of aerosol extinctions above 10 km, as model results are compared with the SAGE II measurements. The discrepancies between the simulated and observed aerosol extinction coefficient are within 8% in the UTLS (averaged over 14–16 km). Simulated vertical profiles of aerosol extinction coefficients with and without nitrate aerosol show large discrepancies from 6 km to tropopause, indicating the important role of nitrate in aerosol layer in the UTLS over the TP/SASM region.

The contribution of $\mathrm{NO_3^-}$ to aerosols in the TP/SASM region is quantified by C_{NIT} (the ratio of nitrate concentration to $PM_{2.5}$ concentration). Over the TP/SASM region, the C_{NIT} values in summer are 5–35% at the surface, 25–50% at 200 hPa, and could exceed 60% at 100 hPa. The mechanisms for the accumulation of nitrate in the UTLS over the TP/SASM region include vertical transport and the gas-to-aerosol conversion of HNO_3 to form nitrate. Such gas-to-aerosol conversion occurs during the upward transport and in the UTLS. The high relative humidity and low temperature associated with the deep convection over the TP/SASM region are favorable for nitrate formation.

Results from the present study indicate that nitrate is an important aerosol species in the UTLS over the TP/SASM region. Considering the scarce measurements of nitrate in the UTLS and the model uncertainties, more observational and modeling studies are needed to further explore the aerosol composition in the Asian tropopause aerosol layer. Further simulations of nitrate aerosol in the UTLS also need to account for NAT formation at low temperatures (Kirner et al., 2011) and the roles of natural aerosols, including the transport of mineral dust and sea salt to the UTLS as well as nitrate formation on these natural particles (Ma et al., 2003).

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Table 1. Summary of Annual Emissions of Aerosols and Aerosol Precursors in Asia (60°E-155°E, 10°N-55°N)

Species	Global	Asia
NO_x (Tg N yr^{-1})		
Aircraft	0.5	0.08
Anthropogenic	28.6	9.96
Biomass burning	4.7	0.27
Fertilizer	0.7	0.31
Lightning	5.9	0.87
Soil	5.9	0.96
Total	46.3	12.45
SO_2 (Tg S yr^{-1})		
Aircraft	0.1	0.01
Anthropogenic	52.6	23.46
Biomass burning	1.2	0.07
Volcanoes	4.4	1.04
No_eruption	8.9	1.78
Ship	7.4	0.94
Total	74.6	27.30
NH_3 (Tg N yr^{-1})		
Anthropogenic	34.9	17.83
Natural	14.2	2.01
Biomass burning	3.5	0.21
Biofuel	1.6	0.71
Total	54.2	20.76
OC (Tg C yr^{-1})		
Anthropogenic	3.1	1.42
Biomass burning	18.7	1.10
Biofuel	6.3	3.28
Biogenic	9.7	1.22
Total	37.8	7.02
BC (Tg C yr^{-1})		
Anthropogenic	3.0	1.43
Biomass burning	2.2	0.12
Biofuel	1.6	0.86
Total	6.8	2.41

Table 2. Simulated seasonal mean concentrations of aerosols and their contributions to $PM_{2.5}$ (in percentages in parentheses) during summertime (June-August) of 2005 for the TP/SASM, TP, and SASM regions. The unit is μg m^{-3} for concentrations at the surface, and 10^{-2} μg m^{-3} for concentrations at 200 hPa and 100 hPa.

	$PM_{2.5}$	SO_4^{2-}	NO_3^-	NH_4^+	OC	BC
TP/SASM						
Surface	4.73	1.70(35.9%)	0.94(19.8%)	0.85(18.1%)	0.94(19.8%)	0.30(6.4%)
200 hPa	16.19	3.27(20.2%)	7.57(46.8%)	2.67(16.5%)	2.22(13.7%)	0.44(2.7%)
100 hPa	12.14	2.60(21.4%)	6.90(56.8%)	1.43(11.8%)	1.05(8.6%)	0.16(1.3%)
TP						
Surface	5.44	2.12(39.0%)	1.05(19.3%)	1.08(19.9%)	0.88(16.1%)	0.31(5.7%)
200 hPa	19.80	4.16(21.0%)	9.43(47.6%)	3.25(16.4%)	2.49(12.6%)	0.47(2.4%)
100 hPa	10.58	2.60(24.6%)	5.51(52.0%)	1.35(12.7%)	0.99(9.4%)	0.14(1.3%)
SASM						
Surface	4.02	1.28(31.8%)	0.83(20.5%)	0.63(15.6%)	1.00(24.8%)	0.29(7.2%)
200 hPa	12.57	2.38(18.9%)	5.72(45.5%)	2.10(16.7%)	1.95(15.5%)	0.41(3.3%)
100 hPa	13.71	2.60(19.0%)	8.30(60.5%)	1.52(11.1%)	1.11(8.1%)	0.18(1.3%)

Table 3. List of gas-phase reactions and heterogeneous reactions involve HNO_3 in the GEOS-Chem model (Version 9-01-03)

	Reactants		Products
	Chemical productions		
R1	NO_2 + OH + M	→	HNO_3 + M
R2	NO_3 + CH_2O	→	HNO_3 + prod.
R3	ALD_2 + NO_3	→	HNO_3 + prod.
R4	RIO_1 + NO	→	HNO_3 + prod.
R5	IAO_2 + NO	→	$0.08HNO_3$+ prod.
R6	ISN_1 + NO	→	$0.05HNO_3$ + prod.
R7	VRO_2 + NO	→	HNO_3 + prod.
R8	MRO_2 + NO	→	HNO_3 + prod.
R9	INO_2+NO	→	$0.85HNO_3$+ prod.
R10	ALK_4 + NO_3	→	HNO_3 + prod.
R11	RCHO + NO_3	→	HNO_3 + prod.
R12	MEK + NO_3	→	HNO_3 + prod.
R13	INO_2 + MO_2	→	$0.425HNO_3$ + prod.
R14	GLYX + NO_3	→	HNO_3 + prod.
R15	MGLY + NO_3	→	HNO_3 + prod.
R16	MACR + NO_3	→	HNO_3 + prod.
R17	C_2H_6 + NO_3	→	HNO_3 + prod
R18	INO_2 + MCO_3	→	$0.85HNO_3$ + prod.
R19	NO_2 + (aerosol)	→	$0.5HNO_3$+ prod.
R20	NO_3 + (aerosol)	→	HNO_3 + prod.
R21	N_2O_5 + (aerosol)	→	$2HNO_3$
R22	DMS + NO_3	→	HNO_3 + prod.
R23	$BrNO_3$ + H_2O(l,s)	→	HNO_3 + prod.
	Chemical loss		
R24	HNO_3 + OH	→	H_2O + NO_3
R25	HNO_3 + hv	→	OH + NO_2

Table 4. 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%	

Figure Captions

Figure. 1. Regions examined in this study: the Tibetan Plateau region (TP, 70–105°E, 25–40°N), the SASM region (SASM, 70–105°E, 10–25°N), and the anticyclone region of (20–120°E, 10–40°N).

Figure. 2. Monthly variations in emissions of NO_x (Tg N month $^{-1}$), SO_2 (Tg S month^{-1}), NH_3 (Tg N month $^{-1}$), OC (Tg C month $^{-1}$), and BC (Tg C month $^{-1}$) over Asia. Values shown are the total emissions (anthropogenic plus natural emissions listed in Table 1).

Figure. 3. Simulated global distributions of surface-layer HNO_3 (pptv) and O_3 (ppbv) averaged over June-August, 2005.

Figure. 4. Comparisons of simulated HNO_3 concentrations (pptv) with observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated HNO_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem HNO_3 simulations are smoothed by the corresponding averaging kernels (AK). White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

Figure. 5. Comparisons of simulated O_3 concentrations (ppbv) with observations (ppbv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated O_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem O_3 simulations are smoothed by the corresponding averaging kernels. White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

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).

Figure. 7. Simulated seasonal mean concentrations (μg m^{-3}) of sulfate, nitrate, ammonium, organic carbon, black carbon, and $PM_{2.5}$ at (a) the surface layer, (b) 200 hPa, and (c) 100 hPa, during summer (June-August) of year 2005. Note that color bars are different for concentrations at the surface, 200 hPa, and 100 hPa.

Figure. 8. (a) Locations with measured aerosol concentrations from previous studies. Also shown are surface winds during summertime. (b)–(f) show the comparisons of simulated seasonal mean concentrations of sulfate, nitrate, ammonium, OC, and BC with measured values, respectively. Also shown in (b)–(f) are the 1:1 line (dashed), linear fit (solid line and equation), correlation coefficient between simulated and measured concentrations (R), and normalized mean bias (NMB) (defined as NMB=$\frac{\sum_{i=1}^{n}(P_i-O_i)}{\sum_{i=1}^{n}O_i}$ ×100%, where P_i and O_i are predicted and observed concentrations at station *i* for each aerosol species).

Figure. 9. (a) Monthly mean distribution of aerosol extinction coefficients (km^{-1}) at 100 hPa for July of 2005. (b) Monthly mean vertical distributions of aerosol extinction coefficients (at 525 nm for SAGE II and 550 nm for GEOS-Chem) (km^{-1}) averaged over the Asian monsoon anticyclone region (20–120°E, 10–40°N) for July of 2005. The horizontal dashed line represents the tropopause averaged over the Asian monsoon anticyclone region simulated by the GEOS-Chem model.

Figure. 10. Simulated contributions of nitrate to $PM_{2.5}$ (C_{NIT}= [NIT]/ [$PM_{2.5}$] ×100%) averaged over summer (June-August) of year 2005 at (a) surface-layer, (b) 200 hPa, and (c) 100 hPa. (d) The latitude-altitude cross section of simulated C_{NIT} (%) averaged over 70–105°E.

Figure. 11. Latitude-altitude cross sections of simulated concentrations (color shades, $\mu g\ m^{-3}$) of SO_4^{2-} and NO_3^- averaged over 70–105°E in June-August of 2005, together with the wind vectors obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data. The black line is the tropopause simulated by the GEOS-Chem model.

Figure. 12. The net chemical production of HNO_3 by gas-phase reactions and heterogeneous reactions (black dotted line) and the production of nitrate from gas to aerosol conversion of HNO_3 (red dotted line) over the TP/SASM region during summertime of year 2005.

Figure. 13. (a)-(b) Distributions of RH (%) and temperature (K) at 100 hPa. (c)-(d) The latitude-altitude cross sections of RH (%) and temperature (K) averaged over 70–105°E. RH and temperature are from the GEOS5 assimilated meteorological fields, and all the values are the averages over June-August of year 2005.

Figure. 14. Mass budget for nitrate aerosol within the selected box of (70–105°E, 10–40°N, 8–16 km). E/W transport indicates net mass flux through the east and west lateral boundaries, N/S transport indicates net mass flux through the north and south lateral boundaries, and upward transport is the net mass flux through the top and bottom sides of the box. The mass flux is positive if it increases nitrate mass within the box. Unit of fluxes is Tg $season^{-1}$. All the values are the averages over June-August of 2005.

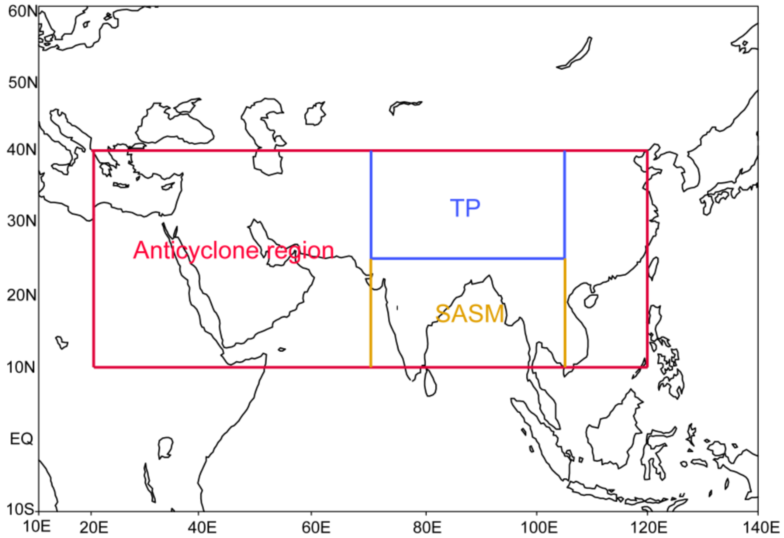


Figure. 1. Regions examined in this study: the Tibetan Plateau region (TP, 70–105°E, 25–40°N), the SASM region (SASM, 70–105°E, 10–25°N), and the anticyclone region of (20–120°E, 10–40°N).

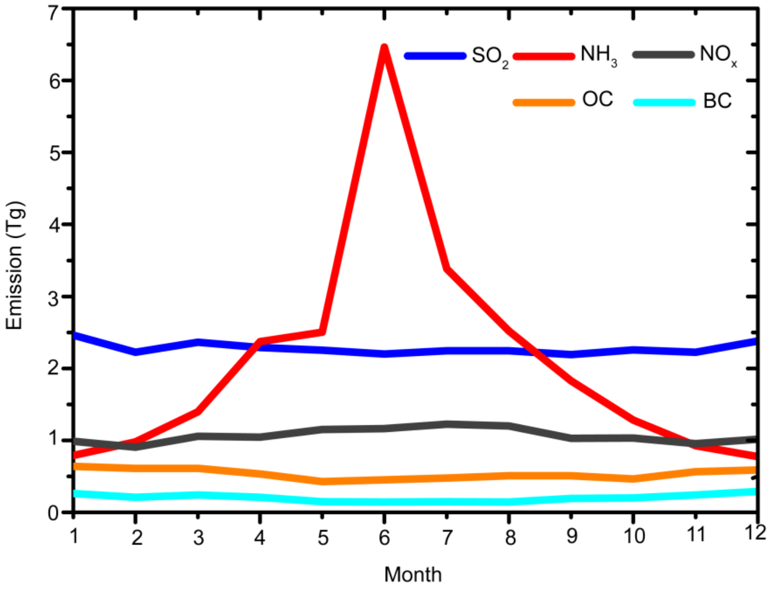


Figure. 2. Monthly variations in emissions of NO_x (Tg N month^{-1}), SO_2 (Tg S month^{-1}), NH_3 (Tg N month^{-1}), OC (Tg C month^{-1}), and BC (Tg C month^{-1}) over Asia. Values shown are the total emissions (anthropogenic plus natural emissions listed in Table 1).

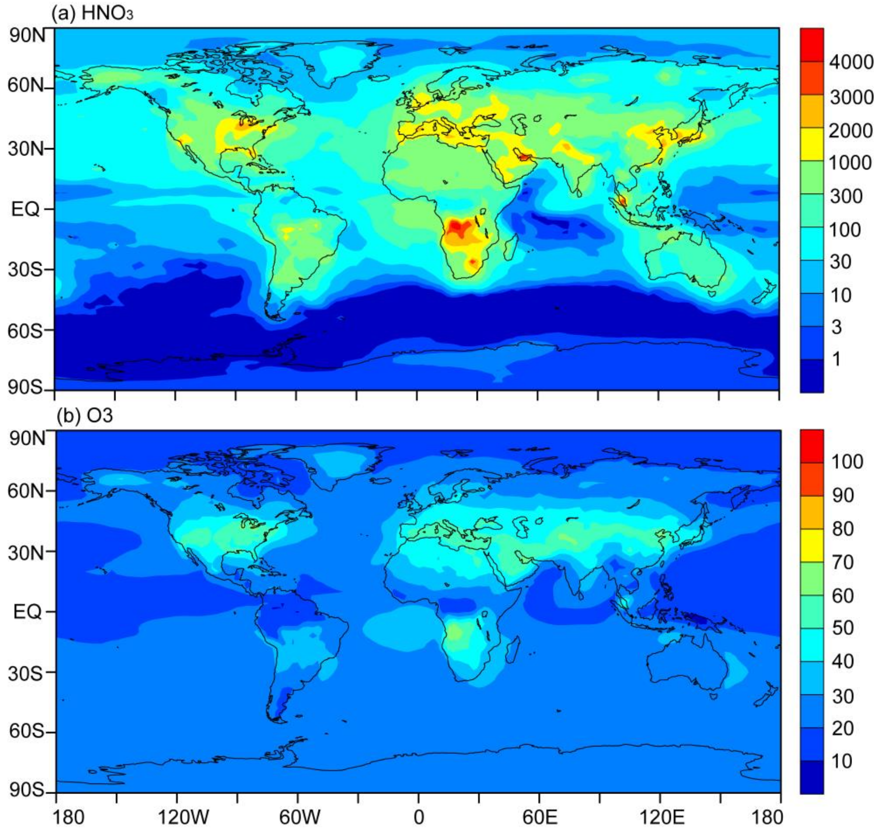


Figure. 3. Simulated global distributions of surface-layer HNO_3 (pptv) and O_3 (ppbv) averaged over June-August, 2005.

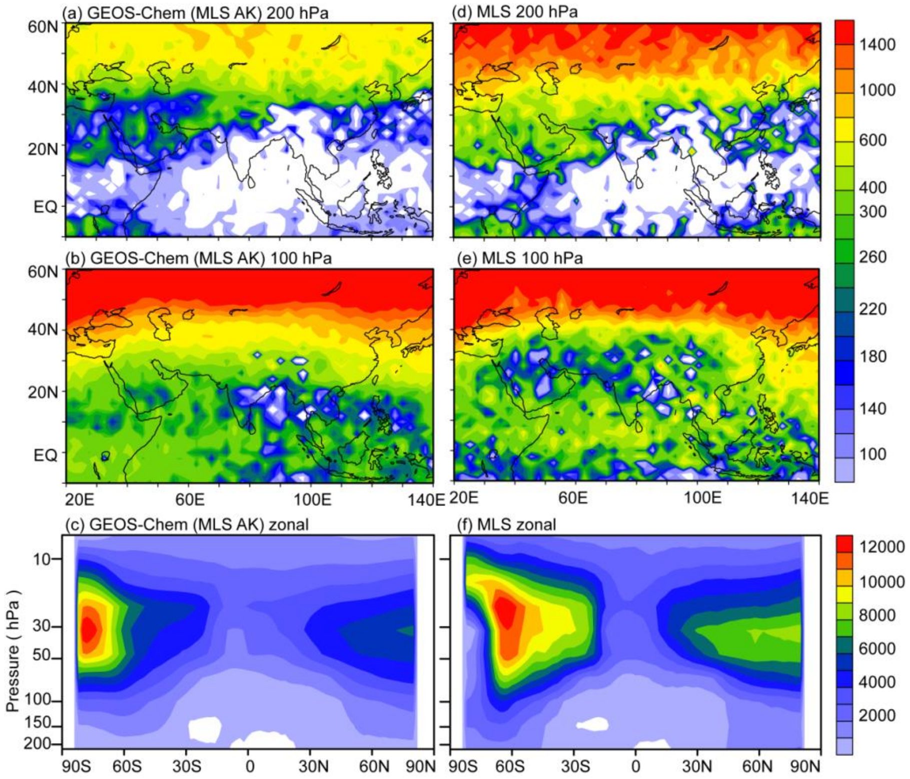


Figure 4. Comparisons of simulated HNO_3 concentrations (pptv) with observations (pptv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated HNO_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem HNO_3 simulations are smoothed by the corresponding averaging kernels (AK). White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

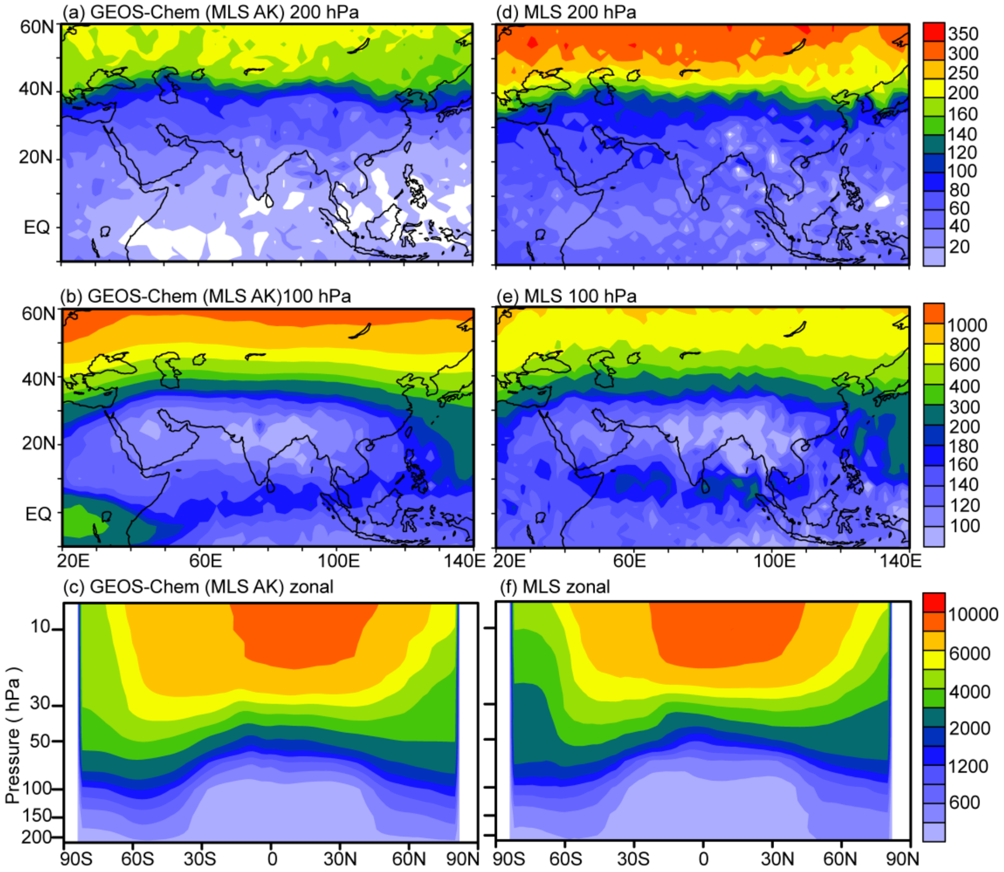


Figure. 5. Comparisons of simulated O_3 concentrations (ppbv) with observations (ppbv) from MLS. (a) and (b) are simulated concentrations at 200 hPa and 100 hPa, respectively. (c) is the latitude-altitude cross section of simulated O_3 concentrations averaged over 70–105°E. (d)-(f) are the same as (a)-(c), except that (d)-(f) are observations from MLS. The GEOS-Chem O_3 simulations are smoothed by the corresponding averaging kernels. White areas indicate lack of data meeting the retrieval quality criteria. All the data are averaged over June-August of 2005.

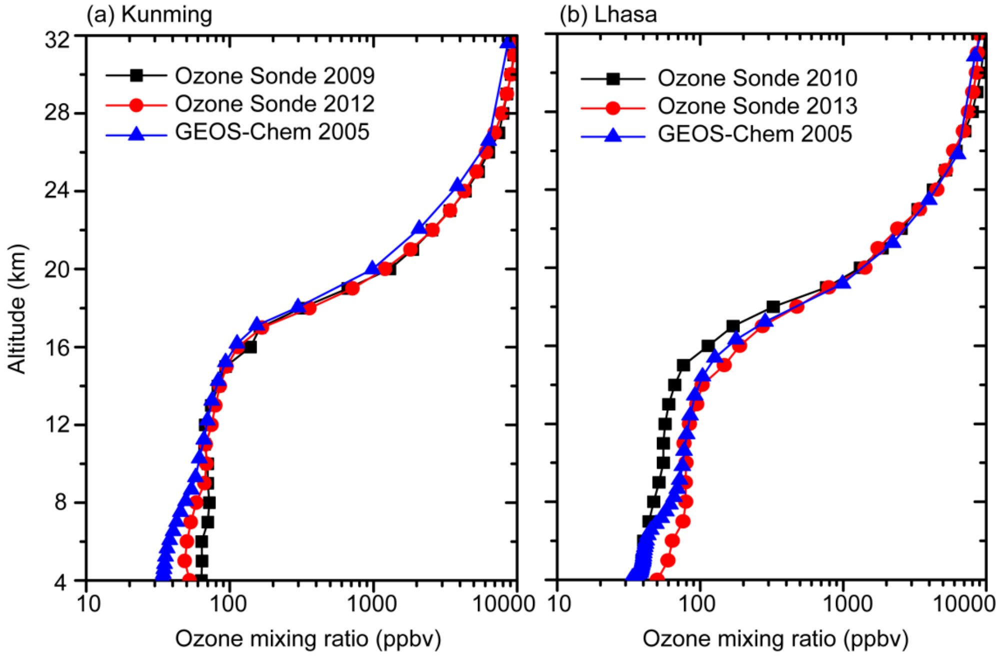


Figure 6. The simulated and observed vertical profiles of monthly mean O_3 mixing ratios at (a) Kunming and (b) Lhasa in August. The model results are from the simulation of year 2005. The observations in Kunming were conducted during August 7–13 (11 profiles of O_3 collected) in 2009 and during August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

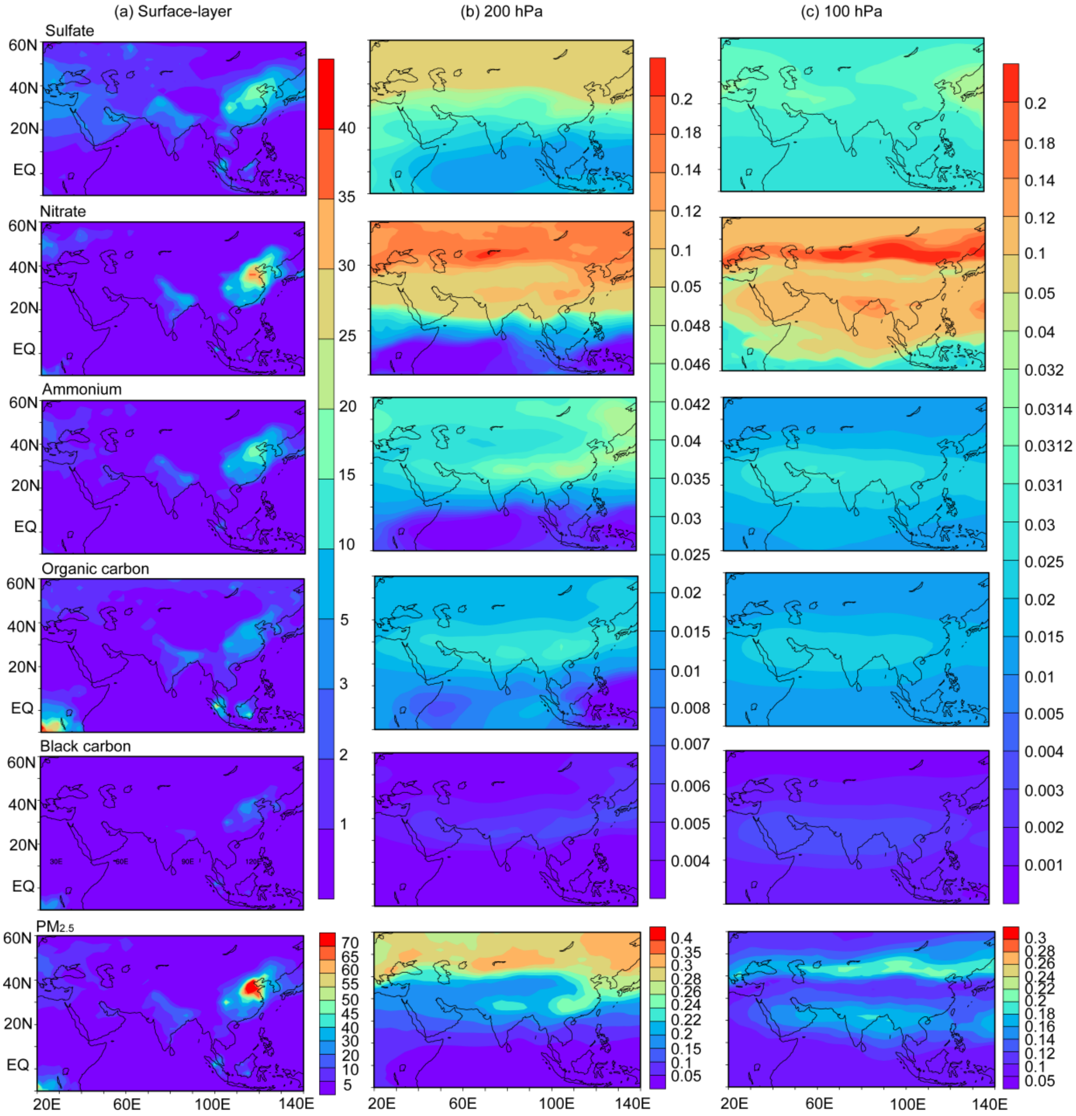


Figure. 7. Simulated seasonal mean concentrations (μg m^{-3}) of sulfate, nitrate, ammonium, organic carbon, black carbon, and PM$_{2.5}$ at (a) the surface layer, (b) 200 hPa, and (c) 100 hPa, during summer (June-August) of year 2005. Note that color bars are different for concentrations at the surface, 200 hPa, and 100 hPa.

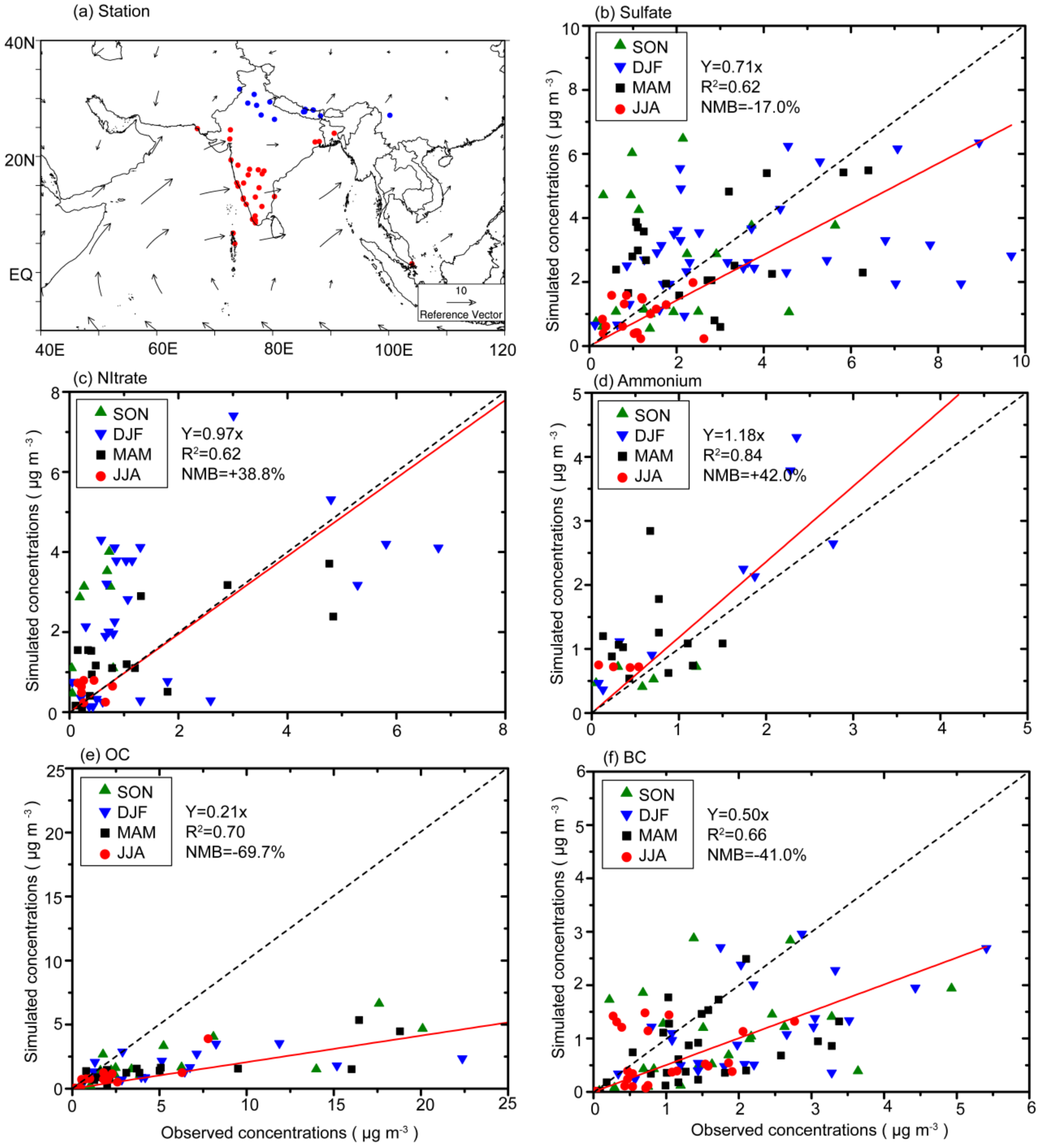


Figure. 8. (a) Locations with measured aerosol concentrations from previous studies over the TP (blue) and SASM (red) region. Also shown are surface winds during summertime. (b)–(f) show the comparisons of simulated seasonal mean concentrations of sulfate, nitrate, ammonium, OC, and BC with measured values, respectively. Also shown in (b)–(f) are the 1:1 line (dashed), linear fit (solid line and equation), correlation coefficient between simulated and measured concentrations (R), and normalized mean bias (NMB) (defined as $\mathrm{NMB}=\frac{\sum_{i=1}^{n}(P_i-O_i)}{\sum_{i=1}^{n}O_i}\times 100\%$, where P_i and O_i are predicted and observed concentrations at station i for each aerosol species).

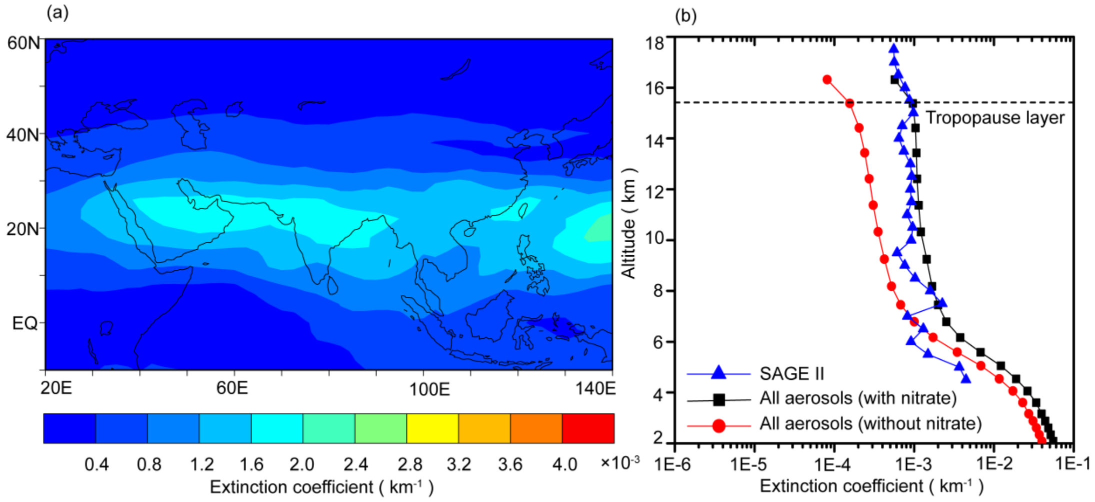


Figure. 9. (a) Monthly mean distribution of aerosol extinction coefficients (km^{-1}) at 100 hPa for July of 2005. (b) Monthly mean vertical distributions of aerosol extinction coefficients (at 525 nm for SAGE II and 550 nm for GEOS-Chem) (km^{-1}) averaged over the Asian monsoon anticyclone region (20–120°E, 10–40°N) for July of 2005. The horizontal dashed line represents the tropopause averaged over the Asian monsoon anticyclone region simulated by the GEOS-Chem model.

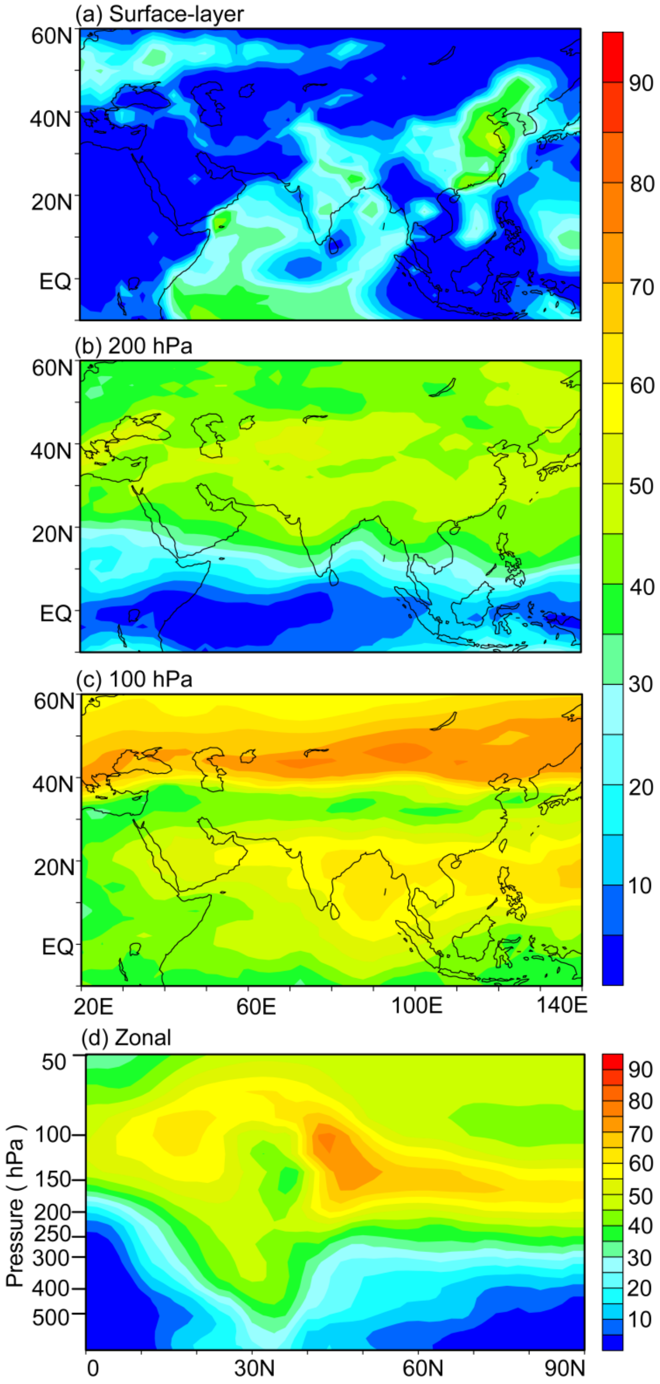


Figure. 10. Simulated contributions of nitrate to $PM_{2.5}$ (C_{NIT}= [NIT]/ [$PM_{2.5}$] ×100%) averaged over summer (June-August) of year 2005 at (a) surface-layer, (b) 200 hPa, and (c) 100 hPa. (d) The latitude-altitude cross section of simulated C_{NIT} (%) averaged over 70–105°E.

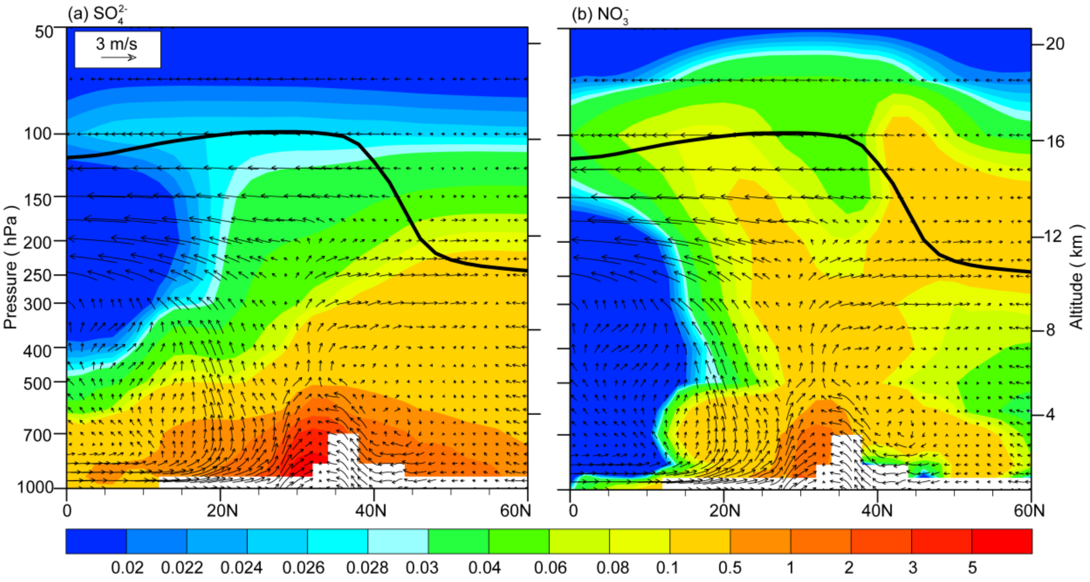


Figure. 11. Latitude-altitude cross sections of simulated concentrations (color shades, μg m^{-3}) of SO_4^{2-} and NO_3^- averaged over 70–105°E in June-August of 2005, together with the wind vectors obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis data. The black line is the tropopause simulated by the GEOS-Chem model.

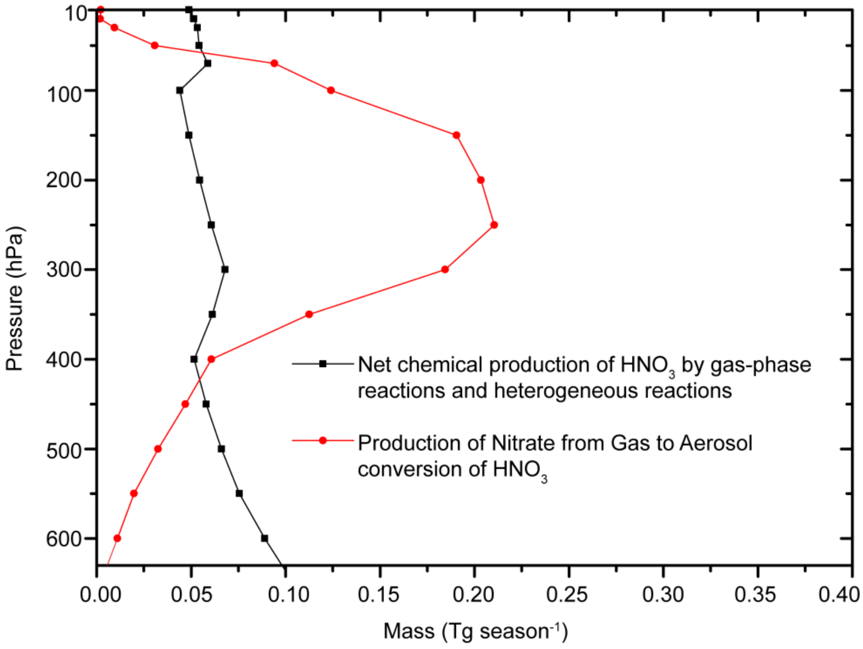
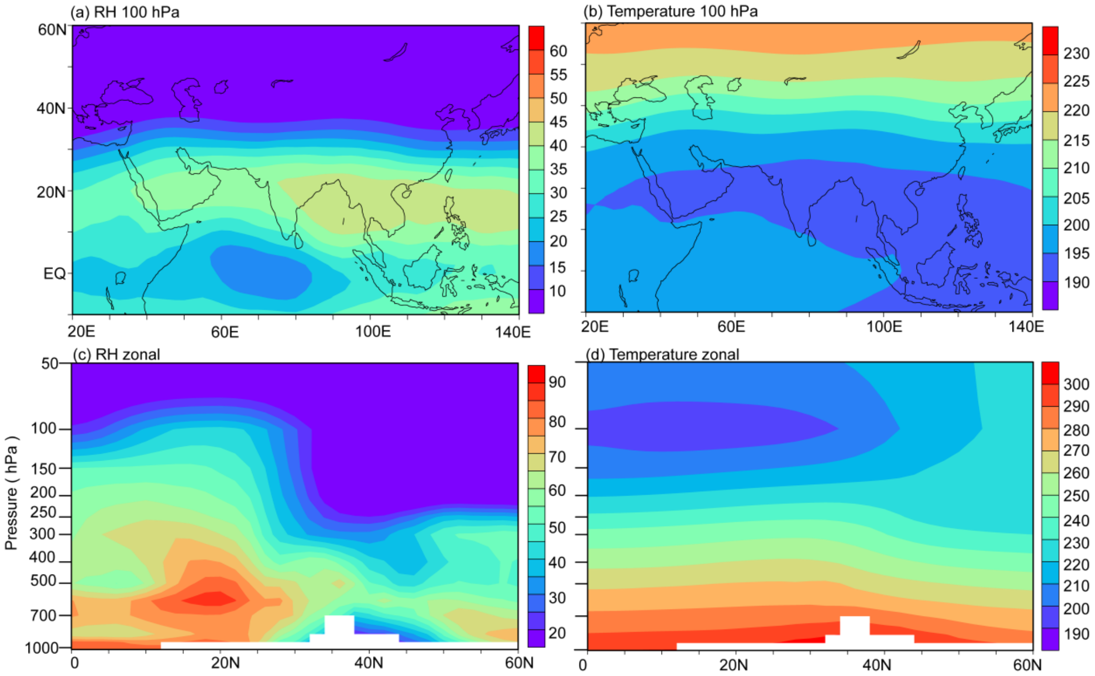


Figure 12. The net chemical production of HNO_3 by gas-phase reactions and heterogeneous reactions (black dotted line) and the production of nitrate from gas to aerosol conversion of HNO_3 (red dotted line) over the TP/SASM region during summertime of year 2005.



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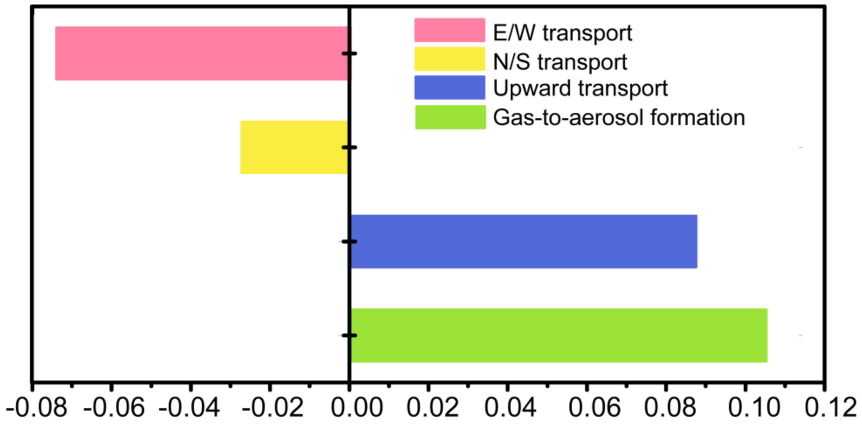

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