

We thank this reviewer for his/her constructive comments.  
Response to the Specific comments by this reviewer

**General comment:** This study investigates the role of mixing of Asian dust with anthropogenic pollutants using a three-dimensional regional chemical transport model (NAQPMS). An extremely strong dust event occurred during 19–22 March 2010 was chosen for the analysis. Several observations of remotely sensed data and surface measurement data are used for model validation. The model result suggests that major portion of anthropogenic aerosols are mixed with dust. 60% of the sulfate and 70–95% of the nitrate in the downwind regions was derived from active mixing processes during transport. Fe solubility rose from 0.5% in the Gobi region to 3–5% in the northwestern Pacific. The surface concentrations of other gaseous pollutants are greatly reduced due to the heterogeneous reactions. The finding in this study is interesting to ACP and the method is reasonable. I would recommend the publication of this paper with major revision.

Response: We greatly thank Reviewer for his/her encouragement and insightful comments concerning the manuscript. We have reworded this manuscript to consider all the comments.

**Comment 1:** Aerosol mixing is highly complex process that depends on the size, shape, chemical compounds, etc. This study relies on a simple reaction rate as presented in Table 1, without considering other effects. It should provide how this simplification affect to mixing in modeling and what are the uncertainty of the reaction rate.

Response: The reviewer is right. Quite a few studies have shown that many Asian dust particles contain sulfate and nitrates which are formed by heterogeneous reactions involving  $\text{SO}_2$ ,  $\text{HNO}_3$  and  $\text{NO}_x$  (Formenti et al., 2012). Aerosol mixing through heterogeneous reactions is high complex. Considering insufficient characterization of the condensed phase, Jacob (2000) recommended reaction probability ( $\gamma$ ) that a molecule impacting the aerosol surface undergoes reaction to describe the reactive uptake of a gas by an aerosol.  $\gamma$  is defined as the net loss rate of a gas due to reaction in the aerosol. Same as current most of 3-D models, we use reaction probability to simulate the mixing process of dust and reactive gases. Further, Han et al., (2011) stated that uncertainties in simulating in the extent on chemical aging of mineral dust are caused by 3 critical issues: (i) the mixing state of aerosols, (ii) the magnitudes of reaction probability (or uptake coefficient) onto mineral dust and anthropogenic urban aerosols and (iii) the chemical component ratio of dust particles. Additionally, size distribution and shape of dust particles are also able to affect the mixing extent in the transport. In the revised manuscript, we discuss the impacts of these factors on our estimation on aerosol mixing.

(1) The mixing state is very critical because it affected the reaction probability of gases on dust. For example, in the assumption of internal mixing of urban (nitrate and sulfate) and dust aerosols, an identical magnitude of reaction probability ( $\gamma$ ) on both aerosols is applied. In the external mixing, different order of  $\gamma$  is applied. Fig.1 shows three categories concerning the mixing states between mineral dust and pollutants in current most of air quality models. In this study, the “external mixing with bulk mineral dust” type is applied.

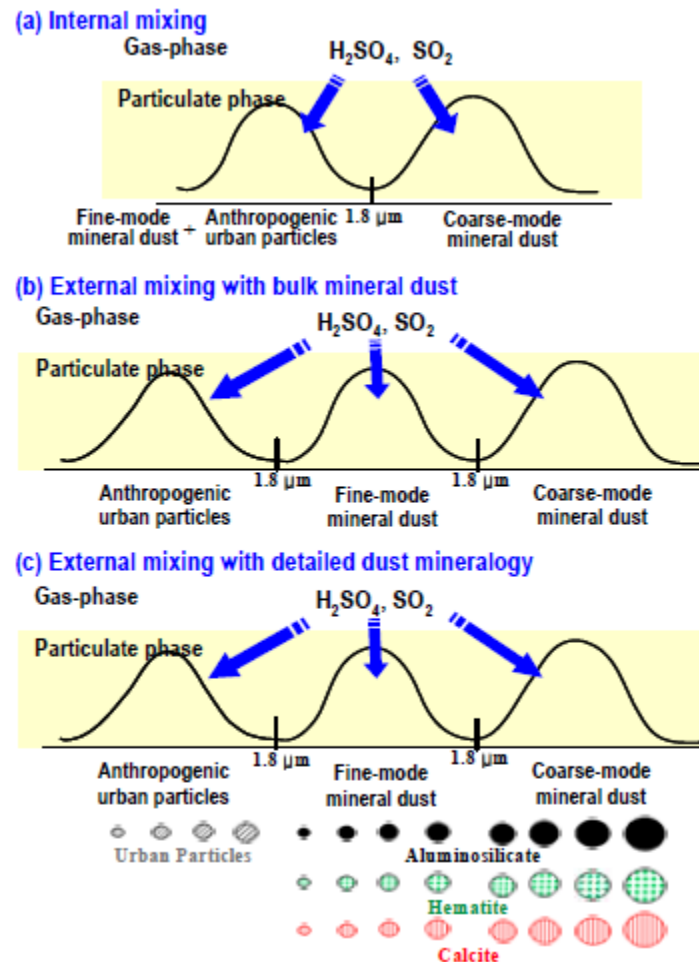


Fig.1 Aerosol mixing states a) internal; b) external; c) external mixing with detailed dust mineralogy (Han et al., 2011)

Most studies of Asian outflow indicate that anthropogenic and dust aerosols are mostly externally (Meskhidze et al., 2004; Tang et al., 2004; Song et al., 2005). Furthermore, Han et al. (2011) used observed size-resolved inorganic aerosol composition at South Korea in Asian duststorms and a photochemical Lagrangian model to investigate the impact of mixing states of urban and dust aerosols on heterogeneous reactions. They found that the model with assumption of internal mixing of aerosols cannot predict the formation rate of sulfate on mineral dust and anthropogenic urban particle. The externally-mixed anthropogenic urban particles and mineral dust can successfully re-generate the size distributions of sulfate measured at South Korea. The detailed dust mineralogy (Type c in Fig.1) does not

bring dramatic progresses comparing with external mixing state between urban aerosols with bulk mineral dust.

The above-mentioned results indicate that our assumption of mixing states in this study is suited to assess the interaction between gaseous pollutants and Asian mineral dust.

(2) Depending on types of dust sample, experimental and analytical methods, reaction probability ( $\gamma$ ) varies greatly, in some cases several orders of magnitude. This could bring uncertainties to assess the mixing extents of dust and urban aerosols. Table1 in this reply shows the upper and lower limit of reaction probability ( $\gamma$ ) in previous studies and their values in this study. To reduce the uncertainties due to  $\gamma$ , we select those values on China losses and Gobi desert which are potential sources considering impact of dust mineralogy. For example, the reaction  $\text{HNO}_3$  on dust particles in this study is experimental  $\gamma$  on China Losses at 0%, 40% and 80% RH (Wei, 2010).

In the revised manuscript, we make use of the sensitive analysis to examine the uncertainty in the results. In order to cover the factor of uncertainties reported by Zhu et al (2010) (Table 1), simulations in which one heterogeneous reaction is calculated individually with either the “lower limit” or “upper limit” are performed. In this study, uncertainties of reaction probabilities ( $\gamma$ ) in HR19 ( $\text{HNO}_3$ +dust,  $\gamma_{\text{hno3}}$ ) and HR12 ( $\text{SO}_2$ +dust,  $\gamma_{\text{so2}}$ ) are analyzed. The results at Shanghai and Xiamen are shown in Fig.2 in this reply. Clearly, the simulated nitrate and sulfate strongly depends on the reaction probability. At Shanghai, the simulated nitrate and sulfate in base case where  $\gamma$  values are taken from table 1 in the manuscript are very close to observations. This indicates  $\gamma_{\text{hno3}}$  and  $\gamma_{\text{so2}}$  in this study is suitable for this dust event. The upper limit of  $\gamma_{\text{hno3}}$  and  $\gamma_{\text{so2}}$  overestimates sulfate and nitrate in Shanghai, whereas the lower limit is not efficient enough. At Xiamen, although the simulated nitrate and sulfate concentrations in upper limit of  $\gamma$  agree with observations, the fraction of these inorganic components in dust particles indeed overestimates the observations. This is because the simulated dust concentrations are lower than observation (Fig.3 in the manuscript).

In this study, we apply the following formulation to calculate the uncertainties:

$$U = \left| \frac{y_{\gamma(\text{upper})} - y_{\gamma(\text{lower})}}{y_{\lambda(\text{base})}} \right|$$

Where  $y_{\gamma(\text{upper})}$ ,  $y_{\gamma(\text{lower})}$  and  $y_{\lambda(\text{base})}$  represents the impacts of mineral dust on pollutants under conditions with upper limit, lower limit and values of  $\gamma$ ., respectively. Uncertainties of nitrate reach 143% and 127% at Shanghai and Xiamen, respectively. For sulfate, Shanghai and Xiamen are 218% and 209%.

Fig.3 in this reply shows the uncertainties of impacts on  $\text{HNO}_3$  and  $\text{SO}_2$ .

Table 1 Heterogeneous reaction, lower and upper limits of reactive uptake coefficients and their values in this study (base)

No.	base	upper limit	lower limit
$O_3 + \text{dust} \rightarrow \text{products}$	$2.7 \times 10^{-5}$	$1 \times 10^{-4}$	$1 \times 10^{-6}$
$HNO_3 + \text{dust} \rightarrow NO_3^-$	$6 \times 10^{-5} \sim 1.8 \times 10^{-1}$	$2 \times 10^{-1}$	$1 \times 10^{-5}$
$N_2O_5 + \text{dust} \rightarrow 2HNO_3$	$3 \times 10^{-2}$	$1 \times 10^{-1}$	$1 \times 10^{-2}$
$OH + \text{dust} \rightarrow \text{products}$	$1 \times 10^{-1}$	1	$4 \times 10^{-3}$
$HO_2 + \text{dust} \rightarrow 0.5H_2O_2$	$1 \times 10^{-1}$	1	$1 \times 10^{-2}$
$H_2O_2 + \text{dust} \rightarrow \text{products}$	$3 \times 10^{-4} \sim 6 \times 10^{-4}$	$1.8 \times 10^{-1}$	$8 \times 10^{-4}$
$SO_2 + \text{dust} \rightarrow SO_4^{2-}$	$1 \times 10^{-4}$	$2.6 \times 10^{-4}$	$5.0 \times 10^{-7}$
$HCHO + \text{dust} \rightarrow \text{products}$	$1 \times 10^{-5}$	$1.1 \times 10^{-4}$	$2.6 \times 10^{-7}$

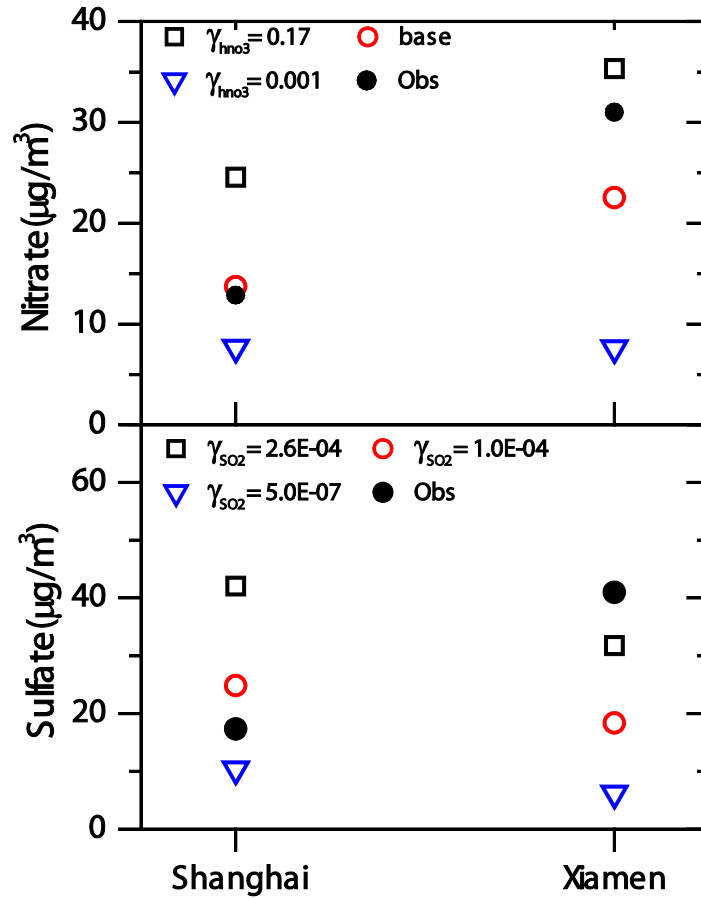


Fig.2 observed and simulated sulfate and nitrate at Shanghai and Xiamen during the dusty period in simulations with base, upper limit and little limit of  $\gamma_{hno3}$  and  $\gamma_{so2}$ .

(3) Size distribution and shape of dust particles are thought to be important physical properties affecting heterogeneous chemistry. In this study, dust particles are divided into 4 size-bins (0.43-1 $\mu$ m, 1-2.5 $\mu$ m, 2.5-5 $\mu$ m and 5-10 $\mu$ m), and assumed to be spherical particles. Table 2 in the manuscript suggests that the simulation reproduce mass size distribution of dust particles in the transport. In

this study, the heterogeneous reactions on dust are calculated in each bin. That is, dust-sulfate and dust-nitrate also have 4 bins.

It is known that mineral dust particles have irregular shapes. So the assumption of spherical particles in this study likely causes errors because irregular particles could have different surface areas with sphere particles. In the manuscript, we use observed aspect ratio of Asian dust particles (1.5) to correct simulated surface areas and analyze the impacts of shape on dust mixing extent.

Ginoux (2003) reported a method to correct surface area of irregular shapes. In his method, surface area of irregular particle can be calculated by a sphericity factor ( $\phi_p$ ) in the formula:

$$S_p = S / \phi_p$$

$$S = \pi \left( \frac{6V_p}{\pi} \right)^{2/3}$$

Where  $S$  and  $S_p$  represent the surface area of spherical and irregular particle, respectively.  $V_p$  is the particle volume. When  $\phi_p$  is equal to 1.0, the particle is sphere, and when it is equal to 0.1, the particle is elongated.

The sphericity factor ( $\phi_p$ ) is expressed in terms of aspect ratio ( $\lambda$ ) (ratio between longer and short axis of an ellipse fitted to the particle outline) as the following formula:

$$\phi_p = \frac{2\lambda^{2/3}}{1 + \frac{\lambda^2}{\sqrt{\lambda^2 - 1}} \arcsin\left(\sqrt{1 - \frac{1}{\lambda^2}}\right)}$$

In China deserts (Taklamakan desert, Gobi), observed aspect ratio of dust is ~1.5 (Okada et al., 2001). Fig.4 in this reply shows the relationship between  $\phi_p$  and  $\lambda$ . Clearly, the  $\phi_p$  is about 0.97 in Asian dust, which means surface areas errors caused by irregular shape is less than 5%.

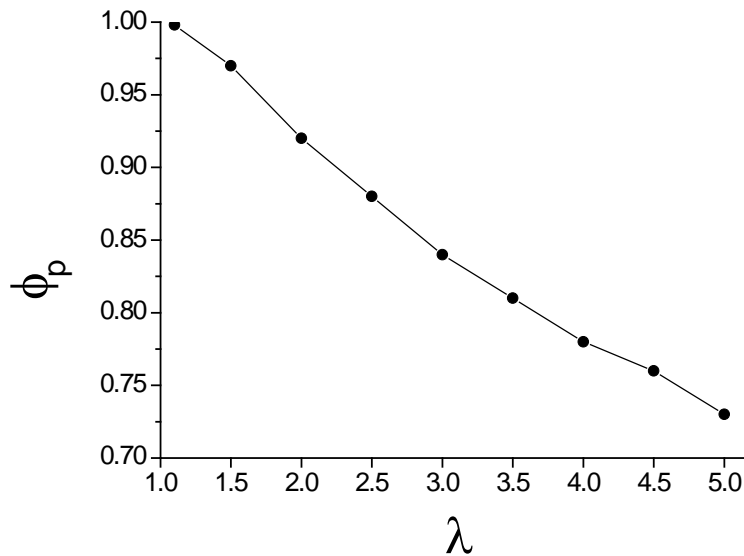


Fig.4 The relationship between sphericity factor ( $\phi_p$ ) and aspect ratio ( $\lambda$ ).

To address this comment, we have added the discussion on uncertainties in the revised manuscript.

**Comment 2: The number of vertical layers in the simulation setting is 20. I think it is very coarse to cover the column atmosphere. How many levels cover the dust layer? There are two domains of 1 coarse domain and 1 nested domain according to Figure 1. Since the nested domain does not include most dust source regions, the dust in the nested domain is from the lateral boundary condition. How does this impact to the simulation and analysis?**

Response: We agree that vertical resolution is key to simulate the long-range transport of dust storms. Previous studies shows that there are two large-scale dust and pollutants transport patterns (Patterns I and II) over Eastern Asia (90°E-140°E) during the spring by a synergetic analysis of both ground-based and spaceborne lidar observations and dust/pollutant transport models (Itahashi et al., 2010). The Pattern I dust layer is 2.5-4 km AGL vertically, and Pattern II is within the PBL (1-2 km thick). The dust event in this study (19-23th March 2010) belongs to Pattern II, as suggested lidar observations in the manuscript.

In this simulation, the model uses 20 terrain-following layers from the surface to 20 km a.s.l, with the lowest 10 layers below 3 km. Table 2 in this reply shows the height of all layers. Compared with previous studies (Li et al., 2011) where only 12 layers from surface to 10km, vertical resolution in dust layers in NAQPMS is more fine, and enables the model to reproduce the profile of dust particles well.

Table 2 Height of 20 layers in NAQPMS model

Layer	Height(m) above sea level
1	50
2	160
3	300
4	470
5	680
6	930
7	1220
8	1570
9	1990
10	2500
11	3110
12	3840
13	4710
14	5760
15	7030
16	8540
17	10350
18	12530

In this study, two nested domains are conducted. The coarse domain (80km) simulates dust emissions and provides dust lateral conditions for the fine domain. The fine domain simulates anthropogenic pollutants more accurately. The analysis on mixing processes between anthropogenic pollutants and dust particles are taken from the simulated results in the fine domain. Because most of dust source regions are not in the fine domain, it is key if dust concentrations in the fine domain from lateral conditions are consistent with observations or coarse domain. Fig.5 in this reply shows dust concentrations in coarse and fine domain. Clearly, simulating PM10 in fine domain is very similar with coarse domain.

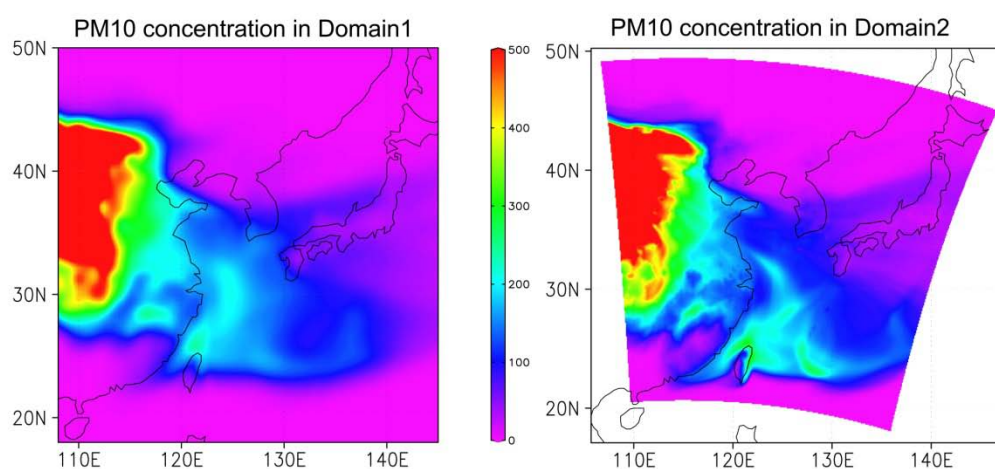


Fig.5 Simulated mean dust concentrations ( $\mu\text{g}/\text{m}^3$ ) in coarse (left) and fine domain (right) during 19-23th March 2010.

**Comment 5: The simulated aerosol validation relies on the bulk mass or aerosol optical depth and therefore there is no constrain the model results especially to the sulfate, nitrate, and Fe in mixture with dust. It must be difficult to have data that distinguishes mixed and non-mixed portion of aerosols but it is worthy showing how model behaves when mixing is allowed and prohibited. I would suggest to conduct an additional simulation without mixing with dust and to compare the result with the current results. For example, adding the no-mixing simulation result in Figure 6 would better show the impact of the mixing with dust.**

Response: We agree and did a sensitivity simulation without mixing with dust as suggested by the reviewer. In the revised manuscript, we show the no-mixing simulation result in Fig.6. As shown in the following figure in this reply, simulated sulfate and nitrate without mixing processes is much less than observations, and show a similar level as anthropogenic aerosols in full simulation. This indicates that our estimated the impact of the mixing with dust is reasonable.

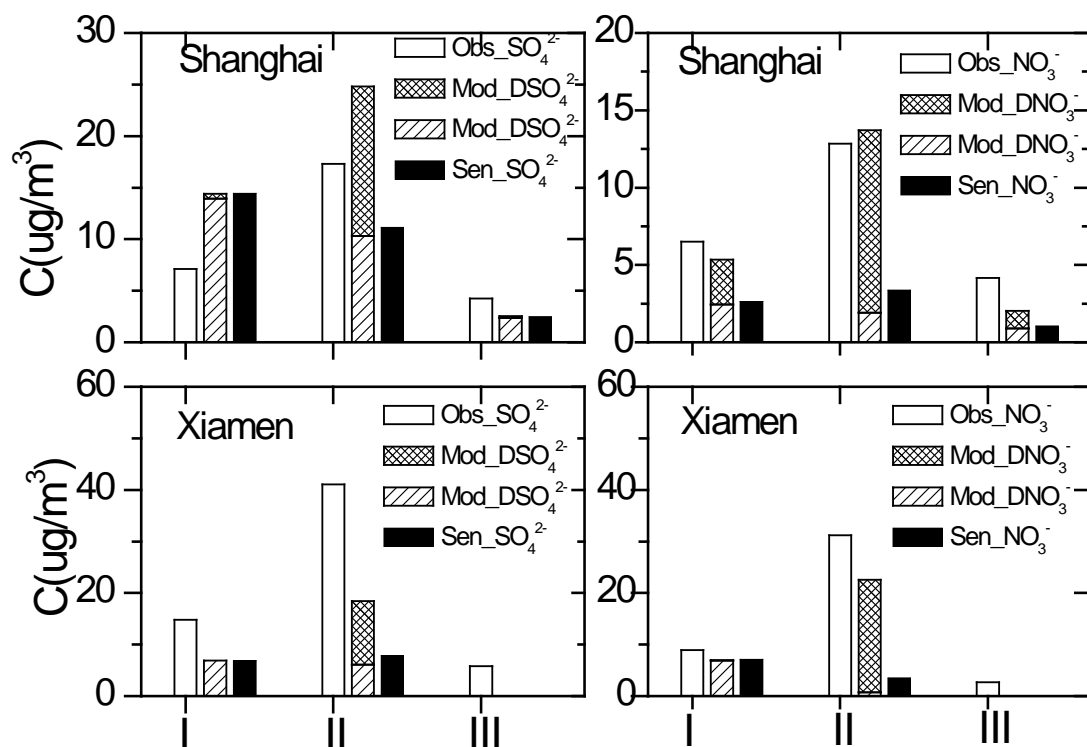


Fig.6 Observed and simulated sulfate (left panel) and nitrate (right panel) at Shanghai (upper) and Xiamen (lower) during pre-dust (I), dust (II) and after-dust (III) period. DSO<sub>4</sub><sup>2-</sup> and DNO<sub>3</sub><sup>-</sup>(same as below) are sulfate and nitrate captured by dust particles by heterogeneous chemistry, respectively. ASO<sub>4</sub><sup>2-</sup> and A NO<sub>3</sub><sup>-</sup> represent the sulfate and nitrate particles by anthropogenic emissions. Sen\_SO<sub>4</sub><sup>2-</sup> and Sen\_NO<sub>3</sub><sup>-</sup> represent simulated sulfate and nitrate in sensitivity simulation (no mixing processes between dust and anthropogenic pollutants). Observations at Xiamen are from Zhao et al. (2011).

**Comment 4: This study is focusing on mixing between dust and anthropogenic pollutants but brief estimation of deposition is presented without thorough examination. I would suggest to delete “: and its impact on regional atmospheric environmental and oceanic biogeochemical cycles over East Asia.” from the title**

Response: We agree and delete “and its impact on regional atmospheric environmental and oceanic biogeochemical cycles over East Asia” in the revised manuscript.

**Comment 5: Page 2744, Line 6: Write Full name of NAQPMS**

Response: We add the full name of NAQPMS. “Nested Air Quality Predicting Modeling System.”



**Comment 6: Page 2745, First Paragraph: It is well known that China is heavily polluted however it is not unique. Several places such as Western US, Mediterranean, Northern India, Korea, and Japan are impacted by heavy air pollution and dust in the globe.**

Response: We delete “unique” in the revised manuscript.

**Comment 7: Page 2746, Line 18: Please change (China) to (East of China) and delete close in “a close downwind region of China”**

Response: We agree and revise it.

**Comment 8: Page 2747, Line 19-27: Please specify how great used several times in sentence.**

Response: We agree. In this revised manuscript, we pointed out that simulated dust emissions by 8 models ranged from 27 to 336Tg in an Asian dust event.

**Comment 9: Page 2748, Line 6-7: Dust impact on nitrate by Yuan et al. (2008) is little which is opposite to other studies. Please add a sentence on that.**

Response: We agree, and revised in the revised manuscript.

**Comment 10: Page 2749, Line 8: Please specify what is “It”, either NAQPMS or WRF.**

Response: it means NAQPMS here.

**Comment 11: Page 2750, Line 2-3: Please provide the optical properties in number.**

Response: We agree. In the “reconstructed extinction coefficient method”, the light-extinction coefficient,  $b_{ext550}$  (expressed as inverse megameters, 1/Mm, at 550nm), was calculated by equation (1):

$$\begin{aligned} b_{ext550} = & 3.0 \times f(RH)\{[(NH_4)_2SO_4] + [NH_4NO_3]\} \\ & + 4.0 \times [OMC] \\ & + 10.0 \times [LAC] \\ & + 1.0 \times [SOIL] \end{aligned}$$

$$+ 0.6 \times [CM] \quad (I)$$

Where the parameters enclosed in the brackets were the mass concentrations of each species. The numbers in front of each species were the optimized specific (or dry mass) extinction efficiency ( $\text{m}^2\text{g}^{-1}$ ). A dry scattering efficiency of  $3 \text{ m}^2/\text{g}$  for sulfates, nitrates and ammoniums was a nominal scattering efficiency based on a literature review by Trijonis et al [1990] and a review by White [1990]. To address the water uptake by hygroscopic species, hygroscopic growth factor or extinction enhancement factor,  $f(\text{RH})$ , was calculated.  $4 \text{ m}^2/\text{g}$  for organic carbon (OMC), and  $1 \text{ m}^2/\text{g}$  and  $0.6 \text{ m}^2/\text{g}$  were the respective scattering efficiencies for soil (SOIL) and coarse mass (CM). The efficiencies for fine soil and coarse mass were taken from a literature review by Trijonis and Pitchford [1987]. We assumed OMC, SOIL and CM were only weakly hygroscopic. A dry absorption cross section of  $10 \text{ m}^2/\text{g}$  for LAC (Light-absorbing carbon) was used from the suggestion by Malm (2000).

**Comment 12: Page 2750, Line 12: “soil dust”. Do you mean “mineral dust”?**

Response: “Soil dust” means “mineral dust”. In the revised manuscript, we change “soil dust” to ‘mineral dust’

**Comment 13: Page 2750, Line 22-25: How to observe  $U^*_0$ ? Why  $U^*_0$  varies by region?**

Response: In the dust models,  $u_*^0$  (threshold friction velocity) represents resistant factor of dust particles from surface to the atmosphere and is the critical factor for dust emission into the atmosphere. When the friction velocity is less than  $u_*^0$ , dust particles cannot be lifted up to the atmosphere.  $u_*^0$  influences the vertical flux and dust transport.

In previous studies,  $u_*^0$  is usually set to a unique value. For example, Westphal et al. (1988) set  $u_*^0$  to be 0.60 in Sahara desert. Recently, Shao et al. (2001) thought that  $u_*^0$  is related to soil type, mineral particle size distribution, surface roughness and soil moisture. This means that there are different  $u_*^0$  in different deserts. In and Park (1996) has used three different values of the threshold friction velocity estimated according to three distinctive surface soil types (Sand, Gobi and Loess soils) in the source regions:  $67 \text{ cm/s}$  for the Gobi,  $50 \text{ cm/s}$  for the Sand and  $40 \text{ cm/s}$  for the Loess. However, these threshold friction velocities are achieved by taking statistical analysis

of physical parameters in the source regions based on routinely available reporting data instead of direct micrometeorological observations.

In this study, we use the direct observed  $u_*^0$  by Zhu et al. (2011) at three deserts in China. This is helpful to improve the model ability in dust storms. Zhu et al. (2011) observed micrometeorological parameters and dust concentrations at high time resolution at towers in three deserts (Table 3). They calculated friction velocities by eddy covariance method or Monin Obukhov similarity theory. When dust concentrations at 3m exceed  $200\mu\text{g}/\text{m}^3$ , the friction velocities are thought as threshold friction velocity. Fig. 7 in this reply shows relationship between and dust concentration and the friction velocity during dust events on Losses Plateau. Clearly,  $0.35\text{ cm/s}$  is the threshold value.

Table 3 Major observational quantities and their respective specification

Metrological parameters	Height (m)	Time Interval	Precision
Wind speed and direction	2,4,16,20	10 min	0.1m/s, 3°
Temperature	2,4,8,16	10 min	0.2°C
Humidity	2,4,8,16	10 min	3%
Sun Radiation	2	10 min	Max:5%
reflected radiation	2	10 min	Max:5%
Net radiation	2	10 min	10 W/(W/m <sup>2</sup> )
Turbulent: wind and temperature	8	10 Hz	0.01m/s 0.01 °C
Dust concentrations	3	10 min	1 $\mu\text{g}/\text{m}^3$

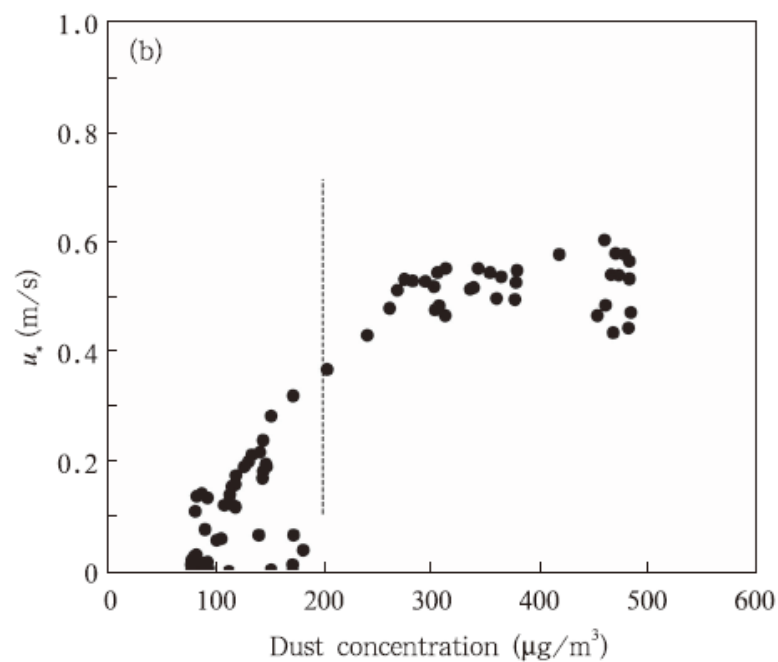


Fig.7 Relationship between and dust concentration and the friction velocity during dust events on Losses Plateau

**Comment 14: Page 2751, Line 11: Please specify radius of black carbon, sulfate, and dust. Also please explain how to handle radius of mixed aerosol.**

Response: In this study, the mean radius and mass density of anthropogenic sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) and black carbon are taken from Tie et al. (2009) (sulfate: 0.24μm, 1.7g/cm<sup>3</sup>; black carbon: 0.04μm, 1.0 g/cm<sup>3</sup>) with a logarithmic normal distribution. Mineral dust (2.65 g/cm<sup>3</sup>) is divided into 4 bins as follows.

Table 4 Aerosol Properties Adopted in NAQPMS

Bins	Radius ranges (μm)	Mean radius (μm)
Bin1	0-1	0.25
Bin2	1-2.5	1.58
Bin3	2.5-5	3.54
Bin4	5-10	7.07

In this study, interactions between the dust and anthropogenic modes can occur *via* gas phase only, so coagulation processes are not accounted for in the model in this study as suggested by Fairlie and Jacob et al (2010). Dust-sulfate and dust-nitrate ranges from 0.43-10μm with 4 bins shown in Table 4. We assume that dust-sulfate and dust-nitrate does not alter the change of mineral dust particles as done in previous study (Fairlie et al., 2010). This assumption is reasonable because concentrations of dust-sulfate and dust-nitrate are much less than mineral dust in each bin. For example, dust-sulfate and dust-nitrate only account for 3-5% of mineral dust particles, which has little effect on the radius of particles.

**Comment 15: Page 2752, Line 21: (OTTE, 2008). Reference is missing.**

Response: We agree and revised it.

**Comment 16: Page 2753, Line 14: Please quantitatively specify how successful the model is.**

Response: We agree. In the revised manuscript, statistical numbers for simulated results with satellite and surface observations are added. In general, correlation coefficients between model and observations range from 0.5 to 0.9. This implies that the model reproduces observed patterns well. Table 5 in this reply shows statistical results.

Table 5 Statistical summary of comparisons of the model results with observations<sup>a</sup>

		$N$	$\overline{C_m}$	$\overline{C_o}$	$r$	$MB$	$RMSE$	$NME$ (%)
AOD550	Modis	5701	0.60	0.65	0.64	-0.05	0.52	56.0
SO <sub>2</sub>	Shanghai	211	8.3	13.1	0.56	-4.8	9.78	42.4
	Beijing	231	15.3	11.1	0.58	4.2	9.36	57.3
	Shanghai	240	13.2	35.9	0.49	-22.0	26.1	62.0

	Beijing	197	21.0	21.2	0.67	-0.2	9.93	42.7
PM <sub>10</sub> <sup>b</sup>	Beijing	9	217.5	236.6	0.91	-19.1	174.9	43.9
	Jinan	9	213.2	160.6	0.82	52.6	203.0	47.6
	Shanghai	9	137.7	182.2	0.94	-44.5	89.4	42.6
	Taipei	117	64.8	70.4	0.80	-5.6	38.5	52.0
	Xiamen	9	116.9	154.7	0.91	-37.8	110.1	67.6
	Nanchang	9	204.7	238.8	0.86	-34.1	129.7	73.5

<sup>a</sup> N is the number of observed samples,  $\overline{C_m}$ ,  $\overline{C_o}$ ,  $r$ ,  $MB$ ,  $RMSE$  and  $NME$  represent the mean modeled and observed values, correlation coefficient, mean bias, root mean square error and normalized mean error. Units of SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>10</sub> are ppbv, ppbv and µg/m<sup>3</sup>, respectively. AOD550 is unitless.

<sup>b</sup> Observed PM<sub>10</sub> is daily mean values, except Taipei which is an hourly mean value.

**Comment 17: Page 2753, Line 18: Please add a brief description on MODIS. Is it dark ocean product or Deep Blue?**

Response: We agree. In the revised manuscript, MODIS, Lidar and API (Chinese Air quality Index) are described in section 3.1. In this study, MODIS data is deep blue product.

**Comment 18: Page 2754, Line 6 and 11: Is it Figure 2 or Figure 5?**

Response: it should be Figure 2. We have revised it in the revised manuscript.

**Comment 19: Page 2754, Line 20-22: Is API in Taipei the same system of China or an independent system? If it is same system why Taipei station has different saturation level?**

Response: In this study, daily mean PM<sub>10</sub> concentrations at 5 stations in China Mainland are converted by Chinese API. PM10 at Taipei is independent from the 5 stations in China mainland. At Taipei, hourly PM10 are directly observed without saturation level. This is why PM10 at Taipei exceeds 600µg/m<sup>3</sup> on 22<sup>th</sup> March 2010.

**Comment 20: Page 2755, Line 22: (personal communication). With Whom? It needs more specific description.**

Response: In the revised manuscript, a more specific description is added. The revised sentence is “However, other observations by Zhang (Zhang, H., Peking University, China) revealed a peak of 3000 µg/m<sup>3</sup> hourly concentrations (unpublished data), which are consistent with this simulation (3300 µg/m<sup>3</sup>).”

**Comment 21: Page 2756, Line 7-20 & Figure 6: See major comment 3.**

Response: we have already revised it.

**Comment 22: Page 2757 and Figure 7: Figure 7 is unclear and hard to follow. Please improve the figure presentation.**

Response: We agree and revised it.

**Comment 23: Page 2758, Line 5: Please specify the period.**

Response: In this study, we used the method by Tang et al. (2004) to classify dusty periods. When the simulated total dust are greater than  $100\mu\text{g}/\text{m}^3$ , we define it as dusty periods. So it's clear that the dusty period at different sites depends on the arriving and residence time of the dust event. For example, For Beijing city, a megacity in northern China, the dusty period was 14:00 (Beijing Time) 18<sup>th</sup> March 2010 – 09:00 20<sup>th</sup> March 2010. For Xiamen, the dusty period was 07:00 (Beijing Time) 21<sup>th</sup> March 2010 – 16:00 23<sup>th</sup> March 2010.

**Comment 24: Page 2758, Line 12: What is the range of uncertainty due to the error in the reaction HR12 in Table 1.**

Response: We discussed it in response to comment 1.

**Comment 25: Page 2758, Line 22-25: Please clarify. It looks a sudden jump to me. What is the uncertainty of the estimated Fe (II)? Is the fraction by mass or volume?**

Response: We reworded it and moved it to section 4.2 in the revised manuscript. It is mass fraction.

**Comment 26: Page 2759, section 4.2.1: The manuscript not shows discussion about NO<sub>2</sub>. Please add it.**

Response: Impact of dust on surface NO<sub>2</sub> is shown in Fig.9 in the revised manuscript. In this study, surface NO<sub>2</sub> are decreased 5-60% due to the heterogeneous reactions (HR13). The band of large decrease is in the western Pacific which is consistent with SO<sub>2</sub>.

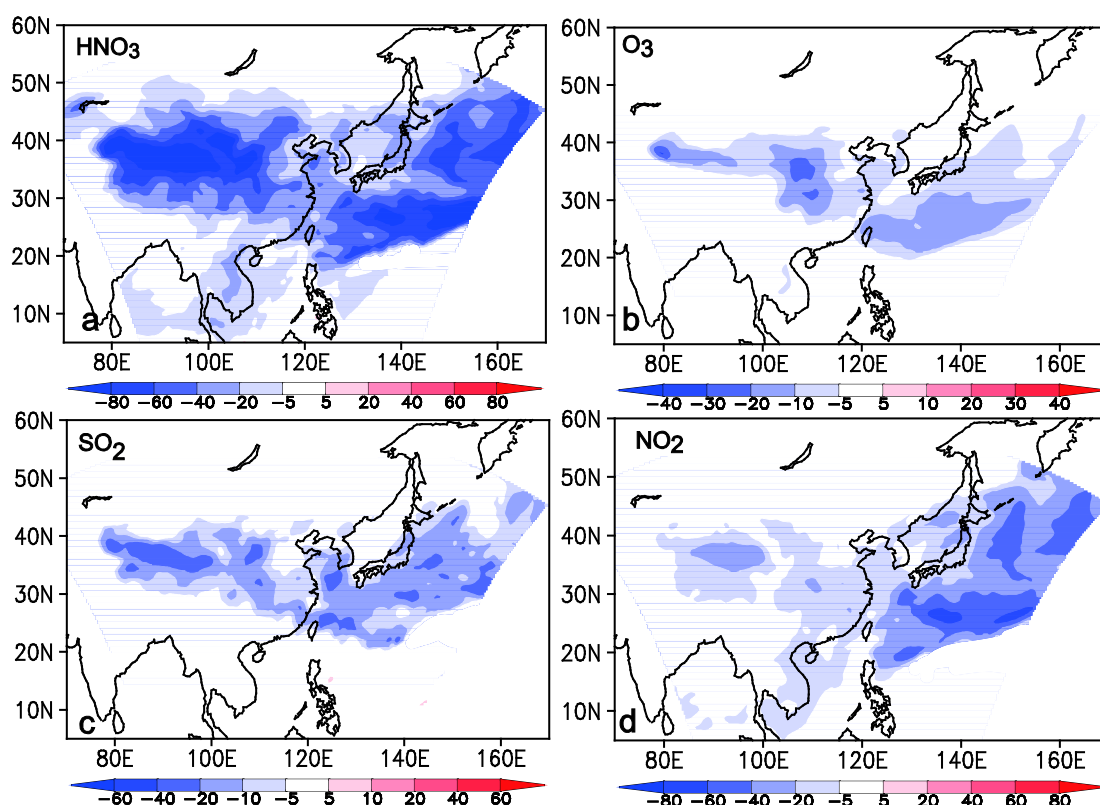


Fig. 8 Averaged dust influences (%) on surface  $\text{HNO}_3$  (a),  $\text{O}_3$  (b),  $\text{SO}_2$  (c) and  $\text{NO}_2$  (d) concentrations over East Asia during 19–22 March 2010

**Comment 27: Page 2759, Line 25: Please add reference.**

Response: We agree and revise it.

**Comment 28: Page 2758, Line 20-24: The dust event is the largest in the history but the Fe is comparable with observation. Please clarify why it is.**

Response: Thanks a lot for your comment. The introduction section in this study affirms that this event on 19-23<sup>th</sup> March 2010 is very special in all dust events because of its transport pathway. In history, transport pathways of most dust events include three types: U type, L type and D type (Tsai et al., 2008). As shown in Fig.9a-c in this reply, they usually occurred in northern China (the north of 30°N), South Korea and Japan. However, dust particles in this event were transported towards to the southern China, even reached the southernmost tip of China. Beijing, Shanghai, Hongkong and Dongsha (Fig. 9d in this reply) observed high  $\text{PM}_{10}$  concentrations (more than  $500\mu\text{g}/\text{m}^3$ ). This means that more regions in China are influenced by this dust storm.

Although there was a larger range of influence, the mass concentrations in this event kept the same level with previous dust events. For example,  $\text{PM}_{10}$  concentrations Beijing in this study was 1000-2000  $\mu\text{g}/\text{m}^3$ , which was similar as observations in 2003 ( $1600\mu\text{g}/\text{m}^3$ ) (Zhuang et al., 2001). This is why Fe in this study is

comparable with observations.

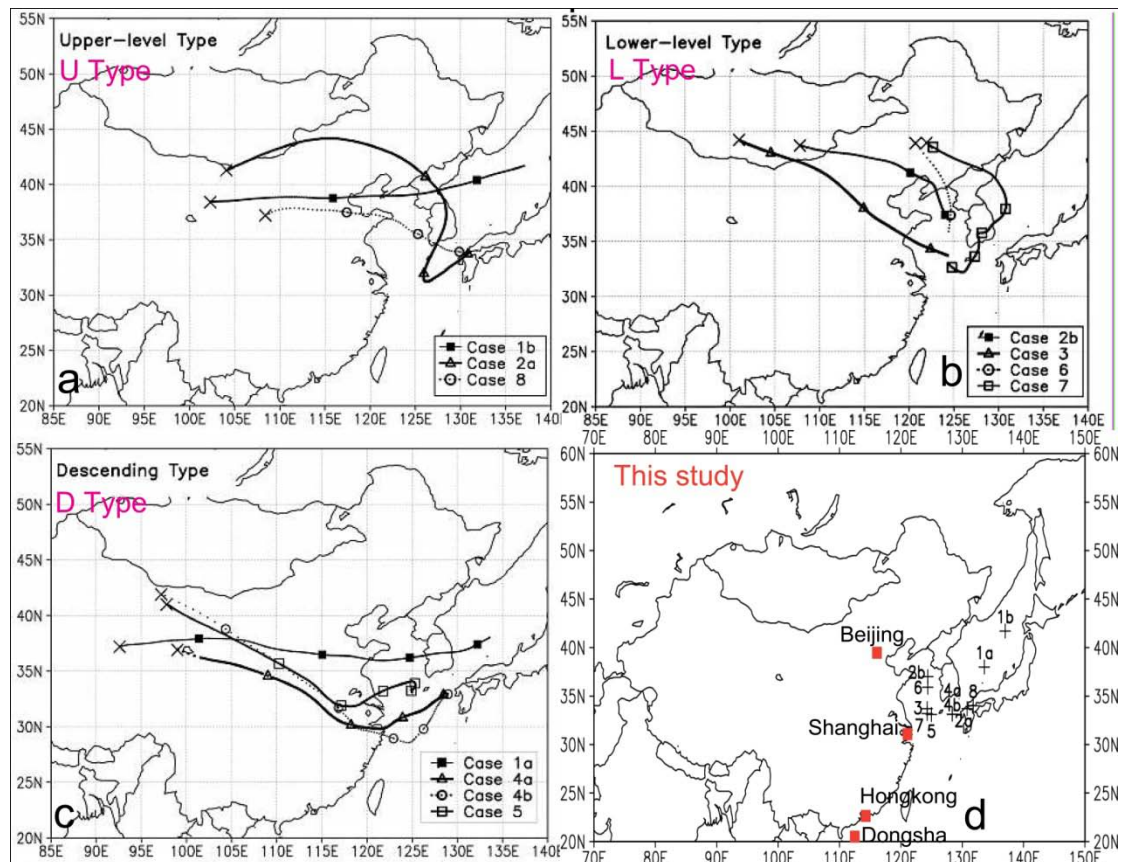


Fig.9 Transport pathways of dust studies in ACE-Asia (a, b, and c) (Tsai et al., 2008)

### Comment 29: Figure 3: What are the mass by dust?

Response: Fig.10 in this reply shows the masses of dust and anthropogenic aerosols. Clearly, dust particles covers more than 85% of total PM10 masses at all sites during dusty days. The estimated mass fraction of dust particles is consistent with previous observations, where soil dust contributed 80-95% of total aerosols (Yuan et al., 2008).



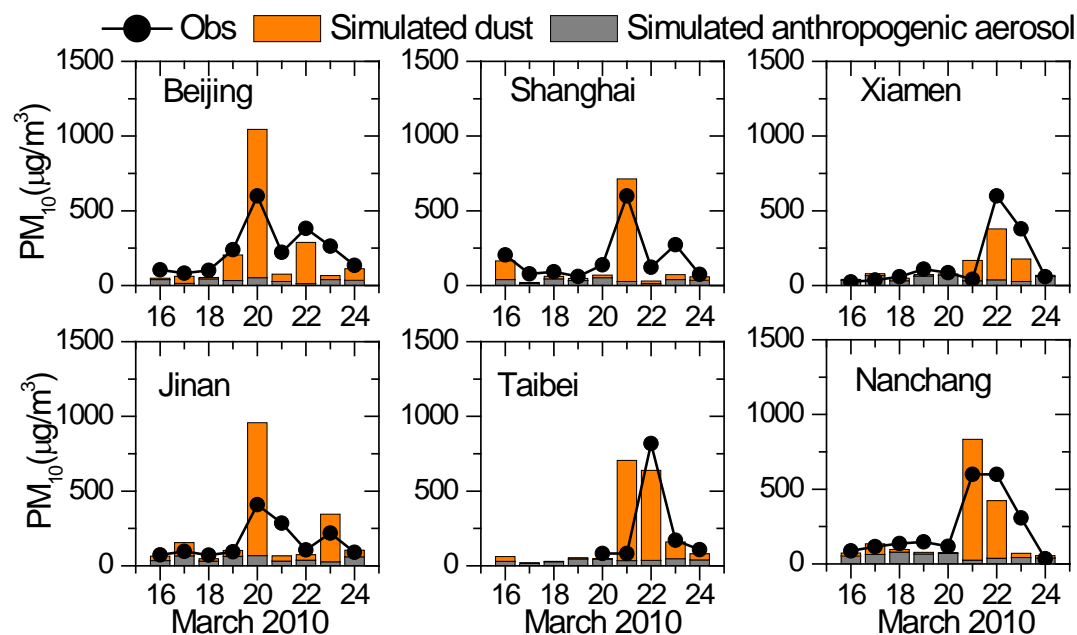


Fig.10 observed daily mean  $PM_{10}$  ( $\mu g/m^3$ ) and simulated daily mean dust and anthropogenic aerosols ( $\mu g/m^3$ ) in  $PM_{10}$  at six stations in China during 15–24 March 2010.

**Comment 30: What is the unit?**

Response: Here AOD is unitless.

**Comment 31: Figure 5: I see diurnal pattern even during dust period. Does that mean the impact of dust is negligible?**

Response: Fig.11 in this reply shows simulated  $SO_2$  mixing ratios at Shanghai in full and no mixing simulations during 18–22 March 2010. It's found that dust decreases 3–8 ppbv  $SO_2$  concentrations. In particular, dust suppresses the increase of  $SO_2$  during 0:00–12:00 LST 20<sup>th</sup> March.

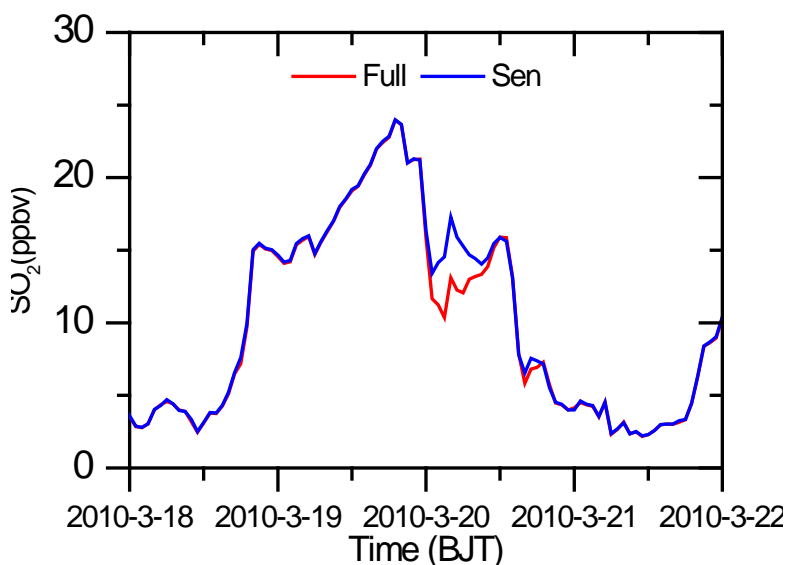


Fig.11 Simulated  $\text{SO}_2$ (ppbv) at Shanghai in full and sensitive (no mixing of dust) simulation during 18<sup>th</sup> -22<sup>th</sup> March 2010.

**Comment 32: Figure 7: To complicate and unclear. What are the colored lines? Please improve the figure and caption.:**

Response: We appreciate this comment. The blue contours represent the  $\text{SO}_2$  (a, c, d and f) and  $\text{NO}_x$  (b and e) emission rate ( $\mu\text{gm}^{-2}\text{s}^{-1}$ ). The lines in colored lines represent the locations of air masses arrived at Shanghai and Xiamen during dusty days. They are calculated by the Hysplit back-trajectories model. The lines reveals where air masses in dusty days come from. For example, Fig. 7a reveals dust events at Shanghai came from Mongolia. The colors in colored lines represent the concentration of sulfate (a and d), nitrate (c and e) ( $\mu\text{gm}^{-3}$ ) captured by dust particles by heterogeneous chemistry and the mass fraction of dissolved Fe (e and f). For example, in Fig.7a, sulfate captured by dust particles by heterogeneous chemistry in Mongolia are only  $0.4\text{-}0.5\mu\text{gm}^{-3}$ . At Shanghai, sulfate reaches  $15\text{-}20\mu\text{gm}^{-3}$ .

**Comment 33: Figure 9: How about  $\text{NO}_2$ ?**

Response: We already added in the revised manuscript. The details can be found in the reply to Comment 26.

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