We thank Prof. C.-H. HO for his constructive comments.
Response to the Specific comments by Prof. C.-H. HO

General comment: This paper has examined the mixing degree between mineral dust and anthropogenic pollutants, and the associated regional atmospheric environmental and oceanic biogeochemical cycles in the duststorm event using a NAQPMS model. The model results are validated with satellite and ground observations, which is comparable each other. Consequently, the authors suggested that the mixing processes result in increasingly polluted particles, which has a considerable impact on the regional-scale atmospheric composition and oceanic biogeochemical cycle. While the paper is well organized, some aspects of the paper are not clear. For potential publication in ACP, some answers to following comments need to be required.

Response: We greatly thank Prof. Ho for his encouragement and insightful comments concerning the manuscript. In the revised manuscript, we have already considered all comments and add the discussion on the description of observations (satellite, lidar et al.,) and model. Particularly, quantitative statistics results between model and observation were provided.

Comment 1: In the paper, the authors used several observational data (e.g., MODIS, API, Lidar) to validate the model performance. It would be preferable to have a section that describes all data used.

Response: We agree that observations should be described in the revised manuscript.

In this study, the Moderate Resolution Imaging Spectroradiometer (MODIS) field of AOD at 550 nm was obtained from the MODIS/AQUA Deep Blue Collection 005. The daily level 3 AOD product at 550 nm (Ver5.1) was retrieved as an average value for an area defined by a $1^o \times 1^o$ square cell.

The Lidar data at Nagasaki, Chiba and Hedo was acquired from the National Institute of Environmental Studies (NIES) lidar network (http://www-lidar.nies.go.jp/). Vertical profiles averaged for 5 min are recorded every 15min. The height resolution was 6m and the profiles are recorded up to 6-km.

The definition of Chinese API and conversion between API and PM_{10} mass concentrations have also been added in the revised manuscript (Equation 4 in the new version).

Comment 2: The authors estimated aerosol optical depth (AOD) by the converted method proposed in Malm et al. (2000). It is questionable how exact or uncertain the method to estimate AOD.

Response: Thanks for this comment. In theory, AOD is calculated by integrating the aerosol extinction coefficient with respect to altitudes. In most air quality models, there are two methods to estimate extinction coefficient from the simulated particulate concentrations. The first is based on the Mie-Theory in which the coefficient is calculated as a function of density of particulate species, mass of particulate species, and extinction efficiency of particulate species and effective radius of particulate species. The second is called as "reconstructed extinction coefficient method" and was used in this study. This method is proposed by Malm (2000) as a part of the Integracy Monitoring of Projected Visual environment (IMPROVE) program.

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):

$$\begin{aligned} b_{ext550} = {} & 3.0 \times f(RH)\{[(NH_4)_2SO_4] + [NH_4NO_3]\} \\ & + 4.0 \times [OMC] \\ & + 10.0 \times [LAC] \\ & + 1.0 \times [SOIL] \\ & + 0.6 \times [CM] \end{aligned} \qquad (1)$$

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 (m^2g-1). A dry scattering efficiency of 3 m^2/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(RH), was calculated. 4 m^2/g for organic carbon (OMC), and 1 m^2/g and 0.6 m^2/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 m^2/g for LAC (Light-absorbing carbon) was used from the suggestion by Malm (2000).

We agree that none of both methods is superior. The Mie-theory is able to estimate accurate AOD, but requires more aerosol microphysical properties mentioned in above paragraph. In particular, the size distributions of different type of aerosols are very important because of their impacts on extinction efficiency and effective radius. However, it's difficult for current 3-D models to provide accurate aerosol microphysical properties in East Asia. For example, the consideration of NH_4NO_3 is of primary importance in East Asia, but its size distribution has not been sufficiently characterized to enable the use of Mie-theory calculation (Song et al., 2008). So the reconstructed extinction coefficient method was recommended in East Asia by Song et al. (2008) rather than Mie-theory.

An uncertain of the reconstructed extinction coefficient method is the assumption that the aerosol characteristics in East Asia are the same as those in USA. To reduce errors caused this assumption, extinction enhancement factor, f(RH), was calculated

based on observations at a site in Central Eastern China during April-May 2006 by Pan et al. [2009] and Liu et al. [2007].

Another uncertain is the assumption of external mixture. Although external mixture was the dominant mixture state in CEC (80-90%), internal mixture or coating mixture contributed 20-40% absorption. The coating soot can enhance the light extinction. Pure external, internal or coated mixture can lead to a discrepancy with the reality (Cheng et al., 2009).

The reconstructed extinction coefficient method has been widely applied in many 3-D chemical transport models (MM5-CMAQ, RAMS-CMAQ and NAQPMS) (Song et al., 2008; Han et al., 2010) and showed a good performance in East Asia, particularly in China (Han et al., 2010). In a 1-year simulation in East Asia by CMAQ (Song et al., 2008), estimated AOD shows good correlations with MODIS satellite in Central East China, South China, Korea and Japan, with correlation coefficients ranging from 0.43 to 0.73. At 20 sites in China, correlation coefficients between ground observations and simulations reach 0.4-0.8.

Comment 3: In Fig. 1, there is no a value for weighting factor over other regions except the Gobi desert. Isn't there dust loading in the regions? Additionally, it is necessary to describe weighting factor determined by soil or land type. For example, what is the range of weighting factor? What is a meaning for its small or large values?

Response: We agree that weighting factor should be described clearly. In this study, weighting factor (E) represents the uplifting capability of land surface, and reflects the impact of landuse categories, vegetation fractions and snow/ice coverage on dust fluxes. In the desert without vegetable and snow cover, dust particles are easily uplifted to the boundary layer, and E is set to 1.0. On the contrary, E is set to 0,0 in evergreen forest.

E is calculated by Equation (1) in this reply:

$$E = A_{soil} A_{veg} A_{s n o} \quad (2)$$

where A_{soil} represents the emissions potential in ratio to that of desert soils, which is listed in Table 1 in this reply. A_{veg} represents the fraction of vegetable in the entire grid cell. A_{snow} is a coefficient reflecting the influence of snow. When the snow amounts is more than 5 kg/m^2, A_{snow} is set to 0.0.

Table1 Ratio factor of potential emission coefficient in this study

Landuse Description	A_{soil}
Evergreen Needleleaf Forest	0.0
Evergreen Broadleaf Forest	0.0

Deciduous Needleleaf Forest	0.0
Deciduous Broadleaf Forest	0.0
Mixed forests	0.0
Closed Shrublands	0.1
Open Shrublands	0.1
Woody Savannas	0.3
Savannas	0.3
Grasslands	0.3
Permanent Wetlands	0.0
Croplands	0.0
Urban	0.0
Cropland Vegetation Mosaic	0.0
Snow and Ice	0.0
Barren or Sparsely Vegetated	1.0
Water	0.0
Wooded Tundra	0.0
Mixed Tundra	0.0
Barreb Tundra	0.0

In this study, soil categories, vegetation fractions and snow/ice coverage are derived from Moderate Resolution Imaging Spectrometer (MODIS) data. Fig.1 in this reply shows the distribution of weighting factor in this study. Fig.2 shows Desert and sandy land of North China and Mongolia. Clearly, weighting factor covers both Gobi deserts in Northwest China and Mongolia (e.g. Taklimakan, Gurban Tonggutm Kumtag, Qaidan, Badain Jaran deserts, Otindaq Sandy land) and some other regions like grasslands (the north of Mongolia et al.). In Evergreen forests and croplands in east China, there is not dust loading.

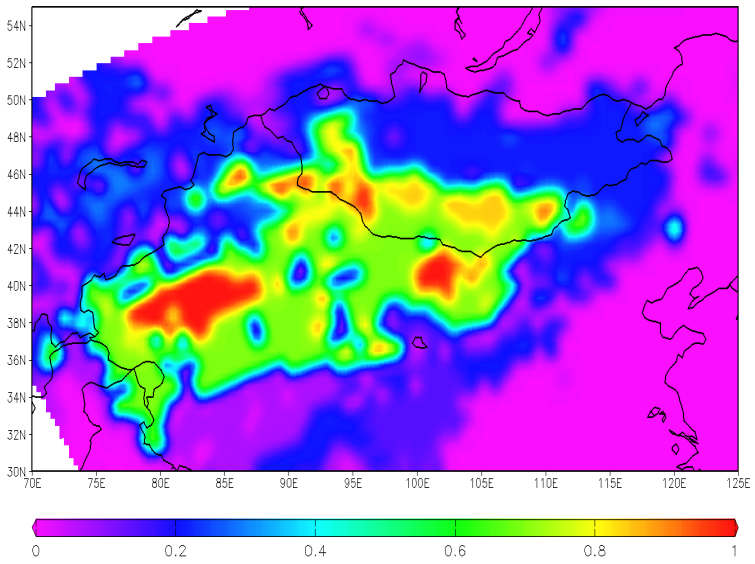

Fig.1 The distribution of weighting factor in this study

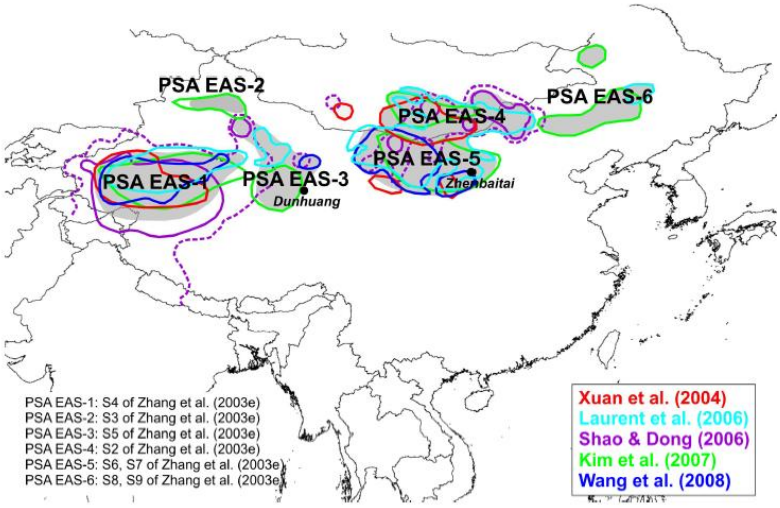

Fig. 2 Potential source areas in Eastern Asia (Formenti et al., 2011)

Comment 4: The authors explained that the reason for significantly overestimated model result of fine particles in Shanghai is due to locally re-suspended dust particles by strong wind. I thought it would be required to show wind fields.

Response: We totally agree. As shown in Fig. 3 and table 2 in the manuscript, the simulated Shanghai PM_{10} on 23th March 2011 underestimated the observed values. This underestimation caused by locally re-suspended dust particles made the fraction of fine particles were overestimated in the simulation. Fig. 3a and 3b in this reply shows the spatial distribution of simulated surface wind velocity on 8:00am 23th March and its time series at Shanghai. Clearly, strong wind on 23th March prevailed at Shanghai and its surrounding regions. This could uplift local deposited dust particles at Shanghai.

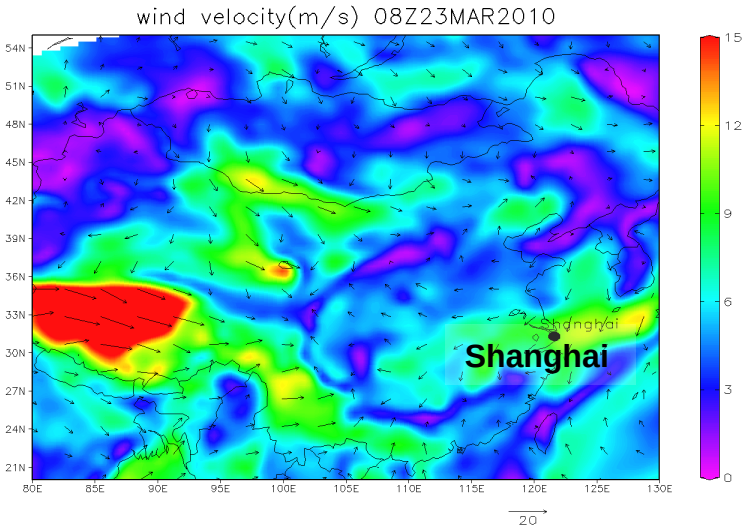

Fig.3a Spatial distribution of simulated surface wind velocity (m/s) at 08:00am 23th March 2010

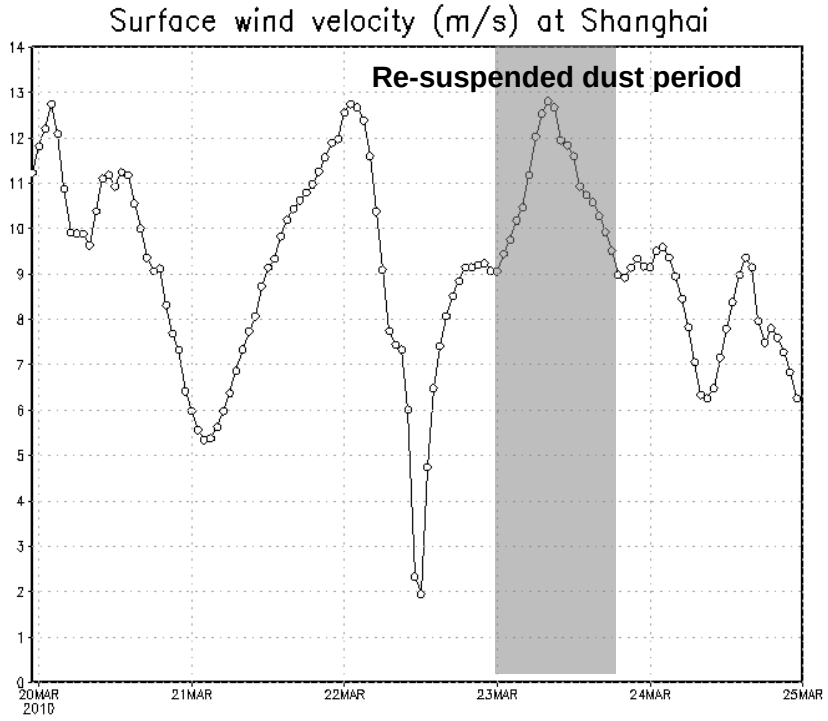

Fig.3b simulated surface wind velocity at Shanghai during 20th -24th March 2010

Comment 5: According to the paper, it is agreeable that the overall patterns are well matched between model results and observations. However, it is also important to determine whether quantitative range between the two values.

Response: Thanks for this insightful comment. In this revised manuscript, a series of statistical measures of agreements or disagreements between the simulation and observation are made (Table 1 in this reply).

Table 1 Statistical summary of comparisons of the model results with observations[a]

		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_2	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
NO_2	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_{10}[b]	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

[a] 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_2, NO_2 and PM_{10} are ppbv, ppbv and μg /m^3, respectively. AOD550 is unitless.

[b] Observed PM_{10} is dauly mean values, except Taipei which is an hourly mean value.

Comment 6: In the Fig. 8, is there any reason for the high concentration of DNO3/NO3 over the Gobi desert with relatively low RH conditions?

Response: In this study, DNO_3/NO_3 means the fraction of nitrate captured by dust particles in all nitrates (the sum of transformation to HNO3 on the surface of the dust aerosol and anthropogenic nitrate). Compared with DSO_4/SO_4 (10-20%) in Gobi regions, the fraction of nitrate on dust particles is much higher in Gobi region (70-80%). This is mainly caused by two factors. Firstly, the reaction probability of HNO_3 on dust is much higher than SO_2, even in Gobi regions. Fig.4 in this reply shows the reaction probability of HNO_3 as a function of relative humidity. Clearly, the reaction probability of HNO_3 (γ_{hno3}) ranges from 0.006 to 0.015 in Gobi RH conditions (10-20%), which is 60-150 times higher than γ_{so2} (0.0001, HR19). This means much more HNO_3 are converted into nitrate than the conversion of SO_2 into sulfate. In this study, 60-80% HNO_3 in Gobi regions is captured by dust particles, which is consistent with previous studies (Tang et al., 2004). Secondly, besides anthropogenic sulfate and mixing between anthropogenic SO_2 and dust particles by heterogeneous chemistry, the SO_4^{2-} is partially from the "crustal" source, i.e. from the emitted mineral dust (Wang et al., 2011). The sulfate from the emitted primary dust in the desert largely decreases DSO_4/SO_4.

Finally, although DNO_3/NO_3 remains high levels, the absolute concentrations of DNO_3 and NO_3 (μg/m^3) are still low in Gobi region than east of China.

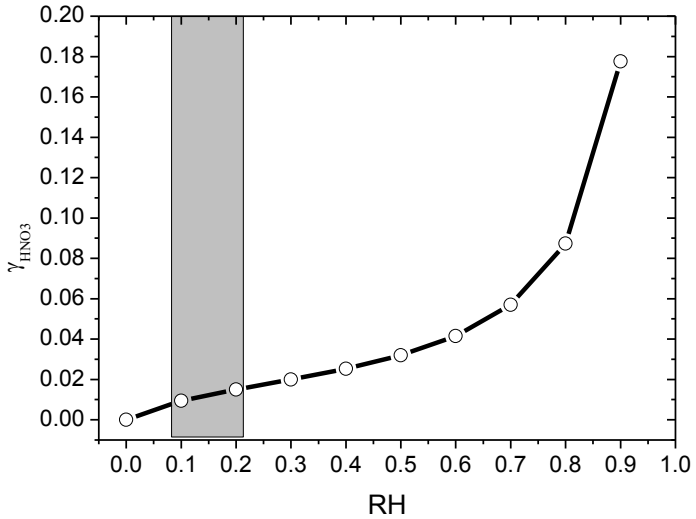

Fig.4 Reaction probability of HNO_3 to dust particles as a function of relative humidity. The gray shade represents the RH ranges in Gobi region.

Comment 7: In section 4.2, due to the mixing between dust and anthropogenic aerosols, the concentration of HNO3, SO2, and O_3 is decreased. What do the authors think about the meaning of these decreasing trends in view of atmosphere in the region?

Response: Thanks for this insight comment. The importance of heterogeneous reactions involving SO_2, NO_2, HNO_3 and O_3 on mineral dust surfaces needs to be evaluated in view of regional atmospheric environment.

From a photochemical perspective, heterogeneous chemistry on mineral dust surfaces that directly affects gaseous (NO_2, HNO_3 and O_3) is of great importance .By altering the complex set of photochemical reactions of O_3 and its precursors, the heterogeneous pathway perturbs photochemical oxidant (OH and HO_2) and thus atmospheric oxidation capacity(Bian et al., 2003). Furthermore, the heterogeneous pathway may impact the self-cleaning of many atmospheric primary pollutants (VOCs) and formation of secondary pollutants. In this study, mineral dust decreased HNO_3 and NO_2 concentrations by up to 80%-90% and 40%, respectively. Indirect ozone due to HNO3 uptake likely exceeds direct heterogeneous uptake of O_3. This indicates that heterogeneous chemistry on mineral dust surfaces may be of importance in the chemical balance of the troposphere over East Asia.

The chemical interactions between the gas and dust particles are able to result in increased amounts of sulfate and nitrate in the aerosol, and a change in size distributions of these anthropogenic pollutants. Under non-dust conditions, the sulfate formation is controlled by the nucleation of H_2SO_4 and H_2O and coagulation processes, and then concentrates in the fine particles. At a large dust loading, the sulfate formation due to surface uptake becomes dominant and its mass exhibits a maximum in coarse mode. As shown in Fig.8 in the manuscript, sulfate coating dust particles by heterogeneous reactions covered 60-80% of total sulfate in this study. This would indicate that dust particles shifted sulfates from fine to coarse mode over Eastern China and Pacific in this study. The coating processes also leads dust particles an increase in their hygroscopicity. The change of size distributions of sulfate and nitrate and hygroscopicity could alter aerosol optical properties, direct radiative forcing characteristics and has important implications for regional climate in East Asia (Tang et al., 2004; Liao et al., 2005). Previous study showed that coating or mixing of dust particles by hygrocopic species with increasing their solubility significantly enhances dust contribution to global CCN.

Comment 8: In the figures showing map (e.g., Figs. 2, 4, 7, 8, 9, and 10), the size of characters denoted on the x- and y-axis is somewhat small.

Response: We revised it.

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