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

General comment: This manuscript does not sound scientific because, described in this manuscript; the NAQPMS is used to calculate gas chemistry. NAQPMS employs an aerosol thermodynamic model (ISORROPIAI1.7) to calculate the composition and phase state of an ammonia-sulfate-nitrate-chloride-sodium-water inorganic aerosol. However, during the dust event, huge amounts of particles in large size existed in the atmosphere, the gas-aerosol. Equilibrium definitely cannot be achieved. Forcing that the gas-aerosol equilibrium has been achieved would result in more nitrates partitioning in the particulate phase, further increases the amount of liquid water and eventually promotes sulphate formation and Fe solubility. This reviewer suggested that the authors should choose a model in which the gas-particle kinetic equations are used to calculate the mass transfer.

Response: Thanks for this comment. The reviewer focused on the treatment of our model (NAQPMS) on dust and anthropogenic aerosols. So we firstly describe the details of aerosol module which was not described at full length. We are sorry for confusing the reviewer in old manuscript.

As shown in Fig.1 in this reply, five categories of aerosols are explicitly treated in our model: 1) anthropogenic aerosols (sodium-chloride-sulfate-nitrate -ammonium) 2): dust particles; 3) sea salt, 4) dust-sulfate and dust-nitrate, 5) sea salt-nitrate. The aerosol and gas gases are coupled through 1) gas chemistry module, 2) aqueous chemistry module, 3) heterogeneous chemistry module, 4) gas-aerosol partitioning of inorganic compounds.

In anthropogenic aerosols, sulfate, nitrate and ammonium are assumed to be internally mixed, meaning that all particles of the same size have the same composition. In this category, sulfate, nitrate and ammonium are in the form of NaCl, Na_2SO_4, $NaNO_3$, NH_4NO_3, $(NH4)_2SO_4$, NH_4Cl, NH_4HSO_4, $NaHSO_4$ and $(NH_4)_3H(SO_4)_2$). They are in fine mode with a lognormal size distribution. Anthropogenic aerosols are external mixed with the other categories of aerosols (dust and sea salt). Previous studies shows that the external mixture of anthropogenic aerosols with dust or sea salt is reasonable (Maxwell-Meier et al., 2004; Tang et al., 2004; Song et al., 2005).

Dust and sea salt are calculated by the dust and sea salt module described in the old manuscript. In this study, 4 bins are divided (0.43-1μm, 1-2.5μm, 2.5-5μm and 5-10μm).

Dust-sulfate and dust-nitrate (DSO_4 and DNO_3) are defined by Fairlie and Jacob (2010), and represents sulfate and nitrate on the surface of dust particles by heterogeneous chemistry. Uptakes of acidic gases on alkaline components (Ca^{2+} and Mg^{2+}) are the pathway of DSO_4 and DNO_3 formation. Formulation (1) shows the DNO_3 formation pathway (Kelly and Wexler et al., 2005). Reaction (1) is reversible in theory,

but the calcium nitrate is often associated with water molecules for relative humidity in the range of 9 and 50% with n=1, 2, 3 or 4 in usual atmospheric conditions. Such hydrous components are very stable and in most of the cases the forward direction of reaction (1) is preferred (Kelly and Wexler, 2005). So formulation (1) in simplified as formulation (2) which is irreversible (Sun et al., 2005).

$$2HNO_3(g) + CaCO_3(s) + nH_2O \leftrightarrow Ca(NO_3)_2(s)(nH_2O) + H_2O + CO_2(g) \qquad (1)$$

$$2HNO_3(g) + CaCO_3 \rightarrow Ca(NO_3)_2(s) + H_2O + CO_2(g) \qquad (2)$$

Clearly, DNO_3 are in forms of stable $Ca(NO_3)_2$. This means that these heterogeneous reactions prefer to be irreversible. So they are assumed to reside in the aerosol phase only.

In this study, DSO_4 and DNO_3 on each dust bin are calculated by heterogeneous chemistry module with a kinetic approach, which is distinct from gas-aerosol partitioning module where reactions are reversible. Same as dust particles, DSO_4 and DNO_3 also have 4 bins. These dust-nitrate and dust-sulfate coat mineral dust particles by a shell on the surface of dust particles, and experience the same transport and deposition processes as the corresponding dust particles. This method in this study is also applied in the study of Fairlie and Jacob et al. (2010) where GEOS-CHEM is applied to check the impact of dust on sulfate and nitrate.

Sea salt nitrate are internal mixed with sea salt and calculated by HR27 and HR28 in table 1 in the manuscript.

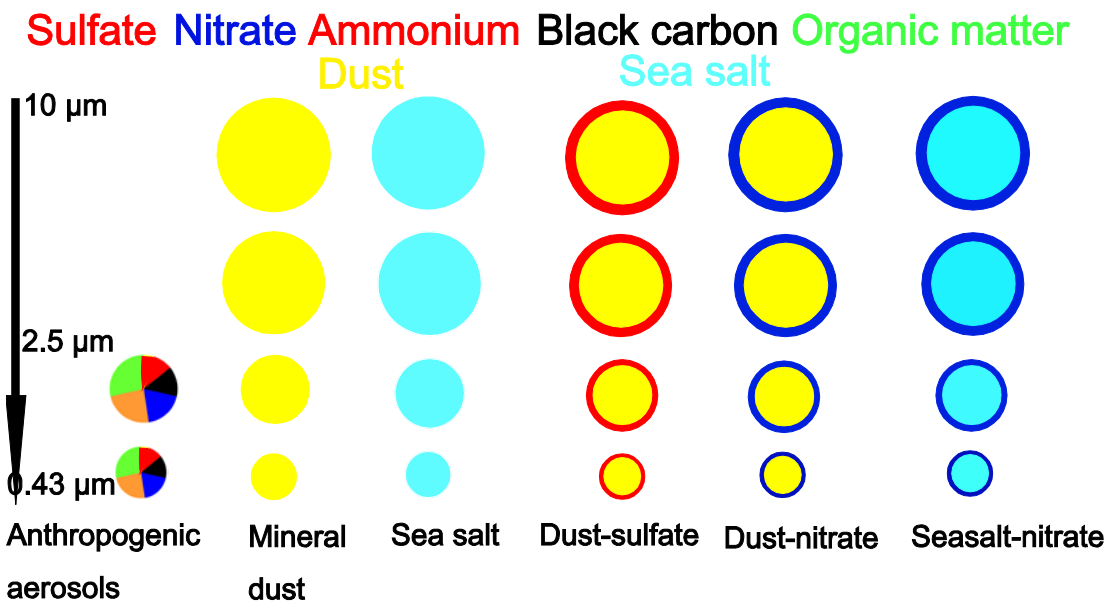

Fig.1 Five categories of aerosols in this study and their size ranges

In this study, anthropogenic aerosols are calculated by an aerosol thermodynamic model (ISORROPIAI1.7). Quite a lot studies show that a gas-aerosol equilibrium for volatile species determines the gas-aerosol partitioning process. ISORROPIAI1.7 deals with thermodynamic equilibrium among NaCl, Na_2SO_4, $NaNO_3$, NH_4NO_3, $(NH4)_2SO_4$, NH_4Cl, NH_4HSO_4, $NaHSO_4$, $(NH_4)_3H(SO_4)_2$, and successfully

reproduces this process in wide simulations (Nenes et al., 1998). As stated by the reviewer, large particles might exist in nonequilibrum transition states and break this equilibrium (Meng et al., 1996). The large particle requires more time to reach the final equilibrium state. However, this might not affect estimated results in this study, because:

1) What ISORROPIAI1.7 in this study deals with is the equilibrium of NaCl, Na_2SO_4, $NaNO_3$, NH_4NO_3, $(NH4)_2SO_4$, NH_4Cl, NH_4HSO_4, $NaHSO_4$ and $(NH_4)_3H(SO_4)_2$. During the dust periods, particles in large sizes are mostly dust particles which are consisted of $CaCO_3$, $MgCO_3$ and Al_2O_3. These non-volatile and stable compounds don't involve the gas-aerosol equilibrium in this model.

2) In this study, dust-sulfate and dust-nitrate are calculated by a heterogeneous module with a kinetic approach assuming a total reaction in the forward direction. Dust-nitrate is in forms of stable $Ca(NO_3)_2$ and assumed to reside in the aerosol phase only. In the model, $Ca(NO_3)_2$ are externally mixed with anthropogenic aerosols and does not involve in the equilibrium system in ISORROPIAI1.7. This treatment is consistent with observations. 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). The work of Fairlie and Jacob et al. (2010) also support uptakes of HNO_3 and SO_2 on bulk minerals do not involve in the rapid sodium-chloride-sulfate-nitrate-ammonium equilibrium.

3) 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). 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).

Overall, the anthropogenic sulfate and nitrate (NH_4NO_3, $(NH4)_2SO_4$ etc) are calculated by ISORROPIAI1.7 model in this study, and mixed sulfate and nitrate (e.g. $Ca(NO_3)_2$) between mineral dust and reactive gases (HNO_3 , SO_2 etc.) are calculated by the heterogeneous chemistry module with a kinetic approach assuming a total reaction in the forward direction described in section 2.3 in the manuscript. The mixing process does not involve in the gas-aerosol equilibrium. This method has also been widely applied in other regional and global models like STEM, CHIMERE, GEOS-CHEM, MOZART and LMDz-INCA to investigate the interactions between Asian mineral dust and anthropogenic pollutants (Fairlie et al., 2000; Bauer et al., 2004; Tang et al., 2004; Tie et al., 2005; Hodzic et al., 2006).

Comment 1: what does it mean "complicated air pollution?"

Response: In the past several decades, air pollution in China was caused mainly by the burning of coal as fuel for industrial and domestic purposes; sulfur dioxide (SO_2) and particulate matter were among the major pollutants. Unfortunately, owing to rapid

urbanization and industrialization, before the existing problems caused by coal combustion could be resolved, other emission sources have become increasingly important. The rapid increase in the number of vehicles in some Chinese megacities and economically developed regions, such as Beijing, Shanghai, Guangzhou, Pearl River delta, and Yangtze delta, has led to a sharp increase in concentrations of nitrogen oxides (NO_x), volatile organic compounds (VOCs), particulate matter (PM), and ozone. The combination of coal smog and traffic exhaust results in serious pollution characterized by high levels of photochemical smog, high concentration of fine particles, and poor visibility. **We call the coexistence of numerous air pollutants with high concentrations and the complicated interactions among them as "complicated air pollution". Air pollution of this kind causes both local and regional problems.**

Comment 2: This reviewer suggested the authors presented more analysis and discussion on their results, now the important part which this reviewer believes only accounts for _25% of content in the whole manuscript

Response: In the revised manuscript, we added a "discussion" section and did uncertainties analysis and implications of mixing of Asian dust with pollutants on regional atmospheric environment. The uncertainties of dust shape, uptake probabilities (γ) are discussed.

Comment 3: Page 2745, lines 20 and line 23; Page 2748, line 10, line 19; Page 2749, line 10: Cited reference should be listed in a right order.

Response: We revised it.

Comment 4: Page 2746, lines 10-15, the citation is questionable, please double check.

Response: We re-write this part. In their hypothesis, the heterogeneous oxidation of SO_2 to sulfate drives the acidification of dust particles. In highly acidic ambient aerosol solutions, Fe (III) in atmospheric dust particles is reduced to be Fe (II) with OH radical production. More Fe(II) could result in the marine production of more DMS which in turn provides more SO_2 into the atmosphere. The more SO_2 result in more acidic SO_4^{2-} aerosol on mineral dust and again more Fe(II). Charlson et al. (1987) suggested that the heterogeneous oxidation of SO_2 to sulfate is a key step in this feedback between iron and sulfate. Furthermore, Mesekhidze et al. (2003) proposed that the SO_2 from urban and industrial centers plays a key role in Fe mobilization in Asian mineral dust. Zhuang et al. (1992) believed that this hypothesis can explain high Fe(II) and non-sea salt sulfate (nss-sulfate) observed in aerosol samples at a remote pacific station (Midway, Hawaii).

Comment 5: From Page 2751, line 15 to Page 2752, line 12, the part is totally misleading and pleases double check.

Response: We are sorry for confusing the reviewer. This part is mainly described to explain how to treat iron mobilization resulting from dust acidification driven by SO_2 and HNO_3 in this study. Usually, mixing of urban acidic gases with mineral dust result in an enhancement of iron mobilization in mineral dust plumes (Mesekhidze et al. 2003). To the extent that this mobilized iron is then acting as a limiting marine primary productivity. This would imply that iron mobilization is one of important indicators reflecting impacts of mixing processes on regional environment. Air pollution controls in China which reduced SO_2 and NO_x emissions might actually depress C-uptake in pacific waters, and thus affect the regional climate.

A large number of studies have demonstrated that dissolved iron (Fe (II)) in surface seawater is the limiting nutrient factor for primary productivity in high-nitrate low-chlorophyll (HNLC) regions of ocean. However, Fe in surface soils of the Gobi deserts is found mainly in the form of undissolved hematite (α-Fe_2O_3). Acid mobilization which reduce undissolved hematite to dissolved Fe(II) is thought as one of possible mechanisms (Mesekhidze et al. 2003). Fan et al. (2006) proposed a two-step mechanism for the acid-mobilization of iron in mineral dust particles. In this mechanism, formation of dissolved Fe (Fe(II)) experiences two processes: sulfate coating and dissolution of iron (hydr)oxide on the coated dust aerosols. The sulfate coating process can be achieved by the heterogeneous oxidation of SO_2 and HNO_3 (HR12 and HR19 in table 1), and represents the acidification of mineral dust surface. Dissolution of iron (hydr)oxide represents the mass transfer from insoluble Fe to dissolved Fe in acid highly acidic ambient aerosol solutions. Obviously, three types iron/dust are included: fresh, coated and dissolved (for Fe). In this study, we use this two-step mechanism to reproduce the impact of mixing processes on Fe mobilization.

In the first step, gas uptakes convert dust particles from fresh to coated by the heterogeneous reactions HR12 and HR19. The rate coefficient represents how many fresh dust particles are converted into coated particles and is calculated from $k_s[SO_2]+k_n[HNO_3]$, where k_s and k_n represent the first order rates of HR19 and HR12. In the second step, Fe(III) in the coated dust is transferred to dissolved Fe (Fe(II)). The production rate of Fe(II) is calculated from $R_{Fe} = R_d AnM / w$, where R_{Fe} is grams of Fe dissolved per gram of Fe(III) per second, R_d is the dissolution rate per unit surface area, A is the specific surface area of hematite ($m^2 \cdot g^{-1}$), n=2 (moles Fe/moles hematite), M is the gram-molecular weight of Fe, and w is the mass fraction of Fe in hematite (i.e., 0.7). Here, R_d and A are set as

$1\times10^{-10} mol\cdot m^{-2}\cdot s^{-1}$ and 100 $m^2\cdot g^{-1}$ as suggested by Fan et al. (2006) and Duckworth and Martin (2001). These values correspond to an e-folding time of 6-10 days for dissolution. Besides the heterogeneous chemistry, the effects of radiation process and cloud process on Fe-mobilization are also considered based on the scheme devised by Luo et al. (2005). Here the Fe solubility is assumed to have a lower limit of 0.5% (Fan et al., 2006).

Comment 6: Page 2756 line 18, what's the difference between TSP and total TSP?

Response: They are the same. We have revised it in the revised manuscript.

Comment 7: Page 2757, how to distinguish Dust-SO42- from Anthropogenic -$SO4^{2-}$ in the total $SO4^{2-}$. More discussion is needed.

Response: As discussed in the response to General Comment, dust-sulfate is explicitly calculated by the heterogeneous chemistry module using uptake probability shown in Table 1 in the manuscript.

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