Response to Anonymous Referee 1:

1. Comment: I find the literature review on previous studies that used GEOS-chem (Page 24481:lines:3-9) very limited in the sense that although you have cited an adequate amount of papers, you do not go on to -at least briefly- summarize the main conclusions of each of these studies. You need to do that in order to show why your study is different or more advanced compared to the other studies, ***i.e. what is the originality of this work****.*

Response: We have changed the first sentence of the appropriate paragraph to reflect the overarching conclusions of the studies cited. The originality of this work, in the context of GEOS-Chem, is that we focus on the impact of acidic gas uptake on dust. The amended paragraph appears below. See also response to Ref.#2's first comment.

"GEOS-Chem has been used previously to show that Asian emissions can lead to enhanced concentrations of CO, ozone, sulfate and dust aerosols in North America (Heald et al., 2003; Jaegle et al., 2003; Hudman et al., 2004; Park et al., 2004; Heald et al., 2006; Park et al., 2006; Fairlie et al., 2007; Nam et al., 2009). The model has been applied previously to simulation of the INTEX-B data by L. Zhang et al. (2008), focusing on ozone, and by van Donkelaar et al. (2008), focusing on sulfate and organic aerosol. Here, we focus on the impact of dust on nitrate and sulfate partitioning in transpacific transport."

2. Comment: More details about the flights are needed. E.g. flight trajectories, etc..

Response: We refer to Singh et al. (2009) who show details of the individual flights, and have added the sentence "We consider flights of the NASA DC8 out of Honolulu, Hawaii, and Anchorage, Alaska (Regions 3 and 4 in Fig.2a of Singh et al., 2009)" to the text. The locations of data taken from the DC8 are also shown in Fig.2 of our paper.

3. Comment: It is obvious that the scaling of the dust emissions (for winds) that you have applied is very important. I would suggest the authors to consider doing some sensitivity runs of different scaling. This will show if and in what extend the strength of your results is affected by this assumption/scaling.

Response: We find that particulate nitrate and sulfate respond quasi-linearly to scaling of dust emissions, because of the linear dependence of the uptake rates on dust surface area (Equation 1). Because of this, we find the ratio of nitrate to calcium (Figure 4b), and the percent consumption of dust alkalinity, relatively insensitive to scaling of dust emissions, even while calcium and surface area increase or decrease with the scaling. Increased dust emissions result in a larger bias in model calcium (Fig. 3). Lower dust emissions would reduce the calcium bias, and an apparent high bias in particulate nitrate, but at a cost of increasing the negative bias of AOD compared with MODIS (Fig. 2). Note, the assumed percent calcium in the bulk dust is consistent with the estimate of

McNaughton et al. (2009). We have inserted the following in our discussion of HNO_3 levels in section 4:

"HNO_3(g) can also be reduced by increasing dust surface area in the model by scaling emissions (section 2.2), but this causes both excess nitrate and excess calcium. Particulate nitrate and sulfate, and dust alkalinity consumption, respond quasi-linearly to scaling of dust emissions, because uptake rates depend linearly on dust surface area (Equation 1)."

We also now refer to additional sensitivity experiments we have conducted to assess the impact (i) on $\gamma(HNO_3)$ of eliminating dust alkalinity from the clay-fraction of the model dust size distribution, and (ii) on ozone of using higher rates of HNO_3 uptake. The following are now included in section 5:

"The smallest size bin accounts for ~70% of the alkalinity consumption. Some studies show that the clay fraction of Asian dust has much smaller calcite content than the bulk (Shi et al., 2005). If we eliminate dust alkalinity from the smallest size bin, eliminating it as a sink for HNO_3, we must approximately double $\gamma(HNO_3)$ to match the bulk consumption rate."

"Use of higher uptake coefficient for $\gamma(HNO_3)$ increases the impact on ozone in the model. We find ~5% reductions in column ozone over the Northern Pacific and up to 4 ppb reductions in surface ozone over North America when we multiply $\gamma(HNO_3)$ by a factor of 10. However, this results in a low bias compared with the ozone observations shown in Fig. 8 and nitrate levels inconsistent with the INTEX-B data (Fig. 5). Models that use higher values for $\gamma(HNO_3)$ may overestimate the impact of dust on ozone."

4. More details are needed about the thermodynamic model used in GEOS-chem since it is playing an important role in this study. Isn't the MARS-A model a little "outdated" for a study like this considering that other thermodynamic models treat more species (e.g. Ca2+, K+, Mg2+, etc.) and are more up to date? Dr. Meskhidge seems to support the same.

Response: We have expanded the reference to the MARS-A model. We have added the following to the text:

"MARS-A does not include mineral ion components; we treat the uptake of acidic gases by dust separately, and maintain fine dust-nitrate and dust-sulfate components distinct from the fine-mode sulfate-nitrate-ammonium system, as described below. Some thermodynamic models, e.g. ISORROPIA II (Fountoukis and Nenes, 2007), do include metal and chloride ions, but our results indicate that rapid equilibrium of HNO_3, SO_2 and bulk minerals is not appropriate, at least for coarse-mode dust."

5. I was wondering what the limitations of a study like this are, considering the use of a global CTM versus a regional CTM (with higher resolution). Can the authors comment on that?

Response: Finer resolution could provide improved representation of dust and pollution sources, and transport mechanisms. For example, use of a non-hydrostatic model to generate the meteorological fields could improve vertical mass and constituent exchange. Nevertheless, to the extent that transpacific transport and mixing of pollution and dust plumes is controlled at the synoptic scale, we would not expect a fundamental change in the conclusions, based solely on a change in resolution.

Response to Reviewer 2

1. I second anonymous reviewer#1 that brief discussion on the originality of the presented work as compared to previous modeling studies of mineral dust needs to be added to the introduction.

Response: We include the following to distinguish that aspect of our method:

"*Global and regional-scale modeling studies have shown significant consequences for sulfate and nitrate partitioning, tropospheric oxidants, and aerosol size distributions (Dentener et al., 1996; Song and Carmichael, 2001a,b; Liao et al., 2003; Bauer and Koch, 2005; Hodzic et al. 2006; Shindell et al., 2007*)" is intended to briefly summarize past modeling studies. We have also added a statement on our specific objectives in the following paragraph: *"We focus particularly on explaining observed particulate nitrate levels, ramifications for the lifetime of* $HNO_3(g)$ *with respect to uptake on dust, and whether dust can account for the high bias of* $HNO_3(g)$ *in models."* We have also added, in section 2.3: *"Previous model studies tend to lump dust into one or two size bins for gas-particle interactions (Song and Carmichael, 2001b; Liao et al., 2003; Solmon et al., 2009). Here, we compute the acid uptake separately for each dust size bin (plus the 4 sub-bins within the smallest size bin), and maintain dust nitrate, dust sulfate, and dust alkalinity as separate size-segregated constituents in the model."*

2. Brief discussion of the airborne measurements needs to be presented to indicate what method/s and at what time resolution were used for chemical speciation of dust and gas species discussed in the manuscript.

Response: We have added the following short section (2.4) to the text:

2.4 Bulk aerosol and gas data

> *Bulk aerosol,* HNO_3*, and fine sulfate data shown here were obtained from the University of New Hampshire (UNH) Soluble Acidic Gases and Aerosol (SAGA)*

> *instruments on board the NASA DC8. Paired filter samples of bulk aerosol were collected with the UNH dual aerosol sampling probe, and analyzed for soluble ions including NO_3^-, $SO_4^=$, NH_4^+, Mg^{2+}, Cl^-, Na^+, K^+, and Ca^{2+}(Dibb et al., 2003). Mean exposure times for the filters were ~ 5 - 6 mins. for altitudes below ~ 6 km., 10 -14 mins. at higher altitudes. HNO_3 and fine sulfate are measured in a mist chamber (Scheuer et al., 2003); collection intervals were typically 2 mins or less. Ozone data shown here were obtained from the NASA Langley FASTOZ instrument at 1 Hz.*

3. Figs 4a and 4b. It is not clear what does each point mean. Is it the time averaged data or data averaged over specific geographic area? Also, it is hard to distinguish between blue and black points. Use of colors with better contrast between them is recommended.

Response: The black points are the observed data. Each point represents one SAGA bulk aerosol measurement (see above for exposure times). In the case of HNO_3, the data are averaged over the collection times of the bulk aerosol data. The colored points are from the model interpolated to the DC8 flight tracks. These are shown at 1-minute intervals. We compare the relationships found in the SAGA data vs. the model. We have replaced blue with red symbols in the figure for better clarity.

4. page 24486, lines 12-14: I'm wondering if the discussed bias can be explained by formation of NH4Cl?

Response: We find there is insufficient Cl- to account for the NH4+ bias (statement included in the text).

5. page 24490, 23-24: the statement is not correct. Uptake coefficient of 0.1 was reported in laboratory experiments for uptake of nitric acid on calcium carbonate, which is only a fraction of mineral dust. Therefore, use of smaller coefficient for dust is not in conflict with laboratory measurements.

Response: We note the reviewer's implicit support for one of our basic conclusions. We have simply deleted the words "on the basis of initial uptake rates measured in the laboratory." The remaining statement that previous models have used $\gamma(HNO_3) = 0.1$ is correct.

C. Fountoukis and A. Nenes, ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K+–Ca2+–Mg2+–NH+–Na+–SO2–NO3 –Cl−–H2O aerosols, Atmos. Chem. Phys., 7, 4639-4659, 2007.

Scheuer, E., R. W. Talbot, J. E. Dibb, G. K. Seid, L. DeBell, and B. Lefer, Seasonal distributions of fine aerosol sulfate in the North American Arctic basin during TOPSE, J.

Geophys. Res., 108(D4), 8370, doi:10.1029/2001JD001364, 2003.

Additional responses to comments from Dr. Meskhidze:

1. We provide the following summary of the papers by Meskhidze et al. (2005) and Solmon et al. (2009), which we hope is more representative of the content:

   "Meskhidze et al. (2005) indicated that dust alkalinity must be titrated for the iron content to be soluble and bio-available upon deposition. They found that calcite strongly buffers dust acidification in Asian outflow, limiting significant iron mobilization to dust plumes with high initial acid-to-dust ratios. Solmon et al. (2009) implemented the dust iron dissolution scheme of Meskhidze et al. (2005) in GEOS-Chem and predicted deposition of significant soluble iron during Asian outflow in Spring 2001."

We have also had very helpful interaction with Dr. Zongbo Shi at University of Leeds, and have qualified our discussion of the titration of dust alkalinity, to distinguish the bulk from the submicron components, which approach alkalinity titration in our model because of higher surface area to mass ratio. We include the following statement in that discussion:

"*Our results indicate that iron mobilization may be limited to the smallest dust particles (radius < 1 μm) and to those with much lower initial alkalinity.*"