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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Intercontinental transport of pollution and dust aerosols: implications for regional air quality" *by* Mian Chin et al.

Mian Chin et al.

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Reply to Anonymous Referee #1 and Editor Murphy's comments

We thank Referee #1 and Editor Jennifer Murphy for their time in reviewing this manuscript and for their very constructive comments. The comments are copied below and start with "C" while our answer start with "A".

Answers to Anonymous Referee #1

C: This paper describes the long-range transport of aerosol particles from pollution and natural sources and the contributions of these sources to surface fine particulate concentrations at both regional and global scales. Fine particular matter has significant

implications for health, and therefore study of its transport is valuable for improving understanding of its environmental impacts, and is a necessary precursor to any attempt to control its concentrations. This paper provides a clear composition and source attribution of fine particulate matter over different parts of the US, including assessment of model performance against surface observations, and a wider assessment of the effects of intercontinental transport. The study is competent and interesting, the results are valuable, and the paper is worthy of publication in ACP.

C: The paper is well written, clearly organized, and appropriately illustrated. My principal reservation is that it is not clear which elements of the study are genuinely new and original and which echo the findings of previous studies. Previous studies are suitably acknowledged, but the paper would be stronger if the novel results were clearly highlighted. I believe that this should be relatively easy to address, and that the manuscript will then be suitable for publication in ACP once the following minor comments have been addressed.

A: We have revised the manuscript to make a clearer distinction between this study and previous studies (e.g., Park et al., 2004; Hadley et al., 2007; Koch et al., 2007). Our major findings are in the context of surface air quality and we consider all major aerosol types from different source categories and/or locations. We have emphasized that (1) North American regional sources are mostly responsible for the surface fine PM concentrations in the U.S. while the intercontinental transport has only minor impact; (2) European anthropogenic sources and Asian dust sources have the largest "impact potential" on other regions" air quality; (3) what have been very much overlooked in previous studies but are quantified in this study is the European influence on Africa and Asia surface PM; (4) we have added a new section 5.3 which not only explains the link between the transported plume height and the surface PM concentrations in trans-Pacific transport, but also reveals the fraction of European pollution and African dust in the Asia outflow which had hardly addressed in the literature before.

General Comments

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C: The paper is heavily weighted towards the US in its focus, but this is not reflected in the title. While it is clear that there are better measurement data to compare against over the US, little attempt has been made to extend the study to other regions. If the main focus is on surface air quality over the US (as is stated on page 9025, l.2), then it would be more appropriate to alter the title to "... implications for US regional air quality". The paper would be improved if the new contributions of this study were more clearly highlighted. Earlier studies from Park et al. [2004] and Koch et al. [2007] are referenced (although more recent papers such as Hadley et al. [JGR, 2007] have been missed) but it is not clear where the present study adds substantial new insight. Is the paper merely supporting the conclusions of these studies, or is it adding significant new results? I believe that a number of the conclusions are original, and these need to be brought out more strongly at the end of the discussion in sections 4 and 5 and again in the conclusions.

A: We have more clearly highlighted our findings in sections 4, 5, and in conclusions. We choose not to specifically limit the title to "US air quality" although we have done detailed comparison with the observations over the U.S. and assessed the aerosol source types and origins over the U.S., a very large fraction of the text is devoted to discuss the relationship between different source-receptor regions in the NH, which takes about the same length as the discussion for the U.S.

Specific Comments

C: p.9016, l.3-7: East Asia is immediately upwind of the US, and this plays a large part in the recent interest.

A: We have added the sentence "Situated upwind of North America across the Pacific Ocean" after the sentence "Recently, much attention has been focused on ...Asia... to put the recent interest into a geographical context.

C: p.9021: l,13: It would be worth noting the successful simulation of the April dust event (also shown in Fig 2) at both sites in the western US in Fig 4.

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A: We have pointed out this success by adding "The model successfully captures the seasonal variations of dust"

C: p.9021-9024: The contribution and source attributions shown in Figures 6 and 7 are interesting, but the arguments would be strengthened if the authors also showed the seasonal variation of the total observed RCFM (a monthly mean of all stations in each sector).

A: We have made a new Figure 6 which now shows the side-by-side comparisons of seasonal variation of RCFM and its components at the four sectors, averaged over the IMPROVE sites located in each sector. (Our previous Figure 6 showed the model results averaged over the entire sector domain instead of the locations of IMPROVE sites.)

C: p.9025: Do the results here confirm or contradict the conclusions of Koch et al [2007] that Arctic BC is dominated by Asian sources rather than US or European sources? Transport to the Arctic is of particular interest at the present time (due to IPY), and any further insight into transport to this region would be valuable.

A: It is difficult to directly compare with the Koch et al. study as that study showed only column BC optical thickness, not the surface concentrations. Yes transport to the Arctic is a very interesting topic and deserves in-depth studies. We have pointed out in our study that the European pollution sulfate has the largest impact on the Arctic surface sulfate concentrations (Figure 8a). However, since this study focuses on air quality on major continents, we decide not to discuss in detail the transport to the Arctic in this paper. We plan to deal with this topic in our next manuscript in the context of the recent model intercomparison study of HTAP (Hemispheric Transport of Atmospheric Pollutants), in which the transport to the Arctic is one of the focused topics for more than dozens of models.

C: p.9030, l.10: This is an important conclusion, and the implications should be stated clearly here (they are currently buried at the end of the fourth paragraph.)

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A: We have revised the text to move this finding into the beginning of a new paragraph devoted to the discussion of the pollutants transported from outside of the US.

C: p.9031, last paragraph: How sensitive are the results described here to the treatment of dust lifting processes or to the model wind speeds? How robust are these conclusions?

A: The results of course are sensitive to both the dust uplifting processes and the model wind speeds which determine how much dust is emitted to the atmosphere and consequently transported in the atmosphere. Since this model version for 2001 has been tested and verified with a variety of observations (MODIS, MISR, AERONET, ACE-Asia, etc.) in our previous studies, we believe that the conclusions presented in this paper are quite robust.

C: p.9032, l.8-16: This paragraph does not appear to add anything new to the conclusions, and should be cut.

A: Paragraph deleted.

C: Table 3: What are the annual mean RCFM from the observations? It would be very useful to add one row to the table showing the mean RCFM over all stations in each domain.

A: The annual means of RCFM and its compositions from model and observations are now shown in the new Figure 6 for appropriate comparisons. The numbers in Table 3 are the model results for the entire domains so they are somewhat different from the values shown in Figure 6, which were averaged over the IMPROVE sites located in each domain. Because there is no domain-averaged values from IMPROVE and because the appropriate comparisons have been shown in Figure 6, we do not list the IMPROVE averaged values in Table 3.

C: Fig 3: It would be helpful to highlight the four sites shown in Figs 4 and 5 with a different symbol.

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A: We now use different symbol in Figure 3 for the locations of the four selected sites shown in Figs 4 and 5.

C: Fig 6: The caption should state that the data shown here are from the model.

A: We caption has been re-written for the new Figure 6 which shows both the model and IMPROVE averages.

Typos

C: p.9013: "Parck" -> "Park"; p.9022, l.2: "agrees" -> "agree"; p.9023, l.14: "is" -> "are"; p.9029, l.10: "detecting" -> "detection"; p.9029, l.1a: add "the" before "North Pacific"; p.9033: "2pp2" ? p.9035, l.2: "Jeffe" -> "Jaffe".

A: All these typos have been corrected.

Answers to Editor Murphy's comments:

C: This paper compares observations of reconstructed fine mass (RCFM) at IMPROVE sites in the continental U.S. with predictions of a comparable product (sulfate+OC+BC+dust) from the GOCART model. The authors find that sulfate predictions match well with observations whereas carbonaceous aerosol is frequently underpredicted, and fine dust is frequently overpredicted. In general, the model captures the seasonal and spatial variations observed in RCFM at IMPROVE sites quite well. The model can therefore be used to attribute aerosol sources, and this analysis is applied not only to aerosol observed over North America, but also other continents and regions in the Northern Hemisphere. The topic of this paper is appropriate for ACP and I recommend that it be published after addressing the following comments:

General Comments

Abstract

C: Are you defining the sum of ammonium sulfate, black carbon and organic matter as "pollution"? Since you use this term as a distinction from "dust" you should define it. Is

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sulfate with a volcanic source or organic matter with a biogenic source still defined as "pollution"? This issue arises again in Sections 2 and 4.

A: The "pollution" aerosols are defined as aerosols originating from "pollution&" sources, i.e., from fossil fuel and biofuel combustions and transportation. The term is defined in the first paragraph of section 4. To clarify further, we have added the definition in the abstract, section 2 (2nd paragraph), and Table 1. Volcanic and biogenic aerosols (sulfate and OM) have been referred as "natural" aerosols in Table 1 and Figure 7.

Section 2

C: No emissions from the Southern Hemisphere are listed in Table 1, yet the GOCART results in Figure 2 clearly demonstrate emissions. Does Figure 2 represent a model run with emissions not listed in Table 1? This should be explained more clearly.

A: The model (and Table 1) includes global emissions, not just emissions in the northern hemisphere, and Figure 2 and other figures show the model results with all emissions. It was an error in section 2 saying that Table 1 only listed emissions in the Northern Hemisphere. We have corrected that error.

Section 3

C: While RCFM is perhaps the most appropriate quantity to compare to GOCART output, which also doesn’t consider nitrate, some estimate should be given regarding the importance of neglecting nitrate. In the U.S., nitrate may only be significant in California, however the analysis then goes on the consider Europe and Asia, where nitrate concentrations may be more significant. Furthermore, since the formation of particulate nitrate can occur more quickly that the formation of sulfate for NOx and SO2 emissions respectively, the relative importance of intercontinental transport is likely to be different. What kind of impact might this have on your conclusions regarding the extent to which pollution from major source regions affects PM surface concentrations

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on a hemispheric scale? By neglecting nitrate, you may be biasing the results towards attributing more of the aerosol loading to distant sources.

A: We use RCFM, which does not include nitrate, in this study, not only because it is convenient for GOCART but also because nitrate data quality is questionable – as Malm had acknowledged that there were substantial losses of nitrate during sampling. RCFM may not be suitable as a proxy for PM2.5 in some locations over the U.S., such as California where local nitrate is important; however for other area the source attribution for RCFM would be similar for PM2.5. Regarding the long-range transport of nitrate, the study of Park et al. 2004 has shown that the long-range transport of nitrate has little impact on receptor region’s surface concentrations; therefore our conclusion about long-range transport of pollutants is not affected by the exclusion of nitrate. We have added the following paragraph in section 6: "It is noted that we have not considered nitrate aerosols in this analysis; therefore the total amount of RCFM does not necessarily represent that of PM2.5 which is a commonly used indicator for air quality. Yet, because intercontinental transport of nitrate seems to have negligible effects on the surface particle concentrations over the U.S. (Park et al., 2004), our source attributions for RCFM are applicable for PM2.5 in general, except California where the local pollution nitrate is an important components of PM2.5."

C: Is it appropriate to assume that the sulfate is fully neutralized, even at sites downwind of power plants? Additionally, the analysis essentially assumes that the sources of ammonia and sulfur are collocated, for the purposes of attributing an observed mass loading to an original source region. Is it not possible that acidic sulfate could be advected from one continent and neutralized by ammonia from another?

A: We understand that sulfate can exist in the forms of fully neutralized sulfate, bisulfate, and sulfuric acid, even though the IMPROVE data (e.g. Malm et al., 1994) and many other observations show that neutralized sulfate is the most common form. Because in the IMPROVE RCFM the sulfate is assumed to be fully neutralized by ammonia and the mass of ammonium sulfate is converted from the measured sulfate ion, we follow the

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same convention in constructing the model RCFM to be consistent. It is possible that the acidic sulfate could be advected from one continent and neutralized by ammonia from another; however this will not affect the sulfate budget or any of our conclusions, because we are not specifically tracking the ammonium budget or individual sulfur (VI) forms (sulfate-bisulfate-sulfuric acid). Rather, we just track "sulfate".

C: The relatively coarse spatial resolution of the model is probably another source of discrepancy between the model results and measurements. The degree to which we can expect the model output to match the observations at any given site could perhaps be quantified by comparing pairs or group of sites within the same model grid cell to assess the actual range of measurements within that area which is assumed to be homogeneous in the model.

A: Yes this is certainly an issue. We have examined the variability of data from different IMPROVE sites that are located in the same model grid, and done the comparisons based on the data and model from the same model grid locations instead of IMPROVE sites. The agreement has improved slightly but not significantly, because there are not many grid cells that contain multiple IMPROVE sites (135 sites spreaded in 92 grid cells: 19 grid cells have 2 sites, 8 cells 3 sites, and 2 cells 5 sites; the rest 63 cells each contains just 1 site). We have added some discussions in this matter and listed the comparisons based on the model grid in Table 2.

Section 4

C: The distinction between different aerosol sources seems different here to earlier in the paper. It seems to discount the possibility of intercontinental transport of aerosol from natural sources other than dust. Is it not possible that volcanic sulfate or biogenically derived organic matter can contribute to aerosol loadings on downwind continents?

A: Although the RCFM from natural sources, i.e. volcanic or biogenic aerosols, can also undergo long-range transport, the influence of intercontinental transport of natu-

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ral aerosols is much smaller than that of pollution and dust aerosols (considering the magnitudes of the emissions) unless major volcanic eruptions occur. We have added this discussion in section 4.

Section 5

C: It might be useful to note that colour scales for Figures 8a and 8b are different.

A: We have added that note at the beginning of section 5.2.

Technical comments

Section 1

C: P 9016, line 6, Insert "an" in the sentence beginning with "Recently", that is "from which the trans-Pacific transport can bring "an" increasingly significant amount of pollution and dust to North America."

A: Done.

C: P 9016, line 12, Consider breaking up the sentence starting with "Previous global modeling studies" as it contains several ideas. Also, "peaked" should be "peak".

A: We have made this sentence into two separate ones to be more readable.

C: P 9016, line 18, Change some prepositions and don't capitalize "haze": It is estimated that the Southeast Asia is the largest contributor "of" black carbon over the Arctic and is also partially responsible "for" the "Arctic haze" problem.

A: Done.

C: P 9016, line 18, Explain Arctic haze in Paragraph 2, rather than the second time it is discussed, in paragraph 3.

A: Done.

Section 3

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C: P 9020, line 11 Remove comma after e.g. in sentence starting with "While sulfate"

A: Done.

C: P 9020, line 11 Remove both instances of "would" in sentence starting with "As such"

A: Done.

C: P 9023, line 13, remove "the" before air quality; P 9023, line 13, change "those" to "the amount"

A: Done.

Figures

C: Figure 2 - Aug 22 is printed twice by colour bars for no reason.

A: This is puzzling. I do not see "Aug 22" anywhere on Figure 2 from the manuscript file I downloaded from the ACPD site. I wonder if the editor was looking at an "un-official" version before it was published on ACPD, as I discovered several errors (including Aug 22 on Figure 2) after I submitted the manuscript and then resubmitted the figure before ACPD publication. In any case, the "Aug 22" is not on the Figure.

C: Figure 6 caption - specify that this is the model output

A: We have a new Figure 6 per reviewer #1's suggestion. Now Figure 6 has both model and data with a new caption.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 9013, 2007.

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