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Atmospheric
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Discussions

Interactive comment on "Impacts of aerosol indirect effect on past and future changes in tropospheric composition" *by* N. Unger et al.

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Response to Reviewer #2

We thank the reviewer for the valuable comments and insights. We now include explicit discussion of the impacts of aerosol-cloud interactions on precipitation and temperature and the resulting influence on the composition parameters, which has considerably improved the manuscript.

1. We have changed the second paragraph of the Introduction to include aerosol direct radiative forcing impacts on chemical composition though changes in meteorological fields:

'Aerosols affect tropospheric chemical composition through a number of pathways. In

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polluted regions, aerosols usually act to suppress photochemistry by decreasing available photochemically active radiation and providing surfaces for heterogeneous reaction processes, effects that have been reasonably well studied (e.g. Dentener et al., 1993; Tie et al., 2003; Martin et al., 2003; Liao and Seinfeld, 2005). It is essential to include such effects of aerosols in global atmospheric chemistry models in order to reconcile simulations of O3 and precursors with observations. Aerosol direct radiative forcing may also influence chemical composition by driving changes in meteorological parameters including temperature, precipitation and winds (e.g. Chen et al., 2007). Whilst research is emerging to quantify the climate response to changes in aerosol loading (e.g. Jones et al., 2007; Koch et al., 2009), feedbacks to chemical composition and surface air quality are less well studied.'

In the third paragraph of the Introduction, to the following: 'Changes to available radiation fields may impact photolysis frequencies and changes to cloud lifetime and precipitation may alter rates of in-cloud processing and wet deposition of pollutants and precursors' we have added: 'Furthermore, ACI can influence meteorological factors such as surface air temperature due to cloud cover change.'

2. We cannot here provide a completely satisfactory answer to this question because it would require additional model simulations due to the complex feedbacks in place. Our contribution has been to assess the impacts of aerosol-induced cloud changes on chemistry for past and present 'real-world' emissions perturbations. However, the reviewer has raised an important suggestion for potential further experiments that extend/continue exploration of the impact of aerosol on atmospheric chemistry through their modification on clouds. Quantifying the sensitivity to the fixed CDNC assumptions would reveal important insights into the aerosol-cloud-chemistry interactions. Valuable sensitivity experiments could include controlling iterative sets of parameters (cloud droplet number, etc.) during model runs, slowly adding in links to aerosols and investigating the impacts on chemistry in clouds. In this way, the results would not be specific to particular emissions scenarios. This particular approach has been outlined in Activ-

ity 3 of the multi-model Atmospheric Chemistry and Climate Initiative.

3. We amend the sentence to clarify: ‘the first 2 years of the simulations allow for the chemistry to adjust from initial conditions and are discarded as spin-up’.

3. The nitrate aerosol module was not available in the GISS atmospheric composition-climate model when these experiments were run. However, we do not expect the inclusion of nitrate aerosol to affect the results presented significantly but we do expect nitrate aerosol distribution to be sensitive to the inclusion of ACI.

We add to Section 2.1: ‘Nitrate represents only a small component of the anthropogenic inorganic aerosol (e.g. Forster et al., 2007) and is not included in this study. Since nitrate aerosol is formed from HNO3, this omission likely leads to only modest overestimates of the impacts on HNO3 and therefore O3 based on our previous work (Bauer et al., 2007).’

4. We have added the following information into the model description Section 2.1:

‘Photolysis rates are calculated using the FAST-J2 scheme (Bian and Prather, 2002). The wet deposition scheme is described in detail in Koch et al., (1999). Briefly, soluble gases and aerosols can be removed into either moist convective plumes or large-scale clouds as derived from the GCM’s internal cloud scheme (Del Genio and Yao, 1993). During convection, all chemical species are transported along with the convective plumes, with scavenging of soluble species within and below cloud updrafts. In large-scale stratiform clouds, soluble gases are removed based on the fraction of the grid box over which precipitation is taking place. Washout of soluble species is calculated below precipitating clouds. The model includes a dissolved species budget (Koch et al., 2006) for stratiform clouds that makes the simulation more faithful to the cloud scheme and more physically realistic.’

5. The purpose of this study is not an assessment of the aerosol indirect effect, which is addressed in numerous other studies with the GISS climate model and many other

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model studies. Our focus is probing the impacts of modeled aerosol-cloud interactions on the chemistry. We believe the confusion is because of a misplaced statement in Section 3.2 (page 4701, lines 25-29 and pg 4702 lines 1-3) that we have now removed. It is useful for the purposes of this study to separate the discussion of aerosol impacts on cloud properties that will affect the composition and the global mean radiative forcing due to those changes. In order to improve the logic, we have defined a new Section 3.1: 'Past and future aerosol-cloud interactions' that includes 3 subsections: (3.1.1) Aerosol distributions and impacts on cloud properties, (3.1.2) Aerosol direct radiative forcing and AIE, and a new subsection, (3.1.3) Impacts of ACI on precipitation and temperature.

It is clearly stated in the model description (Section 2.1) that aerosols are assumed to be externally mixed.

6. We have changed to more accurate number: 'by a factor of 1.7'.

7. We include 2 new figures (now Figure 3 and Figure 4) that show the impacts of ACI-induced changes in precipitation and surface air temperature. The impacts on other meteorological variables including horizontal winds are negligible/insignificant.

We have added in Section 2.3 (Simulations): 'Switching on ACI will influence dynamics and meteorological factors such as precipitation and temperature due to cloud cover change that can also affect composition. Thus, in the experimental setup employed here, we have essentially decoupled the meteorological changes driven by ACI from climate changes driven by greenhouse gases (GHGs) and direct aerosol effects. In reality, these effects are coupled to each other, although recent work suggests that the climate response to GHG and aerosols forcings may be additive (Kloster et al., 2009).'

We have added a discussion of the ACI impacts on temperature and precipitation (Section 3.1.3 Impacts of ACI on precipitation and temperature): 'Figure 3 shows precipitation changes due to the prescribed SST and sea ice boundary conditions as well as the impacts on those precipitation changes when including on-line ACI. Without ACI, global

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annual mean precipitation increases by +0.02 ±0.006 mm/day for the past case and +0.13 ±0.008 mm/day for the future case. The well-known spatial pattern of the precipitation changes expected due to GHG-induced warming (from the prescribed SSTs) is evident: increases in tropics and drying in subtropics. In both the past and future cases, inclusion of ACI tend to increase the global annual mean precipitation change slightly although neither result is statistically significant even at the global scale. For the past case, the impact of ACI is +0.005 ±0.009 mm/day and for the future case is 0.004 ±0.014 mm/day. None of the regional impacts in Figure 3 are 95% statistically significant, however, there are suggestions of ACI influence that tends to either reinforce or counteract the rainfall changes induced from the prescribed SSTs (driven by GHG increases). Tropical precipitation changes correlate with the cloud cover changes (Figure 1). In general, where aerosol pollution increases across the time period, ACI cause increases in liquid water path (not shown) and impose decreases in rainfall and vice versa, Thus, ACI appear to have a drying influence on the PD-PI change on the U.S. east coast region of around 0.3-0.5 mm/day. Drying also occurs over the biomass burning regions due to inclusion of ACI up to 1-2 mm/day. The PD-PI drying over South and Southeast Asia is strengthened when ACI are included, which also may be related to a southward shift in the Intertropical Convergence Zone (e.g. Jones et al., 2007). In contrast, over Europe, ACI cause increases in the PD-PI change by around 0.1-0.3 mm/day that are mainly due to increases in convective precipitation there (not shown). In the future case, there is evidence of ACI leading to precipitation increases over the southeastern U.S. up to 0.5 mm/day. ACI cause increased precipitation over the Middle East and South Asia by 0.3-0.5 mm/day and decreased precipitation in Pacific outflow regions by around 0.5 mm/day.

Figure 4 shows surface air temperature (SAT) changes due to the prescribed SST and sea ice boundary conditions and the impacts of ACI on the SAT changes. Without ACI, the global mean SAT change for the past is 0.63 ± 0.02 °C and for the future is 1.1 ± 0.06 °C. The effects of ACI on SAT are expected to be only small because prescribed SSTs largely determine the SAT (ACI effects on SAT would be larger if full climate feedbacks were allowed in the simulations). Thus, largest ACI effects on SAT occur over continents where SSTs have less influence. Including ACI for the past case generally decreases SAT over the continental northern hemisphere where the ACI impacts may be -1 to -2 °C. The global average impact on the past change amounts to about a 20% reduction of -0.13 ± 0.07 °C. The global impact of including ACI on the future SAT change is small +0.03± 0.05 °C and features more complex regional changes including SAT increases in regions where aerosol emissions have decreased (N. America, Eurasia) and decreases in regions where aerosol emissions have increased (Middle East, North Africa). However, the ACI impacts on future SAT are not statistically significant.'

We include explicit discussion of the meteorological impacts of ACI on the composition, which is most relevant for relating wet deposition changes to changes in precipitation. Thus Section 3.4 (Impact of ACI on sulfate in-cloud production and wet deposition) now reads:

'The influences of ACI on the past and future changes in sulfate in-cloud production, wet deposition and HNO3 wet deposition are shown in Figure 7. Inclusion of ACI in the PD – PI case drives larger increases in in-cloud sulfate production across polluted regions where there have been aerosol-induced increases in CDNC, LWP and COD. This enhancement in sulfate production in clouds amounts to around 15-20% of the PD-PI change across eastern U.S. and East Asia ($\sim$ +50-100 mg m-2 yr-1) due to ACI effects alone.

In general, though not in every case, the impacts of ACI on changes in wet deposition map the influence on precipitation (Figure 3) such that in the regions where aerosol pollution emissions increase and ACI drive decreases in rainfall, precursor wet deposition decreases. There are compensating increases in wet deposition downwind and where the rainfall has increased. Exactly the opposite pattern occurs in regions where aerosol emissions decrease, especially evident in the future case. Since the tracer fate is ultimately wet removal from the atmosphere, the ACI effects on wet deposition appear to displace precursor washout away from source regions.

Thus, for the past case, in the immediate location of the pollution sources in the eastern U.S. and China, the ACI drive decreases in sulfate wet deposition that represent only 5-10% of the PD-PI change. However, in response to the enhanced in-cloud production, there are ACI-driven increases in sulfate wet deposition further north of the source regions and across Europe (∼ +50-100 mg m-2 yr-1). The PD-PI increase in HNO3 wet deposition rate is dampened due to ACI effects up to 10-15% across polluted source regions (∼ -100 mg m-2 yr-1). Over Europe, where ACI causes increases in rainfall, HNO3 wet deposition decreases. Therefore, this effect is most likely due to a decrease in the production rate of HNO3 due to reductions in J(O1D) that deplete OH levels there. A corollary is that the notorious acid deposition problem due to industrialization in the past century in the northern forests across Europe and northeastern U.S was made worse by ACI in the case of sulfate but lessened by ACI in the case of HNO3. Unlike for sulfate aerosol, if the HNO3 lifetime with respect to washout is lengthened, the HNO3 may be photolyzed such that the reactive nitrogen is re-released and available to participate in O3 photochemistry.

For the future 2050 A1B scenario that we examine, the projected decreases in aerosol-loading and ACI over North America and Europe are only modest and correspondingly the model suggests weaker impacts than for the past change on precursor washout and sulfate in-cloud production in these regions (Figure 7). There is a dampening of in-cloud sulfate production across the U.S. and movement of HNO3 wet deposition back to the source regions due to ACI effects on the 2050 – PD change that amounts to around 10% of the future change. ACI driven enhancements in in-cloud sulfate production (∼ +20% of future change) are localized in source regions along the northeastern African coastline. Large and significant effects occur over Africa, the Middle East and South Asia where there are large increases in SO2 emissions and thus aerosol loading. Fossil fuel and biofuel aerosol-driven increases in CDNC and COD in Central Africa cause marked dampening (∼ -20% of the 2050-PD change) in the local wet deposition of sulfate and HNO3, which is displaced across the Middle East and South Asian regions where there are enhancements up to 50-100% of the future change due entirely to ACI effects. This ACI-driven displacement of acid deposition adds new dimensions and complications to transboundary pollution problems.

8. Done.

9. We agree with the reviewer and have moved this discussion to a separate section at the end of the results: Section 3.6 ‘Impact of ACI on O3 and aerosol direct radiative forcing’.

10. Our simulations do all include the aerosol (and ozone) direct radiative effects, however, we don’t specifically isolate the direct effects on composition here as the purpose of this study is probing potential impacts of aerosol-driven changes in clouds on chemistry, feedbacks that have been previously neglected in other studies.

11. Done. Figure 1 now includes PD-PI changes in addition to 2050-PD.

12. In Section 3.2 ‘Impact of ACI on J(O1D) photolysis rate’ we have added: ‘Changes in cloud optical depth (COD) provide the dominant mechanism for the J(O1D) response to including ACI. Thus, decreases (increases) in J(O1D) when ACI are included are largely driven by increases (decreases) in COD (Figure 1). However, in some tropical and southern hemisphere regions such as South America and South Africa in the past and future cases and southeastern China in the future case, J(O1D) increases due to ACI even though COD has increased in those regions. The J(O1D) response in these regions is determined by a co-located decrease in cloud cover (CC) that exposes greater incoming radiation (Figure 1).

13. In Figure 5, we have added plots showing the past and future impacts of ACI on J(O1D) higher up in the troposphere at 500mb, which explains in part the increases in OH in the zonal mean OH change plot that is now Figure 6. We explain the OH increases in Section 3.3: ‘The ACI-induced increases in the OH changes in the north-

ern hemisphere subtropics are partly driven by increases in J(O1D) at higher altitudes as discussed in the previous section. Increased availability of NOx (not shown) due to reduced HNO3 wet deposition (discussed in the following section) also likely plays a role in the OH enhancements.'

14. We have included a brief description: 'The global mean CH4 lifetime (calculated as [global mean tropospheric CH4 mass (kg) / global mean tropospheric loss rate with respect to OH (kg/s)]'.

15. Done. We have combined the [PD-PI] and [2050-PD] figures.

16. Done. We now use the unit μgm-3 for sulfate aerosol changes.

17. We remove the phrase 'push precursor washout away from source regions', which was inappropriate/confusing choice of language. ACI-induced changes in horizontal winds are negligible and do not contribute to the pattern of changes in wet deposition. Tracer wet deposition is decreased in polluted regions due to ACI-induced increases in cloud lifetime and decreases in precipitation. Since the tracer will ultimately be removed from the atmosphere by wet (or dry deposition) downwind away from the source regions, ACI appears to cause displacements in precursor washout. We include in Section 3.4: ' Since the tracer fate is ultimately wet removal from the atmosphere, the ACI effects on wet deposition appear to displace precursor washout away from source regions.'

18. Please see point (3) above. We already have included in Section 2.1 a discussion about the potential issues from not including nitrate aerosol in the simulation HNO3.

19. The positive changes in (the new merged) Figure 7 for future versus past ACI influences on wet deposition are not necessarily more significant but they tend to be larger in absolute magnitude across a wider spatial extent for the future case. The reason is that the future case features large increases in precursor emissions in the North African coastline, Middle East and South Asian regions. All these subtropical

regions have high solar radiation and year round intense photochemical activity versus the past case where the emissions increases took place in the northern mid-latitudes (less sunlight, less photochemical activity). These plots are all showing annual mean changes.

20. We prefer not to introduce ACI-induced impacts on carbonaceous aerosols since the paper is already examining a large number of composition variables. More importantly, carbonaceous aerosols are treated as source emissions only in the model and thus their impacts are reflected in precipitation changes whereas the impacts on sulfate are more complex because they involve mechanisms that affect both the in-cloud production and loss by wet deposition. Therefore, we chose to focus on sulfate aerosol.

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