

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

Anonymous Referee #2

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This manuscript examines the impacts of aerosol indirect effect (AIE) on past and future changes in concentrations of some important atmospheric species such as OH, O₃, and sulfate aerosol. It is generally a very good study and can provide useful information to the scientific community. My major concern with the manuscript is that the authors try to explain the predicted changes in concentrations of chemical species by AIE-induced changes in photolysis rates and wet deposition, with the roles of AIE-induced changes in meteorological parameters (for example, temperature, water vapor content, and winds) neglected. I would expect more thorough explanations in the manuscript with respect to how AIE-induced changes in meteorological parameters influence chemical species by altering natural emissions, chemical reactions, transport, and deposition. My specific comments are as follows.

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1. The second paragraph of Introduction: The sentence that “The direct effects of aerosols on global and regional tropospheric composition have been reasonably well studied (e.g. Dentener et al., 1993; Tie et al., 2003; Martin et al., 2003; Liao and Seinfeld, 2005)” is not accurate. Aerosols can influence chemical species not only by influencing photolysis rates and by serving as sites for heterogeneous reactions, but also by changes in meteorological parameters resulted from aerosol direct radiative forcing. The studies on the latter should also be reviewed here.
2. Page 4698: In all simulations without AIE, CDNC are fixed at 60 cm⁻³ over oceans and 174 cm⁻³ over land. How sensitive are the predicted AIE-induced changes in concentrations to this assumption?
3. P4699: SST and ice climatologies are used in both experiments in each time period, which are expected to allow the climate simulations to reach equilibrium state quickly. However, the manuscript should mention briefly how the 2-yr spin up time was chosen.
3. Nitrate aerosol is not considered in this work, which can influence predicted AIE, NO_x, HNO₃, O₃, etc. Give some discussions in the text how the missing nitrate would influence model results of this work.
4. Since the paper focuses on the impacts of AIE on photolysis rates and wet deposition, model description should include the schemes for calculating photolysis rates and wet deposition.
5. Section 3.2 is logically confusing. Why is aerosol direct effect discussed here? The discussions in the bottom paragraph of Page 4701 indicate that AIE is related closely to aerosol direct forcing, which should be explained more clearly. Further more, global mean forcings are discussed here in this section. If you would like to compare AIE with aerosol direct radiative forcing, I would suggest comparing also values over populated and biomass burning areas. For calculation of aerosol direct radiative forcing, are aerosols internally or externally mixed?

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6. Lines 23-24 on Page 4701: "a factor of 2" can be replaced by a more accurate number.
7. The predicted horizontal distributions of AIE-induced changes in temperature, precipitation, and water vapor content should be given in the manuscript. These are important parameters that can help to explain the predicted changes in O₃ and aerosols.
8. Line 4 on Page 4702: Replace "annual mean forcings" by "annual mean direct forcings".
9. Lines 4-13 on Page 4702: It might be better to present this paragraph after you have discussed the impacts of AIE on aerosol concentrations (or burdens).
10. Aerosol direct radiative effect may couple with indirect effect through convection (or precipitation). Why not considering aerosol direct effect in all simulations listed in Table 2?
11. Figure 1: Besides the plots for 2050-PD, give the same plots for PD-PI.
12. It seems to me that changes in cloud optical depth (bottom of Figure 1) do not agree with changes in photolysis rates (bottom of Figure 3). The increases in COD are expected to reduce J(O¹D), but we see increases in J over South America, South Africa, and southeastern China, although CODs increase in these regions. Please explain.
13. Page 4703: Explanations are needed for the increases in OH shown in Figure 4.
14. Page 4704: Describe briefly how you calculate lifetime of CH₄, since you have fixed CH₄ concentrations for PI, PD and 2050 as listed in Table 1
15. You can combine Figure 5 with Figure 6. Use left column for PD-PI and right column for 2050-PD.
16. Figure 7: Please use the unit of $\mu\text{g m}^{-3}$ instead of pptv for changes in sulfate; $\mu\text{g m}^{-3}$ is a more commonly used unit for aerosols.

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17. Lines 21 and 22 on Page 4704: Please explain the sentence here ("At the same time, the effect of the AIE is to push precursor washout away from source regions as indicated for sulfate and HNO₃ in Figs. 5 and 6.") more clearly. You may need to examine the predicted changes in precipitation. Do AIE-induced changes in winds contribute to the pattern of changes in wet deposition? Is this result robust?
18. Page 4705: It should be mentioned at the end of Section 3.5 that the absence of nitrate aerosol in simulations may influence results of HNO₃.
19. Figure 6: The maximum positive changes are much more significant in all panels of Figure 6 than those in panels of Figure 5. Why?
20. Sulfate, BC, and OC are simulated in this work, but only sulfate is discussed in the manuscript. I would suggest presenting AIE-induced changes in carbonaceous aerosols also.

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