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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and North Indian Oceans: estimates based on in-situ chemical and optical measurements and chemical transport modeling" *by* T. S. Bates et al.

Anonymous Referee #2

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This paper gives a good summary of the characteristics of aerosols from INDOEX, TRACE-P/ACE-Asia, and ICARTT field campaigns. It also shows that using measured aerosol optical properties in CTMs can help to reduce uncertainties associated with aerosol forcing calculations. The manuscript includes much useful information. It should be accepted after addressing the following minor comments:

- Abstract: It should be described clearly that the constrained TOA DCF values of

-3.3±0.47, -14±2.6, -6.4±2.1Wm^{-2} are clear-sky forcings.

- Abstract: It is inaccurate to conclude that "Constraining the radiative transfer calculations by observational inputs reduces the uncertainty range in the DCF in these regions relative to global IPCC (2001) estimates by a factor of approximately 2". Forcing estimates in IPCC (2001) are based on cloudy-sky calculations. Including clouds in DCF calculations is expected to lead to larger uncertainties.

- Line 19 on Page 188: List specifically what "other substances" are.

- Line 11 on Page 190: It is stated that "Observed small increases in aerosol light scattering coefficient under dusty conditions are attributed primarily to the presence of accumulation-mode aerosol (Howell et al., 2004)." Do you mean the increases in light scattering with RH?

- Line 18 on Page 196: It should be described whether sub-μm sea salt is accounted for in this study.

- Line 18 on Page 196: "The concentrations of these aerosol components calculated by the chemical transport models are compared to measurements, which are categorized in the same way, and the optical properties and radiative effects of the aerosol are likewise calculated for these four components." – Is this applied to "a priori" calculations and/or constrained calculations?

- Table 5: It would be better to list aerosol concentrations in both sub-μm and sup-μm modes if such data are available.

- Section 3.3.1: In the closure studies summarized in Table 10, what were the assumptions about aerosol mixing state? A brief description is necessary so that the readers can know how the assumptions in those studies differ from the assumptions in this work. This might also help to understand why measured

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absorption is usually larger than that calculated, as pointed out by authors on Page 208. Have the closure studies examined surface DRE? Surface forcing should be very sensitive to the bias with single scattering.

- From line 19 on Page 212 to the first line on Page 213: This paragraph has been presented earlier in the paper and should be removed.
- Line 12 on Page 213: I don't agree with the argument of "This simplification is justified given that the emphasis here is on the radiative effects of anthropogenic aerosol and to lesser extent on differences due to differing treatments of the optical properties of natural aerosol components." The fixed f(RH) of sea salt would influence the comparisons of calculated optical depth (Figure 24) and surface forcing (Figure 25) , and influence calculated "a priori" and constrained DRE in the models. Chin et al [2002] have shown that sea salt f(RH) is very sensitive to RH.
- Line 17 on Page 214: It should be mentioned that that no additional water uptake above RH of 90% can lead to underestimates of optical depth and forcing. Adams et al. [2001] showed in a global study that such assumption led to an underestimation of 35% in DCF of sulfate.
- Page 216: Equation (6) should be justified by some calculations using the Mie Theory.
- Table 11: In the constrained calculations, mass scattering efficiency in the presence of dust is calculated using the parameterization given in the table caption. Such treatment should be described in the text and the associated references should be cited.
- Line 4 on Page 225: What is the excess POM?
- Equation (11): δ_vP should be Δ_aP.

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- Lines 1 and 6 on Page 231: Why do MOZART runs start from December of 1994? To spin up for several years?
- Page 237: Does STEM or MOZART consider the formation of sulfate and nitrate on sea salt and dust? Since the importance of sea salt and dust uptake is mentioned at several places in the text, the modeling studies of Dentener et al. [1996], Song and Carmichael [2001], Bauer et al. [2004], and Liao and Seinfeld [2005] should be cited to demonstrate the effect of sea salt and dust on the amount of sub-μm sulfate and nitrate.
- Line 9 of Page 238: "Remarkably similar results" is not appropriate. Based on the results given in Table 17, about 1/3 of mean column amounts from two models differ by more than 50%.
- Page 251: It is a good idea to calculate clear-sky DRF, since measurements were mostly taken under cloud-free or with very little cloud conditions. However, it is unusual to calculate just clear-sky DCF. I would suggest presenting both clear- and cloudy- skying DCF in the paper.
- Line 17 of Page 262: "has as large an effect" should be "has a large effect".
- Figures 24 and 25: It would be more interesting if AOD and DRE calculated based on measured aerosol concentrations and constrained optical properties can also be shown in these two plots. This could help to isolate the uncertainties associated with the predictions of aerosol chemical composition and loading, and would also demonstrate why predictions using constrained optical properties do not agree better with measurements as compared with "a priori" estimates.
- Line 1 on Page 266: "Ace Asia" should be "ACE-Asia".
- Line 20 on Page 266: It should also be noted that the IPCC forcings are cloudy-sky values, while those calculated in this work are estimates under clear-sky

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conditions.

- Line 28 on Page 266: It seems to me that an expression of uncertainty associated with sulfate forcing is missing.
- Page 269: It will be helpful to state clearly the extent to which the categorization of aerosol mixing state, parameterization of aerosol hygroscopic growth, and measured optical properties can be applied globally.
- Line 25 on Page 269: Again, it is not appropriate to compare clear-sky aerosol DCF with cloudy-sky forcing by greenhouse gases.
- Line 3 on Page 270: It is summarized in the text that "Constraining the aerosol properties employed in the radiative transfer calculations based on measurements resulted in AODs that were, on average, 34$\pm$8% larger than those obtained using the "a priori" optical properties. The effects of constraining the aerosol properties on calculated TOA DRE and DCF were similar (32$\pm$12% and 37$\pm$7% increase, respectively) but were less for SFC DRE and DCF (14$\pm$8% and 12$\pm$14% increase, respectively). These results imply that AOD and TOA DRE and DCF in these areas are greater than previously estimated." Since the "a priori" optical properties are limited to those in the models used in this work, one can not draw a general conclusion that AOD and TOA DRE and DCF in these areas are greater than previously estimated.
- Because large uncertainties of aerosol forcing are associated with sea salt and mineral dust, the authors might offer some suggestions about how to improve the modeling and measurements of their optical properties in the conclusion section.

References:

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Chin, M., et al., 2002: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, J. Atmos. Sci., 59, 461-483.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 175, 2006.