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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.

T. S. Bates et al.

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We thank reviewer 2 for their careful reading of the manuscript. We have addressed the minor points as follows:

Abstract: It should be described clearly that the constrained TOA DCF values of -3.3ś0.47, -14ś2.6, -6.4ś2.1Wm−2 are clear-sky forcings.

-Corrected.

Abstract: It is inaccurate to conclude that "Constraining the radiative transfer calcula-

tions 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.

-The sentence has been changed to: "With the use of constrained quantities (extensive and intensive parameters) the calculated uncertainty in DCF was 25% less than the "structural uncertainties" used in the IPCC-2001 global estimates of direct aerosol climate forcing.

Line 19 on Page 188: List specifically what "other substances" are. "(e.g., methansulfonate, sulfate)" has been added to the sentence.

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?

-Yes, corrected.

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

-We consider sea salt to exist in one predominately supermicrometer mode and assign one set of optical properties to this mode (Table 14). We state on page 211 line 9 that this sea salt "tails" into the submicrometer mode. For calculating submicrometer mass fraction we state on page 222 line 26 that the sea salt mass is considered to be 6% submicrometer and 94% supermicrometer.

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

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and/or constrained calculations?

-Both.

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

-The data from the long term monitoring stations were collected with high volume samples which sample only the total aerosol. The data from Sable Island were collected with impactors which measured the sub-micrometer fraction.

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 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.

-For Table 10, only studies that used microphysical/chemical properties to calculate optical properties require an assumption about mixing state. The direct optical measurements (in-situ or remote) make no such assumption. The studies using microphysical data (Clarke96, Collins2000, Quinn2002, Quinn2004) all used an internal mixture assumption. Note, however, that different particle sizes have different proportions among the components. This is very similar to the mixing assumptions made in this paper. The Collins et al (2000) paper examined sensitivity of calculated layer extinction to the mixing assumption and showed that external mixture assumption could cause increased or decreased extinction, most often increase, maximum of 25%, and maximum for polluted conditions of only 5%. For absorption, an external mixture assumption would cause the calculated absorption to be smaller by up to several 10's of percent. As Quinn2004 used an internal mixture, the mixing state can not account for the calculated low single scattering albedos. In reviewing this issue we find that we overstated the dis-

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crepancy and have changed the text to read: “Findings by Quinn et al.˜(2004) that the measured absorption was systematically greater than calculated by 50-100{\%} suggest that modeled single scattering albedo may be overestimated”.

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. Most of the paragraph has been removed. We have retained one sentence:

-As discussed above, the contribution of nitrate and sulfate to RH growth of the light scattering coefficient of dust particles is negligible even when dust contributes substantially to scattering (Carrico et al., 2003; Howell et al., 2004).

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.

-We agree that sea salt f(RH) is very sensitive to RH and at high values of RH the uncertainties in f(RH) are large. As our goal was to compare direct radiative forcing with the a priori and constrained optical properties we chose to hold sea salt f(RH) constant so as not to confuse the results. The reviewer is correct however that this biases the comparison in figures 24 and 25. We have added a sentence to the captions of these figures to highlight this problem. We have also taken out the word “justified” as the reviewer may be correct that it wasn’t.

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.

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-We state on line 23 of Page 214: “RHamb was capped at 95% because of the large uncertainty in measurements above this RH. This somewhat arbitrary cap may result in an underestimation of the scattering coefficient and thus DRE and DCF.”

Page 216: Equation (6) should be justified by some calculations using the Mie Theory.

-Such a relation is supported also by measurements during INDOEX (Howell et al., 2005 - Fig. 11).

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.

-The treatment is described in the text on pages 217 and 218. This parameterization is a product of this paper and hence there are no other references.

Line 4 on Page 225: What is the excess POM?

-In the University of Michigan radiative transfer model (UMich RTM), fossil fuel sulfate, POM, and black carbon (BC) are assumed to be internally mixed. In order to calculate the optical properties, we assumed that the mass ratio for POM to black carbon was 4.4 according to the global average value used in the inventories from the 2001 IPCC model inter-comparison [Penner et al. 2001]. However, when POM/BC is not equal to 4.4 from CTMs (MOZART and STEM), we use the CTM-produced BC to calculate a POM (denoted as POM_4.4). The CTM-produced POM minus the POM_4.4 is called the excess/insufficient POM here. This excess/insufficient POM is treated as externally mixed dry sulfate aerosol. This has been clarified in the manuscript.

Equation (11): _vP should be _aP.

-Corrected.

Lines 1 and 6 on Page 231: Why do MOZART runs start from December of 1994? To spin up for several years?

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-Yes.

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.

-MOZART does not consider any interaction between sulfate/nitrate aerosols and seasalt/dust. The Stem model does include these considerations. The details are presented in Youhua Tang, G. R. Carmichael, G. Kurata, I. Uno, R. J. Weber, C.-H. Song, S. K. Guttikunda, J.-H. Woo, D. G. Streets, C. Wei, A. D. Clarke, B. Huebert, and T. L. Anderson Antony D. Clarke, The Impacts of Dust on Regional Tropospheric Chemistry During the ACE-ASIA Experiment: A Model Study with Observations, Journal of Geophysical Research, 109, D19S21, doi:10.1029/2003JD003806, 2004. The presence of dust for the east Asia high dust case was shown to enhance sulfate production by 10-40% in dust-rich regions, and to result in an increase in the sulfate mass in the coarse fraction. The observational data also found similar amounts of coarse-mode sulfate. Reactions involving NO2 and nitric acid were shown to result in the accumulation of nitrate into the aerosol, and this occurs mainly in the coarse mode (however, appreciable amounts may also appear in the fine mode). The suggested references have been added to the subsection on primary dust aerosol.

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%.

-“Remarkably similar results” has been removed from the manuscript.

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

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to calculate just clear-sky DCF. I would suggest presenting both clear and cloudy- sky-ing DCF in the paper.

-The manuscript is already very long. Consequently we choose to include only the clear sky data in this paper.

Line 17 of Page 262: “has as large an effect” should be “has a large effect”.

-Corrected.

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.

-This is a difficult comparison since these Figures and Tables 26 and 27 are comparing a model grid box region over the time period of the experiment. A comparison with measured aerosol concentrations would be the point/single time closure experiments. Examples are given in Table 10.

Line 1 on Page 266: “Ace Asia” should be “ACE-Asia”.

-Corrected.

Line 20 on Page 266: It should also be noted that the IPCC forcings are cloudysky values, while those calculated in this work are estimates under clear-sky conditions.

-We have amended the sentence to read: “However such comparison is subject to the caveat that the previous estimates were for global average total-sky direct radiative forcing, whereas the present estimates are clear-sky direct radiative forcing for specific oceanic domains and during specific periods that are unlikely to be representative of the global mean.”

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Line 28 on Page 266: It seems to me that an expression of uncertainty associated with sulfate forcing is missing.

-Line 28 states that uncertainty for sulfate is a factor of 2.

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.

-This is a difficult question. I personally think these categories/properties could be anywhere over the oceans but this needs to be tested further.

Line 25 on Page 269: Again, it is not appropriate to compare clear-sky aerosol DCF with cloudy-sky forcing by greenhouse gases.

-We state beginning on line 28: “However it must be stressed that such forcings are overestimates of the actual aerosol forcings because they do not take into account the fraction of the domain that is covered by clouds, for which aerosol direct effects will be minimal.

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, 34ś8% 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 (32ś12% and 37ś7% increase, respectively) but were less for SFC DRE and DCF (14ś8% and 12ś14% 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.

-Agreed. We have changed “are greater” to “may be greater”.

Because large uncertainties of aerosol forcing are associated with sea salt and mineral

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dust, the authors might offer some suggestions about how to improve the modeling and measurements of their optical properties in the conclusion section.

-Sea salt and dust are considered natural and thus are not forcings. Improvements in estimates of DRE will require improved parameterizations of source and sink fluxes of these components. The second to the last paragraph of the manuscript has been edited to include this point.

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