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Global-scale black carbon profiles observed in the remote atmosphere and compared to models

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[1] Refractory black carbon (rBC) aerosol loadings and mass size distributions have been quantified during the HIPPO campaign above the remote Pacific from 80N to 67S. Over 100 vertical profiles of rBC loadings, extending from ~0.3 to ~14 km were obtained with a Single-Particle Soot Photometer (SP2) during a two-week period in January 2009. The dataset provides a striking, and previously unobtainable, pole-to-pole snapshot of rBC mass loadings. rBC vertical concentration profiles reveal significant dependences on latitude, while associated rBC mass size distributions were highly uniform. The vertical profiles averaged in five latitude zones were compared to an ensemble of AEROCOM model fields. The model ensemble spread in each zone was over an order of magnitude, while the model average over-predicted rBC concentrations overall by a factor five. The comparisons suggest that rBC removal in global models may need to be evaluated separately in different latitude regions and perhaps enhanced. **Citation:** Schwarz, J. P., J. R. Spackman, R. S. Gao, L. A. Watts, P. Stier, M. Schulz, S. M. Davis, S. C. Wofsy, and D. W. Fahey (2010), Global-scale black carbon profiles observed in the remote atmosphere and compared to models, *Geophys. Res. Lett.*, 37, L18812, doi:10.1029/2010GL044372.

1. Introduction

[2] Black carbon (BC) is the most strongly light-absorbing component of combustion-generated aerosols. There is much interest in confidently determining the extent to which it affects the radiative balance of the atmosphere – estimated as a large fraction of that of carbon dioxide (CO₂) [Ramanathan and Carmichael, 2008]. The mechanisms through which BC is believed to affect the radiation balance are numerous and complex; hence there are large uncertainties in estimates of BC forcing. As BC is clearly recognized as a major contributor to climate change with a lifetime in the atmosphere that is relatively short (*i.e.*, < weeks for most emitted BC), it has been identified as a

possible species for reduction as part of short-term climate control strategies [Levy *et al.*, 2008]. Thus, accurate modeling of BC mass loadings is of substantial interest to climate scientists and policymakers.

[3] There are large differences among model estimates of BC spatio-temporal loadings in the atmosphere in comparison to observations [Koch *et al.*, 2009]. While a recent study [Textor *et al.*, 2007] has shown that differences in emissions inventories used by models only contribute weakly to inter-model variability in BC predictions, another study [Vignati *et al.*, 2009] has identified wet deposition parameterization as a major contributing factor to such variability. Here we focus on new aircraft measurements of BC aerosol mass loadings and mass (size) distributions carried out in the remote Pacific troposphere and lower stratosphere. With flights that incorporated nearly continuous vertical profiling over a global latitude range in a brief 2-week period, a unique dataset was obtained for evaluating and constraining global aerosol models.

2. Experimental Measurements

[4] The measurements were made with a Single-Particle Soot Photometer (SP2) [Schwarz *et al.*, 2008; Schwarz *et al.*, 2010] operated on board the NSF/NCAR GV [www.hiaper.ucar.edu] aircraft in the first deployment of the HIAPER Pole-to-Pole Observations campaign (HIPPO1). HIPPO1 flights provided extensive vertical coverage of the remote troposphere in the Pacific, Arctic, and near-Antarctic regions. Latitudes from 80N to 67S were reached in the brief period of 9–23 January 2009 (Figure 1). Sampling occurred during 105 vertical profiles extending from ~0.3 to 14 km altitude. At the higher altitudes, air was sampled in the lower stratosphere in middle and polar latitudes, and the upper troposphere in the tropics.

[5] The SP2 measures the mass of a specific absorbing component of individual aerosol particles defined as “refractory BC” (rBC) [Schwarz *et al.*, 2010]. In the context here rBC is interchangeable with the more general term “BC” as defined in model parameterizations, which is also the quantity needed for emission inventories [Dentener *et al.*, 2006; Schwarz *et al.*, 2010]. The SP2 quantifies rBC mass in individual aerosol particles independently of particle morphology and mixing state [Cross *et al.*, 2010]. Differences in the optical model used for BC aerosol in global aerosol models (*i.e.*, the choice of mass absorption efficiency, accounting for enhanced absorption by internal mixing, etc.) are unimportant in the mass loading intercomparisons presented below.

[6] Mass distributions of the rBC component of ambient aerosol are formulated by converting the individual rBC

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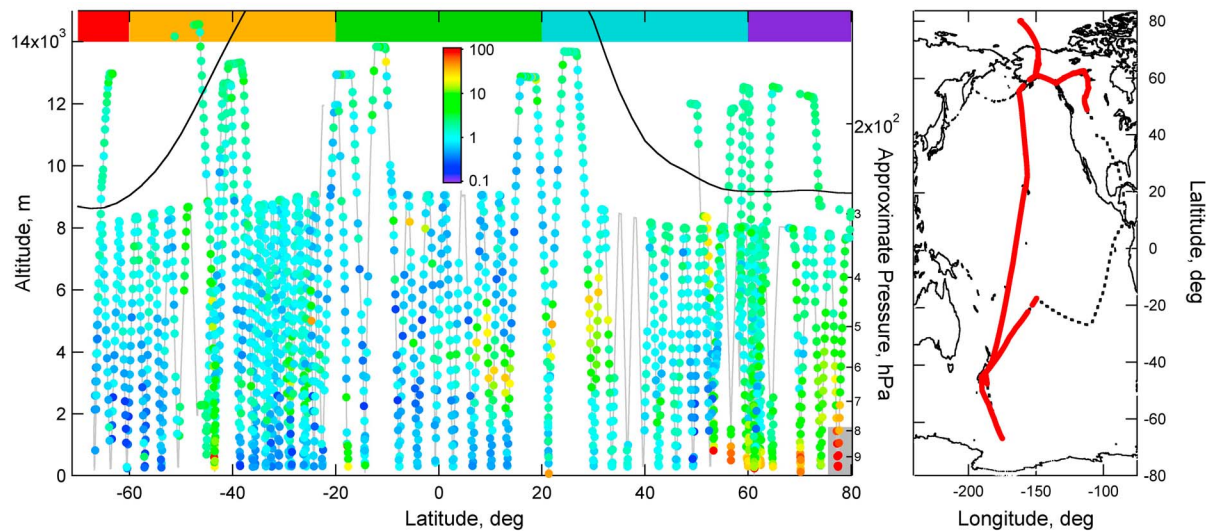


Figure 1. (left) Location of the 105 vertical profiles of rBC MMR analyzed here. Points indicate rBC MMR values using the vertical color scale (60 s averages, units of ng-rBC/kg-air). Colors at the top indicate the five latitude zones highlighted in the text and Figure 3. The grey-shaded region indicates the Arctic pollution observations. The black line is the NCEP retrieved tropopause height averaged over longitude -200 to -100 [Kalnay *et al.*, 1996]. (right) Map showing the entire HIPPO1 flight path and the flight legs included here (red solid line, 9–23 January, 2009).

masses detected to volume-equivalent diameters (VED) by assuming a density of 2 g cm^{-3} for the rBC material (Figure 2). The distributions show that $\geq 90\%$ of the rBC mass in the log-normal accumulation mode was detected. rBC mass mixing ratios (MMRs) were calculated by integrating the individual rBC masses in a volume of ambient air and scaling the result upwards by 10% to better represent the total rBC mass in the accumulation mode. rBC MMR values in HIPPO1 varied over four orders of magnitude, from $\sim 300 \text{ ng-rBC/kg-air}$ in Arctic pollution to the cleanest ambient air yet sampled with an SP2 ($\sim 0.03 \text{ ng-rBC/kg-air}$ in the upper tropical troposphere). The MMR values are linked to calibration standards at the level of 20%. Details of the SP2 operation including uncertainties and critical operating parameters during these measurements are included in Text S1.¹ SP2 data from horizontal flight legs were excluded from the rBC MMR analysis, along with data taken in polluted air encountered below 2 km near airports. Finally, data in water- or ice clouds was excluded from analysis.

3. The rBC Vertical Profiles

[7] Individual vertical profiles of rBC MMR were produced by averaging MMR values observed in 1-km altitude intervals for each ascent or descent of the aircraft. Profiles located in five latitude zones (67–60S, 60–20S, 20S–20N, 20–60N, and 60–80N) were then separately averaged with equal weighting to obtain a representative profile for each zone (Figure 3, top left). The number of profiles in each zone is indicated in the legend. The atmospheric variability of rBC MMR within each altitude interval for each latitude zone was estimated as the standard deviation of the included 1-km average MMRs, and is shown as a set of whiskers on

each average profile in Figure 3. Although the observations were made at long distances from BC sources, the latitudinally averaged profiles show significant variations across zones at all altitudes.

[8] The range of rBC MMRs in the UTLS ($\sim 0.1 - 3 \text{ ng/kg}$) was somewhat less than that at near-surface levels ($0.05 - 50 \text{ ng/kg}$), suggesting that mixing and transport has blended polluted and clean, remote air. The lowest values in the upper troposphere are observed in the tropics, possibly evincing highly efficient rBC removal in convection before detrain-

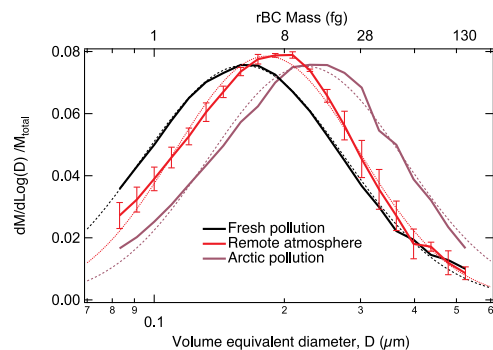


Figure 2. Measured mass distributions of the rBC component of ambient aerosol particles normalized by total rBC mass observed in each case. The remote atmosphere distribution is the average of the distributions found in four latitude bands (60–20S, 20S–20N, 20N–60N and 60N–80N) and excluding both the far southern region (where too few particles were observed to provide a robust distribution) and the Arctic pollution. Whiskers are the standard deviation of the four values within each size interval. Fresh pollution was observed primarily near airports visited during HIPPO1. The dashed lines are lognormal fits to the observations (see text).

¹Auxiliary materials are available in the HTML. doi:10.1029/2010GL044372.

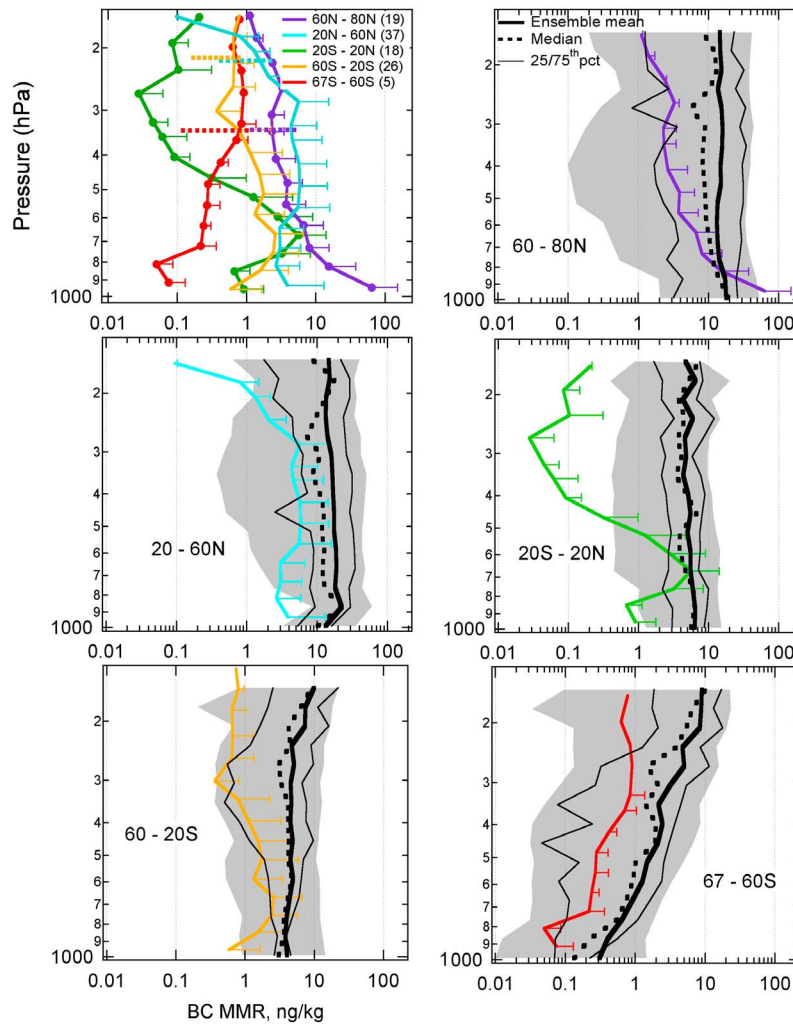


Figure 3. (top left) The average vertical profile of rBC MMR observed in each latitude zone with horizontal dashed lines indicating the approximate tropopause location (which varied from one profile to the next, in general). The whisker for each 1-km interval represents the MMR standard deviation of the separate profile values (see text). The numbers in parentheses are the number of individual profiles incorporated in each average. Colored lines in additional panels repeat the observed average profiles. The range of rBC mass-mixing ratio from the AEROCOM model set is indicated by the grey shading. The model mean (median) is shown with the solid (dashed) black line and the 25th and 75th percentile model results are shown with the thin black lines. Note that Figure 1 provides vertical profiles as a function of altitude, with an accompanying pressure-altitude scale.

ment, while the Northern and Southern polar UTLS show higher (although still low) levels. This implies that either the polar UTLS regions are influenced by a different source of rBC than the clean air uplifted from the tropics, or by transport from different times with higher Pacific tropical UTLS loadings. The LS concentrations are very similar in the polar regions, suggesting a common source.

[9] In the mid-troposphere the rBC loadings are distinctly different in different latitude zones. Higher loadings of rBC here are likely due to long range transport from continental regions. For example, profiles observed around 30N contained $\sim 20\text{--}30$ ng/kg enhancement of rBC between $\sim 700\text{--}400$ hPa (not shown separately) with respect to other nearby profiles. This enhancement contributed to the increased rBC MMR in the 20–60N band. HySplit back trajectories [www.arl.noaa.gov/HYSPLIT_info.php] indi-

cate that northern Asia was a likely source of pollution for these enhanced values.

[10] The contrast between the Arctic and Antarctic rBC MMR profiles is greatest in the lowermost troposphere where Arctic loadings are larger by a factor of ~ 1000 . This is likely due to the stronger rBC sources in the northern hemisphere, differences in advection of pollution to the two regions, and the interhemispheric phase difference in seasonal removal. The distinct enhancement at the lowest altitudes in the northern polar region was due to Arctic pollution observed only on the northernmost descent, where rBC MMR reached values of 240 ng/kg in 60-s averages (Figure 1).

4. AEROCOM Model Intercomparison

[11] We compare the averaged vertical profiles of rBC MMR to results from the Aerosol Comparisons between

Observations and Models (AEROCOM) suite of global aerosol models (<http://dataipsl.ipsl.jussieu.fr/AEROCOM>) [Textor *et al.*, 2007], in Figure 3. 14 models [LMDzT-INCA (LSCE); ECHAM5(MPI); GCM/CAM; MIRAGE; CTM2; CCM-Oslo; LMDzT (LOA); GOCART; MATCH; IMPACT/DAO; ECHAM-MADE (DLR); GISS; TM5; MOZART-GFDL-NCAR] produced global rBC MMRs based on an emissions inventory for the year 2000 as part of the Phase I experiment A [Dentener *et al.*, 2006]. The models generated average “January” rBC MMRs using similar climatologies. BC MMR vertical profiles from each model were first interpolated to the average latitude and longitude of each aircraft profile, and then averaged to the same fifteen altitude intervals used in the averaging of the observational data (vertical resolution was not constant between models). For each latitude zone, these profiles were then averaged together to produce a mean profile for each model, closely following the treatment of observational data.

[12] The model ensemble successfully replicates some features of the observational dataset, but the mean and median model absolute values clearly tend to overestimate rBC MMRs, on average by a factor 5 in the entire dataset analyzed here. The vertical structure of the model profiles appears to have the best agreement with that of the observations in the far southern zone, where both models and observations show rBC concentrations increasing with altitude. Poorer agreement exists outside of this region where the models generally do not show the decreases of rBC MMRs with altitude. This spread may be caused by an underestimation of removal of presumably less-aged rBC by convective precipitation.

[13] In the UTLS the model ensemble has a clear high bias in the tropics and subtropics with less obvious high bias in the poleward zones, where ~25% of the models predict lower mixing ratios than observed. Here model assumptions about mixed and ice cloud scavenging ratios and convective scavenging have important consequences. Thus, the comparison suggests that the tropical/subtropical and extra-tropical/polar removal rates need separate evaluation and perhaps improvement in the model parameterizations.

[14] In the mid-to-lower troposphere, the model ensemble still shows higher MMRs than observed, but to a lesser extent; a decrease from mid to upper tropospheric levels was observed but not simulated on average by the models. However, particularly in this altitude range, this slightly better agreement may be an artifact of local conditions during sampling; it is possible that the observed rBC MMRs were influenced by specific pollution events that were missed by models simulating conditions in an idealized year 2000 (e.g., the Asian pollution event mentioned above). In the case of the measurements in the latitude range 60S–20S, southbound and northbound measurements were separated in time by approximately one week. For those measurements the vertical structure was very similar, and model overestimation of rBC MMR was consistent over both sets of measurements.

[15] The increase in rBC MMR from the boundary layer to the UTLS in the southern polar latitudes may be a result of loadings from very remote sources reaching steady state equilibrium with removal processes at high latitudes; the UTLS rBC MMR is higher due to a longer lifetime in those dry levels, while mid and low-level clouds more

effectively reduce loadings at lower altitudes. The source of rBC-containing particles may be circumpolar transport of pollution in South America and/or South Africa, or the Brewer–Dobson general circulation. The profile form is well represented by the model ensemble even though a high bias exists at all levels in most models. The bias suggests, again, insufficient wet removal in the models, most specifically in the convection that is assumed to loft rBC to the UTLS for transport into the region.

5. The rBC Mass Distributions

[16] Distributions of rBC mass quantified in individual particles provide a valuable constraint on models for the integrated removal from the time of emission to detection. Here average rBC mass distributions were calculated in three different air mass types and fit with lognormal functions (Figure 2). In local pollution observed near airports the rBC median mass diameter (MMD) of the mass distribution was $0.16 \mu\text{m}$ VED with a geometric standard deviation, σ , of 1.71. This mode is virtually identical to the rBC mode observed in fresh urban emissions during a Texas study [Schwarz *et al.*, 2008]. Note that local pollution observations are excluded from the vertical profile/MMR analysis above. At low altitudes in the northern-most Arctic explored (~80N, Figure 1) rBC was observed with larger MMD ($0.220 \mu\text{m}$ VED, $\sigma = 1.68$) consistent with fresh biomass burning emissions observed in Texas (MMD = $0.21 \mu\text{m}$ VED, $\sigma = 1.43$ [Schwarz *et al.*, 2008]). rBC MMD in remote air masses excluding Arctic pollution was seen to lie in a very narrow intervening size range: MMD = $0.181 - 0.183 \mu\text{m}$ VED ($\sigma = 1.64 \pm 0.06$); i.e., the rBC mass distribution was essentially constant in clean remote air masses.

6. Discussion

[17] The uniformity of rBC mass (size) distribution in the remote regions is an interesting result indicating that removal processes are not strongly sensitive to the rBC mass incorporated in individual particles: after long integration times the mass distributions have not changed dramatically. A common “final” mass distribution indicates that rBC from many different sources (and thus, presumably, of different original sizes) has undergone removal processes that shift it all towards a single ultimate mass distribution (note that the remote mass distribution is too narrow to be a superposition of rBC from urban/biomass burning mass sources). The fact that the remote distribution is intermediate in size between typical sources (median-mass diameter of $0.182 \pm 0.001 \mu\text{m}$) implies that more than one removal process with different size dependencies reach a steady-state, balanced condition; one process removing particles with small amounts of rBC, and another removing those with larger rBC amounts. As a particle’s total size is determined by more than rBC content, these may be different processes than those (coagulation/removal) that size-select the accumulation mode of total aerosol. The immediate value of these observations is in providing a benchmark for models that can directly simulate the evolution of rBC mass distributions.

[18] Compared to our observational data, fourteen AEROCOM models overestimate the rBC concentrations in remote regions by a factor of five on average. Koch *et al.*

[2009] also noted model overestimations of rBC mass over continents for a similar set of AEROCOM models. We attribute our results to insufficient wet removal of rBC-containing aerosol in the models because we believe that neither overestimation of rBC emissions nor errors in advection processes are responsible for the entirety of the large positive bias of model results over observations.

[19] In the present study, the models used 1996 emissions updated to the year 2000, with an overall uncertainty of a factor of two [Bond *et al.*, 2004]. Actual emissions during HIPPO1, especially from East Asia, may be higher. The inventory did not include aircraft emissions, which can only accentuate the observed data/model bias. To first order rBC concentrations scale linearly with emissions, hence the emissions inventory would have to be unrealistically high to explain differences between the models and the observations that are as large as two orders of magnitude – or even a factor 5 on average.

[20] Advection processes are also unlikely to explain the global-scale bias seen here. The measurements were performed in a short two-week time interval and thus form a “snapshot” of rBC loadings; additionally they provide a “curtain” of wide latitudinal and vertical coverage. Thus large-scale features that are likely to undergo long-distance zonal transport have little opportunity to bypass the sampling region (this requiring transport around the poles, or at altitudes above those sampled). This is a powerful feature of this data set. However, model simulations using coherent meteorology for the measurement period could provide a more direct comparison to the observed vertical structure of rBC MMR. Note that advection and vertical mixing affects the interaction time between aerosols emitted at the surface and precipitation events, and can thus influence modeled aerosol removal rates.

[21] The lowest rBC concentrations and highest model bias occurred in the tropical upper troposphere, suggesting that increasing removal rates through wet deposition in convection is a possible route to improving model skill in predicting rBC MMRs in the tropics and subtropics — this could be accomplished by accelerating treatment of rBC aging. Better model-data agreement in the latitudes further poleward suggests that mixed- and ice phase cloud removal may need to be addressed separately in the extratropical regions.

[22] The results obtained in the remote atmosphere do not allow one to infer a similar possible model bias closer to rBC source regions (although the results of Koch *et al.* [2009] support such a conclusion). Thus, increasing modeled removal rates during long-range transport would not exclude the possibility that, closer to the source regions in clear skies and anticyclonic weather, models underestimate BC levels. Koch *et al.* [2009] proposed just such a scenario for South America and South Africa based on Aeronet absorption retrievals. However, that comparison was based on a full year of observations, while the January measurements used here do not include the contributions of rBC

from the extensive biomass burning that typically occurs in the southern-hemisphere dry season (October–January).

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