AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER
10.1002/2016GL071241

Key Points:
- A new large data set of black carbon aerosol vertical profiles has been collected and analyzed with comparison to global models
- AeroCom global models do not underestimate Arctic BC loads
- Upper tropospheric BC loadings provide a powerful constraint on model transport and removal processes

Supporting Information:
- Supporting Information S1

Correspondence to:
J. P. Schwarz,
joshua.p.schwarz@noaa.gov

Citation:
Schwarz, J. P., B. Weinzierl, B. H. Samset, M. Dollner, K. Heimerl, M. Z. Markovic, A. E. Perring, and L. Ziemba (2017), Aircraft measurements of black carbon vertical profiles show upper tropospheric variability and stability, *Geophys. Res. Lett.*, *44*, 1132–1140, doi:10.1002/2016GL071241.

Received 23 SEP 2016
Accepted 23 NOV 2016
Accepted article online 9 DEC 2016
Published online 26 JAN 2017

Aircraft measurements of black carbon vertical profiles show upper tropospheric variability and stability

Joshua P. Schwarz[1], Bernadett Weinzierl[2,3,4], Bjørn H. Samset[5], Maximilian Dollner[2,3,4], Katharina Heimerl[2,3], Milos Z. Markovic[6], Anne E. Perring[1,7], and Luke Ziemba[8]

[1]Chemical Sciences Division, NOAA Earth System Research Laboratory, Boulder, Colorado, USA, [2]Deutsches Zentrum für Luft- und Raumfahrt, Oberpfaffenhofen, Germany, [3]Faculty of Physics, Meteorological Institute, Ludwig-Maximilians-Universität, München, Germany, [4]Aerosol Physics and Environmental Physics, University of Vienna, Vienna, Austria, [5]Center for International Climate and Environmental Research–Oslo, Oslo, Norway, [6]Piccaro, Inc., Santa Clara, California, USA, [7]Cooperative Institute for Research in Environmental Science, Boulder, Colorado, USA, [8]NASA Goddard Flight Center, Greenbelt, Maryland, USA

Abstract We present new data sets of black carbon (BC) aerosol mass mixing ratio (MMR) obtained from aircraft missions over North America, Europe, the Arctic, and the outflow region of Saharan Africa before and after trans-Atlantic transport. The data, collected from 2011 to 2013 with single-particle soot photometers, provide new insight into the variability and distribution of BC over global scales and refine understanding of AeroCom global model ensemble performance. The results indicate extensive global-scale longitudinal mixing of BC above altitude pressures as low as 400 hPa. They also constrain the absolute and temporal variability of upper tropospheric BC MMR and point to opportunities for new tests of global aerosol models in the upper troposphere. A comparison to the AeroCom Phase II results generally reinforces previous estimates of the ensemble performance, except that it also strengthens confidence that the ensemble actually is biased high in the Arctic in all seasons.

1. Introduction

Black carbon (BC), an aerosol material with strong anthropogenic sources, generates a substantial and highly uncertain impact on climate. The Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report estimate of fossil fuel and biofuel BC direct radiative forcing (DRF) is 0.4 W m^{-2}, with a range (0.8 W m^{-2}) that contributes a large fraction of the overall uncertainty in total aerosol DRF ($\sim -0.9 \pm 1$ W m^{-2}) [*Myhre et al.*, 2013]. There are several reasons for such a large uncertainty contribution from BC although it is only a small (on the order of 1%) component of total aerosol mass. First, BC is a highly efficient absorber of light, to the extent that it strongly contributes to aerosol absorption of shortwave radiation globally [*Chin et al.*, 2009]. Second, its ability to absorb light can evolve substantially in time from emission [*Wang et al.*, 2014] and also depends strongly on altitude [*Samset and Mhyre*, 2011]. Third, the global abundance and distribution of BC (especially in the vertical direction) are poorly known, in large part because BC's net optical perturbations are difficult to distinguish from the large light-scattering and extinction effects of total aerosol.

In light of BC's importance, the performance of the AeroCom suite of models, which were an integral starting point for IPCC estimates of BC DRF, has been broadly assessed from available measurements [*Koch et al.*, 2009; *Schwarz et al.*, 2013; *Wang et al.*, 2014; *Samset et al.*, 2014, and more]. The model suite consists of up to 16 global climatological models run with similar emissions inventories and unified meteorological conditions. Currently, a simple narrative of its performance is now fairly well established: it underpredicts BC concentrations in some source regions and the Arctic and overestimates them in the remote and at high altitude, while more broadly underestimating column aerosol absorption optical depth [*Koch et al.*, 2009; *Bond et al.*, 2013]. To address these biases, AeroCom BC DRF has been alternately increased and decreased depending on the location and source for the bias. Over the populated world and source regions the model estimate was increased to improve agreement of model and Aeronet column absorption aerosol optical depth by *Bond et al.* [2013]. Over North America, Europe, and the outflow from Africa, regions represented in the data set to be presented here, the BC optical depth was scaled upward by factors of ~1 to 6, resulting in DRF annual average increases of ~0.4, ~0.5, and ~1 W m^{-2}, respectively. Conversely, to address high model bias in the remote atmosphere and at high altitudes [*Schwarz et al.*, 2010, 2013], a 15% reduction globally has been applied by *Bond et al.* [2013].

Here we present new in situ airborne data obtained by researchers from Germany and the U.S. using single-particle soot photometers (SP2s). The data indicate stronger global relevance for middle and upper tropospheric measurements of BC concentration than previously understood and provide new constraints on BC's global distribution and temporal variability. The measurements are compared to AeroCom and are generally found to be in-line with the existing narrative presented above, except in the Arctic region.

The experimental details of the measurements and vertical profile-focused analysis are outlined in section 2. The observations are presented and discussed in section 3. Section 4 summarizes the results and their relevance to estimates of global atmospheric BC distributions, and to future global model evaluations.

2. Experimental and Analytical Technique

The data were obtained with single-particle soot photometers (SP2s) operated by two different groups on two different aircraft. The Deutsches Zentrum für Luft- und Raumfahrt (DLR) SP2 flew on board the DLR Falcon Research Aircraft as part of campaigns focused on four different regions: Europe (CONtrail and Cirrus ExpeRimenT (CONCERT), September 2011 [*Dahlkötter et al*., 2014]), the Arctic (Arctic Climate Change Economy and Society (ACCESS), July 2012 [*Roiger et al*., 2015]), North America (Deep Convective Clouds and Chemistry (DC3), June 2012 [*Barth et al*., 2015]), and the northern hemisphere Atlantic (Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE), June/July 2013 [*Weinzierl et al*., 2016]). The NASA DC-8 research aircraft carried a NOAA SP2 system over North America in both the 2012 DC3 mission (for a longer period than the Falcon participated, over the period of May–June) and the 2013 Studies of Emissions, Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC4RS) effort (August/September [*Toon et al*., 2016]).

The SP2 instruments provide accumulation mode refractory black carbon (rBC) mass mixing ratios with an absolute uncertainty of 25% [*Laborde et al*., 2012; *Schwarz et al*., 2013]. "Refractory black carbon" (rBC) is the accepted nomenclature for the material quantified by the SP2 [*Petzold et al*., 2013], hence used in the detailed discussion here. rBC is experimentally equivalent to "elemental carbon" (the term applied to thermal decomposition measurements of BC broadly used in model inventories) with uncertainty of only 15% [*Kondo et al*., 2011]; Both instruments were calibrated in the same way, using the calibration material (Fullerene soot, as produced, Lot #F12S011, Alfa Aesar Inc., Ward Hill, MA) recommended for the SP2 measurement community [*Baumgardner et al*., 2012]. Adjustments for accumulation-mode rBC mass undetected by the SP2s were calculated on a per-flight basis by fitting a lognormal curve to observed rBC mass size distributions and integrating over the detection range of the instruments to identify the fraction of the total mass that was detected. These adjustments were typically on the order of 20%. This is an approach commonly used over the last decade to account for accumulation mode rBC mass not detected by the SP2 [*Schwarz et al*., 2006; *Reddington et al*., 2013] and allows us to better represent the total rBC mass concentration. During the flights described here, there was no evidence of additional modes of rBC at smaller sizes (i.e., rBC mass concentrations were always decreasing with decreasing particle rBC size at the lower detection limit of the instrument). Data obtained in clouds was excluded from analysis to avoid inlet artifacts. During the DC3 campaign, the research aircraft Falcon, carrying the DLR SP2, flew wing-tip to wing-tip with the NASA DC-8 research aircraft, which was carrying a NOAA SP2 system for the mission (over the period of May–June); this provided a convincing demonstration that the performance of the two instruments is well matched: accumulation-mode rBC aerosol mass mixing ratios (MMRs) agreed within 5% over the range of altitude and concentrations sampled. On the basis of this result and the variability in calibration sensitivity for the instruments, we estimate that the measurements of rBC MMR from the different instruments and at different times to be equivalent at the 10% level, well below the absolute uncertainty of 25%.

Vertical profile analysis was conducted following *Schwarz et al*. [2010, 2013] by identifying each ascent and descent of the aircraft and treating each as an independent measurement of vertical profile of rBC concentration. One-kilometer altitude bins were used for averaging rBC MMR; within each bin corollary averages were calculated for ambient pressure. The pressure data obtained at times that rBC data were removed due to cloud interferences were not included in either of the averages. For example, if the bottom half of a 1 km altitude range of an ascent was obstructed by clouds, the average rBC MMR and the average ambient pressure associated with the corresponding bin would only have data from the upper half.

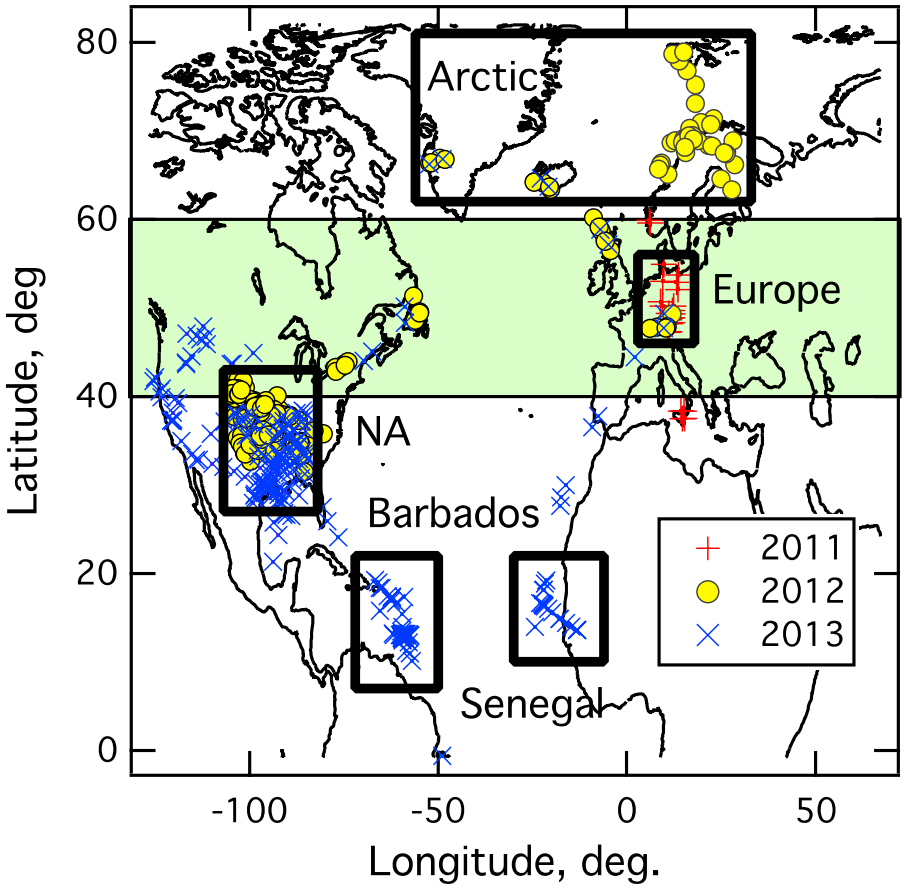

Figure 1. Map showing the location of measured vertical profiles (markers, segregated by year of observation) and the analysis regions (rectangles). The light horizontal bar shows the 40–60°N latitude band discussed in the results section.

Vertical profiles representing the focus regions shown in Figure 1 were generated by averaging, within each altitude level, the individual profile rBC MMR values with unity weighting. Only values representing a minimum of 3 contributing profiles in each 1 km bin were considered valid; this requirement was only relevant at the highest altitudes in smaller collections of profiles, because it reduced the number of points shown at the very tops of the averaged profiles, where perhaps just one or two contributing profiles extended higher than the other in a given analysis area. The value of this approach is that it produces both representative vertical profiles and an estimate of variability between measurements (i.e., the standard deviation of rBC MMR observed between different profiles at the same altitudes). Hence, aircraft sampling strategy (which, for these missions, often targeted specific events) only weakly influences averaged vertical profiles. This is demonstrated by comparison of vertical profiles from the same region (North America, 2012) when the two SP2s flew on aircraft with markedly different sampling strategies (the DLR Falcon targeting both biomass burning emissions and thunderstorm outflow, while the NASA DC8 focused on convective pumping of boundary layer air to the upper troposphere (UT)). The two strategies produced vertical profiles that were very consistent at the level of the analysis presented here (Figure S1 in the supporting information). Hence, we assume that the regional averages produced from this analysis approach are unbiased by aircraft flight-plan choices.

Comparison to AeroCom Phase II BC vertical profile data was performed by using the same 12-model ensemble of opportunity as in *Schwarz et al*. [2013]. Briefly, predictions for total BC MMR were extracted for model runs using year 2000 emissions and year 2006 meteorology, for the model bin closest in time and three-dimensional location to each of the measured vertical profiles, and subsequently averaged in the same way as the observational data. An unavoidable limitation of our model-observation comparison is that the model emissions predate the measurements by more than a decade. In the meantime, BC anthropogenic emissions have risen by 10% over 2000–2010 globally and in Africa, while declining in Europe (−10%) and

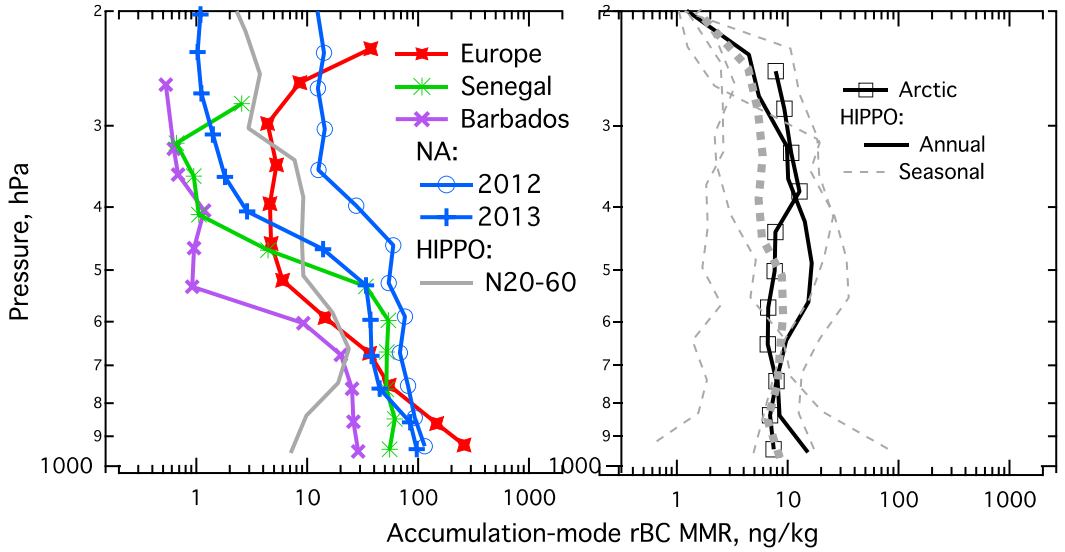

Figure 2. (left and right) Vertical profiles of rBC mass mixing ratio (MMR) measured in the analysis regions, with profiles measured in the remote as part of the HIPPO campaign included. The HIPPO data extend to lower pressures than shown here. The Arctic profiles are shown alone on the right (light dashed lines) for clarity, with only the HIPPO4 profile in heavy dashed line.

the U.S. (−30%) over the same period [*Klimont et al.*, 2016]. However, annual differences in biomass burning emissions are expected to dwarf the possible bias in anthropogenic emissions, meaning that detailed comparisons need to use total BC emissions and meteorology for the year of measurement, and high spatial and temporal resolution, to improve on the comparisons in the present work. Note that here only ensemble performance is tested; individual models can produce MMRs that vary over more than an order of magnitude in some regions (see Figure S2). More detailed discussion of individual model structure, and the impact of aerosol lifetime on their performance, can be found in *Samset et al.* [2014].

A known caveat of the comparisons is that AeroCom only provided model information on monthly time resolution, and on a horizontal grid typically of $2 \times 2°$ in resolution, limiting the value of direct comparisons on small length/time scales. See *Schwarz et al.* [2013] and *Schutgens et al.* [2016] for further discussions of the possible impacts of this on the comparison. To the extent that the averaged measured profiles do indeed represent the regional-scale climatological values, we do not consider these to be significant limitations to our conclusions about model performance: the models show relatively small changes over these length and time steps outside of strong seasonal source regions.

3. Results and Discussion

Approximately 500 vertical profiles were collected over the 3 years, as shown in Figure 1. Mission dates and profile breakout are provided in Table S1 in the supporting information. Most of the data were obtained in North America in 2012/2013, on either side of the subtropical Atlantic in 2013, in the European Arctic in 2012, and over Europe in 2011. Additionally, in 2012 and 2013 there were profiles associated with a total of three trans-Atlantic flights of the DLR Falcon between Germany and the U.S. with stops (i.e., landings and takeoff s leading to vertical profiles) in Great Britain, Iceland, Greenland, and Newfoundland. Table S2 summarizes the contributing missions and profile statistics for each region. This section first presents the average profiles associated with each region, with a focus on temporal and spatial trends, and then compares them to AeroCom model suite climatological predictions.

Measured vertical profiles of rBC MMR averaged over each focus region are shown in Figure 2 (traces with lines and markers). The number of profiles from each mission that contributed to each region are provided in Table S2. Additional previously published profiles from five seasonal measurement series over the remote Pacific (20–60°N) and the Arctic (60–85°N), obtained as part of the High-performance Instrumented Airborne

Platform for Environmental Research Pole-to-Pole Observations (HIPPO) campaign in 2009–2011 [*Schwarz et al*., 2013], are also shown (traces without markers). The Arctic profiles are broken out in Figure 2 (right); here the individual seasonal measurements from HIPPO are shown with dashed lines. Note that the HIPPO data extend to higher altitudes than the current data set; only the portions of those profiles in the altitude range of the new data sets are shown.

The new Arctic data consist of a 3 week period in 2012 (the ACCESS mission), as well as other data from 2012 and 2013 (i.e., the DC3 and SALTRACE transit profiles). The newly measured profiles in the Arctic, at very different longitude from the HIPPO results, are in excellent agreement with the HIPPO4 profile that was obtained at nearly the same season (heavy dashed line in Figure 2, left). The result lies within the envelope of seasonal variability seen in HIPPO, which was wider than seen here and ~1 order of magnitude in rBC MMR through the troposphere. The results show a rBC MMR of ~10 ng/kg that is nearly independent of altitude, consistent with weak local sources/sinks and widespread distant source regions.

An order of magnitude variability in UT rBC MMR was observed in the North American (NA) profiles (blue markers), which because of excellent statistics could be separated between 2012 and 2013. The rBC MMRs associated with the two time blocks are quite consistent at ~100 ng/kg in the lowest 2 km (~10% difference in MMR, which is at the limit of our precision), likely reflecting high consistency in boundary layer values due to slowly changing anthropogenic sources over the broad North American region. Above this range, and starting at ~500 hPa pressure, the 2013 profile sharply transitions toward background free tropospheric loadings (1–2 ng/kg), while the 2012 profile undergoes a much weaker transition to ~13 ng/kg. The 2013 profile is the more typical even though it was mostly obtained in the traditional NA biomass burning season. The difference is not simply due to seasonal changes between the May/June 2012 and August/September 2013 measurements based on examination of past near-season NA data including those in *Murphy et al*. [2014] and *Spackman et al*. [2008]. Examination of the Global Fire Emissions Database database [*Giglio et al*., 2013] did not reveal any dramatic differences in larger-scale northern hemisphere biomass burning specific to those years that was likely to explain the factor of 10 change in loadings. Hence, this observation suggests additional very significant differences in transport/removal to the UT between the two measurement periods in addition to seasonal changes. Within each measurement set, which lasted ~1.5 months, there is no clear temporal trend in UT rBC loads in this region (Figure S3); the two measurements represent two different "states" of the atmosphere and do not provide information about the transition between these states. These results in particular point to the value in establishing the time scales for changes in UT rBC loads. To be correctly modeled, they will require improving treatment of injection and removal processes in this region. Modelers can therefore focus on this to test mechanisms, which may be highly relevant to improving model performance in the upper troposphere-lower stratosphere, where many presently tend to overestimate rBC by an order of magnitude.

The European vertical profile contains data from 2011 to 2013. Hence, the MMRs at altitudes above the ~500 hPa level reflect an average of those different years and our best estimate of typical free tropospheric loads from the additional data sets introduced here. These values (5 ng/kg) are highly consistent with the remote HIPPO values for the northern midlatitudes, indicating the likely value of both sets of data as (1) climatologically relevant and (2) indicating substantial mixing at time scales shorter than injection/removal processes in each region. In the lowest altitudes, higher loadings in Europe (~250 ng/kg) than over NA (~100 ng/kg) likely reflect local source strength differences, and in particular the influence of local emissions from Munich—a major industrial center—which was the departure and/or arrival city for many of the European flights. The clear enhancement at the highest levels in the European profile (up to ~40 ng/kg) were caused by biomass burning emissions from North America that had been transported across the arctic with little removal. When profiles with those influences are removed, the resulting vertical profile is consistent with rBC-conserved mixing above the 500 hPa level (at 5 ng/kg).

The profiles over the subtropical Atlantic were obtained in 2013 from the SALTRACE mission focused on Saharan dust transport and aging. Both the profiles, from the "near-field" measurements (Senegal) and the "aged" measurements after about 5 days of transport (Barbados), reveal a common air mass constrained below the ~500 hPa level that originated over the Sahara and contained significant loadings of rBC (~55 ng/kg in the near field and ~25 ng/kg after transport). This air mass extended to higher altitudes in the fresher plume. Above this air mass, more typical UT loads (~1 ng/kg) were seen. Although the low-altitude

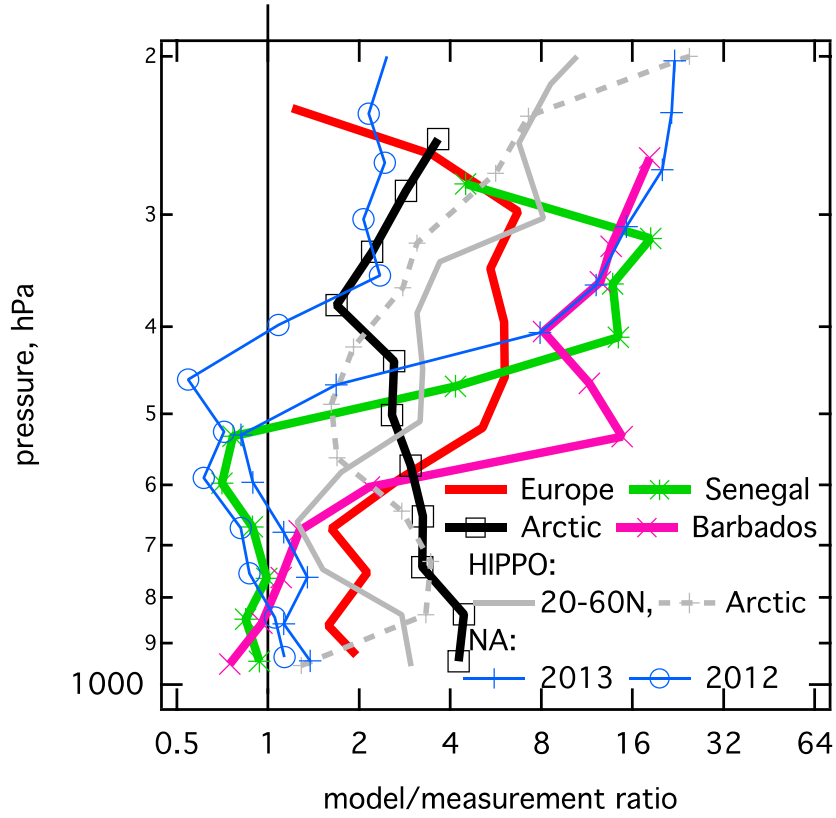

Figure 3. Model/measurement ratios from the AeroCom/SP2 comparison. At low altitudes, the model ensemble matches measurements in NA and on both sides of the Atlantic near 20S latitudes, with some overestimation in Europe and larger high bias in the Arctic. At high altitudes, the models overpredict rBC loads in all the analysis regions, with the best performance in the Arctic and poorer performance closer to the tropics.

data reflect dilution and removal with transport that strongly influence rBC loads, this dependence disappears above the plume, reflecting differing air parcel history and the longer time scales of removal and injection in the UT even in the subtropics. The lower concentrations in the UT are consistent with HIPPO results from the Pacific basin that unequivocally showed the impacts of convective removal of BC with large-scale mixing. At the highest altitude measured (~280 hPa), the Senegalese profile statistics were poor and strongly influenced by a single measurement of rBC MMR higher than any other profile measurement in the region at altitudes over the 500 hPa level; hence, we consider the enhancement (2.5 ng/kg compared to a background of ~1 ng/kg) at that altitude less representative of the broadly distributed rBC burden in the region and more a reflection of limited statistics and the presence of high-altitude plumes in this region.

Figure 3 shows the vertical profiles of the ratios of the AeroCom model ensemble results to the observations for the profiles shown in Figure 2. Hence, values below 1 reflect model ensemble underestimations and values above 1 show a high model bias compared to the measurements. Figure S2 shows the (often large) diversity of the individual models contributing to the AeroCom average, as well as the AeroCom median value, and the standard deviation of rBC MMR in the observed average profile (a measure of actual diversity in the air masses detected). Consistent with previous AeroCom evaluations mentioned in the introduction, and increased data availably at low altitudes, the ensemble shows relatively small biases over Europe and North America as expected since constraints on models are generally strong in or near source regions. Over the subtropical Atlantic, where measurements are sparse, the AeroCom suite performs well on the African outflow during the summer dust season; this result is not relevant to the winter biomass-burning season at this latitude range, when higher rBC emissions are expected. At high altitudes, again, the model bias is very similar to that already observed in the remote regions. However, based on these SP2 measurements in conjunction with those of HIPPO we note that the ensemble does significantly overestimate rBC in the Arctic over much of the column on average. In particular, the combined measurements represent different

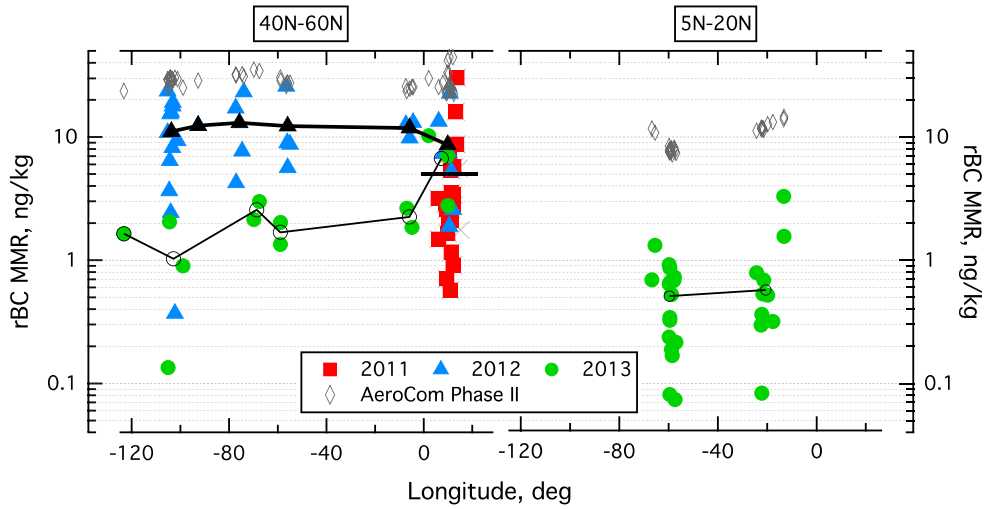

Figure 4. rBC MMR averaged between 8 and 10 km altitude (~375–250 hPa) and plotted against longitude for each vertical profile measured (markers). All results from 3 years are shown (by marker shape in grey: squares for 2011, triangles for 2012, and circles for 2013.), as well as AeroCom ensemble averages corresponding to each profile position. (left) Northern mid-latitude results; (right) the northern tropics.

locations and seasons in the Arctic, yet regularly show substantially lower loads than the model suite. The two seasonal measurements from HIPPO that show the highest loadings (March/April and November measurements, shown by thicker traces in Figure 2) are approximately on par with AeroCom. *Eckhardt et al*. [2015] identify the same seasonality from surface measurements but identify approximately a factor of 2 springtime low bias for a non-AeroCom model ensemble based largely on equivalent BC measurements (i.e., BC concentrations deduced from aerosol light absorption). There is no evidence from the new data and HIPPO that AeroCom substantially underestimates rBC in the Arctic in any season.

The relatively sparse availability of profiles associated with the U.S./Europe transits provides a novel opportunity to explore UT rBC variability over wide range of longitudes. Previous work quantifying BC over large length scales with the SP2 tended to explore latitudinal trends, yet produced results that were speculated to be relevant on global scales of longitude (at high altitude) [*Schwarz et al*., 2013]. To explicitly explore longitudinal trends, Figure 4 presents UT rBC MMR from individual profiles averaged over 8–10 km altitude from the data, with the AeroCom Phase II value shown for comparison. Observational results are segregated by year and highlight two different latitude bands. Although the statistics of the observations are limited, several features of the data set appear reasonably robust. In the 40–60°N band, a shift of an order of magnitude in loadings was observed between the measurements of 2012 (in May–July) and in 2013 (June–September). This difference, which was discussed in the context of the NA measurements, clearly extended over a wide longitudinal range. In the tropical data on either side of the Atlantic, only a smaller latitude range is covered, but again, the background UT rBC loads are very similar. As shown in the supporting information, clear trends at approximately monthlong time scales were not captured in the data. This set of features provides some direct experimental evidence that zonal mixing of the middle/upper troposphere occurs on time scales much shorter than rBC lifetime in that altitude range. It follows that UT background rBC loads are indeed representative of global loads and not just local conditions. These are features that are already clearly represented in the AeroCom model ensemble, which, in climatological predictions, shows very little longitudinal dependence at these altitudes. Note that we cannot estimate the rBC lifetime in the UT from these data.

4. Conclusions

The new BC MMR data presented here represent several years of measurements extending on both sides of the Atlantic, covering latitudes from 5 to 80°N, and extending from near the surface to above 11 km (~200 hPa), and provide extensive vertically resolved SP2 measurements of BC in areas where they were previously not available. They provide measurement constraints on BC's abundance and distribution which we have used to strengthen and refine understanding of AeroCom Phase II model suite performance. Consistent

with expectations, the model predictions were reasonable accurate at lower altitudes over North America and the tropical Atlantic, and less so over Europe. At higher altitudes in all the analysis areas the ensemble overpredicts rBC loads. However, the new data taken in conjunction with 2009–2011 HIPPO data help crystallize the lack of evidence for any significant AeroCom underestimations of BC concentration throughout the sampled altitude range in the Arctic.

The results also reveal opportunities to identify some of the sources of the ensemble high bias at altitude and broaden the community's awareness of BC as a uniquely valuable aerosol tracer without secondary production or loss processes other than wet and dry removal (for example, as in *Schwarz et al*. [2013]). Over North America, in two different years (and seasons), the upper tropospheric rBC loads differed by an order of magnitude, yet over ~1 month time scales did not change. This indicates the variability of loadings and, perhaps more importantly, suggests that aerosol sources and sinks affecting this region act quite slowly relative to mixing. This conclusion is supported by the consistency of the newly observed UT BC loadings over wide extents of longitude. This supports the conjecture that background rBC concentrations measured in the UT are zonally relevant. This conclusion can be tested via longer-term measurements in this part of the atmosphere (for example, from the In-service Aircraft for a Global Observing System (IAGOS) project [*Bundke et al*., 2015]) and from global-scale measurements including better statistics over wide ranges of longitude (for example, the presently occurring NASA Atmospheric Tomography project).

Acknowledgments

The NOAA SP2 data were obtained and analyzed with the support of the NOAA Atmospheric Composition and Climate Program, the NASA Radiation Sciences Program, and the NASA Upper Atmosphere Research Program. The DLR SP2 data were obtained and analyzed with the support of the Helmholtz Association under grant VH-NG-606 (Helmholtz-Hochschul-Nachwuchsforschergruppe AerCARE), the DLR projects CATS and VolcATS, and the European Union project ACCESS under grant agreement 265863. Additional analysis was supported in part by the Center for Advanced Studies at LMU, LMU Munich's Institutional Strategy LMUexcellent within the framework of the German Excellence Initiative, and by the European Research Council under the European Community's Horizon 2020 research and innovation framework program/ERC grant agreement 640458-A-LIFE. Data are available at http://www-air.larc.nasa.gov/missions/seac4rs/index.html for the SEAC4RS mission, http://www-air.larc.nasa.gov/cgi-bin/ArcView/dc3 for the DC3 mission (both DLR Falcon and NASA DC8 data), and by request to the DLR for the CONCERT, ACCESS, and SALTRACE missions; the PI for the Falcon BC data can be reached at Bernadett.Weinzierl@univie.ac.at. AeroCom model data are freely available upon application, see aerocom.met.no for details. We thank the modeling groups for use of their results. B.H.S. acknowledges funding by the Research Council of Norway through the grants AEROCOM-P3, AC/BC (240372), and NetBC (244141).

References

Barth, M. C., et al. (2015), The Deep Convective Clouds and Chemistry (DC3) field campaign, *Bull. Am. Meteorol. Soc.*, doi:10.1175/BAMS-D-13-00290.1.

Baumgardner, D., et al. (2012), Soot reference materials for instrument calibration and intercomparisons: A workshop summary with recommendations, *Atmos. Meas. Tech.*, *5*, 1869–1887, doi:10.5194/amt-5-1869-2012.

Bond, T. C., et al. (2013), Bounding the role of black carbon in the climate system: A scientific assessment, *J. Geophys. Res. Atmos.*, *118*, 5380–5552, doi:10.1002/jgrd.50171.

Bundke, U., M. Berg, N. Houben, A. Ibrahim, M. Fiebig, F. Tettich, C. Klaus, A. Franke, and A. Petzold (2015), The IAGOS-CORE aerosol package: Instrument design, operation and performance for continuous measurement aboard in-service aircraft, *Tellus, Ser. B*, *67*, doi:10.3402/tellusb.v67.28339.

Chin, M., T. Diehl, O. Dubovik, T. F. Eck, B. N. Holben, A. Sinyuk, and D. G. Streets (2009), Light absorption by pollution, dust, and biomass burning aerosols: A global model study and evaluation with AERONET measurements, *Ann. Geophys.*, *27*, 3439–3464, doi:10.5194/angeo-27-3439-2009.

Dahlkötter, F., M. Gysel, D. Sauer, A. Minikin, R. Baumann, P. Seifert, A. Ansmann, M. Fromm, C. Voigt, and B. Weinzierl (2014), The Pagami Creek smoke plume after long-range transport to the upper troposphere over Europe—Aerosol properties and black carbon mixing state, *Atmos. Chem. Phys.*, *14*, 6111–6137, doi:10.5194/acp-14-6111-2014.

Eckhardt, S., et al. (2015), Current model capabilities for simulating black carbon and sulfate concentrations in the Arctic atmosphere: A multi-model evaluation using a comprehensive measurement data set, *Atmos. Chem. Phys.*, *15*, 9413–9433, doi:10.5194/acp-15-9413-2015.

Giglio, L., J. T. Randerson, and G. R. van der Werf (2013), Analysis of daily, monthly, and annual burned area using the fourth-generation Global Fire Emissions Database (GFED4), *J. Geophys. Res. Biogeosci.*, *118*, 317–328, doi:10.1002/jgrg.20042.

Klimont, Z., K. Kupiainen, C. Heyes, P. Purohit, J. Cofala, P. Rafaj, J. Borken-Kleefeld, and W. Schöpp (2016), Global anthropogenic emissions of particulate matter including black carbon, *Atmos. Chem. Phys. Disc.*, doi:10.5194/acp-2016-880.

Koch, D., et al. (2009), Evaluation of black carbon estimations in global aerosol models, *Atmos. Chem. Phys.*, *9*, 9001–9026, doi:10.5194/acp-9-9001-2009.

Kondo, Y., L. Sahu, N. Moteki, F. Khan, N. Takegawa, X. Liu, M. Koike, and T. Miyakawa (2011), Consistence and traceability of black carbon measurements made by laser-induced incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques, *Aerosol Sci. Technol.*, *45*, 295–312.

Laborde, M., et al. (2012), Single particle soot photometer intercomparision at the AIDA chamber, *Atmos. Meas. Tech.*, *5*, 3077–3097, doi:10.5194/amt-5-3077-2012.

Murphy, D. M, K. D. Froyd, J. P. Schwarz, and J. C. Wilson (2014), Observations of the cheical composition of stratospheric aerosol particles, *Q. J. R. Meteorol. Soc.*, *140*, 1269–1278, doi:10.1002/qj.2213.

Myhre, G., et al. (2013), Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, *Atmos. Chem. Phys.*, *13*, 1853–1877, doi:10.5194/acp-13-1853-2013.

Petzold, A., et al. (2013), Recommendations for reporting "black carbon" measurements, *Atmos. Chem. Phys.*, *13*, 8365–8379, doi:10.5194/acp-13-8365-2013.

Reddington, C. L., G. McMeeking, G. W. Mann, H. Coe, M. G. Frontoso, D. Liu, M. Flynn, D. V. Spracklen, and K. S. Carslaw (2013), The mass and number size distributions of black carbon aerosol over Europe, *Atmos. Chem. Phys.*, *13*, 4917–4939.

Roiger, A., et al. (2015), Quantifying emerging local anthropogenic emissions in the Arctic region: The ACCESS aircraft campaign experiment, *Bull. Am. Meteorol. Soc.*, *13*, 441–460, doi:10.1175/BAMS-D-13-00169.1.

Samset, B., and G. Mhyre (2011), Vertical dependence of black carbon, sulphate and biomass burning aerosol radiative forcing, *Geophys. Res. Lett.*, *38*, L24802, doi:10.1029/2011GL049697.

Samset, B. H., et al. (2014), Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, *Atmos. Chem. Phys.*, *14*, 12,465–12,477, doi:10.5194/acp-14-12465-2014.

Schutgens, N. A. J., E. Gryspeerdt, N. Weigum, S. Tsyro, D. Goto, M. Schulz, and P. Stier (2016), Will a perfect model agree with perfect observations? The impact of spatial sampling, *Atmos. Chem. Phys.*, *16*(10), 6335–6353, doi:10.5194/acp-16-6335-2016.

Schwarz, J. P., et al. (2006), Single-particle measurements of midlatitude black carbon and light-scattering aerosols from the boundary layer to the lower stratosphere, *J. Geophys. Res.*, *110*, D16207, doi:10.1029/2006JD007076.

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.

Schwarz, J. P., B. H. Samset, A. E. Perring, J. R. Spackan, R. S. Gao, P. Stier, M. Schulz, F. L. Moore, E. A. Ray, and D. W. Fahey (2013), Global-scale seasonally resolved black carbon vertical profiles over the Pacific, *Geophys. Res. Lett*, *40*, 5542–5547, doi:10.1002/2013GL057775.

Spackman, J. R., J. P. Schwarz, R. S. Gao, L. A. Watts, D. S. Thomson, D. W. Fahey, J. S. Holloway, J. A. de Gouw, M. Trainer, and T. B. Ryerson (2008), Empirical correlations between black carbon aerosol and carbon monoxide in the lower and middle troposphere, *Geophys. Res. Lett.*, *35*, L19816, doi:10.1029/2008GL035237.

Toon, O. B., et al. (2016), Planning, implementation, and scientific goals of the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC^{4}RS) field mission, *J. Geophys. Res. Atmos.*, *121*, 4967–5009, doi:10.1002/2015JD024297.

Wang, Q., R.-J. Huang, J. Cao, Y. Han, G. Wang, G. Li, Y. Wang, W. Dai, R. Zhang, and Y. Zhou (2014), Mixing state of black carbon aerosol in a heavily polluted urban area of China: Implications for light absorption enhancement, *Aerosol Sci. Technol.*, *48*(7), 689–697, doi:10.1080/02786826.2014.917758.

Weinzierl, B., et al. (2016), The Saharan Aerosol Long-range Transport and Aerosol-Cloud- Interaction Experiment (SALTRACE): Overview and selected highlights, *Bull. Am. Meteorol. Soc.*, doi:10.1175/BAMS-D-15-00142.1, in press.