



## Abstract

Atmospheric black carbon (BC) absorbs solar radiation, and exacerbates global warming through exerting positive radiative forcing (RF). However, the contribution of BC to ongoing changes in global climate is under debate. Anthropogenic BC emissions, and the resulting distribution of BC concentration, are highly uncertain. In particular, long range transport and processes affecting BC atmospheric lifetime are poorly understood. Here we discuss whether recent assessments may have overestimated present day BC radiative forcing in remote regions. We compare vertical profiles of BC concentration from four recent aircraft measurement campaigns to simulations by 13 aerosol models participating in the AeroCom Phase II intercomparison. An atmospheric lifetime of BC of less than 5 days is shown to be essential for reproducing observations in remote ocean regions, in line with other recent studies. Adjusting model results to measurements in remote regions, and at high altitudes, leads to a 25 % reduction in AeroCom Phase II median direct BC forcing, from fossil fuel and biofuel burning, over the industrial era. The sensitivity of modeled forcing to BC vertical profile and lifetime highlights an urgent need for further flight campaigns, close to sources and in remote regions, to provide improved quantification of BC effects for use in climate policy.

## 1 Introduction

As an absorber of solar radiation, anthropogenic BC emissions can contribute positively to global radiative forcing through the aerosol direct effect, they can affect clouds through the aerosol indirect and semidirect effects, change albedo of snow and ice, and influence precipitation by changing atmospheric stability and the surface energy balance (Myhre et al., 2013a; Ramanathan and Carmichael, 2008; Haywood and Shine, 1995). Presently both the magnitude of anthropogenic BC emissions and the resulting global distribution of BC concentrations are highly uncertain. In particular the vertical profile of BC concentration, which strongly affects its total impact on the energy balance

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of the atmosphere, is poorly constrained (Koffi et al., 2012; Textor et al., 2007; Samset et al., 2013). Comparisons of measurements with model results, with emphasis both on total BC mass, spatio-temporal distribution and vertical structure are therefore essential for constraining estimates of BC effects on climate.

The IPCC AR5 (Boucher et al., 2013) assessed the direct aerosol effect radiative forcing due to anthropogenic BC (defined here as BC from anthropogenic fossil fuel and biofuel sources, BC FF + BF) to be  $+0.40$  [range:  $+0.05$  to  $+0.80$ ]  $\text{W m}^{-2}$  over the period 1750–2010. That assessment took into account both model based and observational studies. Recently, Phase II of the AeroCom model intercomparison project (Myhre et al., 2013b) also evaluated BC FF + BF radiative forcing (RF), based purely on 15 global aerosol models (Myhre et al., 2013b), and found it to be  $+0.23$  [ $+0.06$  to  $+0.48$ ]  $\text{W m}^{-2}$ . The uncertainty ranges (5–95 %) show that BC is still a major contributor to the total uncertainty on anthropogenic radiative forcing. Further, it has recently been shown that, because the direct radiative forcing per unit mass BC increases strongly with altitude (Ban-Weiss et al., 2011; Zarzycki and Bond, 2010; Samset and Myhre, 2011), the diversity in modeled vertical profiles of BC concentration in the AeroCom Phase II models may account for up to 50 % of the model diversity in anthropogenic BC RF (Samset et al., 2013). Climate model simulations, however, indicate that, while direct BC forcing strengthens with altitude, its climate efficacy may decrease, i.e. the surface temperature response to BC in the middle and upper troposphere may be small or even negative (Ban-Weiss et al., 2011; Flanner, 2013). Further, Samset et al. (2014) concluded that an upward adjustment on the model based uncertainty on total aerosol forcing may be necessary, in an analysis that looked at multi-model variability in per-species aerosol burdens and optical parameters. BC forcing diversity was found to be a significant component in this analysis.

Recently, several single model studies have investigated other factors that may underlie the intermodel variability in AeroCom Phase II, or assessed its multi-model mean results in light of observations. E.g. Wang et al. (2014) compared results from the GEOS-Chem model (<http://geos-chem.org>) with results from the HIPPO flight

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campaign, and concluded that to reproduce HIPPO, more wet removal was required than is represented in most present models. Based on this, and on revised estimates on direct radiative forcing of BC when taking HIPPO constraints (Schwarz et al., 2013) into account, they argue that previous model estimates may be biased high due to elevated BC concentrations in the free troposphere. Bauer et al. (2013) studied the atmospheric lifetime of BC, which is a combined measure of transport and removal processes, by comparing simulations with the GISS-MATRIX model to HIPPO (Bauer et al., 2010), using CMIP5 emissions. They found that under present day conditions, BC lifetime should be no more than 4 days, which is significantly shorter than what is used in some present models. Based on the above, it is natural to investigate both whether the full AeroCom Phase II RF estimate is biased high, as was found for GEOS-Chem, and what contribution to multimodel variability may be due to differences in modeled BC lifetime.

In the following, we compare vertical concentration profiles from the AeroCom Phase II models to recent aircraft campaigns. The results are used to set multi-model constraints on BC lifetime, and to find limits on the possible high bias of AeroCom Phase II BC RF if constraining to HIPPO results.

## 2 Methods

### 2.1 Flight data and BC definition

We have used data from flight campaigns HIAPER Pole-to-Pole Observations (HIPPO) 1–5 (Schwarz et al., 2013), Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) SP2 (Jacob et al., 2010), Polar Airborne Measurements and Arctic Regional Climate Model Simulation Project (PAMARCMiP) (Herber et al., 2012; Stone et al., 2010) and Aerosol Radiative Forcing in East Asia (A-FORCE) (Oshima et al., 2012). (See Table 1.)

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All flights measured BC concentrations using the single particle soot photometer (SP2) instrument (Schwarz et al., 2010). Hence, in the present work, “BC” in relation to measured data stands for “refractive BC (rBC)” as quantified by SP2, equivalent to properly measured elemental carbon (Kondo et al., 2011).

HIPPO 1–5 (Schwarz et al., 2013) flew mainly pole-to-pole over the Pacific Ocean at various times during 2009–2011. Combining data from all five campaigns yields an approximate annual average. The HIPPO data have been screened against contributions from fires, so as to be representative of the background concentration of BC over the Pacific. Two of the HIPPO campaigns also flew over the North American mainland, allowing also for comparisons closer to anthropogenic BC source regions. HIPPO covers atmospheric pressures from surface values up to 100 hPa, meaning that its upper range reaches into the lower stratosphere.

The ARCTAS SP2 campaign (Jacob et al., 2010) was flown in two separate time periods in 2008. During spring, flights were conducted over the northern Pacific Ocean, comparable to parts of the HIPPO region, and also over the North Polar regions. During summer, flights were conducted over the North American continent, again comparable to HIPPO. Part of the ARCTAS motivation was to study fires, so the measured concentrations can be expected to have a larger contribution from open biomass burning than the HIPPO dataset. ARCTAS data cover the atmospheric segment from the surface and up to 250 hPa.

The Polar Airborne Measurements and Arctic Regional Climate Model Simulation Project (PAMARCMiP) campaign (Herber et al., 2012; Stone et al., 2010) consists of a series of flights conducted over the North Polar and Northern Pacific regions, from 2009 through 2012. It covers a vertical range up to 500 hPa, and is partially comparable to both ARCTAS and HIPPO. As ARCTAS, PAMARCMiP is partially affected by biomass burning emissions.

Finally, the Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign (Oshima et al., 2012) flew over East Asia (southwestern Japan) in spring 2009, covering a vertical range up to 300 hPa. It is not regionally comparable to the other campaigns,

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but is highly relevant because it represents a region dominated by outflow from mainland China, one of the main sources of anthropogenic BC emissions.

## 2.2 AeroCom Phase II models

The models results in the present study come from 13 state of the art aerosol models, submitted as part of Phase II of the AeroCom model intercomparison project (Myhre et al., 2013b; Samset et al., 2013; Schwarz et al., 2013). Participating models are NCAR-CAM3.5 (Lamarque et al., 2012), CAM4-Oslo (Silver et al., 2013), CAM5.1 (Liu et al., 2012), GISS-MATRIX (Bauer et al., 2010), GISS modelE (Koch et al., 2011), GMI (Bian et al., 2009), GOCART-v4 (Chin et al., 2009), HadGEM2 (Bellouin et al., 2011), IMPACT (Lin et al., 2012), INCA (Szopa et al., 2012), ECHAM5-HAM (Zhang et al., 2012), OsloCTM2 (Skeie et al., 2011) and SPRINTARS (Takemura et al., 2005). See Myhre et al. (2013b) and individual model references for further descriptions.

All models submitted monthly mean 3-D fields of total BC mass mixing ratio, year 2000 emissions (Lamarque et al., 2010), year 2006 meteorology, or model present day (PD) climatology. To calculate BC concentrations from mixing ratios, the models' own monthly mean temperature and pressure fields were also used. When discussing modifications to the BC radiative forcing from the direct effect from anthropogenic fossil fuels and biofuels, the models' own monthly mean 2-D forcing fields were used, in combination with a preindustrial simulation using year 1850 emissions but still year 2006 or PD meteorology (Myhre et al., 2013b). For consistency with recent literature, forcing results are given for 1750–2010, using scaling factors presented in Myhre et al. (2013b).

## 2.3 Analysis

To compare flight campaigns and model output, a series of geographical regions were first selected. See Fig. 1. For the flight data, only measurements that fell within the regions were kept, and for each region an average profile was constructed. For the

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models, all output within the selected region was averaged into a single profile for each model. However, to take the seasonality into account, we produced a model profile for each measurement point or profile from the flights. These were then averaged. The result is a set of model profiles that correspond to the flight profiles both geographically and temporally.

From the concentration profiles, we calculated aerosol burdens for both models and flight data. To ensure comparability, model burdens were calculated only in the same vertical range covered by the flight campaign.

Further, we used the methodology presented in Samset et al. (2013) to calculate BC RF from the concentration profiles. Briefly, we use spatially and temporally resolved normalized forcing efficiency profiles (RF exerted per gram of aerosol at a given altitude) calculated from a single model (Samset and Myhre, 2011), and multiplied with profiles of BC burden per model layer. This yields comparable estimates for total BC forcing within the selected regions, for the seasons covered by the respective flight campaigns; thus all RF calculations are performed with a consistent method. To distinguish the models' own estimates of RF from the RF calculated by this method, we refer to the two as "native RF" and "recalculated RF" respectively. While using the forcing efficiency from a single model (OsloCTM2) will naturally bias the calculated RF towards the forcing strength predicted by that model, it also allows for an estimate of differences in vertical profile shape. Since the forcing efficiency for BC is strongly and monotonically rising with altitude, differences between the overall shape of measured and modeled profiles will cause the ratio of recalculated forcing per burden to differ from unity.

## 2.4 Calculation of RMS values and correlations

To estimate how well a given model reproduces the Pacific HIPPO flight data, as primarily used below, we calculated model bias, root-mean-square (RMS) error values and correlation coefficients. The HIPPO dataset was subdivided into five regions (P1–P5 in Fig. 1), and an annual mean profile was constructed for each region as shown

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Pacific Ocean and for western North America, overlain with AeroCom Phase II results. A common pattern is that the models strongly overpredict the HIPPO measurements. Further, the overprediction is more pronounced at high altitudes. Comprised of five campaigns distributed throughout the year, HIPPO represents an approximate annual average. As recently noted (Schwarz et al., 2013), its Pacific measurements indicate that at the highest altitudes studied, BC concentrations converge towards a common background value, here found to be approximately  $0.1 \text{ ng m}^{-3}$ , with very low seasonality. Here we also find (Fig. 2f) the same background value above western North America.

10 Panels g–i of Fig. 2 show the ARCTAS (Jacob et al., 2010) campaign, which reports significantly higher concentrations than HIPPO. The models mainly underpredict these observations, linked to the fact that ARCTAS encountered biomass burning BC from episodic forest fires (Wang et al., 2011), which HIPPO did not encounter in this region. A notable feature is that above the fire dominated segments, the ARCTAS profiles show a strong decline with altitude. In the P1 (Northern Pacific) region upper tropospheric ARCTAS concentrations are similar to those measured in HIPPO.

15 Panels j–l show PAMARCMiP (Herber et al., 2012; Stone et al., 2010) data, first over the North Pacific region, and then over two North Polar regions. While the altitude range covered by PAMARCMiP is limited compared to ARCTAS and HIPPO, the concentrations found in the lowest few kilometers of the troposphere are consistent with ARCTAS. Over the NP1 and NP2 region, north of America and Greenland, models underpredict the measurements. As for ARCTAS, this is at least partly due to episodic fires. The Arctic region may however have further sources of BC not adequately represented in the emission inventories used by the models (Stohl et al., 2013).

20 Panel m shows A-FORCE (Oshima et al., 2012) data in the sea areas around Japan. Here we find good agreement between models and measured concentrations, both in absolute values and in the shape of the vertical profile. The variability between models is also much lower in this region than for the others. The aerosol in the A-FORCE region is mainly sensitive to outflow from mainland China, Korea and Japan. This indicates that

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in AeroCom Phase II, East Asian BC emissions and outflow are either well represented or, if emissions are still underestimated as discussed for AeroCom Phase I in recent literature (Bond et al., 2013; Chung et al., 2012), the atmospheric lifetime of BC must be compensatingly long in the models to allow enough BC to be transported into the region sampled by A-FORCE.

### 3.2 Consequences for BC atmospheric lifetime

In the following, we assess the implications of the flight observations on modeled BC lifetime and RF. Episodic biomass burning emissions from fires, even though represented in the model emissions, pose challenges when comparing flight campaigns to monthly mean model data. Arguably fires are also difficult to characterize as anthropogenic. Below, we therefore constrain our discussion to the HIPPO dataset, which was less influenced by episodic fires, reached the highest altitudes, covers the largest geographical area, and represents an approximate annual mean.

15 Figure 2 shows that some models more closely reproduce the measurements than others, both in magnitude and shape. Several studies have suggested that to reproduce HIPPO data, a low modeled atmospheric lifetime, or a short ageing timescale, of BC is required (Bauer et al., 2013; Wang et al., 2014). Here we can test this supposition for a larger set of models. Quantifying the difference between models and data requires care, as absolute concentrations range over several orders of magnitude. Common diagnostic variables include model bias and RMS error. Of these, RMS error and model mean bias (see Sect. 2) will be dominated by high absolute concentrations, i.e. low altitudes in the present case. Model mean normalized bias avoids this, but will be more sensitive to model and measurement uncertainties in high altitude, low concentration ranges. In Fig. 3, RMS error and biases are plotted as function of the modeled BC lifetime. Lifetime, also referred to as atmospheric residence time, is here defined as modeled global, annual mean emissions divided by burden (Table 3).

25 Figure 3 shows that, independent of diagnostic variable, a low BC lifetime is a requirement for good reproduction of absolute modeled concentrations. Regressing bias

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or RMS error vs. lifetime (black, dashed line in Fig. 3) gives an intercept at 3 days for RMS error and model mean bias. This value is in line with indications from other recent studies (e.g. Bauer et al., 2013). In the present dataset, a single model with high lifetime (HadGEM2) represents a significant outlier. That model did not include BC ageing and transition to a hydrophilic state, with the consequence that both BC lifetime and burdens over remote areas become high (Bellouin et al., 2011). To test the impacts of single models on the result, Fig. 3 also shows regressions with one model removed (grey lines). For the normalized mean bias, which is less sensitive to high concentrations, the regression with this particular model removed is consistent with the results from RMS error and mean bias.

Bias and RMS error give information on absolute deviations, but less on any covariance in shape. The Pearson correlation coefficient, however, is sensitive to the shape of the BC profiles. In Fig. 3, correlation is indicated by symbol size. (See also values in Table 3.) Several models with low lifetimes also yield low correlations. Regressing only the models with correlation coefficients  $\rho > 0.8$  gives similar slopes and intercepts to what we find using all models (red, dashed line). We note that 12 out of 13 models show correlation with the Pacific HIPPO data at significance  $p > 0.05$ .

Low BC lifetime appears necessary, but not sufficient, to describe the data. Only three models (IMPACT, GMI, GISS-MATRIX) exhibit both a low bias or RMS error and a high correlation, with no single obvious factor linking their aerosol treatments. AeroCom Phase II models use a wide variety of microphysics schemes (Mann et al., 2014). Meteorology and treatment of BC aging and wet scavenging also vary, and will impact the vertical profiles (Kipling et al., 2013; Bauer et al., 2013). Further model experiments, in line with the single model study in Wang et al. (2014), are required to address the reasons behind the relationship found in Fig. 3.

### 3.3 Consequences for modeled BC FF + FB RF

However, the three models that best reproduce HIPPO in the Pacific all report consistent and relatively low BC FF + BF forcing, exerted close to emission sources as 20095

expected from the low lifetime. In these three models very little BC reaches remote ocean regions, or gets lifted above 500 hPa, relative to the other models in the ensemble. While the correspondence to HIPPO cannot be used to extract information close to emission sources, it does suggest that scaling down the average modeled forcing aloft and in remote ocean regions has merit.

Compared with HIPPO, the current model ensemble overestimates BC concentrations at all altitudes in remote regions. The overestimation increases with altitude, and is particularly significant at pressures below 200 hPa. Further, Schwarz et al. (2013) suggest that the minimum concentration consistently observed by HIPPO in the upper troposphere, lower stratosphere and tropical transition layer may be a global feature. Interestingly, the models shown here do, on average, reproduce the general feature of a common background level, but with a concentration that is approximately 20 times higher than indicated by HIPPO.

The HIPPO dataset allows us to test the possible implications of these observations on the multimodel BC RF from AeroCom Phase II, a key basis for BC forcing recently assessed in the IPCCs AR5. We attempt two scalings of the modeled BC concentration fields, to align them with the HIPPO observations. The first assumes that the vertically resolved ratio between models and observations in the Pacific holds for all remote ocean regions, shown as grey shaded areas in Fig. 1. The second assumes that the supposition of a globally uniform high altitude BC concentration from Schwarz et al. (2013) is true. While the present dataset is insufficient to determine if such a supposition is true, it is nevertheless interesting to assess its potential impact to see if efforts to measure high altitude BC concentrations should be prioritized.

Figure 4 shows the implications of these scalings on the multimodel direct RF due to BC from fossil fuel and biofuel burning (BC FF + BF). Unscaled, the multimodel median RF found here is  $+0.24$  [ $+0.17$  to  $+0.47$ ]  $\text{W m}^{-2}$ . (See Table 3.) Applying the remote region scaling reduces the global, annual median BC FF + BF RF to  $+0.22$  [ $+0.16$  to  $+0.39$ ]  $\text{W m}^{-2}$ . The high altitude scaling reduces it to  $+0.19$  [ $+0.15$  to  $+0.33$ ]  $\text{W m}^{-2}$ . Applying both simultaneously, while ensuring that we do not doubly scale in remote,





as outlined above. The obvious feature is that for these three models, forcing is exerted primarily closer to the sources, and at lower altitudes, than in the full ensemble. Very little is exerted in the southern ocean, or above 200 hPa.

Figure 6 shows the results from the scaling analysis above, compared with results for the three HIPPO-corresponding models only. From the outset they have low forcing, and the scaling exercise does not significantly affect them as there is already very little forcing in the scaled regions. The final model median, however, is consistent with that from the scaled full model ensemble. This gives a separate indication that a reduction of 25 % in anthropogenic BC RF relative to the AeroCom Phase II value is reasonable if we take the HIPPO Pacific measurements as guidance.

#### 4 Conclusions

In conclusion, when comparing recent aircraft based measurements of BC concentration with state of the art global aerosol-climate models, there is a tendency for the models to overestimate the aircraft measurements, where and when the effects of fires are small. This overestimation is most pronounced in remote regions, where BC concentrations are dominated by long range transport, as well as at high altitudes. For a region sensitive to Asian emission sources, models reproduce the aircraft measurements remarkably well, with no indication of an underestimation in BC emissions. In remote ocean regions, an atmospheric lifetime of anthropogenic BC of less than 5 days seems crucial, but not sufficient, to be able to reproduce measurement data. Scaling the multimodel results to HIPPO measurements, remotely and aloft, and assuming a globally uniform high altitude BC concentration, leads to a reduction of 25 % in anthropogenic BC direct RF, relative to the models native values. The revised median of  $0.17 \text{ W m}^{-2}$  stands in stark contrast to recent assessments, which report up to 2–3 times stronger present day BC forcing, but is in line with recent single-model studies (Wang et al., 2014; Bauer et al., 2013). This discrepancy underlines the impact of combining measured BC concentration data with model estimates. To resolve these differences, and

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better constrain the climate impact of BC, there is an urgent need for further flight campaigns to provide BC vertical concentration profiles over both source regions, and regions where anthropogenic BC concentrations are dominated by transport and wet scavenging.

*Acknowledgements.* B. H. Samset, G. Myhre and M. Schulz were supported by the Research Council of Norway, through the grants SLAC, AEROCOM-P3 and EarthClim, and the EC Seventh Framework Programme under grant agreement FP7-ENV-2011-282688 (ECLIPSE). R. C. Easter, S. J. Ghan and K. Zhang were supported by through Scientific Discovery through Advanced Computing (SciDAC) program funded by US Department of Energy Office of Advanced Scientific Computing Research and Office of Biological and Environmental Research. The Pacific Northwest National Laboratory (PNNL) is operated for the DOE by Battelle Memorial Institute under contract DE-AC06-76RLO 1830. Y. Kondo, N. Moteki, M. Koike, and N. Oshima were supported by the Global Environment Research Fund of the Ministry of the Environment, Japan (A-0803, A-1101, and 2-1403) and the Japan Society for the Promotion of Science (JSPS) KAKENHI grant numbers 23221001 and 26701004.

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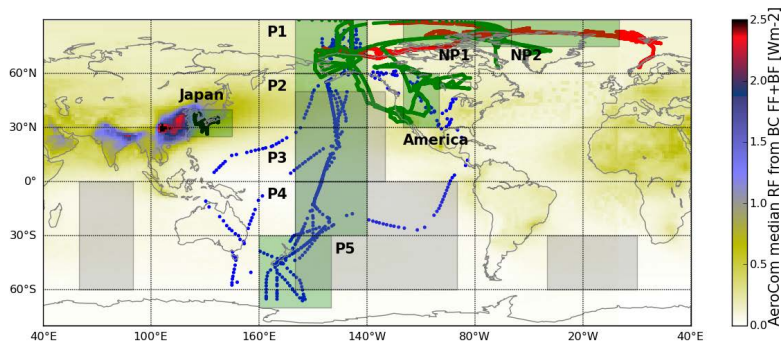
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**Table 1.** The flight campaigns included in the present work, and the times when they flew.

Campaign	Region	Time	Data/web site
HIPPO 1	Pacific	Jan 2009	<a href="http://hippo.ucar.edu/">http://hippo.ucar.edu/</a>
HIPPO 2	Pacific	Nov 2009	
HIPPO 3	Pacific	Mar/Apr 2010	
HIPPO 4	Pacific	Jun 2011	
HIPPO 5	Pacific	Aug 2011	
A-FORCE	Japan	Mar/Apr 2009	<a href="http://wwwair.larc.nasa.gov/missions/arctas/dataaccess.htm">http://wwwair.larc.nasa.gov/missions/arctas/dataaccess.htm</a>
ARCTAS Spring	Northern Pacific, North Polar	Apr 2008	
ARCTAS Summer	Continental North America	Jul 2008	
PAMARCMiP 2009	Northern Pacific, North Polar	Apr 2009	
PAMARCMiP 2011	Northern Pacific, North Polar	Apr 2011	
PAMARCMiP 2012	North Polar	Mar–Apr 2012	

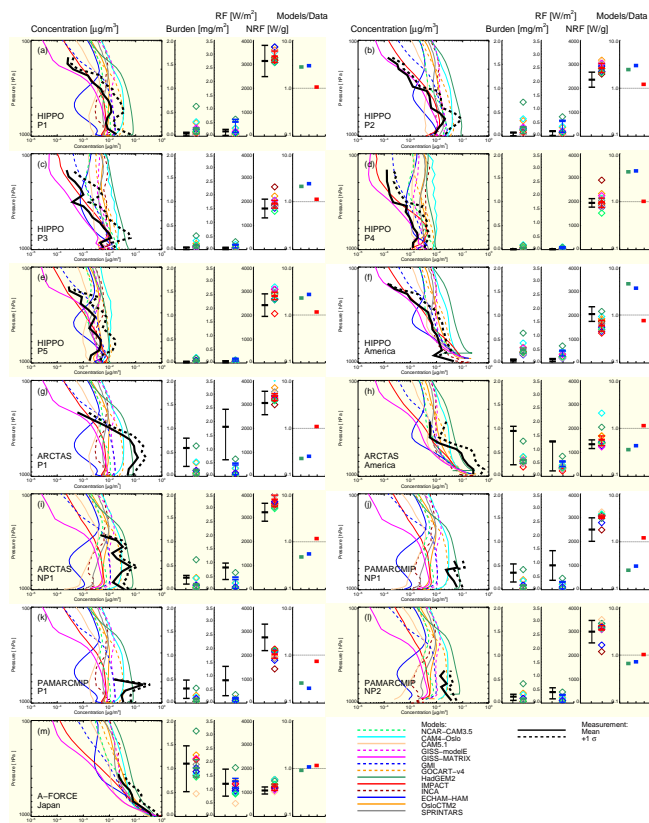
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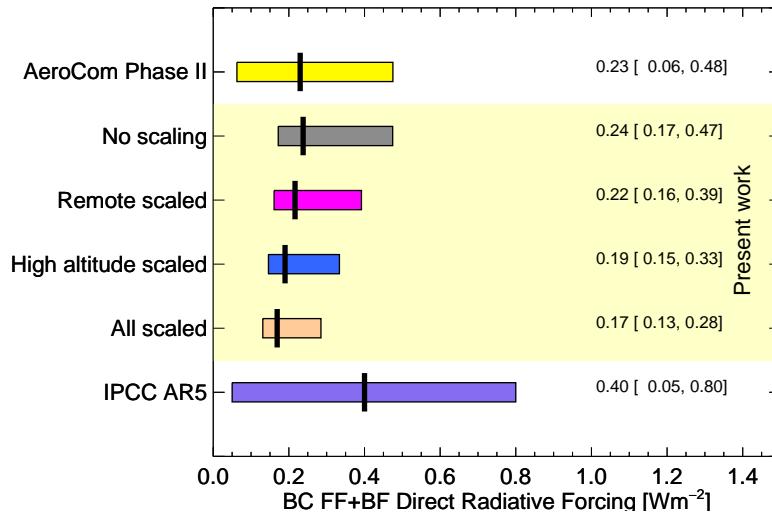
**Figure 1.** Flight tracks and regions selected for the analysis. Dots represent either single measurements (ARCTAS, A-FORCE, PAMARCMiP) or pre-averaged profiles (HIPPO) from the flight campaigns. Blue: HIPPO. Green: ARCTAS. Red: PAMARCMiP. Black: A-FORCE. Green boxes show the geographical regions where model data was averaged for comparison to the flight profiles. Grey boxes show the areas defined as “remote ocean”. The background map shows the median, annual mean RF due to fossil fuel and biofuel burning from the 13 models used in the present study.

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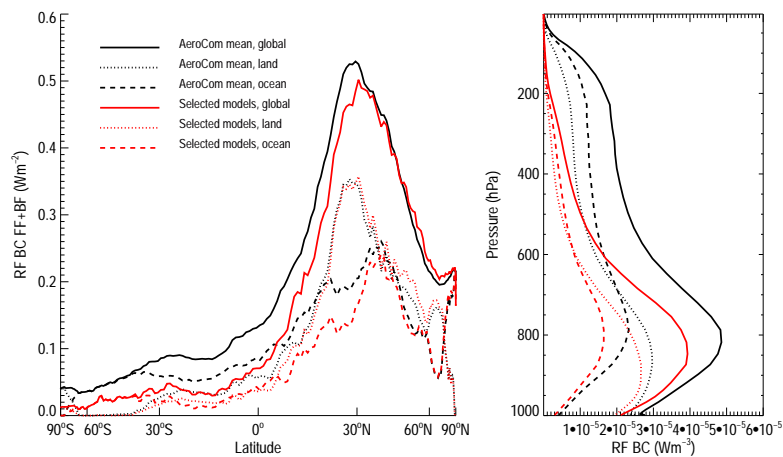
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**Figure 4.** Model median and 5–95% range for BC FF + BF forcing, for 1750–2010, with various scalings applied. The yellow bar shows the AeroCom Phase II result (Myhre et al., 2013b). The grey bar shows unscaled values from the present work, then with remote scaling (pink) and high altitude scaling (blue) applied. The khaki bar shows the lower limit on BC FF + BF forcing from the present work, with both scalings applied. Below we compare with the recent estimate in IPCC WG1’s AR5.

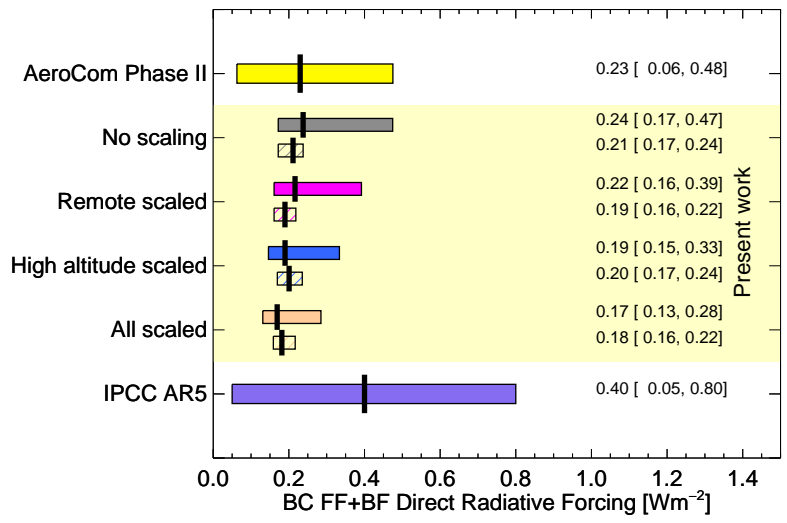
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**Figure 5.** Zonal mean and altitude forcing profiles for AeroCom Phase II models. **(a)** BC FF + BF forcing, zonal mean, for all 13 models in the present study (black, solid), and the three models selected based on RMS and correlation vs. HIPPO (red, solid). Dotted lines show only forcing over land, dashed show forcing over ocean. **(b)** As a, except total BC (FF + BF + BB) forcing, and global, annual mean vertical profile. Forcing is shown per altitude meter (unit  $\text{W m}^{-3}$ ), to avoid dependence on model vertical structure.

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**Figure 6.** As Fig. 4, except also showing the BC FF + BF forcing from the three models selected based on RMS and correlation vs. HIPPO (hatched boxes).