

## 1 Modeled black carbon radiative forcing and atmospheric lifetime in

## 2 AeroCom Phase II constrained by aircraft observations

### 3 Responses to reviewers

4

5 We thank both reviewers for their positive assessment of our paper, and for their helpful comments.  
6 Point-by-point responses and details of the changes made to the paper can be found below (in red).

#### 7 Reviewer #1:

8 The following few points may be helpful.

9 Page 20089 line 14: The models are comparing with data collected between 2008 and 2012, however  
10 the emissions used in the model are from a decade before (2000). Given that some regions used in  
11 the study are subject to rapidly changing emissions, what impact may this have on the results?

12 Page 20093 line 25 to page 20094 line 5: Whilst I agree with the points made, it is also worth pointing  
13 out that the A-FORCE measurements do not extend to the elevated altitudes measured in HIPPO and  
14 to a lesser extent in the other studies. It should be pointed out that there is a significant model to  
15 model variability at altitude in this and other regions but the measurement data is not available to  
16 confirm whether upper tropospheric BC values are similar in this region to the remote Pacific and  
17 continental north America.

18 We agree with both of these related points. In the text, we have added the following sections:

19 *We note, however, that the A-FORCE data do not extend as far up in the atmosphere as HIPPO did,*  
20 *and that we find significant intermodel variability at  $p < 400\text{hPa}$  also for the near-source A-FORCE and*  
21 *HIPPO America regions.*

22 *While the aircraft data in the present study were taken over the period 2008-2012, the models used*  
23 *emissions from year 2000. BC emissions have increased in the intervening period (e.g. Wang et al.,*  
24 *Trend in Global Black Carbon Emissions from 1960 to 2007, Environ Sci Technol, 48, 6780-6787, 2014,*  
25 *indicates a global mean increase of ~10%), indicating that any overestimation of concentrations by*  
26 *the models would have been strengthened had they used a more recent emission inventory. One*  
27 *model (CAM4-Oslo) delivered results for both year 2000 and 2006 emissions, reflecting this increase.*  
28 *In remote regions (e.g. the HIPPO regions in Figure 1), the resulting 20%-30% increase in*  
29 *concentration is found to be evenly distributed throughout the vertical profile, except in the range*  
30 *1000-800hPa where no significant increase was found. It is clear that for future comparisons, model*  
31 *calculations with updated emission inventories are desirable.*

32 Page 20099 lines 11-15: This statement is not true close to polluted regions and is contradicted later  
33 in the paragraph. I suggest rephrasing.

34 We agree. The start of the conclusions section now reads:

35 *We have compared recent aircraft based measurements of BC concentration with state of the art*  
36 *global aerosol-climate models. In remote regions where BC concentration are dominated by long*  
37 *range transport, and at high altitudes, there is a tendency for the models to overestimate the aircraft*  
38 *measurements, where and when the effects of fires are small.*

39 I am not sure how it can be improved, but figure 2 is very hard to read clearly and easily.

40 While we agree that the figure is dense with information, we still wish to present the data on a  
41 unified plot. In the final paper, where the page orientation is standard, the plot will hopefully come  
42 out better. A number of minor fixes have been made to improve clarity (see e.g. response to  
43 comment nr. 2 from Reviewer 2).

44

45 **Reviewer #2:**

46 SPECIFIC COMMENTS

47 1/ The authors should check the numbering of tables and figures, e.g., Fig. 4 is mentioned right after  
48 Fig. 1 and there is no reference to Table 2 in the text.

49 *We thank the reviewer for spotting this. The numbering has been updated, and a reference to table 2*  
50 *that had been lost in editing is reinserted.*

51 2/ Fig. 2 presents average vertical profiles of BC mass concentrations from observations together  
52 with results from models. Observations are presented as average plus 1 standard deviation. In the  
53 current form there is only an upper bound for observation data given, while for most of the cases,  
54 the model BC values are significantly smaller than average observation values. The authors may  
55 consider plotting the observational data as 25-percentile, median, and 75-percentile values. Then  
56 also a lower bound of observational data is given in the figures for comparison with model data.

57 *Median and +1sigma was used for consistency with other studies, however we agree that adding*  
58 *median and percentiles is relevant. In several regions, the measurements vary quite extensively, to*  
59 *the point where the mean is at times above the 75<sup>th</sup> percentile. We added the median and 25<sup>th</sup>/75<sup>th</sup>*  
60 *percentiles to the plots, to ensure this information comes across.*

61 TYPOS

62 1/ Page 20088, line 20: I suggest deleting “and”; then the sentence would read “: : : the atmospheric  
63 segment from the surface up to 250 hPa.

64 *Fixed.*

65 2/ Page 20088, line 22: There is a confusion of past tense and present tense; I suggest rephrasing the  
66 sentence: “The Polar Airborne : : : campaign consisted of : : :”. This would be in accordance with the  
67 sentences describing the other field campaigns.

68 *Fixed.*

69 3/ Page 20089, line 16: I suggest deleting “also”; then the sentence would read “To calculate BC : : :  
70 mean pressure and temperature fields were used.”

71 *Fixed.*

72

73

74 **Full manuscript including TrackChanges:**

75 **Modeled black carbon radiative forcing and atmospheric lifetime in**  
76 **AeroCom Phase II constrained by aircraft observations**

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### 133 **Abstract**

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

150 **Introduction**

151 As an absorber of solar radiation, anthropogenic BC emissions can contribute positively to  
152 global radiative forcing through the aerosol direct effect, they can affect clouds through the  
153 aerosol indirect and semidirect effects, change albedo of snow and ice, and influence  
154 precipitation by changing atmospheric stability and the surface energy balance (Myhre et al.,  
155 2013a; Ramanathan and Carmichael, 2008; Haywood and Shine, 1995). Presently both the  
156 magnitude of anthropogenic BC emissions and the resulting global distribution of BC  
157 concentrations are highly uncertain. In particular the vertical profile of BC concentration,  
158 which strongly affects its total impact on the energy balance of the atmosphere, is poorly  
159 constrained (Koffi et al., 2012; Textor et al., 2007; Samset et al., 2013). Comparisons of  
160 measurements with model results, with emphasis both on total BC mass, spatio-temporal  
161 distribution and vertical structure are therefore essential for constraining estimates of BC  
162 effects on climate.

163 The IPCC AR5 (Boucher et al., 2013) assessed the direct aerosol effect radiative forcing due  
164 to anthropogenic BC (defined here as BC from anthropogenic fossil fuel and biofuel sources,  
165 BC FF+BF) to be +0.40 [range: +0.05 to +0.80] W m<sup>-2</sup> over the period 1750-2010. That  
166 assessment took into account both model based and observational studies. Recently, Phase II  
167 of the AeroCom model intercomparison project (Myhre et al., 2013b) also evaluated BC  
168 FF+BF radiative forcing (RF), based purely on 15 global aerosol models (Myhre et al.,  
169 2013b), and found it to be +0.23 [+0.06 to +0.48] Wm<sup>-2</sup>. The uncertainty ranges (5-95%)  
170 show that BC is still a major contributor to the total uncertainty on anthropogenic radiative  
171 forcing. Further, it has recently been shown that, because the direct radiative forcing per unit  
172 mass BC increases strongly with altitude (Ban-Weiss et al., 2011; Zarzycki and Bond,  
173 2010; Samset and Myhre, 2011), the diversity in modeled vertical profiles of BC concentration  
174 in the AeroCom Phase II models may account for up to 50% of the model diversity in

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175 anthropogenic BC RF (Samset et al., 2013). Climate model simulations, however, indicate  
176 that, while direct BC forcing strengthens with altitude, its climate efficacy may decrease, i.e.  
177 the surface temperature response to BC in the middle and upper troposphere may be small or  
178 even negative (Ban-Weiss et al., 2011; Flanner, 2013). Further, (Samset et al., 2014)  
179 concluded that an upward adjustment on the model based uncertainty on total aerosol forcing  
180 may be necessary, in an analysis that looked at multi-model variability in per-species aerosol  
181 burdens and optical parameters. BC forcing diversity was found to be a significant component  
182 in this analysis.

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183 Recently, several single model studies have investigated other factors that may underlie the  
184 intermodel variability in AeroCom Phase II, or assessed its multi-model mean results in light  
185 of observations. E.g. (Wang et al., 2014a) compared results from the GEOS-Chem model  
186 (<http://geos-chem.org>) with results from the HIPPO flight campaign, and concluded that to  
187 reproduce HIPPO, more wet removal was required than is represented in most present models.

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188 Based on this, and on revised estimates on direct radiative forcing of BC when taking HIPPO  
189 constraints (Schwarz et al., 2013) into account, they argue that previous model estimates may  
190 be biased high due to elevated BC concentrations in the free troposphere. Bauer et al. (2013)  
191 studied the atmospheric lifetime of BC, which is a combined measure of transport and

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192 removal processes, by comparing simulations with the GISS-MATRIX model to HIPPO  
193 (Bauer et al., 2010), using CMIP5 emissions. They found that under present day conditions,  
194 BC lifetime should be no more than 4 days, which is significantly shorter than what is used in  
195 some present models. Hodnebrog et al. (2014) showed that such a reduction in lifetime, when

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196 combined with estimates of the impact from BC on atmospheric stability, can lead to major  
197 reductions in the global mean climate impact of BC emissions. Based on the above, it is  
198 natural to investigate both whether the full AeroCom Phase II RF estimate is biased high, as



199 was found for GEOS-Chem, and what contribution to multimodel variability may be due to  
200 differences in modeled BC lifetime.

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

## 205 **Methods**

### 206 *Flight data and BC definition*

207 We have used data from flight campaigns HIAPER Pole-to-Pole Observations (HIPPO) 1-5  
208 (Schwarz et al., 2013), Arctic Research of the Composition of the Troposphere from Aircraft  
209 and Satellites (ARCTAS) SP2 (Jacob et al., 2010), Polar Airborne Measurements and Arctic  
210 Regional Climate Model Simulation Project (PAMARCMiP) (Herber et al., 2012; Stone et al.,  
211 2010) and Aerosol Radiative Forcing in East Asia (A-FORCE) (Oshima et al., 2012). (See  
212 Table 1)

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

217 HIPPO 1-5 (Schwarz et al., 2013) flew mainly pole-to-pole over the Pacific Ocean at various  
218 times during 2009-2011. Combining data from all five campaigns yields an approximate  
219 annual average. The HIPPO data have been screened against contributions from fires, so as to  
220 be representative of the background concentration of BC over the Pacific. Two of the HIPPO  
221 campaigns also flew over the North American mainland, allowing also for comparisons closer

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222 to anthropogenic BC source regions. HIPPO covers atmospheric pressures from surface  
223 values up to 100hPa, meaning that its upper range reaches into the lower stratosphere.

224 The ARCTAS SP2 campaign (Jacob et al., 2010) was flown in two separate time periods in  
225 2008. During spring, flights were conducted over the northern Pacific Ocean, comparable to  
226 parts of the HIPPO region, and also over the North Polar regions. During summer, flights  
227 were conducted over the North American continent, again comparable to HIPPO. Part of the  
228 ARCTAS motivation was to study fires, so the measured concentrations can be expected to  
229 have a larger contribution from open biomass burning than the HIPPO dataset. ARCTAS data  
230 cover the atmospheric segment from the surface ~~and~~ up to 250hPa.

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

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236 Finally, the Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaign (Oshima et  
237 al., 2012) flew over East Asia (southwestern Japan) in spring 2009, covering a vertical range  
238 up to 300hPa. It is not regionally comparable to the other campaigns, but is highly relevant  
239 because it represents a region dominated by outflow from mainland China, one of the main  
240 sources of anthropogenic BC emissions.

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#### 241 *AeroCom Phase II models*

242 The models results in the present study come from 13 state of the art aerosol models,  
243 submitted as part of Phase II of the AeroCom model intercomparison project (Myhre et al.,  
244 2013b; Samset et al., 2013; Schwarz et al., 2013). Participating models are NCAR-CAM3.5  
245 (Lamarque et al., 2012), CAM4-Oslo (Kirkevåg et al., 2013), CAM5.1 (Liu et al., 2012),

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246 GISS-MATRIX (Bauer et al., 2010), GISS modelE (Koch et al., 2011), GMI (Bian et al.,  
247 2009), GOCART-v4 (Chin et al., 2009), HadGEM2 (Bellouin et al., 2011), IMPACT (Lin et  
248 al., 2012), INCA (Szopa et al., 2012), ECHAM5-HAM (Zhang et al., 2012), OsloCTM2  
249 (Skeie et al., 2011) and SPRINTARS (Takemura et al., 2005). See (Myhre et al., 2013b) and  
250 individual model references for further descriptions.

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251 All models submitted monthly mean 3D fields of total BC mass mixing ratios, using year  
252 2000 emissions (Lamarque et al., 2010), ~~year~~ Meteorological year was 2006-meteorology, or  
253 model internal present day (PD) climatology. To calculate BC concentrations from mixing  
254 ratios, the models' own monthly mean temperature and pressure fields were ~~also~~ used. When  
255 discussing modifications to the BC radiative forcing from the direct effect from anthropogenic  
256 fossil fuels and biofuels, the models' own monthly mean 2D forcing fields were used, in  
257 combination with a preindustrial simulation using year 1850 emissions but still year 2006 or  
258 PD meteorology (Myhre et al., 2013b). For consistency with recent literature, forcing results  
259 are given for 1750-2010, using scaling factors presented in (Myhre et al., 2013b).

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## 260 *Analysis*

261 To compare flight campaigns and model output, a series of geographical regions were first  
262 selected. See Figure 1. For the flight data, only measurements that fell within the regions were  
263 kept, and for each region an average profile was constructed. For the models, all output within  
264 the selected region was averaged into a single profile for each model. However, to take the  
265 seasonality into account, we produced a model profile for each measurement point or profile  
266 from the flights. These were then averaged. The result is a set of model profiles that  
267 correspond to the flight profiles both geographically and temporally.

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

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

#### 285 *Calculation of RMS values and correlations*

286 To estimate how well a given model reproduces the Pacific HIPPO flight data, as primarily  
287 used below, we calculated model bias, root-mean-square (RMS) error values and correlation  
288 coefficients. The HIPPO dataset was subdivided into five regions (P1-P5 in Figure 1), and an  
289 annual mean profile was constructed for each region as shown in Figure 2. For each model,  
290 diagnostics were calculated from the difference between the HIPPO concentration profile at

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291 each of its given altitude levels, and the regionally averaged model concentration value  
 292 interpolated to the corresponding altitude, according to the following equations:

$$Mean\ bias = \frac{1}{N} \sum_{Reg=P1}^{P5} \sum_{Alt=Surf}^{TOA} [C_{HIPPO}(Reg, Alt) - C_{Model}(Reg, Alt)]$$

$$Normalized\ mean\ bias = \frac{1}{N} \sum_{Reg=P1}^{P5} \sum_{Alt=Surf}^{TOA} \frac{[C_{HIPPO}(Reg, Alt) - C_{Model}(Reg, Alt)]}{0.5 \times [C_{HIPPO}(Reg, Alt) + C_{Model}(Reg, Alt)]}$$

$$RMS\ Error = \sqrt{\frac{1}{N} \sum_{Reg=P1}^{P5} \sum_{Alt=Surf}^{TOA} [C_{HIPPO}(Reg, Alt) - C_{Model}(Reg, Alt)]^2}$$

293 Here C denotes a concentration value and N is the total number of data points. In the present  
 294 case, this value is 72, determined by the number of altitude bins where HIPPO reported  
 295 measurements. Further, we calculated the Pearson sample correlation coefficient based on the  
 296 same dataset.

### 297 *Derivation of scaled forcing estimates*

298 Two scalings were applied in the present work to assess the potential impact of adjusting  
 299 models to measured BC concentrations. These scalings were derived by altering the 3D  
 300 concentration fields of total BC (fossil fuel, biofuel and biomass burning) provided by the  
 301 AeroCom models, and then applied to the BC FF+BF forcing fields supplied. This method is  
 302 used to ensure that intermodel variability in RF due to differences in optical parameters of BC,  
 303 cloud distributions and other factors related to the host model are kept unchanged.

304 For the “remote ocean” scaling, the concentration fields were altered within the grey boxes  
 305 shown in Figure 1. Between the surface and 500hPa concentrations were reduced to 1/3, then  
 306 to 1/8 up to 200hPa, and then to 1/15 up to TOA. These factors were derived from the

307 comparison between AeroCom Phase II and HIPPO 1-5 presented in Schwarz 2013 (Schwarz  
308 et al., 2013).

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309 Using the forcing efficiency profile method (Samset and Myhre, 2011), we then calculated  
310 global, annual mean BC RF from both scaled and unscaled concentration fields. The ratio of  
311 these forcing values is taken as the scaling factor for that particular model.

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312 Finally, we constructed the multi-model median BC FF+BF for all 13 models used for the  
313 present study, based on their original 2D forcing fields. These forcing values were scaled with  
314 the derived scaling factors, to produce the revised model median forcing ~~estimates presented~~  
315 ~~in Figure 4.~~

316 For the “high altitude” scaling the same procedure was followed, except that the concentration  
317 fields were scaled to 1/20 at altitudes between 200hPa and TOA globally.

318 For the “all scaled” analysis both scalings were applied to the concentration fields, i.e. the  
319 fields were scaled to 1/20 at altitudes between 200hPa and TOA globally, and then as  
320 described above in the grey marked regions at altitudes below 200hPa.

## 321 **Results and discussion**

### 322 *Comparisons of flights and models*

323 Here, we constrain the model range of global, annual mean direct radiative forcing by  
324 anthropogenic BC, by comparing AeroCom Phase II vertical profiles from 13 models, to  
325 recent aircraft campaigns. Figure 1 shows the flight tracks of the four campaigns, the  
326 AeroCom multimodel median anthropogenic BC forcing field, and the regions selected for  
327 analysis.

328 [Table 2 shows individual model BC RF, recalculated using the forcing efficiency profile](#)  
329 [method, globally and for the regions in Figure 1. We also show the fraction of exerted above](#)  
330 [5km \(500hPa\),](#)

331 Figure 2 compares flight campaign data with AeroCom Phase II model output. Panels a-f  
332 show the HIPPO1-5 campaigns (Schwarz et al., 2013) for five regions in the remote Pacific  
333 Ocean and for western North America, overlain with AeroCom Phase II results. A common  
334 pattern is that the models strongly overpredict the HIPPO measurements. Further, the  
335 overprediction is more pronounced at high altitudes. Comprised of five campaigns distributed  
336 throughout the year, HIPPO represents an approximate annual average. As recently noted  
337 (Schwarz et al., 2013), its Pacific measurements indicate that at the highest altitudes studied,  
338 BC concentrations converge towards a common background value, here found to be  
339 approximately  $0.1 \text{ ng m}^{-3}$ , with very low seasonality. Here we also find (Figure 2f) the same  
340 background value above western North America.

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

348 Panels j-l show PAMARCMiP (Herber et al., 2012; Stone et al., 2010) data, first over the  
349 North Pacific region, and then over two North Polar regions. While the altitude range covered  
350 by PAMARCMiP is limited compared to ARCTAS and HIPPO, the concentrations found in  
351 the lowest few kilometers of the troposphere are consistent with ARCTAS. Over the NP1 and

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352 NP2 region, north of America and Greenland, models underpredict the measurements. As for  
353 ARCTAS, this is at least partly due to episodic fires. The Arctic region may however have  
354 further sources of BC not adequately represented in the emission inventories used by the  
355 models (Stohl et al., 2013).

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356 Panel m shows A-FORCE (Oshima et al., 2012) data in the sea areas around Japan. Here we  
357 find good agreement between models and measured concentrations, both in absolute values  
358 and in the shape of the vertical profile. The variability between models is also much lower in  
359 this region than for the others. The aerosol in the A-FORCE region is mainly sensitive to  
360 outflow from mainland China, Korea and Japan. This indicates that in AeroCom Phase II,  
361 East Asian BC emissions and outflow are either well represented or, if emissions are still  
362 underestimated as discussed for AeroCom Phase I in recent literature (Bond et al.,  
363 2013; Chung et al., 2012), the atmospheric lifetime of BC must be compensatingly long in the

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364 models to allow enough BC to be transported into the region sampled by A-FORCE. We note,  
365 however, that the A-FORCE data do not extend as far up in the atmosphere as HIPPO did, and  
366 that we find significant intermodel variability at p<400hPa also for the near-source A-FORCE  
367 and HIPPO America regions.

368 While the aircraft data in the present study were taken over the period 2008-2012, the models  
369 used emissions from year 2000. BC emissions have increased in the intervening period (Wang  
370 et al., 2014b), indicating that any overestimation of concentrations by the models would have  
371 been strengthened had they used a more recent emission inventory. One model (CAM4-Oslo)  
372 delivered results for both year 2000 and 2006 emissions, reflecting this increase. In remote  
373 regions (e.g. the HIPPO regions in Figure 1), the resulting increase in concentration is found  
374 to be evenly distributed throughout the vertical profile, except in the range 1000-800hPa  
375 where no significant increase was found. It is clear that for future comparisons, model  
376 calculations with updated emission inventories are desirable.

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377 *Consequences for BC atmospheric lifetime*

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

385 Figure 2 shows that some models more closely reproduce the measurements than others, both  
386 in magnitude and shape. Several studies have suggested that to reproduce HIPPO data, a low  
387 modeled atmospheric lifetime, or a short ageing timescale, of BC is required (Bauer et al.,  
388 2013; Wang et al., 2014a). Here we can test this supposition for a larger set of models.

389 Quantifying the difference between models and data requires care, as absolute concentrations  
390 range over several orders of magnitude. Common diagnostic variables include model bias and  
391 RMS error. Of these, RMS error and model mean bias (see Methods) will be dominated by  
392 high absolute concentrations, i.e. low altitudes in the present case. Model mean normalized  
393 bias avoids this, but will be more sensitive to model and measurement uncertainties in high  
394 altitude, low concentration ranges. In Figure 3, RMS error and biases are plotted as function  
395 of the modeled BC lifetime. Lifetime, also referred to as atmospheric residence time, is here  
396 defined as modeled global, annual mean emissions divided by burden (Table 3).

397 Figure 3 shows that, independent of diagnostic variable, a low BC lifetime is a requirement  
398 for good reproduction of absolute modeled concentrations. Regressing bias or RMS error  
399 versus lifetime (black, dashed line in Figure 3) gives an intercept at 3 days for RMS error and  
400 model mean bias. This value is in line with indications from other recent studies, e.g. (Bauer

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401 et al., 2013). In the present dataset, a single model with high lifetime (HadGEM2) represents a  
402 significant outlier. That model did not include BC ageing and transition to a hydrophilic state,  
403 with the consequence that both BC lifetime and burdens over remote areas become high  
404 (Bellouin et al., 2011). To test the impacts of single models on the result, Figure 3 also shows  
405 regressions with one model removed (grey lines). For the normalized mean bias, which is less  
406 sensitive to high concentrations, the regression with this particular model removed is  
407 consistent with the results from RMS error and mean bias.

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

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

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423 *Consequences for modeled BC FF+FB RF*

424 However, the three models that best reproduce HIPPO in the Pacific all report consistent and  
425 relatively low BC FF+BF forcing, exerted close to emission sources as expected from the low  
426 lifetime. In these three models very little BC reaches remote ocean regions, or gets lifted  
427 above 500hPa, relative to the other models in the ensemble. While the correspondence to  
428 HIPPO cannot be used to extract information close to emission sources, it does suggest that  
429 scaling down the average modeled forcing aloft and in remote ocean regions has merit.

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

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

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447 Figure 4 shows the implications of these scalings on the multimodel direct RF due to BC from  
448 fossil fuel and biofuel burning (BC FF+BF). Unscaled, the multimodel median RF found here  
449 is +0.24 [+0.17 to +0.47] W m<sup>-2</sup>. (See Table 3) Applying the remote region scaling reduces  
450 the global, annual median BC FF+BF RF to +0.22 [+0.16 to +0.39] W m<sup>-2</sup>. The high altitude  
451 scaling reduces it to +0.19 [+0.15 to +0.33] W m<sup>-2</sup>. Applying both simultaneously, while  
452 ensuring that we do not doubly scale in remote, high altitude regions, yields a BC FF+BF RF  
453 of +0.17 [+0.13 to +0.28] W m<sup>-2</sup>, or a reduction of 25% from the AeroCom Phase II value  
454 combined with a strong reduction in the model spread.

455 A 25% reduction in the direct radiative forcing of BC would have significant implications,  
456 placing the entire model based 5-95% range below the central BC RF value recently reported  
457 in IPCC AR5 (Boucher et al., 2013). Presently, the remaining uncertainty in BC forcing is  
458 heavily driven by scalings such as the ones attempted above. The IPCC AR5 assessment took  
459 input both from AeroCom Phase II and other studies. One of these studies (Bond et al., 2013)  
460 reported a significantly stronger forcing of 0.51 Wm<sup>-2</sup> from fossil fuel and biomass burning.  
461 That estimate includes both a gross 15% global downscaling of BC forcing efficiency due to  
462 overestimation of BC aloft, and a differentiated regional upscaling of emissions derived by  
463 comparing aerosol absorption optical depth from AeroCom Phase I with that from AERONET  
464 ground based remote sensing. Their downscaling, based on recent model studies (Bond et al.,  
465 2013; Samset and Myhre, 2011; Zarzycki and Bond, 2010) and evaluation of AeroCom Phase I  
466 results (Schwarz et al., 2010), is comparable to our 25% reduction in forcing, though our  
467 reduction is attributed to remote ocean areas. For near source and remote regions covered in  
468 the present study we here find no need for an emission bias related upscaling; however the  
469 present data do not cover the regions where the upscaling in that analysis (Bond et al., 2013)  
470 was most pronounced. Also, the median anthropogenic BC RF in AeroCom Phase II is  
471 already a factor of 2 stronger than in Phase I, in part due to differences in emissions and

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472 modeled aerosol optical properties (Myhre et al., 2013b). We note that the above conclusions  
473 are broadly consistent with recent findings using the GEOS-Chem model, which is not  
474 represented in the present dataset (Wang et al., 2014a). It is clear that further observations of  
475 BC concentrations, vertically resolved and both in situ and remote, are imperative for  
476 constraining the radiative forcing of black carbon.

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477 *Applicability of scaling factors derived from total BC to BC FF+BF fields*

478 A question raised by the scaling analysis is whether we bias the results by deriving scaling  
479 factors based on total BC fields, and subsequently applying them to BC FF+BF forcing. At  
480 present, no measurement exists that can determine systematic differences between the global  
481 distributions of total BC and BC from fossil fuel and biofuel burning. However four of the  
482 models participating here (OsloCTM2, CAM3, CAM5 and IMPACT) also supplied full 3D  
483 concentration fields from BC FF+BF only, and we have used these to test the applicability of  
484 the method.

485 For this subset of models, which spans the range of predicted BC burdens, we found the ratio  
486 of modeled anthropogenic BC FF+BF to total BC concentrations to be approximately constant  
487 with altitude in the regions defined as remote. While trends exist for individual models in  
488 single regions, for the remote regions as a whole the ratio changes by less than 10% through  
489 the atmospheric column. Hence any alteration of the BC vertical profile should equally affect  
490 both fields. Further, the fraction of the total global mean forcing found to be exerted at  
491 altitudes above 200hPa was, for these models, found to be comparable for total BC and BC  
492 FF+BF. (See Table 2.)

493 These two observations lead us to conclude that we do not strongly bias our results by  
494 applying scaling factors derived from total BC fields to BC FF+BF forcing.

495 *Forcing pattern from models with low RMS and good correlation with HIPPO*

496 We have shown that of the models participating in the present comparison, there are three  
497 (GMI, GISS-MATRIX, IMPACT) that both show a low RMS error and a good correlation  
498 with the Pacific HIPPO data. These three models all have low global mean atmospheric  
499 lifetimes of BC, and report among the lowest BC FF+BF RF values in the AeroCom Phase II  
500 ensemble.

501 Figure 5 shows the zonal mean BC FF+BF RF, and total BC forcing density (RF per unit  
502 height) vertical profile, from the full model ensemble, and from the three HIPPO-  
503 corresponding models only. Total BC is used for the vertical profile as not all models  
504 provided full 3D concentration fields, as outlined above. The obvious feature is that for these  
505 three models, forcing is exerted primarily closer to the sources, and at lower altitudes, than in  
506 the full ensemble. Very little is exerted in the southern ocean, or above 200hPa.

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

## 514 **Conclusions**

515 ~~In conclusion, when~~We have comparing recent aircraft based measurements of BC  
516 concentration with state of the art global aerosol-climate models. In remote regions where BC  
517 concentration are dominated by long range transport, and at high altitudes, there is a tendency  
518 for the models to overestimate the aircraft measurements, where and when the effects of fires  
519 are small. ~~This overestimation is most pronounced in remote regions, where BC~~

520 | ~~concentrations are dominated by long range transport, as well as at high altitudes.~~ For a region  
521 sensitive to Asian emission sources, models reproduce the aircraft measurements remarkably  
522 well, with no indication of an underestimation in BC emissions. In remote ocean regions, an  
523 atmospheric lifetime of anthropogenic BC of less than 5 days seems crucial, but not sufficient,  
524 to be able to reproduce measurement data. Scaling the multimodel results to HIPPO  
525 measurements, remotely and aloft, and assuming a globally uniform high altitude BC  
526 concentration, leads to a reduction of 25% in anthropogenic BC direct RF, relative to the  
527 models native values. The revised median of  $0.17 \text{ Wm}^{-2}$  stands in stark contrast to recent  
528 assessments, which report up to 2-3 times stronger present day BC forcing, but is in line with  
529 recent single-model studies (Wang et al., 2014a; Bauer et al., 2013). This discrepancy  
530 underlines the impact of combining measured BC concentration data with model estimates.  
531 To resolve these differences, and better constrain the climate impact of BC, there is an urgent  
532 need for further flight campaigns to provide BC vertical concentration profiles over both  
533 source regions, and regions where anthropogenic BC concentrations are dominated by  
534 transport and wet scavenging.

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558 **Figure captions**

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

565

566 Figure 2: Comparison of measurements and model data for all selected regions. For each panel, the left box shows an  
567 overlay of the observed total BC concentration profiles (black lines: mean (solid), median (dotted) and mean and +1  
568 standard deviation (dashed), and 25<sup>th</sup>-75<sup>th</sup> percentile range (grey band)-black) with the mean BC concentration profiles  
569 from individual AeroCom Phase II models (colored lines, see legend). The three middle boxes show, from left to right, the  
570 BC burden (mg/m<sup>2</sup>), direct radiative forcing (W/m<sup>2</sup>) and forcing efficiency (W/g) for observations (black) and models  
571 (red). The colored diamonds show the individual AeroCom Phase II models. Finally, the rightmost box shows the ratio of  
572 models to observations for the burden (green), radiative forcing (blue) and forcing efficiency (red) within the selected  
573 region.

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576 Figure 3: Modeled global black carbon lifetime plotted versus root-mean-square error, mean bias and normalized mean  
577 bias, between model prediction and observations for the five HIPPO regions (P1-P5). Each RMS or bias calculation is  
578 based on 72 data points. The symbol size indicates the corresponding Pearson correlation coefficient. The black dashed  
579 line shows a least-squares regression line with all models included. The grey lines show regressions with one model  
580 removed. The red dashed line shows a regression using only models with  $\rho > 0.8$ .

581

582

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

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589 Figure 5: Zonal mean and altitude forcing profiles for AeroCom Phase II models. a) BC FF+BF forcing, zonal mean, for all  
590 13 models in the present study (black, solid), and the three models selected based on RMS and correlation vs HIPPO (red,  
591 solid). Dotted lines show only forcing over land, dashed show forcing over ocean. b) As a, except total BC (FF+BF+BB)

592 forcing, and global, annual mean vertical profile. Forcing is shown per altitude meter (unit  $\text{Wm}^{-3}$ ), to avoid dependence  
593 on model vertical structure.

594

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

597

598 **Supplementary tables:**

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

<b>Campaign</b>	<b>Region</b>	<b>Time</b>	<b>Data/web site</b>
<b>HIPPO 1</b>	Pacific	January 2009	<a href="http://hippo.ucar.edu/">http://hippo.ucar.edu/</a>
<b>HIPPO 2</b>	Pacific	November 2009	
<b>HIPPO 3</b>	Pacific	March/April 2010	
<b>HIPPO 4</b>	Pacific	June 2011	
<b>HIPPO 5</b>	Pacific	August 2011	
<b>A-FORCE</b>	Japan	March/April 2009	
<b>ARCTAS Spring</b>	Northern Pacific, North Polar	April 2008	<a href="http://wwwair.larc.nasa.gov/missions/">http://wwwair.larc.nasa.gov/ missions/</a>
<b>ARCTAS Summer</b>	Continental North America	July 2008	<a href="http://arctas/dataaccess.htm">arctas/dataaccess.htm</a>
<b>PAMARCMiP 2009</b>	Northern Pacific, North Polar	April 2009	
<b>PAMARCMiP 2011</b>	Northern Pacific, North Polar	April 2011	
<b>PAMARCMiP 2012</b>	North Polar	March - April 2012	

600

601

602 **Table 2: Modeled mean forcing and fractions for the regions used in the present analysis (see Figure 1). Mean RF is the**  
 603 **forcing within that region. Fraction of global RF is defined as the fraction of energy deposited, on annual mean, within**  
 604 **that region. "Remotes" represents the total of all grey marked regions in Figure 1.**

605

<b>Region</b>	<b>Mean RF</b> <i>[Wm-2]</i>	<b>Fraction of global area</b> <i>[1]</i>	<b>Fraction of global RF</b> <i>[1]</i>	<b>Fraction of RF above 200hPa</b> <i>[1]</i>
<b>Global</b>	0.26	1.00	1.00	0.24
<b>America</b>	0.32	0.01	0.01	0.30
<b>Japan</b>	0.98	0.01	0.04	0.20
<b>P1</b>	0.28	0.01	0.01	0.31
<b>P2</b>	0.39	0.02	0.03	0.28
<b>P3</b>	0.17	0.03	0.02	0.32
<b>P4</b>	0.06	0.03	0.01	0.43
<b>P5</b>	0.06	0.02	0.01	0.47
<b>NP1</b>	0.26	0.003	0.003	0.30
<b>NP2</b>	0.25	0.003	0.003	0.31
<b>Remotes</b>	0.17	0.26	0.15	0.35

606

607

608 **Table 3: BC FF+BF RF and BC atmospheric lifetime of BC for the models used in the present study. Bias, RMS error and**  
609 **correlation coefficients are for comparisons of each model with HIPPO data for all Pacific regions (P1-P5). Each**  
610 **calculation is based on 72 data points.**

<b>Model</b>	<b>BC FF+BF RF</b>	<b>BC Lifetime</b>	<b>RMS error</b>	<b>Correlation</b>	<b>Mean bias</b>	<b>Normalized bias</b>
	<i>[Wm<sup>-2</sup>]</i>	<i>[days]</i>	<i>[ngm<sup>-3</sup>]</i>	<i>[1]</i>	<i>[ngm<sup>-3</sup>]</i>	<i>[1]</i>
<b>INCA</b>	0.18	7.1	5.6	0.12	0.002	0.79
<b>GOCART</b>	0.18	7.2	13.3	0.86	0.010	1.34
<b>OsloCTM2</b>	0.28	6.0	4.7	0.64	0.003	0.87
<b>CAM4-Oslo</b>	0.37	8.2	15.9	0.72	0.014	1.44
<b>SPRINTARS</b>	0.21	6.7	4.4	0.62	0.002	0.78
<b>ECHAM-HAM</b>	0.14	5.5	4.5	0.39	-0.002	0.01
<b>HadGEM2</b>	0.19	17.1	34.1	0.84	0.025	1.56
<b>GISS-modeIE</b>	0.21	5.9	6.1	0.67	0.004	0.87
<b>IMPACT</b>	0.14	3.8	2.7	0.82	0.000	0.10
<b>GMI</b>	0.17	4.4	4.1	0.86	0.003	0.68
<b>CAM5</b>	0.2	3.8	4.6	0.37	-0.001	0.15
<b>NCAR-CAM3.5</b>	0.15	5.8	4.1	0.80	0.002	0.71
<b>GISS-MATRIX</b>	0.19	3.5	2.9	0.87	-0.001	-0.49

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