

**Supporting Information: Derivation of GWP for black carbon**

***Definition***

IPCC (1) defines the global warming potential as:

$$\text{GWP}_s = \frac{\left[ \int_0^\tau a_s \cdot r_s(t) dt \right]}{\left[ \int_0^\tau a_{\text{CO}_2} \cdot r_{\text{CO}_2}(t) dt \right]} \quad (\text{S.1})$$

where  $\tau$  is the time frame of interest,  $a$  is the forcing efficiency,  $r(t)$  is the atmospheric burden at time  $t$  after a pulse injection, and the subscripts  $s$  and  $\text{CO}_2$  refer to the substance of interest and  $\text{CO}_2$ , respectively.

***Black carbon***

We estimate  $a_{\text{BC}}$  and  $r_{\text{BC}}(t)$  by using the results of modeling studies from seven independent groups, summarized in Table S1. The average of 5.5 days is taken as a central value for the decay time in an exponential to represent  $r_{\text{BC}}(t)$ . Two studies (2,3) examined various removal processes and provided lifetime estimates ranging from 2.4 to 8.4 days, a range we adopt for these calculations. Our analysis excludes biomass burning sources, so we do not consider the much longer lifetimes reported by Penner *et al.* (4) for aerosol injected into upper atmospheric layers.

**Table S1. Published estimates of direct forcing by BC.**

Reference	Emission (Tg/year)	Global		Forcing (W/m <sup>2</sup> )				Unmixed BC properties			Mixed BC
		burden (Tg)	Lifetime (days)	Unmixed BC	Shell Core	Internal Homog.	calc	Absorp. (m <sup>2</sup> /g)	Scat. (m <sup>2</sup> /g)	Norm DRF (W/g)	Norm DRF (W/g)
	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)
Chung & Seinfeld (5)	A/ 11.5	0.22	6.6	0.51	-	0.80	g	7.8	4.8	1300	1900
Cooke et al. (6)	F/ 5.1	0.073	5.2	0.17	-	-	c	8.5	2.5	1200	-
Haywood & Rama- swamy (7)	A/ 14	0.28	7.3 <sup>a</sup>	1.06	-	-	c	7.4	1.8	1900	-
" larger particles	"	"	"	0.38	-	-	"	3.1	2.7	670	-
Jacobson (8, 9)	A/ 11.9	0.23	7.1	0.27	0.54	0.78	c	4.2	2.1	600	1200, 1700
Koch (3)	A/ 12.2	0.15	4.8	0.35	-	-	g	6.2	2.8	1200	-
Myhre et al. (10)	F/ 5.1	0.07	5.0	0.16	-	0.42	c	7.4	1.8	1200	3100
Penner et al. (11)	F/ 5.64	0.16	5.3	0.20	-	0.40 <sup>b</sup>	c	7.4	1.8	1200	2500
Wang (12)	A/ 14	0.17	4.4	0.22	-	-	g	5.9	2.1	1100	-
Best estimate								7.5 <sup>c</sup>	2.5 <sup>c</sup>	1200	1800

*Explanation of columns:* (1) emission sources used, F=fossil fuel, A=all (both fossil-fuel and biomass burning); (2) global burden simulated by transport model; (3) lifetime estimated assuming simple box model; (4, 5, 6) TOA direct forcing from the model results; (7, 8, 9) mass-specific absorption and scattering of pure, unmixed soot (c) calculated from data given by authors, using Mie theory, or (g) given by authors directly; (10) normalized direct radiative forcing (DRF) for pure, externally mixed aerosol obtained by dividing forcing by burden; (11) same as column 10 but for BC aerosol mixed with other compounds. *Notes:* (a) Study used the aerosol climatology of an earlier study. (b) Forcing by mixed aerosol inferred from values given in the paper. (c) From review of measured optical properties (13).

The choice of  $a_{BC}$  requires some discussion of models and particle optics. The TOA forcing divided by the atmospheric burden gives the normalized direct radiative forcing (NDRF, column 10). The median value is around 1200 W/g, and another pair of values lies around 600-660 W/g. The value of NDRF depends on both the optical properties of the BC and its location over surfaces of different albedos. Sensitivity studies (3, 8) show that NDRF is strongly related to the absorption cross-section of the particles, and the table demonstrates that differences between the two groups are caused at least partly by that factor. The single high value of NDRF comes from a study (7) in which much of the aerosol was situated above clouds, so that it had a long lifetime and a large forcing because of the reflective underlying surface. Later estimates by this research group reduced estimates of both lifetime and forcing (2). We use the highest value only in calculating the high bounds of GWP.

For the remaining values, we calculate the relationship between NDRF (column 10) and absorption cross-section (column 8) using a linear regression ( $R^2 = 0.83$ ). We use only unmixed BC results, because the absorption cross-section used in each model is not provided for mixed BC. We then choose an absorption cross-section for pure BC,  $7.5 \text{ m}^2/\text{g}$ , based on an extensive review of optical properties (13). We use this value in the regression relationship to obtain our best estimate of NDRF, which is  $1200 \text{ W/g}$  for unmixed BC.

Models that produced the median values of NDRF ( $1200 \text{ W/g}$ ) do not use measured refractive index, size, and density. However, it appears that the combined set of parameters used in these models was chosen to match the observed optics of freshly-emitted light-absorbing aerosol. For that reason, these values of NDRF are probably representative of pure soot. In contrast, the lower NDRF are calculated from more supportable assumptions, but do not match measured values. Such a mismatch could be caused by the Mie-theory assumption of spherical particles, in contrast to the actual aggregate morphology which could increase absorption by about 30% (14).

Coating the light-absorbing particles with non-absorbing material enhances both light absorption and positive forcing, and particles observed in the atmosphere are usually mixed in this fashion. We accept the most physical representation of this process modeled by Jacobson (8), which predicts that positive forcing increases by about a factor of two due to coagulation during the aerosols' lifetime. That study used realistic refractive indices, particle sizes and densities and predicted an NDRF of about  $600 \text{ W/g}$  for uncoated soot. The *median* NDRF of  $1200 \text{ W/g}$  already incorporates an increase in absorption by about 30% to match observed values. Detailed calculations of coated, aggregate soot (14) show that the "coating enhancement" offsets, rather than enhances, this 30% increase. This consideration would predict an enhancement factor of two divided by 1.3, or 1.5. The resulting NDRF

is about 1800 W/g. As Table S1 shows, this value lies between the estimates of forcing by unmixed BC and the estimates for aerosol that is mixed in an internally-homogeneous fashion. The enhancement factor of 1.5 also agrees with calculated increases in absorption cross-section (13).

Finally, we estimate uncertainties in direct BC-NDRF. Standard errors of the regression parameters represent variation in NDRF that cannot be ascribed to differences in aerosol optical properties, most likely the transport and resulting location of aerosol within three-dimensional models. These uncertainties ( $2\text{-}\sigma$ ) are included in our upper and lower bounds of NDRF. We also include low and high estimates of absorption cross-section, coating enhancements, and impact of vertical location, combining these uncertainties in quadrature. The resulting low and high boundaries are 900 and 3200 W/g.

In this analysis, we have deliberately not addressed climate forcing due to “sooty snow” (15), reasoning that such albedo-changing deposits possibly result from specific emission regions. Addressing such regions individually could yield a larger climatic benefit than that estimated here.

### ***Carbon dioxide***

Low, medium and high removal rates of CO<sub>2</sub> after emission were given by the Bern carbon-cycle model as reported to the UNFCCC (F. Joos, “CO<sub>2</sub> Impulse Response Function of Bern SAR and Bern TAR models,” <http://unfccc.int/program/mis/brazil/carbon.html>). Forcing of atmospheric CO<sub>2</sub> was 0.000994 W g<sup>-1</sup> according to the recommendations of the IPCC (1). The impulse response represented in this model has a more rapid initial decay, and results in a higher GWP, than does a simple exponential.

## ***Global warming potential***

We calculate values of  $GWP_{BC}$  for 20- and 100-year time periods using Equation S.1. Central estimates assume the intermediate value of  $a_{BC}$  (1800 W/g), an exponential decay in  $r_{BC}(t)$  using a decay constant of 5.5 days, and the intermediate equation for  $CO_2$  concentration to represent  $r_{CO_2}(t)$ . High estimates for  $GWP_{BC}$  assume the higher value of  $a_{BC}$ , a longer removal time for BC, and a more rapid decay in  $CO_2$  concentration. Similarly, the lower estimates for  $GWP_{BC}$  assume lower forcing by BC and a longer lifetime for  $CO_2$ . The uncertainties in each parameter are added in quadrature.

The central value for  $GWP_{BC,100}$  is 680, with a range of 210-1500. The estimate of  $GWP_{BC,20}$  is 2200, with a range of 690-4700.

## **References**

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