Assessing the climatic benefits of black carbon mitigation

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To limit mean global warming to 2 °C, a goal supported by more than 100 countries, it will likely be necessary to reduce emissions not only of greenhouse gases but also of air pollutants with high radiative forcing (RF), particularly black carbon (BC). Although several recent research papers have attempted to quantify the effects of BC on climate, not all these analyses have incorporated all the mechanisms that contribute to its RF (including the effects of BC on cloud albedo, cloud coverage, and snow and ice albedo, and the optical consequences of aerosol mixing) and have reported their results in different units and with different ranges of uncertainty. Here we attempt to reconcile their results and present them in uniform units that include the same forcing factors. We use the best estimate of effective RF obtained from these results to analyze the benefits of mitigating BC emissions for achieving a specific equilibrium temperature target. For a 500 ppm CO₂e (3.1 W m⁻²) effective RF target in 2100, which would offer about a 50% chance of limiting equilibrium warming to 2.5 °C above preindustrial temperatures, we estimate that failing to reduce carbonaceous aerosol emissions from contained combustion would require CO₂ emission cuts about 8 years (range of 1-15 years) earlier than would be necessary with full mitigation of these emissions.

aerosols | air pollution | climate change | radiative forcing

n the Copenhagen Accord, more than 100 countries have recognized "the scientific view that [to avoid dangerous anthropogenic interference with the climate system] the increase in global temperature should be below 2 degrees Celsius." Efforts to limit climate change have to date focused almost exclusively on reducing emission of long-lived greenhouse gases (GHGs), which are responsible for the majority of positive anthropogenic radiative forcing (RF). Air pollutants, however, not only have direct adverse impacts on human health but also impact climate. Reductions in air pollutants that form scattering aerosols with negative RF (e.g., sulfate, nitrate, and organic carbon) can warm the climate, whereas reductions in absorbing aerosols with positive RF (e.g., black carbon) can cool the climate. One recent study (1) estimated that the RF of black carbon (BC) was about about half the RF of CO₂ and larger than the forcing of other GHGs such as CH₄, halocarbons, N₂O, and tropospheric ozone (2). Moreover, because aerosols are powerful forcers with short atmospheric lifetimes (about a week), the climate effects of emission changes are realized more rapidly than for GHGs. Cobenefits for both climate and health therefore exist from strategic coordination of policies to address air pollution and climate change.

The term "carbonaceous aerosol" conjoins two categories of aerosols with distinct radiative properties: black carbon and organic carbon (OC). Both are coemitted in the incomplete combustion of organic matter. Their principal sources are contained combustion (CC) of fossil fuels and biofuels and open biomass burning (BB) (3). Chemically, BC is elemental carbon in a variety of structures (4). Radiatively, BC is defined as the absorbing component of carbonaceous aerosols and contributes to warming; in this context, the term includes some organic molecules as well as elemental carbon (1). OC aerosols are radiatively defined as the scattering component of carbonaceous aerosols and contribute to cooling. The OC/BC ratio of carbonaceous aerosol emissions varies among combustion sources, with CC sources (e.g., diesel engines, low-temperature coal combustion, and biofuels) typically having a lower OC/BC ratio than open fires. Because OC has a cooling effect, carbonaceous aerosol sources with lower OC/BC ratios generally have a greater warming effect per unit mass of BC emitted than sources with higher ratios. (See *Targets for Mitigation Efforts* below.)

Carbonaceous aerosols have strong local and regional effects on both climate and human health. For example, BC emissions from China and India, which in 1996 produced about 40% of total world CC BC emissions (3), contribute disproportionately to the warming of the Himalayas (1), while simultaneously contributing to elevated PM-2.5 concentrations and thereby increasing rates of premature mortality. BC emission reductions thus provide more immediate direct benefits for developing-world emitters than do CO_2 emission reductions and could serve as a fertile area within climate diplomacy for global cooperation between developed and developing countries (5).

Estimates of the importance of carbonaceous aerosols as global warming agents vary greatly. Part of the variability between models is due to the inclusion or exclusion of certain physical effects, including the nature of the mixing of absorbing BC aerosols with other, scattering aerosols, the multiple effects of aerosols on clouds, and the effects of BC on surface albedo. Differences also arise between models based purely on physical theory and those that attempt to fit observational data. An additional challenge in comparing model results is that different authors often present their results in different terms: Some use the net RF of carbonaceous aerosols, others the RF of BC alone, and still others the net temperature change due to carbonaceous aerosols. Furthermore, the efficacy of BC as a climate warming agent (i.e., the global equilibrium temperature response per unit RF by BC relative to the response to RF by CO_2) is highly dependent upon its vertical distribution (6), which also varies between models.

The magnitude of the direct absorptive and scattering effects of carbonaceous aerosols depends in part upon their atmospheric mixing state. Aerosols can be emitted as either an external mixture or as an internal mixture within agglomerated particles and tend to become more internally mixed as they age. The net radiative effect of externally mixed aerosols is equal to the sum of their individual effects in isolation. In internally mixed agglomerations, the absorptive strength of BC depends upon the configuration of the mixture, but tends to be higher than in isolation

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because scattering components of a particle can focus radiation onto absorbing components (2, 7).

Carbonaceous aerosols also exert several indirect effects on cloud formation and albedo. Hygroscopic aerosols serve as cloud condensation nuclei (CCN), promoting cloud formation. BC aerosols are initially hydrophobic, but become hygroscopic with aging; OC aerosols are typically a mixture of hygroscopic and hydrophobic compounds and likewise become more hygroscopic over time (8). By increasing the number of droplets in clouds, carbonaceous CCN increase cloud albedo (2). BC CCN, however, darken clouds, which has the counteracting effect of decreasing their albedo (7). BC particles can further exert a strong warming effect that extends beyond their atmospheric lifetime if they deposit onto snow or ice, thereby reducing surface albedo through the snow albedo effect (9).

Absorbing aerosols like BC also heat the air and lower relative humidity in their vicinity, resulting in a positive RF called the semidirect effect. This decreases cloud cover at the altitude of the aerosols but, by inhibiting convection, increases cloud cover below. At sufficiently low altitudes, this effect causes a reduction in cloud cover without a compensating increase at still lower levels. BC therefore exerts a strong warming effect near the surface but a weaker warming effect at higher altitudes (6). This effect is largely responsible for the altitude-sensitivity of the efficacy of BC forcing.

In this paper, our goal is to reconcile four recent sets of estimates of the warming effects of carbonaceous aerosols and translate them into common terms to permit their direct comparison. We then use our standardized results to address two questions of key policy relevance: (*i*) how large a contribution do carbonaceous aerosols from CC make to the Earth's radiative balance today, and (*ii*) what is the climatic benefit of policies to reduce carbonaceous aerosol emissions from CC?

Results and Discussion

Radiative Forcing Estimates of Carbonaceous Aerosols. We consider four different estimates of the RF strengths of carbonaceous aerosols: (i) the AeroCom intercomparison study (10, 11), which served as the main source of RF estimates of carbonaceous aerosols for the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (2); (*ii*) Hansen et al. (6, 12); (*iii*) Jacobson (7, 9, 13, 14); and (*iv*) Ramanathan and Carmichael (1). These four estimates are henceforth referred to as A, H, J, and RC. A* refers to A adjusted to account for internal mixing, as described below.

To determine the magnitude of the contribution of carbonaceous aerosols to the earth's radiative balance, we must first ensure that each model accounts for the major effects of carbonaceous aerosols, add approximations for omitted effects, and then translate these estimates into a common global denominator using the best estimates of current carbonaceous aerosol emissions. Although we label each estimate by the study on which it is based, we impute values for processes not included in the original study; consequently, our values should be regarded as interpretations of these sources.

Fig. 1 shows the probability distribution function of the effective RF (RFe, the product of RF and efficacy) of carbonaceous aerosol emissions as determined by our interpretation of each of the four studies. For A, H, and RC, we apply the efficacies calculated by ref. 6. (These are: 0.78 for CC BC, 0.58 for BB BC, 1.0 for CC OC, and 0.91 for BB OC; the difference in efficacies between CC and BB arises due to differences in geographic and altitudinal distribution.) Below we summarize our analysis. A detailed description of the method is in *SI Text*, with key parameters summarized in Table S1 and Table S2.

AeroCom (10, 11) (A): The nine models in A report the global RF of BC and particulate organic matter (POM) (which has the same RF as OC but by construction has 1.4× the mass), along with their atmospheric loadings and lifetime. Most of the models do not attempt to account for the effects of internal mixing on the optical properties of aerosols. Following ref. 6, we therefore increased RF in the five models that do not include internal mixing by a factor of two. We designate the corrected values as A*. The models also address only the direct effects of carbonaceous aerosols. We therefore augment their estimates with an estimate of the indirect cloud albedo effect and an estimate of the global snow albedo RF.

To estimate the indirect cloud albedo effect of carbonaceous aerosols, we partition the IPCC's estimate of the cloud albedo effect for all aerosols ($-0.7 \pm \frac{0.4}{1.1}$ W m⁻²) (2) among aerosol



Fig. 1. Probability distributions for the total effective RF of carbonaceous aerosols from (*A*) contained combustion (CC) and (*B*) biomass burning (BB) from the four sets of analyses considered here. The four estimates are: (*i*) A*, AeroCom intercomparison study with internal mixing adjustment; (*ii*) H, Hansen et al. (6, 12); (*iii*) J, Jacobson (7, 9, 13, 14); and (*iv*) RC, Ramanathan and Carmichael (1). The nine models in A* are shown individually and collectively. The efficacies from H are applied to A and RC. Solid lines incorporate the snow albedo effect, assuming that it is caused by CC and BB in proportion to their BC emissions, whereas dashed lines exclude the snow albedo effect. The upper x-axis on (*A*) indicates for each RF value the number of years earlier CO₂ emissions must be cut to 50% of 2005 levels in a 500 ppm CO₂e stabilization scenario where CC carbonaceous aerosols emissions are kept constant rather than cut to zero. For example, the CC RFe from H (with 90% confidence range) is 0.23 ± 0.18 W m⁻²; if CC emissions are not cut, CO₂ will have to reach 50% of 2005 levels 8.9 years (1.5–16.7 years) earlier than otherwise. See Fig. 2 for illustration.

species in proportion to their atmospheric load. This approximation assumes that each POM and BC particle contributes as much negative indirect forcing as a sulfate particle. Due to their variable hygroscopicity, carbonaceous aerosols are on average less effective CCN than sulfate and BC also has an associated darkening effect on cloud albedo. Consequently, this assumption will lead to an underestimate of the total positive RF of carbonaceous aerosols. As an estimate of the effect of BC emissions on snow albedo, we employ that of ref. 12, which is in agreement with ref. 15 ($0.05\pm 0.03 \text{ Wm}^{-2}$, with an efficacy of 2.7).

- Hansen et al. (6, 12) (H) calculate effective RFs that explicitly incorporate indirect effects and the snow albedo effect and distinguish the fraction of the indirect effects due to carbonaceous aerosols. (Ref. 12 corrects an error in the snow albedo calculation of ref. 6.) They approximate the effects of internal mixing on optical properties by doubling BC absorption. They find that carbonaceous aerosols from fossil fuels and from biomass burning have net positive and negative effective forcings, respectively.
- Jacobson (9, 13, 14) (J) includes the most sophisticated attempt to estimate the effects of internal mixing, as he tracks the growth and agglomeration of aerosol particles in air, clouds and precipitation, and the ensuing changes in optical properties. He assumes that a core BC particle is coated by other, less absorptive materials that increase the positive RF of the BC. Jacobson (7) showed that a different set of assumptions, allowing multiple BC particles per agglomeration in clouds and precipitation, yields a slightly stronger effect, but the difference between the two cases is <5%. His model explicitly incorporates indirect effects (based on ref. 16) and the snow albedo effect. J reports the globally averaged cooling that results from the elimination of fossil fuel carbonaceous aerosols. Rather than employing the effective RFs of H as with the other models, we transform J's temperature changes into effective RFs by comparing them to the warming produced by doubling CO_2 (13).
- Ramanathan and Carmichael (1) (RC): In contrast to the model-driven estimates analyzed here, RC estimate the global RF of BC and non-BC aerosols by coupling satellite and surface observations of aerosol optical depth, single scattering albedo, and asymmetry parameters with an aerosol chemical-transport model and a radiative transfer model. Their results are thus more directly constrained by data. We apportion the non-BC RF (which includes aerosol indirect effects) among aerosol species in proportion to the atmospheric loads estimated by A. As with the aerosol indirect effect in A, this assumption may overappropriate the negative RF to OC and thus lead to an underestimate of the total positive RF of carbonaceous aerosols. Whereas RC incorporate the aerosol indirect cloud effect, they do not include the snow albedo effect. As with A, we add the snow albedo effect to their calculations using the ref. 12 values. We again use the efficacies of refs. 6 and 12.

Although individual studies within A used the same emission inventories, emissions in H, J, and RC differ. We therefore adjust the results of the studies to use the current best estimate of carbonaceous aerosol emissions Bond et al. (3), although we caution that these emission levels contain uncertainties of a factor of 2 and are based on data from 1996. Bond et al. estimate emissions from CC of 4.6 Tg/y BC and 8.9 Tg/y OC and from BB of 3.3 Tg/y BC and 25.0 Tg/y OC.

To convert the results of A and H into global RFs based on these emissions, we first use the emissions employed in each analysis (Table S1 and refs. 17 and 18) to produce steady-state normalized RFs, expressed in terms of W m⁻² per Gg/y. Because the geographical and altitudinal distribution of biofuel aerosols more closely resembles that of aerosols from fossil fuels than of aerosols from open fires, we assume that the steady-state normalized RFs of BC and OC from biofuels (and thus from CC as a whole) are the same as those from fossil fuels. Because of the short lifetimes of atmospheric aerosols (a few days; ref. 11), the steady-state normalized RFs are the ratios of the global RFs calculated by each model divided by the emissions in the same model. Such steady-state normalized RFs are appropriate for aerosols because their short atmospheric lifetimes means that aerosol loadings are almost directly proportional to their emission rates; this would not be the case for long-lived gases.

J does not separately report the effects of BC and OC; as a consequence, it is not possible to recalculate his results for different emission levels. He does, however, exclusively consider the effects of emissions from CC. Jacobson (9) uses BC emissions of 3.9 Tg/y, 15% less than those of Bond et al. (3), but also employs countervailing OC emissions of 6.0 Tg/y, 33% less. We simply employ his global temperature change value with his emissions, converted into units of effective RF so as to be comparable to the other estimates. We expect that the difference in emissions will produce a relatively small effect on global RF; using the steady-state forcings from A*, H, and RC, we find that the emissions in J produce an effective RF about 0–10% less than the Bond et al. emissions.

To calculate probability distributions for our estimates, we use a Monte Carlo simulation to sample uncertainties in forcings. For A, we assume that each of the nine models has an equal probability. For H, RC, and indirect effects inferred from Forster et al. (2), we use the quoted uncertainties. For the snow albedo effect inferred from (6, 12), we use the 65% 1 σ uncertainty estimate of ref. 6. J provides no uncertainty estimates. The resulting probability distributions for the total effective RF of carbonaceous aerosols are shown in Fig. 1. We emphasize that these distributions incorporate only currently quantifiable uncertainties in existing studies and would change if uncertainties are reevaluated.

A* falls at the low end of estimates of RFe due to CC (median of 0.02 W m⁻², with a 90% range of -0.27 to 0.23 W m⁻²). H gives rise to a median estimate of 0.22 W m⁻² (range of 0.05 to 0.40 W m⁻²), whereas RC gives rise to a median estimate of 0.25 W m⁻² (range of -0.06 to 0.47 W m⁻²). J falls toward the high end of estimates, corresponding to 0.37 W m⁻². The central tendency of these values is roughly half of the IPCC's best estimate for the forcing exerted by methane (0.48 ± 0.05 W m⁻²) (Fig. 1*A*). The median RF estimates for the carbonaceous aerosols produced by biomass burning (not including the effects of carbon dioxide) range from -0.12 to -0.31 W m⁻² (Fig. 1*B*).

Of all the studies we employed, A is closest to that used by the IPCC in its estimate of the RF of carbonaceous aerosols. The IPCC (2) reported total direct RFs of 0.15 ± 0.16 W m⁻² for FF soot and 0.03 ± 0.12 W m⁻² for BB soot. By comparison, the corresponding direct RFs from A* are $0.18 \pm \frac{0.15}{0.20}$ W m⁻² for CC and $0.09 \pm \frac{0.13}{0.10}$ W m⁻² for BB. Our results differ from those reported by the IPCC because we (*i*) use the emissions inventory of ref. 3, (*ii*) apply an internal mixing adjustment, and (*iii*) account for efficacies. The bottom-line effective RFs above further differ because they incorporate indirect and snow albedo effects. (RF probability distributions including or excluding different components of the estimates are shown in Fig. S1, Fig. S2, and Fig. S3.) That the estimates from A* fall at the low end of our estimates indicates that the IPCC may have underestimated the climatic importance of BC.

In the remainder of the paper, we interpret these results in terms of their implications for greenhouse gas mitigation. We focus on three representative estimates of the contribution of carbonaceous aerosols from fossil fuels and biofuels to current global RFe: a "best" estimate of 0.22 W m⁻², based on the means of the four median projections; a "low" estimate of 0.02 W m⁻²,

Table 1. The role of mitigation of carbonaceous aerosols from CC in achieving a 3.1 W m⁻² (500 ppm CO₂e) RF target in 2100.

	2100 CO ₂ targets		Change in timing of 50% CO ₂ target (years)
	RF(W m ^{−2})	Conc. (ppm)	
"Best" RF estimate (0.22 W n	n ⁻²)		
Full mitigation	2.21	420	_
Constant emission	2.00	404	8.4
A2	2.06	408	5.2
B1	2.11	413	3.1
"Low" RF estimate (0.02 W n	n ⁻²)		
Full mitigation	2.21	420	_
Constant emission	2.19	419	0.8
A2	2.19	419	0.8
B1	2.18	418	1.1
"High" RF estimate (0.37 W	m ⁻²)		
Full mitigation	2.21	420	_
Constant emission	1.84	392	14.5
A2	1.94	399	10.9
B1	2.04	407	6.0

For each estimate of carbonaceous aerosol RF (best, low, high), we report the resulting targets for CO_2 RF and concentrations in 2100. In the constant emissions scenarios, carbonaceous aerosol emissions are maintained at 1996 levels. Change in timing of 50% CO_2 target records the number of years earlier than in the full mitigation scenario that CO_2 emissions must be cut to 50% of their 2005 levels to achieve the 3.1 W m⁻² (500 ppm CO_2e) RF target in 2100.

based on the median of A*; and a "high" estimate of 0.37 W m⁻², based on J.

Implications for Greenhouse Gas Targets. The most commonly discussed greenhouse gas emission reduction scenarios, such as the mitigation scenarios employed by Working Group 3 in the IPCC's Fourth Assessment Report, focus on long-lived greenhouse gases and sometimes include ozone precursors and sulfate aerosols, but do not consider carbonaceous aerosol emissions (e.g., 19, 20). Many studies also do not consider changes in scattering aerosols such as sulfate, which currently mask a significant fraction of the positive anthropogenic RF but are expected to decrease with pollution controls over the course of the century (e.g., 21, 22). However, if controls targeted at pollutants such as SO₂ are successful and carbonaceous aerosol emissions are not significantly reduced, then meeting a maximum temperature increase target, such as the 2 °C target endorsed by the Copenhagen Accord, will require greater reductions in CO₂ emissions than indicated by these scenarios. Such large CO₂ reductions may be economically unachievable.

Using the three estimates of the contribution of carbonaceous aerosols from CC to current RF calculated in the previous section, we consider the implications for 21st century CO₂ emission targets of four alternative scenarios for carbonaceous aerosol emissions from CC of fossil fuels and biofuels. In the "constant emissions" scenario, carbonaceous aerosol emissions remain constant through the century at their 1996 levels. In the "full mitigation" scenario, carbonaceous aerosol emissions from CC are eliminated entirely by 2100. In the third and fourth scenarios, carbonaceous aerosol emissions projected for 2050 consistent with the IPCC Special Report on Emissions Scenarios (SRES) A2 and B1 story lines (CC BC [OC] emissions approximately 28% [28%] and 46% [35%] below 1996 levels, respectively; ref. 23) persist until 2100. These two scenarios represent the high and low end projections of ref. 23 and provide a sense of the reductions possible in the absence of directed policy.

We further assume that aerosol emissions from open biomass burning remain constant in all scenarios (generating a RF of -0.23 Wm^{-2} , the mean of the median estimates from A, H, and RC) and that long-lived GHGs other than CO₂ follow path-



Fig. 2. Target CO₂ emissions and concentrations under four CC carbonaceous aerosol emission scenarios using our "best estimate" of RF from carbonaceous aerosols of 0.22 W m⁻², as calculated using our simple atmospheric and economic model. (A) shows CO₂ emissions relative to 2005 levels (on the left axis) and as an annual emission rate (on the right axis). The solid lines in (B) show the associated CO₂ concentrations. The dashed lines in (B) indicate (on the left axis) total RF from all GHGs and carbonaceous aerosols in terms of ppm CO₂e and (on the right axis) the associated equilibrium warming assuming a 3 °C/CO₂ doubling climate sensitivity (24). The arrow (in A) indicates the 8 years difference in timing of the 50% reduction in CO₂ emissions between the full mitigation and constant emissions—1996 carbonaceous aerosol emissions continue through 2100. "A2" and "B1" are high and low projections of carbonaceous aerosol emissions from ref. 23 based on IPCC SRES storylines.

ways projected by the A1B storyline. These two factors together produce a RF of about 0.94 W m⁻² in 2100 (compared to 0.72 W m⁻² in 2005). As a consequence, a RF target of 3.1 W m⁻² (500 ppm CO₂e) in 2100 would allow a CO₂ concentration of approximately 420 ppm in the full mitigation scenario, about 33 ppm higher than in 2009. The most likely equilibrium temperature increase for 500 ppm CO₂e is approximately 2.5 °C (67% range of 1.7–3.8 °C) above preindustrial levels (24).

We do not consider the effects of sulfate aerosols, as our focus is on the opportunity cost of neglecting to address carbonaceous aerosols. Continued sulfate aerosol emissions would allow higher overall emissions for a given target but would have only a limited effect on the difference between scenarios and would have other deleterious environmental effects.

Failing to reduce carbonaceous aerosol emissions requires a greater reduction in CO₂ emissions to meet the same RFe or temperature target. According to our best estimate of the RFe of carbonaceous aerosols, the global RFe resulting from 1996 emission levels of carbonaceous aerosols is about 0.22 W m⁻². If these emission levels continued, they would require that CO₂ concentrations not exceed 404 ppm (a mere 17 ppm higher than in 2009) to meet a 3.1 W m⁻² RF goal in 2100. The high estimate of the RFe of carbonaceous aerosols (0.37 W m⁻²) would require that CO₂ concentrations not exceed 392 ppm, a level likely to be surpassed around 2012 (Table 1), whereas the low estimate (0.02 W m⁻²) would allow CO₂ concentrations to reach 419 ppm.

To demonstrate the implications of these targets, we use the simple model described in *SI Text* to generate corresponding emissions pathways for CO₂. Fig. 2 shows the CO₂ emission pathways possible in the four carbonaceous aerosol emission scenarios given our best estimate of carbonaceous aerosol RF. We summarize these calculations by looking at the year in which they require CO₂ emissions to be cut to 50% below 2005 levels (Table 1). This approach provides a useful alternative to metrics that are designed for long-lived heat-trapping gases, such as Global Warming Potential, for assessing the warming effect of short-lived pollutants. According to the best estimate, continuing CC carbonaceous aerosol emissions at 1996 levels will require cutting CO₂ emissions to 50% of 2005 levels 8.4 years earlier (14.5 years earlier in the high estimate and 0.8 years earlier in the low estimate) than required with complete mitigation of

carbonaceous aerosols emissions from CC and 5.3 years earlier (8.5 years earlier in the high estimate, and 0.3 years later in the low estimate) than if emissions are guided onto the B1 trajectory. The upper x-axis of Fig. 1*A* shows the change in timing of the 50% CO₂ emissions cut for the constant emissions scenario as compared to the full mitigation scenario for a range of RF estimates.

Targets for Mitigation Efforts. From a climatic perspective, not all carbonaceous aerosol sources are equivalent. Sources that emit nearly pure BC have a more positive RF than those that simultaneously emit large amounts of scattering aerosols like OC, though there is a great deal of uncertainty in determining the OC/BC ratio above which a carbonaceous aerosol emission no longer produces a positive RF. In addition, the potential for aerosols to reduce snow albedo depends upon their likelihood of transport to snow or ice-covered regions (e.g., 25). Although aerosol transport is currently an area of considerable uncertainty, carbonaceous aerosols emitted at mid-to-high latitudes or proximal to mountain glaciers likely have a greater impact on snow albedo than those emitted at low latitudes. Further, the magnitude of the Arctic snow albedo effect is highly seasonal, as the effect is unimportant during the dark Arctic winter. Fig. 3 provides estimates of the probability a particular emission is warming based on its approximate OC/BC ratio (but not including the effect of sulfur content, source location or timing). Using the average geographic and altitudinal distribution for BC and including the snow albedo effect, median estimates for this threshold value range from 2.7-5.0; excluding the snow albedo effect, median estimates range from 1.6-3.7. Using the geographic and altitudinal distribution characteristic of CC BC (Fig. S4), median estimates including (excluding) the snow albedo effect range from 2.4-6.2 (1.6-5.1). Using the distribution characteristic of BB BC (Fig. S5), median estimates range from 2.6-3.5(1.3-2.8).

Although health benefits are associated with reduced concentrations of all carbonaceous aerosols, from a climatic perspective, reducing emissions of sources with low OC/BC ratios, such as diesel engines and inefficient coal combustion, will have a greater climatic benefit than reducing emissions from sources with higher ratios, such as biofuels or open biomass burning (e.g., forest fires)



Fig. 3. Probability distribution of the OC/BC threshold above which emissions are no longer net warming, assuming an average geographic and altitudinal distribution for the emitted particles. Typical OC/BC values of different combustion sources from (3) are marked. Solid lines incorporate the snow albedo effect, assuming that it is caused by CC and BB in proportion to their BC emissions, whereas dashed lines exclude the snow albedo effect. For example, residential biofuels have a typical OC/BC ratio of 3.9; based on H, assuming an average geographic and altitudinal distribution, and including the snow albedo effect, there is a 78% chance they produce carbonaceous aerosols with a net warming effect. Although some sources (e.g., diesel engines using high sulfur fuel) coemit SO₂, the RF of the sulfate aerosol thereby formed will depend on the relative quantities of compounds emitted and whether the resulting aerosols are internally mixed (increasing RF) or externally mixed (leading the positive BC RF to be partially cancelled by the negative sulfate RF).

(Fig. 3). In developed countries, diesel engines are the main source of strongly warming carbonaceous aerosol emissions, as coal is primarily used in controlled high temperature point sources, like power plants, which emit little BC. Lower temperature residential and industrial coal combustion, common in developing countries, is another strongly warming BC source. Whereas recent discussion has focused on the residential use of biofuel-based cookstoves in the developing world, from a climatic perspective, a growing transport fleet employing uncontrolled diesel engines and residential and industrial coal use are of greater concern.

Future Directions. The range of estimates of the contribution of carbonaceous aerosols to Earth's radiative budget is considerable, with uncertainty in emission inventories compounded by physical uncertainties, particularly in indirect effects. Improved global emission inventories, reporting of model results in standardized terms, and further research into aerosol and cloud microphysics and aerosol transport to and deposition on snow and icecovered regions would reduce key uncertainties. Nevertheless, our metaanalysis of existing studies highlights the importance of efforts to reduce carbonaceous aerosol emissions from CC. In addition to the health benefits of such a reduction, our results suggest that reaching a 500 ppm CO₂e RF by 2100 in the absence of carbonaceous aerosol emission reductions requires that global CO_2 emissions be cut to half their 2005 levels about 8 years (range of 1-15 years) earlier than with aggressive reductions in carbonaceous aerosol emissions from CC. As the largest sources of carbonaceous aerosols are located in the developing world, this challenge is a clear candidate for cooperative global action.

Materials and Methods

Generation of Probability Distributions for RF. As described above, we first convert the RF estimates from models A, H, J, and RC into common terms of effective RF assuming carbonaceous aerosol emissions equal to the best estimate of ref. 3. To generate the associated probability distributions, we

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perform a Monte Carlo simulation employing the uncertainties reported for each model. The underlying equations for this analysis, as well as key parameter values, are described in *SI Text*.

Calculation of Illustrative CO₂ Emission Pathways. To calculate illustrative CO₂ emissions pathways that could be followed to meet a given RFe target in 2100, we used a simplified atmospheric and economic model. The model employs a static approximation of the Bern Carbon Cycle model (2) to determine the atmospheric lifetime of CO₂ emissions. CO₂ emissions for the 19th and 20th century were calculated to match the observed concentration profile. Reference 21st century CO₂ emissions are those of the IPCC SRES A1B scenario. Non-CO₂ GHGs follow their A1B trajectories, as modeled by the National Aeronautics and Space Administration (NASA) Goddard Institute for Space Studies (GISS). We assume that the cost of CO₂ emissions reductions relative to the reference level is proportional to the fractional reduction raised to the 2.8th power. We discount costs at 5% annually and find the minimum cost path that attains the desired RFe target. Details are provided in *SI Text*.

We note that the economic component of this model is essentially schematic but argue that a simplified model is appropriate for constructing metrics to serve as alternatives to Global Warming Potentials. Whereas the results should be interpreted cautiously, the difference between scenarios in the timing of the 50% reduction in CO₂ emissions from 2005 levels is fairly robust to changes in the discount rate and the exponential factor for mitigation costs and can be regarded with greater confidence than the exact shape and timing of the calculated pathways.

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