

1 **Will black carbon mitigation dampen aerosol indirect forcing?**

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14 If mitigation of black carbon (BC) particulate matter is accompanied by a decrease in  
15 particle number emissions, and thereby by a decrease in global cloud condensation nuclei  
16 (CCN) concentrations, a decrease in global cloud radiative forcing (a reverse “cloud  
17 albedo effect”) results. We consider two present-day mitigation scenarios: 50% reduction  
18 of primary black carbon/organic carbon (BC/OC) mass and number emissions from fossil  
19 fuel combustion (termed HF), and 50% reduction of primary BC/OC mass and number  
20 emissions from all primary carbonaceous sources (fossil fuel, domestic biofuel, and  
21 biomass burning) (termed HC). Radiative forcing effects of these scenarios are assessed  
22 through present-day equilibrium climate simulations. Global average top-of-the-

23 atmosphere changes in radiative forcing for the two scenarios, relative to present day  
24 conditions, are  $+0.13 \pm 0.33 \text{ W m}^{-2}$  (HF) and  $+0.31 \pm 0.33 \text{ W m}^{-2}$  (HC).

## 25 **Introduction**

26 Black carbon (BC) both absorbs and scatters radiation, resulting in heating of the  
27 atmosphere and reduction of solar radiation reaching the Earth's surface. Ambient  
28 measurements indicate that freshly emitted BC eventually becomes co-mixed with other  
29 compounds, such as sulfate, which are invariably less absorbing, but more hygroscopic,  
30 than BC. Whereas particles containing a mixture of BC and non-absorbing, but  
31 hygroscopic, components absorb proportionately more solar radiation than BC particles  
32 alone, the lifetime of such particles against wet deposition decreases relative to that of  
33 pure BC, leading to a complex interplay between enhanced radiative absorption but  
34 shorter lifetime [*Stier et al.*, 2007]. Accounting for the relative increase of BC since pre-  
35 industrial conditions allows reconciliation between satellite-derived and modeled  
36 estimates of direct aerosol forcing [*Myhre*, 2009]. Mitigation of BC leads to a reduction  
37 in (positive) top-of-the-atmosphere (TOA) direct radiative forcing, and therefore has been  
38 suggested as a strategy complementary to reduction of greenhouse gases [*Jacobson*,  
39 2002; *Bond*, 2007].

40 Any perturbation that affects the global aerosol number concentration has the  
41 potential to alter Cloud Condensation Nuclei (CCN) concentrations and cloud properties.  
42 The present study is based on two key assumptions: (1) a decrease in BC mass emissions  
43 would be accompanied by a decrease in primary particulate number emissions, which  
44 would lead to a lower global aerosol number concentration; and (2) by virtue of internal  
45 mixing with hydrophilic aerosol components, BC is assumed to contribute to the CCN

46 population. Depending on the fraction of BC primary particles that eventually become  
47 CCN (a function of their growth and loss rates [*Pierce and Adams, 2007*]), BC mitigation  
48 would affect global CCN concentrations, leading to a change in global cloud radiative  
49 forcing from warm clouds. If such a perturbation were to result in a reduction in TOA  
50 cloud radiative forcing, the amount of that reduction would oppose the amount by which  
51 the TOA direct BC radiative forcing is also reduced.

52 The “aerosol indirect effect” (AIE) generally relates to mechanisms by which  
53 aerosols affect cloud radiative forcing via the availability of CCN, through their size  
54 distribution, composition, and mixing state. The AIE has been divided according to: (1)  
55 the cloud albedo effect; and (2) the cloud lifetime effect. When both cloud albedo and  
56 lifetime effects are considered, the magnitude of the cloud lifetime effect has been found  
57 to range from 0.2 to 1.4 of that of the cloud albedo effect [*Lohmann and Feichter, 2005*].  
58 Although the separation into these two effects has served as a useful way to describe  
59 aerosol-cloud interactions, it is widely appreciated that such a separation is an  
60 oversimplification [*Feingold and Siebert, 2009; Stevens and Feingold, 2009*]. For the  
61 purpose of the present study, we follow an approach to estimating aerosol indirect forcing  
62 similar to that of IPCC [*Forster et al., 2007*].

## 63 **2. Effect of BC Emission Reductions on Aerosol Indirect Forcing**

64 The present-day direct and indirect aerosol forcings associated with the prescribed  
65 reduction in BC emissions have been evaluated by employing the Goddard Institute for  
66 Space Studies (GISS) Global Climate Middle Atmosphere Model III (referred to as GISS  
67 III hereafter), following *Chen et al. [2010]* (see footnotes in Table 1). Effects from the

68 changes in both cloud albedo and lifetime are considered, while indirect effects on  
69 convective clouds and mixed-phase and ice clouds are not included in the present study.

70 The atmospheric aerosol number concentration is established, in principle,  
71 through a combination of primary particle emissions, in-situ nucleation, coagulation, and  
72 removal processes [Adams and Seinfeld, 2002; Pierce and Adams, 2009]. The effect of  
73 BC mitigation on global CCN concentrations is assessed using the GISS-TOMAS model  
74 [Adams and Seinfeld, 2002]. Primary particles considered include sea salt, mineral dust,  
75 “primary” sulfate produced in power plant plumes, and primary BC and organic carbon  
76 (OC). Sea salt includes a portion of ultrafine particles [Pierce and Adams, 2006].  
77 Atmospheric new particle formation is represented by binary H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O nucleation,  
78 accounted for using the model of Vehkamäki *et al.* [2002]. Particle growth occurs by  
79 coagulation and condensation of sulfate and secondary organic aerosol (SOA). Nitrate  
80 formation is not included in the present calculation, as it is likely to exert a small effect  
81 under present-day conditions.

82 According to Köhler theory, the CCN activity of a chemically heterogeneous  
83 particle depends primarily on the moles of solute it contains. Since the majority of the  
84 emitted particles are small, ultrafine particles and initially hydrophobic, the amount of  
85 solute they contain will be dominated by sulfuric acid and organics that condense on  
86 them during atmospheric processing. For example, even modest growth of an initially  
87 hydrophobic diesel particle from 80 nm to 100 nm implies that its volume has  
88 approximately doubled. The solute added by the condensed sulfate and organics will  
89 completely dominate the ~1% co-emitted sulfate of the initial soot particle. Put another  
90 way, Köhler theory predicts that a particle with 1% sulfate would need to have a diameter

91 of ~350 nm in order to activate at supersaturations of ~0.2%. A negligible fraction of the  
92 emissions of interest occur at that size, and sulfate added to soot particles by atmospheric  
93 processing (condensation) is critical to their CCN activity.

94 For a soot particle to function as a CCN: (1) A soot particle must become  
95 internally mixed with other aerosol components (e.g. sulfate and organics), and this is  
96 assumed to occur with a 1.5 day aging timescale. Once internally mixed with other,  
97 soluble aerosol components, the particle may or may not be a CCN depending on its size  
98 and the overall chemical composition of that size bin in that model grid cell; and (2) Once  
99 “internally mixed”, the typical soot particle will still be quite small (e.g. 20-60 nm) and  
100 therefore will not activate regardless of its composition. The particles then have to grow  
101 by condensation to the sizes (~100 nm diameter) where they will activate and function as  
102 CCN [Adams and Seinfeld, 2002; Pierce and Adams, 2009]. Under essentially all  
103 reasonable atmospheric conditions, the second step is the limiting one for a soot particle  
104 to become a CCN. In the BASE simulation, ten bins span the size range from 10 nm to 1  
105  $\mu\text{m}$  dry diameter with an additional two size bins in the coarse mode. Mineral dust is  
106 included according to the treatment of Lee *et al.* [2009], and biomass burning emissions  
107 are based on the GFED inventory [van der Werf *et al.*, 2004].

108 BC particles can act as ice nuclei [Phillips *et al.*, 2008], impacting the  
109 microphysical state of clouds at temperatures below the freezing point of liquid water. In  
110 mixed-phase clouds, decreases in BC ice nuclei concentrations may reduce the glaciation  
111 frequency of supercooled clouds, which can decrease precipitation and enhance  
112 shortwave cloud albedo and longwave emissivity [Lohmann and Feichter, 2005]. BC  
113 reduction in cold cirrus clouds may enhance crystal number concentrations, especially if

114 both heterogeneous and homogeneous freezing are actively competing for water vapor  
115 [*Barahona and Nenes, 2009ab*]. Ice crystal concentration enhancements will increase the  
116 longwave emissivity of cirrus [*Karcher et al, 2007*]. These ice nuclei effects on climate  
117 are not considered in this study.

118 The scenarios considered consist of three cases (Table 1):

119 BASE: present-day conditions; primary emissions from fossil fuel combustion are  
120 assumed to follow a lognormal size distribution with number median diameter =  
121 25 nm and geometric standard deviation = 2.0; biofuel and biomass burning  
122 emissions have number median diameter = 100 nm and geometric standard  
123 deviation = 2.0. (To represent coagulative scavenging of fresh emissions on  
124 subgrid scales, the fresh primary size distributions undergo 10 h of subgrid  
125 processing before being added to GCM grid cells [*Pierce et al., 2010*]).

126 HF: “half fossil fuel”; primary BC/OC mass and number emissions from fossil fuel  
127 combustion are each reduced by 50%.

128 HC: “half carbonaceous”; primary BC/OC mass and number emissions from all  
129 primary carbonaceous sources (fossil fuel, domestic biofuel, and biomass  
130 burning) are reduced by 50%; this scenario represents deeper reductions than HF;  
131 note that emissions reductions in this scenario occur mostly at particle sizes larger  
132 than those of HF.

133 Table 1 gives the annual mean aerosol budgets for the BASE, HF, and HC  
134 scenarios. In the HF scenario, BC mass emissions are reduced by ~20%. Since the fossil  
135 fuel emissions inventory is predominantly BC, with only small amounts of OC, the total  
136 emissions of OC in HF decrease by only about 4%. Global OC emissions are dominated

137 by biomass burning and domestic biofuel combustion. The change in particle number  
138 emitted in scenario HF is also relatively high (~23%) because the fossil fuel particles are  
139 relatively small (~25 nm). In contrast, the HC scenario has (by definition) a 50%  
140 reduction in primary BC and OC mass emissions. Although this represents much deeper  
141 cuts in BC/OC mass than the HF scenario, primary number emissions are reduced only  
142 modestly further (26% in HC vs. 23% in HF) because the biomass burning emissions  
143 involve larger particles (~100 nm). The fact that only a 26% reduction in total primary  
144 particle number occurs, even in the HC scenario, is a reflection of the fact that the global  
145 aerosol number concentration comprises many non-carbonaceous particles. Mineral dust  
146 is a small contribution. Ultrafine sea salt is a somewhat larger contribution, and primary  
147 sulfate is a large contributor since its assumed size distribution peaks ~20 nm.

148         The sources of global aerosol number concentration (for particles larger than 10  
149 nm) are given in Table 2. Note that in Table 2, “nucleation” refers to the number of  
150 particles that grow to 10 nm diameter and are, therefore, tracked explicitly in the model.  
151 The formation rate of critical clusters (~1 nm diameter) is much faster. Biomass and  
152 biofuel combustion are ~6% of the total source of aerosol number. Natural particles (sea-  
153 salt plus mineral dust) are ~2% of the total source of aerosol number, a value that  
154 includes a substantial contribution from ultrafine sea-salt emissions. The fraction of  
155 biomass burning that is natural is uncertain, but generally thought to be small (~10%), so  
156 the ~2% does not include any contribution from biomass burning.

157         Generally, only a small fraction of primary combustion particles is smaller than  
158 10 nm. Typically, atmospheric particles smaller than 10 nm result from homogeneous  
159 nucleation. Particles smaller than 10 nm, however, are extremely short-lived in the

160 atmosphere (see, for example, *Zhang et al.* [2004]). For purposes of CCN calculations,  
161 neglect of particles smaller than 10 nm has only a minor impact. The probability of these  
162 particles growing to be CCN is very low since they undergo efficient coagulation  
163 scavenging in the atmosphere [*Pierce and Adams*, 2007] and even much larger changes in  
164 the nucleation rate of particles smaller than 10 nm has a relatively minor impact on global  
165 CCN [*Pierce and Adams*, 2009]. Finally, for the purposes of emissions book-keeping in  
166 Table 1, the 10 nm cutoff is somewhat arbitrary. It represents, however, a reasonable  
167 balance between including most primary particles and excluding those that are too short-  
168 lived to influence CCN.

169 Particle emissions from fossil fuel sources are sometimes reported as bimodal,  
170 where the smallest mode contains organic/sulfate particles and the second mode contains  
171 black carbon/organic/sulfate particles. Here we have assumed a single mode. We  
172 performed sensitivity simulations to investigate the effect of this assumption. Comparing  
173 the two cases, the resulting change in global cloud drop number concentration between  
174 unimodal to bimodal cases is ~2%.

175 Global average present-day AIE forcing at top-of-the-atmosphere (TOA) for the  
176 two scenarios relative to the BASE case are:  $+0.13 \pm 0.33 \text{ W m}^{-2}$  (HF) and  $+0.31 \pm 0.33$   
177  $\text{W m}^{-2}$  (HC). The magnitude of these (positive) forcings can be compared to the best  
178 guess IPCC [*Forster et al.*, 2007] value of pre-industrial minus present-day AIE forcing  
179 from the cloud albedo effect of  $-0.7 \text{ W m}^{-2}$ . The standard deviations associated with the  
180 predicted values for the two scenarios are a result of the inter-annual variability of cloud  
181 and precipitation. Reduction of BC leads also to decreased heating of the atmosphere via  
182 shortwave absorption. The TOA aerosol direct forcing (assuming an internal aerosol



183 mixture) associated with the two scenarios is:  $-0.07 \text{ W m}^{-2}$  (HF) and  $-0.12 \text{ W m}^{-2}$  (HC).  
184 These direct aerosol forcings due to BC control can be compared with the IPCC  
185 estimated present-day minus pre-industrial direct radiative forcings for fossil fuel and  
186 total BC/OC of  $+0.15$  and  $+0.18 \text{ W m}^{-2}$ , respectively. Noting that direct forcing is  
187 calculated by diagnosing the change in TOA radiative fluxes in the absence of feedbacks,  
188 whereas indirect forcing is calculated from the perturbation in cloud forcing (all-sky  
189 minus clear-sky fluxes) allowing for feedbacks in cloud water and precipitation, we can  
190 estimate the net climatic effect of the two scenarios as:  $+0.06 \text{ W m}^{-2}$  (HF) and  $+0.19 \text{ W}$   
191  $\text{m}^{-2}$  (HC).

### 192 **3. Emissions Scenarios**

193 In the HF and HC emissions scenarios examined here both the mass and number  
194 of emitted carbonaceous particles are reduced from their respective sources by 50%.  
195 These scenarios also assume that the size distribution of emitted particles remains  
196 invariant. In reality, the change in the size and number of emitted particles will depend on  
197 the precise controls adopted [*Kittelson, 1998*]. Recent measurements on a suite of PM  
198 control technologies for heavy-duty diesel vehicles found that number emission factors  
199 ( $\text{particles km}^{-1}$ ) could increase approximately one order of magnitude or decrease by  $\sim 3$   
200 orders of magnitude depending on the control technology in question [*Biswas et al.,*  
201 2008].

202 The relatively simple scenarios considered here can be considered a reasonable  
203 starting point for analysis. First, some control strategies (e.g. replacement of domestic  
204 biofuel burning by a much cleaner alternative) lead to proportional reductions in both  
205 mass and number emissions. Second, concerns over potential health impacts of ultrafine

206 particles may lead to a preference for control technologies that reduce both mass and  
207 number. Third, measurements made in a tunnel near San Francisco in 1997 and 2006  
208 indicate that number emission factors for both light-duty vehicles and diesel trucks have  
209 decreased over that time period by a proportion similar to, but somewhat less than, the  
210 mass emission factor decrease [*Ban-Weiss et al.*, 2010], although these data do not yet  
211 reflect widespread deployment of diesel particulate filters. Nevertheless, one cannot rule  
212 out an increase in number emissions if BC controls are implemented; this would likely  
213 lead to an enhancement of the aerosol indirect effect, precisely the opposite of the result  
214 obtained here.

#### 215 **4. Conclusion**

216 Black carbon emission control is recognized to be an effective strategy for  
217 mitigation of both global warming and air pollution. If BC mass emission reduction is  
218 reflected in an alteration of both the global CCN concentration and cloud droplet number  
219 concentration, such a reduction can lead potentially to a change in indirect aerosol  
220 radiative forcing. We have evaluated the change in aerosol indirect forcing under  
221 present-day conditions for two scenarios of BC control: (1) 50% reduction in fossil fuel  
222 emissions of BC/OC; and (2) 50% reduction in BC/OC emissions from all primary  
223 carbonaceous aerosol sources. The estimated net changes in TOA cloud radiative forcing  
224 corresponding to the two scenarios are  $+0.13$  and  $+0.31 \text{ W m}^{-2}$ , respectively, that is, a  
225 reduction in global cloud radiative forcing. Because of the importance of both BC control  
226 and of understanding aerosol indirect effects, additional studies of the interplay between  
227 these two issues are warranted.

228

229 **Acknowledgment**

230 This work was supported by the U.S. Environmental Protection Agency under Science To

231 Achieve Results (STAR) grant RD833370.

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327 Table 1. Scenarios and Results  
 328

Scenario		Emission <sup>a</sup>	Source <sup>b</sup>	Aerosol Burden <sup>c</sup>	Lifetime (days)	ADE forcing (W m <sup>-2</sup> ) <sup>d</sup>	$N_c$ in surface (cm <sup>-3</sup> ) <sup>c</sup>	Cloud AIE forcing (W m <sup>-2</sup> ) <sup>e</sup>
BASE	BC	7.7 Tg yr <sup>-1</sup>	7.7 Tg yr <sup>-1</sup>	0.17 Tg	7.8	----	196	----
	OC	60.6 Tg yr <sup>-1</sup>	79.7 Tg yr <sup>-1</sup>	1.1 Tg	5.2			
	$N_a$	$2.26 \times 10^4$ cm <sup>-3</sup> yr <sup>-1</sup>	$2.79 \times 10^4$ cm <sup>-3</sup> yr <sup>-1</sup>	$2.83 \times 10^2$ cm <sup>-3</sup>	5.3			
HF	$\Delta$ BC	-19.7%	-19.7%	-21.4%	-0.1	-0.07±0.0017 (int.)	187 (-5.9%)	+0.13±0.33
	$\Delta$ OC	-3.6%	-2.7%	-3.0%	0	-0.03±0.0002 (ext.)		
	$\Delta N_a$	-22.5%	-17.3%	-4.2%	+0.5			
HC	$\Delta$ BC	-50.0%	-50.0%	-49.7%	+0.1	-0.12±0.0017 (int.)	179 (-9.8%)	+0.31±0.33
	$\Delta$ OC	-50.0%	-38.0%	-35.8%	+0.2	-0.04±0.0008 (ext.)		
	$\Delta N_a$	-26.2%	-20.0%	-4.9%	+0.7			

329

330<sup>a</sup> The change in primary particles listed is for *all* primary particles. Primary sulfate (from  
 331 in-plume nucleation processes), sea salt, and mineral dust are unchanged in each of the  
 332 scenarios. Fossil fuel emitted particles are ~ 45% by number of total primary particle  
 333 emissions, so that a 50% reduction in that amount leads to a ~ 23% reduction by number  
 334 in total particle emissions.

335<sup>b</sup> The difference between the “Source” and “Emission” columns represents secondary  
 336 production: SOA in the case of OC and nucleation in the case of particle number. The  
 337 nucleation source is defined as  $J_{10}$ , the number flux of nucleated particles that grow past  
 338 10 nm diameter.

339<sup>c</sup> In the BASE simulation, emissions from fossil fuel and biofuel sources are based on the  
340 emissions inventory of *Bond et al.* [2004], and biomass burning emissions are from the  
341 GFED inventory. Offline, monthly-averaged concentrations of ammonium sulfate,  
342 ammonium nitrate, OC, BC, and sea salt aerosols are derived from the Two-Moment  
343 Aerosol Sectional microphysics model (TOMAS [*Pierce and Adams*, 2009]). The  
344 aerosol activation parameterization of *Nenes and Seinfeld* [2003] and *Fountoukis and*  
345 *Nenes* [2005] is used to calculate corresponding monthly average  $N_c$  fields from a  
346 detailed global aerosol model [*Lee et al.*, 2009]. Updraft velocities representative of  
347 stratiform clouds ( $0.15 \text{ m s}^{-1}$  over ocean and  $0.3 \text{ m s}^{-1}$  over land) are assumed.

348<sup>d</sup> Direct aerosol forcing is determined by the instantaneous change in annual mean TOA  
349 net (shortwave plus longwave) radiative fluxes using GISS-III [*Chen et al.*, 2010]. Each  
350 simulation is integrated for two years with prescribed monthly mean sea-surface  
351 temperature (SST) from HadISST1 observed climatology for 1992 to 2000 [*Rayner et*  
352 *al.*, 2003]. Radiative forcings for internal (int.) and external (ext.) aerosol mixtures are  
353 derived separately. A standard gamma size distribution is assumed for the aerosol with a  
354 surface area-weighted dry radius ( $0.1 \mu\text{m}$  for pure BC in external mixture;  $0.3 \mu\text{m}$  for all  
355 other species and the internal mixture) and variance ( $= 0.2$ ). The wavelength-dependent  
356 refractive indices of dry sulfate, nitrate (assumed the same as that of sulfate), and sea salt  
357 are taken from *Toon et al.* [1976], with those for organic carbon, BC, and water from  
358 *d'Almeida et al.* [1991]. Extinction efficiency, single-scattering albedo, and asymmetry  
359 parameter are then supplied to the radiation scheme of the GISS III [*Schmidt et al.*,  
360 2006; *Rind et al.*, 2007] to calculate aerosol optical depth and radiation fluxes. For an

361 internal mixture, the density of the aqueous aerosol mixture is computed as the mass-  
362 averaged density of water and dry aerosols, and the composite aerosol radiative  
363 properties are derived based on a homogeneous volume-weighted mixing rule. For  
364 external mixing, the Mie calculation is applied for individual species of dry aerosols and  
365 aerosol water.

366<sup>e</sup> To derive the AIE forcing, modifications have been made by *Chen et al.* [2010] to the  
367 formulations of optical depth and autoconversion rates in liquid-phase stratiform clouds  
368 in GISS III to introduce explicit dependence on  $N_c$  fields. Following the common  
369 measure of AIE forcing [*Forster et al.*, 2007], the cloud properties and hydrological  
370 cycle are allowed to respond accordingly, and the AIE forcing is determined by the  
371 change in annual mean TOA net (shortwave plus longwave) cloud forcing (all-sky  
372 minus clear-sky radiative fluxes) between each set of simulations. Each simulation is  
373 integrated for 20 years with prescribed SST and specific levels of offline, monthly-  
374 averaged  $N_c$  values from the TOMAS model. With the first five years as a spin-up  
375 period, the average AIE forcing over the last 15 years is reported.

376 Table 2. Global Sources of Aerosol Number Concentration (Particles larger than 10 nm)  
 377

Process	Rate (particles cm <sup>-3</sup> yr <sup>-1</sup> )
“Primary” (plume nucleation) sulfate <sup>a</sup>	1.02 x 10 <sup>4</sup>
Sea-salt emissions	5.4 x 10 <sup>2</sup>
EC/OC from fossil fuel <sup>a</sup>	1.02 x 10 <sup>4</sup>
EC/OC from biofuel/biomass	1.66 x 10 <sup>3</sup>
Mineral dust emissions	2.08 x 10 <sup>1</sup>
Nucleation ( <i>J</i> <sub>10</sub> )	5.24 x 10 <sup>3</sup>

378

379 <sup>a</sup> That the magnitudes of these two rates are the same is coincidental. The sulfate mass  
 380 emission rate is ~ 2.1 Tg sulfate yr<sup>-1</sup>, and the EC/OC fuel emission rate is ~ 7.2 Tg yr<sup>-1</sup>, a  
 381 ratio of 0.29. The number emission rate per kg of primary sulfate is 3.57 times that per kg  
 382 of EC/OC fossil fuel. Then, 0.29 x 3.27 ~ 1.