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 ± 0.33 W m^{-2} (HF) and + 0.31 ± 0.33 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_2SO_4-H_2O nucleation,

78   accounted for using the model of *Vehkamaki 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 μm 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 coagulational 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 coagulational

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: $+ 0.13 \pm 0.33$ W m^{-2} (HF) and $+ 0.31 \pm 0.33$

177 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 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 W m^{-2} (HF) and -0.12 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 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 W m^{-2} (HF) and +0.19 W

191   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   (particles km^{-1}) could increase approximately one order of magnitude or decrease by ~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 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**

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Table 1. Scenarios and Results

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

329

330 [a] 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 [b] 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[c]   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 m s^{-1} over ocean and 0.3 m s^{-1} over land) are assumed.

348[d]   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 μm for pure BC in external mixture; 0.3 μm 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[e]   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^{-3} yr^{-1})
"Primary" (plume nucleation) sulfate [a]	1.02×10^4
Sea-salt emissions	5.4×10^2
EC/OC from fossil fuel [a]	1.02×10^4
EC/OC from biofuel/biomass	1.66×10^3
Mineral dust emissions	2.08×10^1
Nucleation (J_{10})	5.24×10^3

378

379 [a] That the magnitudes of these two rates are the same is coincidental. The sulfate mass

380 emission rate is ~ 2.1 Tg sulfate yr^{-1}, and the EC/OC fuel emission rate is ~ 7.2 Tg yr^{-1}, 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.