

Strong Direct Forcing by Black Carbon Inferred from the Global Simulation of Its Mixing State

Nature paper

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Supplemental Information

Description and Setup of the Model

The model used was GATOR-GCMM¹⁻³ (Gas, Aerosol, Transport, Radiation, General Circulation, and Mesoscale Model). The model treated gas, aerosol, radiative, meteorological, and transport processes. It resolved subgrid-scale soil classes, water, ice, snow, and urban surfaces for ground-temperature calculations. Radiative processes included photolysis (80 wavelengths) and heating rate calculations (409 wavelengths). The model transported gases and aerosols given winds predicted by the online meteorology. Meteorology was driven by heating rates that accounted for optical properties of gases and size-resolved aerosols, cloud liquid, and cloud ice. The resolution of the global domain was 4° S-N x 5° W-E with 22 sigma-pressure layers from the ground to 0.425 mb (\approx 55 km).

Gas processes included emissions, gas chemistry, dry deposition, transport, homogeneous nucleation, condensational growth, dissolutional growth, and rainout. Size- and composition-resolved aerosol processes included emissions, homogeneous nucleation,

condensation, dissolution, coagulation, chemical equilibrium, transport, sedimentation, dry deposition, and rainout.

Eighty-eight gases, 18 aerosol size distributions, 17 size bins per distribution, and an average of 6.778 components per bin per distribution were treated in each of 69,696 grid cells. Thus, a total of 2468 components [88 gases + 17 sizes x 18 distributions x (1 number concentration + 6.778 mole concentrations)] were tracked in each cell.

The 18 aerosol size distributions (Table 1) consisted of four "primary" distributions [sea spray (A), soil (B), black carbon (E1), and organic matter (F)] into which emissions occurred, one "primary" distribution [sulfate (D)] into which homogeneous nucleation occurred, two additional black carbon distributions (E2 and E3) into which primary BC grew, 10 "binary" distributions (AB, AD, AE, AF, BD, BE, BF, DE, DF, EF) that resulted from heterocoagulation among A, B, D, E1, E2, E3, and F distributions, and a completely-mixed distribution (MX) that resulted from all higher coagulation interactions. Table 2 describes the coagulation interactions among the 18 distributions.

When sulfuric acid gas was present, $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ heterogeneous nucleation was calculated^{11,12} to be sufficiently rapid over all particle types that it was necessary to assume that $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ condensation could occur on all types. Since ammonia, if present, usually dissolves in sulfuric-acid solutions, it was also necessary to treat ammonia dissolution. As such, each distribution contained room for H_2O , H^+ , NH_4^+ , HSO_4^- , and SO_4^{2-} in addition to its base constituents. Although this resulted in some distributions (e.g., A and AD), potentially containing the same components, new particle sources in each distribution differed (e.g., the source in A was emissions and that in AD was coagulation).

Since condensation during and after emissions coated emitted BC particles, it was necessary to create three BC distributions (E1...E3) with different percentages of coatings (0-5, 5-20, and >20 percent shell material by mass, respectively), so that new emissions of

BC always had a relatively pure distribution, E1, to enter. Initially, E1 contained only BC, whereas E2 and E3 contained no material. The only source of new E1 particles was emissions, and those of new E2 and E3 particles were coagulation and growth from E1 and/or E2. If the shell mass of an E1 bin rose above 5 percent, for example, all material from the bin was moved to an E2 bin. If a limit were not placed on E1's shell fraction, condensation would soon cause its shell to dominate particle mass, preventing the realistic simulation of the radiative effects of newly-emitted, externally-mixed BC particles. No distribution aside from E1...E3 contained shell-fraction limits.

Additional species, such as nitrate and sea-spray ions aside from sodium, chloride, and sulfate, were neglected due to computational memory limitations. The inclusion of additional species will only enhance the rate of BC internal mixing, strengthening the conclusions found here.

Only externally-mixed sea spray, soil, sulfate, BC, and organic matter (OM) were initialized (Table 1). The initial tropospheric anthropogenic plus natural sulfate and ammonium loadings were 0.65-Tg S and 0.37 Tg-NH₄⁺, comparable with 0.7-Tg S and 0.39 Tg-NH₄⁺, respectively, from *Adams et al.*¹³. The initial BC loading (0.20 Tg) was within *Cooke et al.*'s¹⁴ range of 0.15-0.25 Tg. The initial OM loading was 1.3 Tg, giving an OM:BC ratio as 6.6, equivalent to that from *Lioussé et al.*¹⁰. The initial soil loading was 15 Tg, close to 16.4 Tg from *Andreae*¹⁵. Initial number concentrations over the poles were 5-100 cm⁻³, oceans were 100-700 cm⁻³, remote continental regions were 500-1500 cm⁻³, and rural regions were 1000-10,000 cm⁻³, all consistent with mean observations¹⁶⁻¹⁸. Number and mass concentrations varied in time in the model.

H₂SO₄-H₂O homogenous nucleation rates were calculated with classical theory^{11,12}. Homogeneous nucleation of H₂SO₄-H₂O into the ammonium sulfate distribution was solved simultaneously with condensation of H₂SO₄-H₂O between the gas

phase and all 18 distributions (and 17 bins per distribution). A mass-conserving, noniterative, and unconditionally stable scheme¹⁹ was used to solve the 308 condensation/nucleation equations and unknowns simultaneously. Allowing competition for available vapor between nucleation and growth allowed for vapor to be distributed among new and existing particles in a physical manner, reducing the impact of errors in predicted nucleation rates. If nucleation and growth were instead operator-split (solved separately), overpredictions of nucleation rates would frequently result in nearly all vapor nucleating to form new particles and little remaining to grow existing particles.

Sulfuric acid was oxidized in the gas phase from SO₂. Non-DMS emissions rates of SO₂(g) were obtained from *Voldner et al.*²⁰. Ocean emissions rates of DMS were calculated with the parameterization of *Liss and Merlivat*²¹, and sea water DMS concentrations were estimated from *Kettle et al.*²². Emissions rates of NH₃ were obtained from *Bouwman et al.*²³. Dissolution equations of ammonia gas into all size distributions and bins (307 equations and unknowns each cell) were solved simultaneously with a mass conserving, noniterative, and unconditionally stable dissolution scheme¹⁹.

Sea spray and spume drop emissions versus size were calculated with the parameterizations of *Monahan et al.*²⁴ and *Wu*²⁵, respectively. Soil emissions versus size were calculated from *Marticorena et al.*²⁶ using soil distribution data from *FAO*²⁷. Emissions rates of biomass-burning and fossil-fuel BC were obtained from *Cooke et al.*¹⁴ and applied over lognormal distributions for fossil fuels²⁸ and biomass burning²⁹. OM emissions rates from biomass burning were set to 7.9 times those of BC, per table 1 of *Liousse et al.*¹⁰ and those from fossil fuels were set to a factor of 3.1 times those of BC [an average OC:EC fossil-fuel ratio given by *Strader et al.*³⁰ is 2.4, and the OM:OC ratio is typically 1.3¹⁰].

The coagulation scheme was derived from an earlier volume-conserving, noniterative, coagulation scheme in which multiple size distributions were considered³¹. When external mixtures coagulated in the original scheme, they were sent into a single internally-mixed distribution. For this work, the scheme was modified to treat coagulation into and among any number of externally-mixed, binary, ternary, etc. combinations of distributions and any number of components in each distribution (maintaining volume conservation). Coagulation kernels included those for Brownian motion, Brownian diffusion enhancement, gravitational collection, turbulent inertial motion, and turbulent shear (e.g., Figure 16.4 and Equations 16.28-16.34 of ref. 32). Aerosol-aerosol coagulation and aerosol-raindrop coagulation in and below clouds were treated, but aerosol-cloud-drop coagulation was not. If treatment of aerosol-cloud-drop coagulation were included, it would increase the rate of internal mixing of aerosols²⁵, strengthening the conclusions found here.

Aerosol liquid water content in all bins and distributions was solved with EQUISOLV II⁵. Sedimentation and dry deposition equations are given in Chapter 20 of ref. 32. Aerosol rainout was calculated by coagulating each size bin of each aerosol distribution with each size bin of a discretized Marshall-Palmer rainfall distribution in each grid cell (within and below clouds) in which precipitation, predicted by the model, was occurring. The five coagulation kernels for rainout were the same as those for aerosol coagulation.

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Table 1. Components in and processes solved over each size distribution.

Distribution name	Symbol	Components	Initia- l- ized?	Sources of new particles	Depositi- on processes affecting	Other processes affecting
Sea spray	A	H ₂ O, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	Yes ^A	Em	Dd, Sd, Rn	Co, Gr, Eq, Tr
Soil	B	Soil, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	Yes ^B	Em	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sulfate	D	H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	Yes ^C	Nuc	Dd, Sd, Rn	Co, Gr, Eq, Tr
BC w/ 0-5% shell	E1	BC, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	Yes ^D	Em	Dd, Sd, Rn	Co, Gr, Eq, Tr
BC w/ 5-20% shell	E2	BC, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co,Gr from E1	Dd, Sd, Rn	Co, Gr, Eq, Tr
BC w/>20% shell	E3	BC, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co, Gr from E1,E2	Dd, Sd, Rn	Co, Gr, Eq, Tr
Organic matter (OM)	F	OM, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	Yes ^E	Em	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sea spray-soil	AB	H ₂ O, Soil, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sea spray-sulfate	AD	H ₂ O, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sea spray-BC	AE	H ₂ O, BC, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sea spray-OM	AF	H ₂ O, OM, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Soil-sulfate	BD	Soil, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Soil-BC	BE	Soil, BC, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Soil-OM	BF	Soil, OM, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sulfate-BC	DE	H ₂ O, BC, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Sulfate-OM	DF	H ₂ O, OM, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
BC-OM	EF	BC, OM, H ₂ O, H ⁺ , NH ₄ ⁺ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr
Internal mixture	MX	H ₂ O, Soil, BC, OM, H ⁺ , Na ⁺ , NH ₄ ⁺ , Cl ⁻ , HSO ₄ ⁻ , SO ₄ ²⁻	—	Co	Dd, Sd, Rn	Co, Gr, Eq, Tr

Em = emissions, Nuc = homogeneous nucleation, Co = coagulation, Gr = condensational growth of sulfuric-acid-water plus dissolutional growth of ammonia, Dd = dry deposition, Sd = sedimentation, Rn = rainout, Eq = internal-aerosol equilibrium chemistry plus water uptake by hydration, Tr = horizontal and vertical transport.

Each distribution contained 17 size bins. Condensation and homogeneous nucleation of H₂SO₄-H₂O were solved simultaneously among the gas phase and all bins of all distributions. Dissolution of NH₃ was solved simultaneously among the gas phase and all bins of all distributions. Differences in distributions A and AD were that the only source of new A particles was emissions, and that of new AC particles was coagulation of A with C, A with AC, or C with AC particles. The differences among distributions E1...E3 and DE were that the only source of new E1 particles was emissions, the only sources of E2 and E3 particles were self-coagulation and

growth from E1 or E2, and the only source of new DE particles was coagulation of E1, E2, or E3 with D or DE and coagulation of D with DE.

^ANear-surface dry sea spray mass and number were initialized with the sub- and supermicron data shown in Table 3 of ref. 8., derived from *Quinn et al.*⁴. Initial sea spray water content was obtained from these masses, relative humidities, and temperatures using EQUISOLV II⁵.

^BInitial near-surface and elevated soil mass was interpolated from *Dentener et al.*⁶ then distributed trimodally with lognormal parameters given in Table 2a of ref. 2.

^CInitial near-surface sulfate mass was interpolated from *Langner and Rodhe*⁷. Elevated sulfate was extracted from SAGE II satellite data⁸. Sulfate mass was distributed trimodally with lognormal parameters given in Table 2a of ref. 2. The initial $\text{NH}_4^+:\text{SO}_4^{2-}$ mole ratio was unity

^DExternally-mixed black carbon was divided into three distributions: those with 0-5 (E1), 5-20 (E2), and 20-99.99 (E3) percent shell material by mass, respectively. Only distribution E1 was initialized (with zero percent shell material). The initial near-surface and elevated BC mass from fossil fuels and biomass burning was interpolated from *Cooke and Wilson*⁹, then distributed trimodally with lognormal parameters given in Table 2a of ref. 2. The initial BC mass was scaled to 0.20 Tg. BC density was assumed to be 1.5 g cm^{-3} .

^EThe initial near-surface and elevated OM mass from fossil fuels, biomass burning, and natural sources was interpolated from *Liousse et al.*¹⁰.

Table 2. Coagulation interactions.

Size distribution name/symbol	Symbol of second distribution																	
	A	B	D	E1	E2	E3	F	AB	AD	AE	AF	BD	BE	BF	DE	DF	EF	MX
Sea spray	A	AB	AD	AE	AE	AE	AF	AB	AD	AE	AF	MX	MX	MX	MX	MX	MX	MX
Soil	B	AB	BD	BE	BE	BE	BF	AB	MX	MX	MX	BD	BE	BF	MX	MX	MX	MX
Sulfate	D	AD	BD	DE	DE	DE	DF	MX	AD	MX	MX	BD	MX	MX	DE	DF	MX	MX
BC w/ 0-5% shell	E1	AE	BE	E1	E2	E3	EF	MX	MX	AE	MX	MX	BE	MX	DE	MX	EF	MX
BC w/ 5-20% shell	E2	AE	BE	E2	E2	E3	EF	MX	MX	AE	MX	MX	BE	MX	DE	MX	EF	MX
BC w/>20% shell	E3	AE	BE	E3	E3	E3	EF	MX	MX	AE	MX	MX	BE	MX	DE	MX	EF	MX
Organic matter (OM)	F	AF	BF	DF	EF	EF	F	MX	MX	MX	AF	MX	MX	BF	MX	DF	EF	MX
Sea spray-soil	AB	AB	MX	MX	MX	MX	MX	AB	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX
Sea spray-sulfate	AD	AD	MX	MX	MX	MX	MX	MX	AD	MX	MX	MX	MX	MX	MX	MX	MX	MX
Sea spray-BC	AE	AE	MX	MX	MX	MX	MX	MX	MX	AE	MX	MX	MX	MX	MX	MX	MX	MX
Sea spray-OM	AF	AF	MX	MX	MX	MX	AF	MX	MX	MX	AF	MX	MX	MX	MX	MX	MX	MX
Soil-AS	BD	MX	BD	MX	MX	MX	MX	MX	MX	MX	MX	BD	MX	MX	MX	MX	MX	MX
Soil-BC	BE	MX	BE	MX	BE	BE	BE	MX	MX	MX	MX	MX	BE	MX	MX	MX	MX	MX
Soil-OM	BF	MX	BF	MX	MX	MX	BF	MX	MX	MX	MX	MX	MX	BF	MX	MX	MX	MX
Sulfate-BC	DE	MX	MX	DE	DE	DE	MX	MX	MX	MX	MX	MX	MX	MX	DE	MX	MX	MX
Sulfate-OM	DF	MX	MX	DF	MX	MX	DF	MX	MX	MX	MX	MX	MX	MX	MX	DF	MX	MX
BC-OM	EF	MX	MX	MX	EF	EF	EF	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	EF
Internal mixture	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX	MX

The table gives the symbol of the size distribution into which two particles from either the same or different distributions coagulate to in the model. For example, when a sea spray particle (A) coagulates with a soil particle (B), the resulting particle is a sea spray-soil (AB) particle. The internally mixed distribution (MX) contains all components in externally-mixed (A...F) and binary (AB...EF) distributions. Thus, the combination of black carbon (E) with sea spray-soil (AB) gives a mixed particle (MX).