A physically-based treatment of elemental carbon optics: Implications for global direct forcing of aerosols

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Abstract. To date, global models of direct radiative forcing have treated elemental carbon (EC) as completely externally mixed or well-mixed internally. No global study has treated EC as a core in an internal mixture. It is hypothesized that the well-mixed treatment is unphysical and reality lies between the externally-mixed and core treatments. It is also suggested, but not proven, that most EC particles are coated to some degree; hence, the core treatment may be more representative than the external-mixture treatment. Global simulations with the core treatment resulted in EC forcing 50% higher and 40% lower than forcings obtained with the externally-mixed and well-internally-mixed treatments, respectively. In the core case, EC's positive forcing more than offset negative forcing due to all other anthropogenic aerosol components combined. Further studies are needed to understand the mixing state of EC and determine the accuracy of the core treatment.

1. Introduction

Since the early 1970s, anthropogenic aerosols have been thought to offset a fair portion of anthropogenic greenhouse gas warming. In the early 1980s, models estimated that anthropogenic aerosols offset more than half the warming. By 1995, the anthropogenic aerosol direct forcing[1] estimate was reduced to -0.5 (-0.25 to -1.0) W m^{-2} (IPCC, 1995) and consisted of forcing due to anthropogenic sulfate [-0.4 (-0.2 to -0.8) W m^{-2}], elemental carbon (EC) from fossil fuels [+0.1 (+0.03 to 0.3) W m^{-2}], and biomass-burning aerosols [-0.2 (-0.07 to -0.6) W m^{-2}]. The EC values were primarily from Haywood and Shine (1995) who calculated clear-sky forcing due to fossil-fuel EC of +0.03 to +0.24 W m^{-2}. The lower and upper estimates were for EC externally mixed from other particle components and EC well-mixed internally with other components, respectively.

Since then, Haywood et al. (1997) estimated forcing due to fossil-fuel EC of +0.20 to +0.36 W m^{-2} for an external mixture and well-mixed internal mixture, respectively, Haywood and Ramaswamy (1998) estimated forcing due to externally-mixed biomass-burning and fossil-fuel EC of +0.40 W m^{-2}, Myhre et al. (1998) estimated forcing due to EC from fossil fuels as +0.16 W m^{-2} for an external mixture and +0.42 W m^{-2} for an internal mixture, and Penner et al. (1998) estimated forcing from externally-mixed fossil-fuel EC as +0.20 W m^{-2}.

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1 Direct radiative forcing is the difference in the net downward (+ is downward) irradiance (W m^{-2}) over a set of wavelengths when a substance is present compared with when it is absent or less concentrated. A positive forcing indicates that the substance initially forces an increase in temperature before a feedback can occur.

2. Optical Treatments of EC

To date, all direct forcing estimates have been based on externally mixed or well-internally-mixed treatments (refractive indices volume averaged) of EC. No other treatment has been used in a global three-dimensional (3-D) model. Of the two treatments, the second appears more realistic, since the only source of EC is emissions from fossil-fuel combustion and biomass burning in the form of soot, and soot contains a mixture of EC, OC, and small amounts of O, N, and H (Chang et al., 1982). Soot emitted from engines contains EC covered with a layer of polycyclic aromatic hydrocarbons (PAHs) under a shell of volatile compounds (Steiner et al., 1992). Soot from biomass-burning contains EC, aliphatic hydrocarbons, and PAHs (Reid and Hobbs, 1998; Fang et al., 1999). Globally, about 54-57% of EC originates from fossil-fuel combustion, and the rest originates from biomass burning (Cooke and Wilson, 1996; Liousse et al., 1996).

Once emitted, soot particles can coagulate and/or grow by condensation. Although EC is hydrophobic, hygroscopic organics in soot increase its ability to attract water and inorganics (Andrews and Larson, 1993). Evidence of condensation and coagulation is abundant, since tunnel studies indicate that 85% of fossil-fuel EC is emitted in particles <0.12 μm in diameter (Venkataraman et al., 1994; Berner et al., 1996), but ambient measurements in Los Angeles, the Grand Canyon, Glen Canyon, Vienna, and the North Sea, show that accumulation-mode EC often exceeds emissions-mode EC (McMurry and Zhang, 1989; Hitzenberger and Puxbaum, 1993; Venkataraman and Friedlander, 1994; Berner et al., 1996). The only way ambient EC can redistribute so dramatically is if EC coagulates and/or grows. Whereas some EC self-coagulation occurs, it cannot account for EC growth from the emissions mode to the upper accumulation or coarse modes, as seen in Figures 2b and 4b of Jacobson (1997a). Heterocoagulation of EC with particles in these modes and condensation can account for such growth (same figures).

Similarly, the measured mean number diameter of biomass-burning smoke less than 4 minutes old is 0.10-0.13 μm (Reid and Hobbs, 1998), yet the mass of such aerosols increases by 20-40% during aging, with a third to half the growth occurring within hours after emissions (Reid et al., 1998). Young smoke contains not only EC and OC, but also K$^+$, Ca^{2+}, Mg^{2+}, Na$^+$, NH$_4^+$, Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ (Reid et al., 1998; Ferek et al., 1998; Andreae et al., 1998). Murphy et al. (1998) found that almost all aerosols >0.13 μm in the boundary layer in the remote Southern Pacific Ocean contained sea salt, indicating that externally-mixed particles were rare. Based on single-particle measurements in the same region and over the North Atlantic, Pósfai et al. (1999) found that "internally mixed soot and sulfate appear to comprise a globally significant fraction of aerosols in the troposphere." Almost all soot particles found in the North Atlantic contained sulfate.

Transmission electron microscopy (TEM) images support the theory that EC particles can become coated once emitted. Katrlnak et al. (1992, 1993) show TEM images of soot from fossil-fuel sources with a sulfates or nitrate coating. Martins et al. (1998) show a TEM image of a coated biomass-burning EC particle. Pósfai et al. (1999) show TEM images of North Atlantic soot particles containing ammonium sulfate.

Although EC may be internally-mixed with other components in a particle, EC cannot be "well-mixed" (diluted) in the particle, since soot, which contains EC, is irregularly-shaped and mostly solid, containing from 30-2000 graphitic spherules aggregated by collision during combustion (Katrlnak et al., 1993). Thus, EC must be a distinct and not a well-mixed component of a mixed particle. Katrlnak et al. (1992) found that coated aggregates have carbon structures similar to uncoated aggregates; thus, individual spherules in EC structures do not readily break off during coating. In cloud drops, several soot particles as a whole may be dispersed throughout the drop (Chylek et al., 1984).

Three treatments of EC optics are available in 3-D models -- the externally-mixed, well-internally-mixed, and core treatments. Of these, the core treatment covers the largest range of particle configurations. Figure 1 shows that external mixtures can be treated with the core method, but the reverse is not true. The core method treats externally-mixed EC by reducing the shell volume of one particle to zero and adding a second particle, consisting of the shell, but with no core. By expanding the number of size bins and allowing bins of the same size to have different composition (Figure 1 of Jacobson et al., 1994), the core treatment can cover any combination of mixtures. Neither of the other two treatments has this ability.

Other treatments of EC are available, but none has been applied to a 3-D model. These include treating a composite of several whole EC particles distributed in an internal mixture (applicable to cloud drops) (Chylek et al., 1988), embedding EC inclusions at random locations in a particle (Chylek et al., 1995), treating EC as a subsurface inclusion or grain on a particle (Fuller et al., 1999), and treating EC as a coated chain aggregate (Fuller et al., 1999). The last treatment appears more realistic than the core treatment, but since real soot take on so many possible shapes, it is unclear whether even a few representations of chain aggregates can result in better absorption coefficient predictions, averaged over all particles, than the core treatment. The TEM images discussed show that when EC is coated, it is enveloped by the coating, indicating that the core treatment is a plausible physical representation.
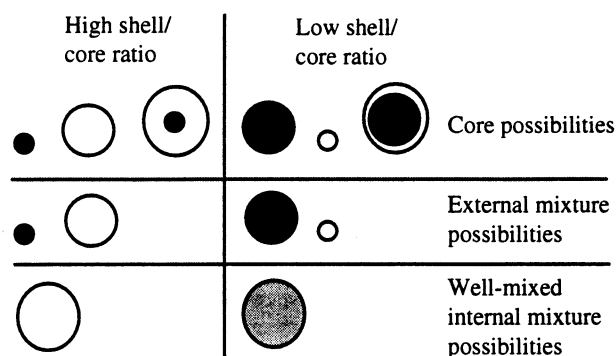

Figure 1. Possible core, external mixture, and internal mixture configurations. The core treatment encompasses all external mixture treatments. The well-mixed internal mixture treatment is unphysical, since EC cannot be diluted through a particle, except in the limiting external-mixture case.
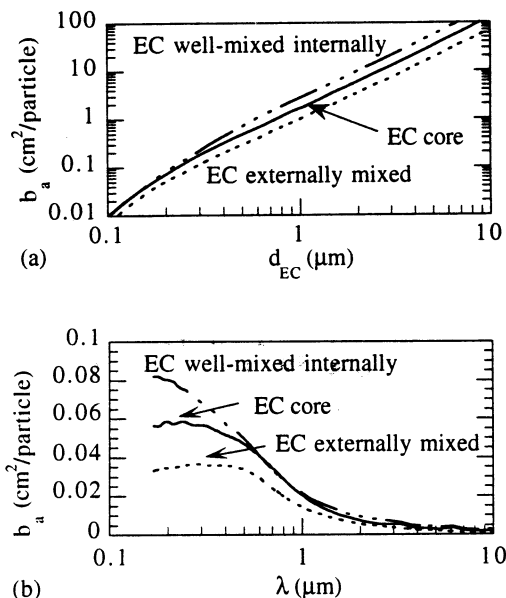

Figure 2. Total particle absorption cross section ($b_a = Q_a \pi d_p^2/4$, where Q_a = single-particle absorption efficiency and d_p = total particle diameter) as a function of (a) effective EC diameter (d_{EC}) when the wavelength (λ) = 0.5 μm and (b) λ when d_{EC} = 0.169 μm for three treatments of EC. In the well-mixed and core cases, EC and liquid water coexist, the volume fraction of EC is 21.6%, and d_{EC}=0.6d_p. In the externally-mixed case, the volume fraction of EC is 100% and d_{EC}=d_p. The Mie code used originated from Toon and Ackerman (1981).

With respect to the core treatment, Toon and Ackerman (1981) developed a stable code to treat light scattering and absorption by concentrically-coated spheres. Ackerman and Toon (1981) applied the code to study the single-scattering albedos of mixtures of absorbing and nonabsorbing particles, including a soot core and a sulfate shell. Jacobson (1997a,b; 1999a) performed the first 3-D simulations using the core treatment to predict, among other parameters, absorption coefficients measured in the Los Angeles basin. Results were obtained by assuming EC was core and all other aerosol components were well-mixed in a shell. Figure 9 of Jacobson (1997a) shows an absorption-extinction prediction error of 9.5% in a box model calculation constrained by an observed size distribution. Table 5 of Jacobson (1999a) shows an absorption-extinction gross error of 10.6% and bias of +4.2%, normalized over eight locations. Figures 68-70 of Jacobson (1997b) show two days of predictions versus observations of absorption extinction from a 3-D simulation.

Simulations in which EC was externally mixed were also performed but not reported. This assumption resulted in normalized gross errors and biases of >50% and <-50%, respectively, indicating significant underpredictions versus observations. When EC was treated with volume-averaged refractive indices in an internal mixture, normalized gross errors and biases were nearly 25% and +25%, respectively, indicating overpredictions. The reasons for differences relative to the core case can be elucidated from Figure 2.

Figure 2a shows that at λ=0.5 μm, absorption efficiencies for particles containing a water shell and an EC core are greater than those for EC particles externally mixed from water particles and less than those for well-mixed EC-water particles. Similar results are seen in Figure 2b, but for one particle size and a range of wavelengths. When refractive indices are volume averaged, the entire particle is assumed to absorb weakly, and all waves passing through the particle

have potential to be absorbed. When EC is core, only waves scattered into the core can be absorbed, resulting in a smaller net absorption efficiency than in the well-mixed case. When EC is externally mixed in air, it is exposed to fewer waves than when in water, since (1) in the case of particles larger than the wavelength, geometric optics implies that n^2 more light is incident on a small sphere when it is at the center of a much larger transparent sphere with real refractive index n than when it is in air (optical focusing effect) (Bohren, 1986; Twohy et al., 1989); (2) in the case of particles near or smaller than the wavelength, enhanced diffraction at the edge of a particle increases the exposure of core material to waves in comparison with exposure of the material to waves in air.

The core treatment results not only in lower absorption coefficients, but also (often) in lower scattering coefficients optical depths, and lower single scattering albedos (ω_0) than the well-mixed treatment. For this reason, ω_0 is misleading, since its low value in the core case relative to the well-mixed case suggests that more absorption occurs in the core case. In fact, less absorption occurs in the core case but so does less scattering, resulting in the lower ω_0.

Results from Los Angeles may be relevant for other regions where EC concentrations are moderate to high. Such regions include biomass-burning regions of Africa and South America and urban regions of Europe and East Asia, where EC concentrations are typically >1.0 μg m^{-3} (Cooke and Wilson, 1996; Liousse et al., 1996), well within the range of Los Angeles data. Since regions of high EC dominate global forcing, it is important to treat EC correctly in these regions.

3. Global simulations and results

To test the effect of the core treatment on global direct forcing, simulations were carried out with GATORG (Gas, Aerosol, Transport, Radiation, and General circulation model). Details of the model and simulations are described in Jacobson, 1999b. The model was first run in time-dependent mode for one year to generate 24 cloud, temperature, water vapor, and relative humidity fields, then run in equilibrium mode 24 times (4 times per day on 6 days of the year) to generate aerosol composition and radiative fields. Aerosol fields were initialized from tracer studies and observations. Initial fields were then applied to temperature- and relative-humidity-dependent thermodynamic equilibrium calculations to determine solid, ion, and water concentrations. The model considered 48 aerosol components in each of 17 size bins and 409 solar and thermal-infrared wavelengths. Concentrations of EC from fossil fuels and biomass burning were obtained from Cooke and Wilson (1996), but reduced by 15% to account for a possible overestimation of this inventory (Bond et al., 1998). The resulting EC burden in the model was 0.23 Tg, within range of Cooke et al.'s (1999) revised estimate of total fossil fuel plus biomass burning EC, 0.15-0.25 Tg.

Table 1 shows that, in the baseline case, in which EC was core and the other 47 components were shell, EC tropopause forcing was +0.54 W m^{-2}. This forcing offset those of sulfate (-0.32 W m^{-2}), OC (-0.034 W m^{-2}), soil (-0.012 W m^{-2}), nitrate (-0.038 W m^{-2}), and ammonium (+0.002 W m^{-2}) to give a total anthropogenic forcing of +0.042 W m^{-2}. Haywood et al. (1997), Haywood and Ramaswamy (1998), and Myre et al. (1998) estimated slight positive forcings when sulfate and soot were well-mixed internally. Since the well-mixed treatment overpredicts EC absorption, such estimates, if correct, may have been correct for the wrong reason.

When EC was treated here as externally-mixed, its mean forcing was nearly cut in half, to +0.27 W m^{-2}. When EC was

Table 1. Tropopause solar plus thermal-infrared forcings, averaged globally and over 24 simulations, for several cases.

Case	Forcing (W m^{-2})
Baseline (EC core, well-mixed shell, $\rho_{EC} = 1.25$ g cm^{-3}, visible complex refractive index $m_{EC}=1.8-0.74i$)	+0.54
EC externally-mixed from other particle components	+0.27
EC well-mixed in internal mixture	+0.78
Baseline, but $\rho_{EC} = 1.75$ g cm^{-3}	+0.41
Baseline, but visible $m_{EC}=2.0-0.66i$	+0.49
Baseline, but +15% relative humidity	+0.56
Baseline, but -15% relative humidity	+0.51
Baseline, but +50% EC concentration	+0.80
Baseline, but -50% EC concentration	+0.27
Baseline, but +15% EC concentration (orig. inventory)	+0.62

well-mixed internally, its forcing increased to +0.78 W m^{-2}. Thus, forcing due to the core treatment was between that of the two other treatments, as expected from Figure 1.

Crystalline graphite has a bulk density of 2.25 g cm^{-3} and an imaginary refractive index (κ) of 1.34 at 0.55 μm (Fuller et al., 1999), but the density of EC in soot is less than that of crystalline graphite. Horvath (1995) suggested that EC consisting of graphite and 50% voids may have a density of 1.25 g cm^{-3}, which is the baseline density used here. The baseline midvisible κ used here is 0.74 (Krekov, 1993), which is coincidentally the κ obtained by scaling the crystalline κ with density down to a density of 1.25 g cm^{-3}. When density was increased to 1.75 g cm^{-3}, forcing decreased from +0.54 to +0.41 W m^{-2}. When refractive index data of Bergstrom (1973) instead of Krekov (1993) were used, EC forcing decreased from +0.54 to +0.49 W m^{-2}. Changes in concentration of ±50% had a large impact on EC forcing. Since EC is nonhygroscopic, changes in relative humidity did not greatly affect EC forcing.

The EC forcing estimate here for the externally-mixed case, +0.27 W m^{-2}, can be compared with the externally-mixed estimate of +0.4 W m^{-2} from Haywood and Ramaswamy (1998), since both studies used the EC distribution of Cooke and Wilson (1995). If the 15% reduction were taken into account by these authors, their external-mixture results (+0.34 W m^{-2} if scaled linearly) would be +0.07 W m^{-2} different from the results presented here. Comparisons cannot be made with other studies, since other studies have not isolated EC forcing due to both biomass burning and fossil fuel EC.

In sum, all direct forcing studies to date have assumed that EC is externally mixed or well-mixed internally. The former treatment appears unsupported by observations, and the latter treatment is physically unreal. These treatments have not been validated in any study to date and were shown not to work in a case study. In the same study, the core treatment compared well with observations. In the current study, treatment of EC as core resulted in positive forcings that more than offset negative forcings of all other anthropogenic aerosol constituents. In reality, particles contain many mixtures and some particles may be externally mixed. Hence, more studies are needed to examine the mixing state of aerosols and test results of the core treatment.

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