Convergence on climate warming by black carbon aerosols

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Scientific interest in the climate effects of black carbon (BC) intensified with the publication of Crutzen and Birks' (1) report dealing with the ejection of large amounts of smoke into the atmosphere after a major nuclear war. A key component of smoke is BC, which is the strongest absorber of visible solar radiation. BC solar absorption became a central issue in climate change research when a synthesis of satellite, in situ, and ground observations concluded (2) that the global solar absorption (i.e., direct radiative forcing, DRF) by atmospheric BC is as much as 0.9 $W{\cdot}m^{-2}$, second only to the CO_2 DRF. BC is also an important component of air pollution, which is plaguing large parts of the world. BC results from poor combustion of fossil fuel, household burning of coal briquettes, wood, and dung as fuel for home heating and cooking practiced by 3 billion people, as well as from agricultural and natural vegetation fires. These fine BC particles thus touch on personal and cultural basics, such as how we cook our food, how we move about, and the quality of the air that we breathe. This air pollution, consisting of BC and other particles, causes worldwide an estimated 7 million premature deaths annually, with most in East and South Asia (3). BC particles are also implicated in large-scale environmental effects, such as melting of the Himalaya and other glaciers (e.g., refs. 4 and 5). BC, along with the coemitted organic aerosols, is a major source of global dimming (2), which has been linked with reduction in precipitation (6).

Despite the established importance of BC to climate forcing, estimates of the DRF for BC, averaged over the globe, still span over a poorly constrained range from about 0.2–1 $W{\cdot}m^{-2}$ (7–10). This absence of consensus on the DRF of BC is one of the grand challenges in atmospheric climate science.

A possible clue to this challenge is the systematic discrepancy between model and observation estimates of the light absorption of atmospheric BC aerosols. Global climate models used in the latest assessment by the Intergovernmental Panel on Climate Change (IPCC) predict an average positive DRF from BC aerosols of about 0.4–0.5 $W{\cdot}m^{-2}$, with a large uncertainty range (7). In contrast, estimates based on large-scale observations of BC aerosol absorption using satellites and ground-based instrumentation, such as AERONET, arrive at a BC DRF of about 0.7–0.9 $W{\cdot}m^{-2}$ (e.g., refs. 5 and 8). In a study funded by the California Air Resources Board to compare models and observations of the aerosol absorption optical depth (AAOD) of BC over California, supposedly a region with one of the best-constrained inventories of BC emissions, the regional model underestimated the observed AAOD by a factor of 2–3 (9). The systematic underestimation by global climate model predictions relative to atmospheric observations are shown in Fig. 1 for East and South Asia.

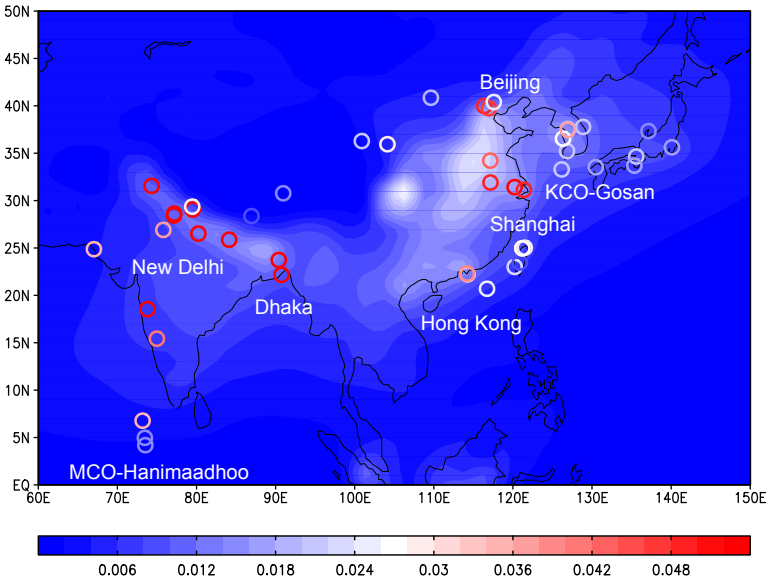

Fig. 1. AAOD for BC (550 nm) predicted by the NASA-GISS global climate model (background grid) compared with observation-based BC-AAOD retrieved for the NASA-AERONET sites (circles). The same color scale applies to both sets of data. The AERONET BC-AAOD is based on average inversion of data sampled during years 2000–2014 (10). The figure illustrates the systematic underestimation by factors 2–3 of the AAOD in current climate models relative to observational programs in the key regions of East and South Asia, which may originate from an uncertain combination of several factors described in the text.

The comprehensive model study by Bond et al. (10) scaled up their simulated AAOD results to agree with

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Author contributions: Ö.G. and V.R. wrote the paper.

The authors declare no conflict of interest.

See companion article 10.1073/pnas.1602310113.

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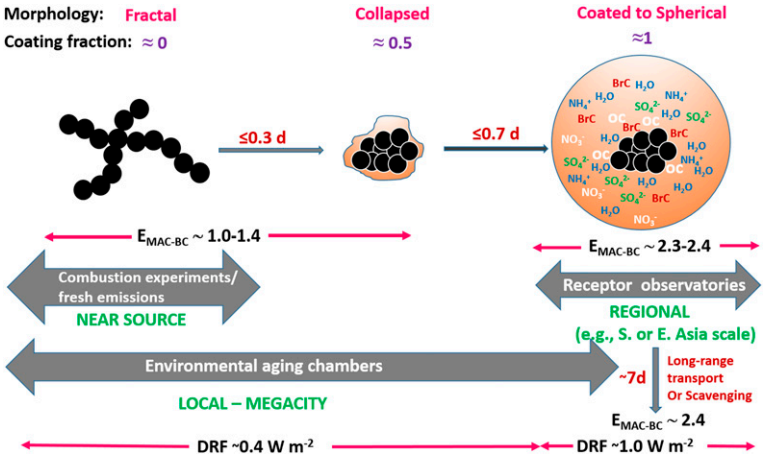

Fig. 2. Time-course evolution of BC aerosol composition, light absorption (where $E_{MAC\text{-}BC}$ is the enhancement because of coatings), and associated climate effects (as DRF).

ground-based AERONET observations of BC light absorption, which then led to a revised model estimate of 0.85 W·m^{-2}, thus converging with the observation-based estimates. However, this study did not offer an explanation for why the model AAODs have to be scaled upwards.

In PNAS, Peng et al. (11) offer compelling experimental evidence and explanation for the underestimation of BC absorption by models. The authors explore how changes in aerosol morphology and coatings affect the absorption of ambient BC and find that aged BC aerosols have an absorption that is enhanced by a factor of 2.4 relative to BC in fresh emissions.

Several hypotheses have been forwarded to account for the underestimation in models versus observations in both AAOD and DRF of BC aerosols. These include: (*i*) underpredictions of how much BC is actually released to the atmosphere; (*ii*) a need to account also for light-absorbing organic aerosols (known as brown carbon, BrC) next to BC; and (*iii*) an underestimate in the coating-enhancement of ambient BC absorption. It is likely that all these factors contribute.

Concerning the third hypothesis, addressed by Peng et al. (11), there are conflicting results of absorption enhancement of BC aerosols during atmospheric aging from available experimental studies, theoretical modeling, and field investigations, which transfer directly into large uncertainties in model estimates of DRF (e.g., refs. 7 and 10). To improve model predictions, it is imperative to better understand both the mass absorption cross-section of the bare BC skeleton (MAC_{BC}) and to constrain the enhancement of BC absorption after mixing with non-BC aerosol components in the atmosphere ($E_{MAC\text{-}BC}$), and to comprehend what values are most relevant to use for radiative transfer calculations over different ranges of temporal and spatial scales (Fig. 2). The coating materials of atmospherically aged BC consist mainly of sulfate, nitrate, ammonium, and organic carbon (OC) (12) (Fig. 2). The climate models vary widely today in their description of this BC mixture. Reports of $E_{MAC\text{-}BC}$ span between negligible effects (value ~1) to >3 (e.g., refs. 10 and 13), causing confusion and correspondingly large uncertainties of this important aspect in climate models. However, generally, climate models appear to have taken parameterizations of $E_{MAC\text{-}BC}$ in the low end of this wide range.

Writing in PNAS, Peng et al. (11) now offer a possible reconciliation of these previous reports on the coating-enhancement of BC aerosols. A specially constructed environmental aging chamber flowing the ambient gaseous mixture of the atmospheres in Houston and Beijing (excluding ambient particles) was fed with well-characterized fresh BC particles, which then were allowed to age in this "real" atmosphere under sunlight exposure. The BC particle mixture was continuously probed by a suite of instruments to characterize the development of, for example, BC aerosol size, density, and optical properties. Peng et al. (11) show that aging brings not only growth of coatings but also dramatic changes in morphology. Their study demonstrates a two-stage process, where the first stage is characterized initially by a highly fractal morphology where the primary BC/soot spheres are held together in aciniform (grape-like) clusters of stretched-out chains, often seen in electron microscopy images of fresh diesel soot (10). In this first stage of aging, which in Houston takes ~9 h and in Beijing ~2.3 h, this chain-like BC structure of low particle-density collapses to form a semispherical particle (Fig. 2). Notably, there is no significant absorption enhancement during this first stage (i.e., $E_{MAC\text{-}BC}$ ~1). In the second stage, monitored for about an equal time duration in the chamber experiments by Peng et al. (11), there is continued coating growth. This leads to formation of spherical internally mixed BC particles, and, importantly, an associated increase in $E_{MAC\text{-}BC}$ to reach about 2.4 for both cities.

The mechanistic insights of the two stages of aging, associated time scales, and relevance for the dynamic evolution of $E_{MAC\text{-}BC}$, aids in understanding earlier reports of the BC absorption enhancement. The vast majority of BC MAC and $E_{MAC\text{-}BC}$ are reported from combustion experiments and from other fresh emissions, such as in classic road tunnel studies aimed to get environmentally realistic MAC measurements (Fig. 2). These near-source studies generally report MAC of pure BC and on freshly aged (time scale of <1 h) BC that correspond to $E_{MAC\text{-}BC}$ of about 1.0–1.5 (e.g., refs. 10, 14, and 15) (Fig. 2). Such studies are often the basis for input to regional and global climate models (e.g. refs. 7 and 10). In the context of the BC aging scheme put forth in PNAS by Peng et al. (11), such optical measurements of near-source BC may catch only the fractal to possibly collapsed stages of aging, where there is still little absorption enhancement by coatings (Fig. 2). Climate models using these canonical MAC and $E_{MAC\text{-}BC}$ values are getting DRF of BC in the lower range (e.g., 0.4 W·m^{-2}). The question is how appropriate it is to use this parameterization of BC optical properties, obtained on less-aged BC, to estimate the regional and even global forcing of BC aerosols that stay in the atmosphere about a week on average, thus rendering the vast majority of atmospheric BC to be aged with fully developed coatings?

An $E_{MAC\text{-}BC}$ of 2.4, as suggested by Peng et al. (11), is in agreement with another recent report based on comparing the MAC of ambient BC aerosols collected at a receptor observatory in China, before and after a decoating method (15). Cui et al. developed a

two-step method to remove both inorganic and organic matter coatings of ambient Chinese aerosols and tested the method in several ways, including demonstrating near-full recovery of BC mass. This study found $E_{MAC\text{-}BC}$ of 2.3 ± 0.5 for aged BC aerosols, with a strong dependency on the amount of both inorganic and organic coatings. The advantage of estimating the optical properties of ambient aged and coated BC aerosols at receptor observatories is that this is likely more relevant for their optical and climate effects on the regional to even global scale (Fig. 2). Another field-observation–based estimate of $E_{MAC\text{-}BC}$ in the cleaner atmosphere of California obtained lower values (16). This result may be caused by the use of thermal denuders that may not fully remove all coatings and/or because the sampled aerosols were not fully aged due to less gaseous air pollution, leading to lower values of SO_4/BC, for example; Peng et al. (11) showed that aging to a coated spherical particle in Houston takes up to 24 h, which is 3–4 times slower than in Beijing. It is encouraging that now two independent studies with completely different technical approaches, yet with the common denominator of probing aged and thus more fully morphologically transformed and coated BC aerosols, get results on $E_{MAC\text{-}BC}$ that are converging.

A reservation here is that the chamber experiment in Beijing is taken to be representative for "developing countries." The mixing ratio between BC and key inorganic coating components, such as sulfate, are much higher in South Asia (e.g., India) than in East Asia (e.g., China) (17), one reason why both effects and time scale of aging may be expected to be different between the two regions. We encourage that effects of aging and coatings on BC absorption enhancement in receptor locations in key regions of South Asia, East Asia, Africa, and elsewhere be addressed experimentally.

In PNAS, Peng et al. (11) assess the implications on climate by the constrained coating-enhancement of BC absorption. The authors demonstrate that the DRF difference between fully aged and fresh BC particles increases by about 0.5 $W{\cdot}m^{-2}$. Their climate model simulations yield a global DRF from coated BC aerosols of ∼1.0 $W{\cdot}m^{-2}$ (Fig. 2). If this were to be confirmed by forthcoming studies, also focusing on properties of aged aerosols most relevant to large-scale effects, this will close the factor of 2–3 gap between model predictions and observations on the effect of BC aerosols on climate.

Sources that emit BC also emit OC aerosols. The issue of solar absorption by OC is a related issue that needs to be resolved. Most, if not all, climate model studies used in IPCC reports assume OC aerosols to be nonabsorbing and assume them to be just scatterers of solar radiation. As a result, OC aerosols lead to a net cooling effect in climate models (7, 10). However, both laboratory and field studies have shown OC aerosols to absorb solar radiation (BrC), particularly in the shorter (<0.5 μ) wavelengths (18, 19). When the BrC solar absorption is included in the treatment of OC aerosols, the net direct radiative forcing of these is close to zero (8) because the heating resulting from BrC solar absorption nearly cancels the cooling effect of other OC.

A convergence of the BC aerosol climate effect toward the upper end of the wide earlier range would make mitigation efforts even more rewarding. For example, 3 billion of the poorest people of the world rely on cooking with solid fuels (20), which is the largest source of anthropogenic BC emissions (10). The fundamental problem is that the poorest 3 billion cannot afford readily available clean cooking technologies. The new findings, as well as recent experimental work on BrC, would yield a global warming potential (40 y) of ∼2,000 for BC and the coemitted OC; the resulting equivalent carbon credits would break the affordability barrier for the poorest 3 billion. There is a need to simultaneously mitigate emission of long-lived CO_2 and short-lived climate pollutants, such as BC aerosols, to avoid passing critical thresholds in the climate system (21). The multiple cobenefits of reducing BC emissions on respiratory health, climate, water, and food security are particularly beneficial to the developing world.

Acknowledgments

The authors' research in this area was supported by the Swedish Research Councils VR and Formas (Ö.G.); the Swedish Energy Agency (Ö.G.); and the US National Science Foundation (V.R.).

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