Exposure to ambient black carbon derived from a unique inventory and high-resolution model

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Black carbon (BC) is increasingly recognized as a significant air pollutant with harmful effects on human health, either in its own right or as a carrier of other chemicals. The adverse impact is of particular concern in those developing regions with high emissions and a growing population density. The results of recent studies indicate that BC emissions could be underestimated by a factor of 2–3 and this is particularly true for the hot-spot Asian region. Here we present a unique inventory at 10-km resolution based on a recently published global fuel consumption data product and updated emission factor measurements. The unique inventory is coupled to an Asia-nested (~50 km) atmospheric model and used to calculate the global population exposure to BC with fully quantified uncertainty. Evaluating the modeled surface BC concentrations against observations reveals great improvement. The bias is reduced from ~88% to ~35% in Asia when the unique inventory and higher-resolution model replace a previous inventory combined with a coarse-resolution model. The bias can be further reduced to ~12% by downscaling to 10 km using emission as a proxy. Our estimated global population-weighted BC exposure concentration constrained by observations is 2.14 μg·m⁻³, 130% higher than that obtained using less detailed inventories and low-resolution models.

In this study we develop and evaluate a unique global BC emission inventory using a zoomed aerosol model, and estimate the global population’s exposure to BC with a focus on Asia. The influence of model resolution and the use of an updated emission inventory on the calculated BC concentration are evaluated against field observations.

Updated BC Emission Inventory

Based on an updated EF_Bc dataset (21, 22) and a recently published 0.1° × 0.1° fuel consumption data product (12), we have drawn up a global 0.1° × 0.1° BC inventory for 2007 (Peking University BC Inventory for 2007 (PKU-BC-2007)) (Methods). According to PKU-BC-2007, the global total and anthropogenic-only BC emission in 2007 was 8.9 and 6.3 Tg, respectively. Asia contributed 4.1 Tg of the total and 3.8 Tg of the anthropogenic BC. A Monte Carlo simulation, varying the parameters of PKU-BC-2007, yields an interquartile range (IQR) of 5.4–14.8 Tg for global annual BC emission. The IQR would be 5.6–14.4 Tg if

Significance

In this study, we have developed a unique global black carbon (BC) emission inventory using a 10-km grid based on the latest source and emission factor information. The inventory is used to model BC concentrations using a global atmospheric aerosol climate model, with a high-resolution grid for Asia, to better resolve the exposure of populations to elevated BC concentration. The model with even higher resolution (10 km) is used for the North China Plain. Finally, the population exposure concentrations are evaluated.


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Global distribution of BC emissions in 2007. (A) The 0.1° × 0.1° distribution of BC emissions by PKU-BC-2007, (B) Difference between the PKU-BC-2007 (downscaled to 0.5° × 0.5°) and MACCity (2007, 0.5° × 0.5°) inventories.

Fig. 1. Global distribution of BC emissions in 2007. (A) The 0.1° × 0.1° distribution of BC emissions by PKU-BC-2007. (B) Difference between the PKU-BC-2007 (downscaled to 0.5° × 0.5°) and MACCity (2007, 0.5° × 0.5°) inventories.
are as high as 6.68, 5.49, and 4.31 μg m\(^{-3}\), respectively. In these regions, a total of 1.02 Tg BC were emitted in 2007, primarily from the domestic burning of biomass fuel (44%) and coal (20%) and from coke production (19%), motor vehicles (9%), and brick kilns (6%). Extremely high BC exposure (>6 μg m\(^{-3}\)) can be found in metropolitan areas with a population density over 1,000 per km\(^2\) (Fig. 3B and C).

Fig. 4 shows the global and regional cumulative frequency distributions of ambient BC exposure concentration. People in East Asia are exposed to a much higher BC concentration than those in other regions. According to Geng et al. (3), in Shanghai, China, for an IQR increase of average BC concentration from 2.2 to 4.9 μg m\(^{-3}\), the cardiovascular, respiratory, and total mortality of the population increase by 3.2%, 0.6%, and 2.3%, and emergency-room and outpatient visits increase 1.33% and 3.35%, respectively. It appears that the percentages of people exposed to an annual mean BC concentration above 4.9 μg m\(^{-3}\) in East Asia and South Asia are as high as 18% and 11%, respectively, suggesting a relatively high health risk which cannot yet be quantified.

As with the modeled concentrations at measurement sites, the calculated exposure concentrations are influenced significantly by inventory and model resolution. Fig. 5 compares the average BC concentrations modeled at monitoring sites and the regional average exposure concentrations derived by the four inventory/model combinations without downscaling and by the two combinations with downscaling (\(E_{PKU/M_{INz}}\) and \(E_{PKU/M_{IN}}\), where \(E\) indicates the downscaling method) in East Asia, South Asia, Europe, and North America. When the estimate resolution is increased from ~200 km (\(E_{PKU/M_{INz}}\) and \(E_{PKU/M_{IN}}\)) to ~10 km, the model-calculated BC concentrations at the monitoring sites approach the observations. With downscaling, the calculated results agree well with the observations (100–104%) except in Europe (61%) where the low bias is due to the relatively coarse model resolution of INCA. Simultaneously, the modeled by \(E_{PKU/M_{INz}}\) and \(E_{PKU/M_{IN}}\) are downscaled to 0.1° × 0.1° using emission density as a proxy (Methods), and then compared with that explicitly modeled by \(E_{PKU/CHIMERE}\). An exponent (\(a\)) is introduced to address the influence of the dispersion of subgrid scale emissions (Methods). The optimal value of \(a\) (0.30) is derived by a trial-and-error method to achieve the lowest mean deviation between calculated and observed BC concentrations at 55 sites in Asia and 174 sites in the rest of the world (Fig. S6). Using this downscaling method, the residual low bias of the model is substantially reduced for both Asia and other regions. Although the exponent can implicitly account for dispersion in a simplified way, the influence of transport within each model grid cell could not be fully resolved by the downscaling method.

**Population Inhalation Exposure to Ambient BC**

These results suggest that realistic BC concentrations downscaled to 10 km can be better modeled by using the combination of the \(E_{PKU/M_{INz}}\) in Asia and \(E_{PKU/M_{IN}}\) elsewhere, together with the emission-based nonlinear downscaling. The resulting surface ambient BC concentrations are converted into population-weighted exposure concentrations. Fig. 3 shows the spatial distribution of these exposure concentrations at 0.1° × 0.1° in 2007. The geographical distribution of the exposure concentrations is similar to that of the emission, due to the short atmospheric lifetime of BC. Because emission sources and population covary spatially, the global average population exposure concentration (2.14 μg m\(^{-3}\)) is significantly higher than the global average surface concentration in ambient air (0.28 μg m\(^{-3}\)). The high-exposure areas are located in the densely populated areas of East Asia and South Asia (Fig. 3B and C). The annual mean BC exposure concentrations in the NCP (China), Sichuan Basin (China), and Indo-Gangetic Plain (India)
exposure concentrations increase dramatically. The four model/inventory combinations without downscaling would underestimate the exposure risk by 30–67% for these regions. If the combination of $E_{\text{MAC}}/M_{\text{IN}}$ were used, the calculated average exposure concentration in Asia would be as low as 1.52 µg m$^{-3}$, compared with 2.12 and 3.39 µg m$^{-3}$ by $E_{\text{PKU}}/M_{\text{IN}}$ without and with downscaling. In Europe and North America, the exposure concentrations would increase by 80% and 110%, respectively, after downscaling (from ~200 to ~10 km). In addition, for the 5% of the most vulnerable population, the exposure concentrations are equal to or higher than 6.72 µg m$^{-3}$ in India and 10.4 µg m$^{-3}$ in China, which would be underestimated as 2.21 and 4.33 µg m$^{-3}$ if the combination of $E_{\text{MAC}}/M_{\text{IN}}$ were to be used.

**Conclusion**

The levels of population exposure to ambient air BC estimated in this study are higher than those previously documented for several reasons: (i) the unique BC emission inventory derived from the recently published fuel consumption data product and updated EF$_{\text{BC}}$ database, (ii) modeling with higher resolution in Asia, and (iii) downscaling to rectify the bias caused by the covariance of emission and population at resolution higher than the model resolution. It is demonstrated that the exposure of populations living in hot spots is diluted and the population-weighted concentration is reduced when a coarse model resolution is applied and the population exposure could be underestimated by an insufficiently fine-resolution model. Although it is demonstrated in this study that downscaling can help reduce the bias, direct calculation using a finer-resolution model (10 km or higher) would be preferred for drawing up future assessments of the exposure and risk of BC, as well as other short-lived air pollutants.

The need for high-resolution inventories and atmospheric models illustrated by this study will become even more critical for health impact studies when accelerated urbanization in developing countries causes populations to become more unevenly distributed in the future. It is predicted that the urban population of China and India will respectively increase from 42% and 29% of the population in 2007 up to 62% and 40% in 2030 (31). If BC concentrations were stable, urbanization alone would cause population exposure to increase by 15% and 14% in China and India, respectively, due to factors of 2.1 and 3.0 differences in the exposure concentrations between urban and rural populations.

**Methods**

**Development of the BC Emission Inventory.** The global 0.1° × 0.1° BC emission inventory PKU-BC-2007 is based on a global fuel combustion database (PKU-FUEL-2007) (12) and an updated EF$_{\text{BC}}$ database (SI Methods). A total of 706 EF$_{\text{BC}}$ data measured in 13 countries from 1985 to 2011 were included (Table S5), including up-to-date measurements, particularly for developing countries. The technical-split method is applied for 16 combustion processes including coal combustion in power plants and industrial boilers, brick kilns, coke production, motor vehicles, residential firewood, residential coal, and agricultural waste combustion (Tables S4 and S5). A 1,000-point Monte Carlo approach is applied to characterize the uncertainty. Detailed information for developing the inventory is provided in SI Methods.

**Atmospheric Models of BC.** The global aerosol model LMDZORINCA and the regional off-line chemistry-transport model CHIMERE are used to calculate the 4D distributions of BC concentrations. These models are briefly introduced here and more details are presented in the literature (28, 30, 32). LMDZORINCA is a global aerosol model that couples a general circulation model (Laboratoire de Météorologie Dynamique) to an aerosol module (Interactions between Aerosols and Chemistry) (28, 32). A zoomed version is run at a resolution of 0.51° × 0.66° in latitude and 0.66° in longitude for a region of 50–130°E, 0–55°N centered over China and India, whereas a regular version is run at a fixed resolution of 1.27° × 2.50°. The same vertical coordinates are used in the two versions with 19 vertical layers from 3.88 to 1,013 hPa. For CHIMERE, the latest version (30) is run at a horizontal resolution of 0.1° × 0.1° with 8 vertical layers in hybrid sigma-pressure coordinates for a domain over the NCP (113°5’-120°E, 36°N-42°N), driven by 0.1° × 0.1° meteorological data from the Weather Research and Forecasting Model (30). In CHIMERE, BC is divided into nine size bins, and major aerosol dynamics, dry deposition, and wet scavenging are considered for BC (30).

**Fig. 4.** Cumulative frequency distributions of BC exposure in different regions. The BC concentrations are derived by downscaling the results given by $E_{\text{PKU}}/M_{\text{IN}}$ in Asia and by $E_{\text{PKU}}/M_{\text{IN}}$ in the other regions to 0.1° × 0.1° grids using emission as a proxy.

**Fig. 5.** Comparison of the calculated surface-air BC concentrations at observation sites and population exposure concentrations in four regions of South Asia, East Asia, Europe, and North America among different methods. The methods compared are four inventory/model combinations ($E_{\text{MAC}}/M_{\text{IN}}$, $E_{\text{PKU}}/M_{\text{IN}}$, $E_{\text{MAC}}/M_{\text{IN}}$, and $E_{\text{PKU}}/M_{\text{IN}}$) and the two with downscaling ($E_{\text{PKU}}/M_{\text{IN}}$ for $E_{\text{PKU}}/M_{\text{IN}}$ and $E_{\text{PKU}}/M_{\text{IN}}$ for $E_{\text{PKU}}/M_{\text{IN}}$). The model-calculated average surface-air concentrations at all observation sites are shown as bars, which are compared with the observed mean concentrations (dashed lines) by the marked percentages (modeled concentrations divided by observed ones). Error bars for individual bars are derived by using the first and the third quartiles of PKU-BC-2007 as inputs, which represent the uncertainty range of modeled surface BC concentrations resulting from the uncertainty in emissions. The population-weighted BC exposure concentrations at these sites are shown as diamonds.

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Downscaling of BC Concentrations to 0.1° × 0.1°. Surface BC concentrations calculated by INCA-2A in Asia and by INCA for the rest of the world are downscaled from the model grid (\( x, y \)) (0.51° × 2.5° grid in INCA-2A or 1.27° × 2.5° grid in INCA) to a 0.1° × 0.1° subgrid (i, j) as

\[
C_{ij}^* = C_{x_i y_j} E_{ij}^* \sum_{i,j} E_{ij}
\]

where \( C_{ij}^* \) is the downscaled concentration of the subgrid (i, j); \( C_{x_i y_j} \) is the model-calculated concentration of grid cell (\( x, y \)); \( E_{ij}^* \) is the emission density in the subgrid (i, j); the exponent \( \alpha \) is a seasonal-constant coefficient describing the nonlinear relationship between \( C_{x_i y_j} \) and \( E_{ij}^* \), and is derived from a trial-and-error method to minimize the mean deviation between calculated and observed BC concentrations at 55 sites in Asia and 174 sites in the rest of the world (Fig. S6; \( F_{ij}^* \) is the fraction of area of each subgrid (i, j) in the model grid (x, y), and the product of \( F_{ij}^* \) and \( E_{ij}^* \) is the area-weighted emission density in the subgrid (i, j).}

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