Global Emission of Black Carbon from Motor Vehicles from 1960 to 2006

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ABSTRACT: Black carbon (BC) is a key short-lived climate change forcer. Motor vehicles are important sources of BC in the environment. BC emission factors (EF_{BC}), defined as BC emitted per mass of fuel consumed, are critical in the development of BC emission inventories for motor vehicles. However, measured EF_{BC} for motor vehicles vary in orders of magnitude, which is one of the major sources of uncertainty in the estimation of emissions. In this study, the main factors affecting EF_{BC} for motor vehicles were investigated based on 385 measured EF_{BC} collected from the literature. It was found that EF_{BC} for motor vehicles of a given year in a particular country can be predicted using gross domestic product per capita (GDP_{c}), temperature, and the year a country’s GDP_{c} reached 3000 USD (Y_{3000}). GDP_{c} represents technical progress in terms of emission control, while Y_{3000} suggest the technical transfer from developed to developing countries. For global BC emission calculations, 87 and 64% of the variation can be eliminated for diesel and gasoline vehicles by using this model. In addition to a reduction in uncertainty, the model can be used to develop a global on-road vehicle BC emission inventory with spatial and temporal resolution.

INTRODUCTION

Based on a 15-year climate response simulation, it was found that black carbon (BC) from fossil fuels is one of the leading causes of global warming, and the elimination of BC from fossil fuels can reduce global surface air temperatures by 0.3−0.5 K.† Due to a strong effect in radiative forcing, the inclusion of BC within the new warming mitigation framework after the Kyoto Protocol was proposed.‡

A technology-based global BC emission inventory was first developed by Bond et al. who estimated that annual emissions of BC from 50 anthropogenic sources were 4669 Gg in 1996, among which 917 Gg were from motor vehicles.§ It was estimated that on-road motor vehicles contributed 20% of the total anthropogenic BC emission in the world. By using a similar approach, Zhang et al. developed a 0.5° × 0.5° BC emission inventory for Asia and estimated that the total BC emissions from on-road vehicles in this region were 294 and 446 Gg in 2001 and 2006, respectively.¶ Because of the increase in the number of motor vehicles in the fast transitioning world, BC emissions from these countries have increased rapidly, and the trend is expected to continue in the future.¶ It was predicted that global BC emissions from motor vehicles might reach 1025−1754 Gg by 2050 under different IPCC (Intergovernmental Panel of Climate Change) scenarios, overriding residential, industry, and power sectors to become the largest anthropogenic source.¶

BC emissions are often estimated based on the amount of fuel consumed and emission factors (EF_{BC}, defined as the amount of BC emitted per mass of fuel consumed) of various activities. EF_{BC} for motor vehicles measured by various laboratories vary in orders of magnitude due to many factors including vehicle type, fuel composition, vehicle model year, marketing country, operation mode, ambient temperature, analytical method, and so on, leading to high uncertainty in emission inventory development.¶ The uncertainty could be reduced significantly by applying different EF_{BC} for diesel and gasoline vehicles and for separating developed and developing countries.§,¶,† Novakov et al. had assumed a linear decreasing trend of diesel EF_{BC} for developed countries over time from 1965 to 1985.‡ In spite of these efforts, large uncertainty remains in emission inventories developed to date since EF_{BC} still vary widely within developed/developing countries and within gasoline/diesel vehicles.§,¶,† In addition, since the majority of EF_{BC} were measured in limited countries, especially developed countries, it is very likely that the direct application of these measurements to other countries can cause prediction bias.

The objectives of this study were to identify the key factors affecting EF_{BC} for motor vehicles based on a thorough literature review and to develop a quantitative relationship between EF_{BC} and these factors. If such a model can be established, the EF_{BC} for various countries in different years where no measurement was available could be better estimated. Subsequently, spatial and temporal variations of global BC emission can be characterized with reduced uncertainty. There is also a potential...
of applying the quantitative approach developed in this study to other pollutants.

**METHODOLOGY**

**Collection of EF_{BC}.** A total of 385 directly measured and originally reported EF_{BC} for on-road diesel and gasoline vehicles from 14 countries during the period from 1985 to 2008 were collected from 66 published papers or reports. Detailed information on vehicle fleet composition is listed in Table S1. The data include both BC (optically measured) and EC (elemental carbon, thermally measured). It was reported that the ratio of EC to BC varied with the nature and aging of the deposited particles.\[^{9,15}\] In this study, EC was used equivalently to BC following the common practice in the literature,\[^{16}\] which is likely to cause a certain degree of uncertainty. Of 385 EF_{BC} collected, around 10% were only reported as emissions per kilometer distance driven, which was converted into emission per kilogram of fuel consumed using a vehicle economic factor of 278 and 97 g/km for heavy-duty and light-duty vehicles produced before 1990 and 265 and 74 g/km for heavy-duty and light-duty vehicles produced after 1990, respectively.\[^{17-19}\]

Information relevant to BC emissions measurements collected together with EF_{BC} included vehicle type, vehicle in-use country, model year, measured year, method for the measurement, run cycle, vehicle odometer, and ambient air temperature. For the EF_{BC} measured for diesel or gasoline vehicle fleets on roads, the vehicle model year was calculated by subtracting the year when the measurement was conducted by the average on-road vehicle ages of 4\(^{20,22}\) and 6\[^{22,23}\] for developing and developed countries, respectively. The collected 385 EF_{BC} together with other relevant information and literature sources are listed in Table S2. Although we tried our best to collect as many data as possible and the data were from various countries including developed, developing, and in-transition economies, the data availability is still the major constrain of this study since approximately 70% of them were from the United States. Hopefully, more measurements in a wider range of countries will be available in the future for validating and improving the model.

**Prediction of EF_{BC}.** The overall frequency distribution of EF_{BC} was tested to confirm the difference between gasoline and diesel vehicles\[^{8}\] and for separating them into two populations. Factors including vehicle in-use country, model year, measured year, method for the measurement, run cycle, vehicle odometer, and ambient air temperature (\(T\)) were tested for their significant influences on EF_{BC} using an analysis of variance for gasoline and diesel vehicles separately. Of these factors, in-use country, model year, and \(T\) were identified as significant. By recognizing that per capital gross domestic product (purchasing power parity) (\(GDP_{c}\))\[^{24}\] is a good indicator representing differences among countries and over years, regression models were developed using \(GDP_{c}\) and \(T\) (gasoline only) as independent variables to predict EF_{BC} of gasoline and diesel vehicles in different countries and at different times. Later, it was found that the technical transfer from developed to developing countries could help to reduce EF_{BC} in developing countries and a new parameter, \(Y_{BC}\) defined as the year when a country’s \(GDP_{c}\) reaching 3000 USD was introduced into the regression models.

**Global Vehicles’ BC Emission.** BC emissions were calculated for various countries from 1960 to 2006 based on the model-predicted EF_{BC} and fuel consumption quantities. The 221 countries/territories involved were classified into 4 categories of OECD90 (members of the Organisation for Economic Co-operation and Development), ASIA (developing countries in Asia), REF (undergoing economic reform), and ALM (Africa, Latin America, and the Middle East countries) according to IPCC.\[^{25}\] Historical data on total diesel and gasoline consumption by motor vehicles for 138 countries from 1970 to 2006 and from 1960 to 1969 were collected from the International Energy Agency and the U.N. Energy Statistics database, respectively.\[^{26-28}\]

For the rest of the 82 countries/territories, their fuel consumptions, which contributed less than 2% of the global total, were predicted using gross domestic product proxy.\[^{24}\] The regression coefficients of the equations are shown in Table S3. Contribution from two-stroke gasoline vehicles (motorcycles) and superemitters to the emissions were taken into consideration in emission inventory development. The fraction of gasoline consumption by motorcycles in a given country was calculated based on vehicle numbers, mean travel distances, and mean mileages of motorcycles and other vehicles.\[^{29,30}\] Without enough measurement, the EF_{BC} of motorcycles was assumed to be 5 times that of four-stroke gasoline engine vehicles.\[^{3}\] It was assumed that there were 5 and 10% of superemitters in developed and developing countries, respectively,\[^{3}\] and their EF_{BC} were 3.8 (median of 2.9–4.8) times those for regular vehicles.\[^{31,32}\]

**Statistical Analysis.** A gauss2 module for bimodal distribution was applied to characterize the frequency distribution of EF_{BC} using Matlab.\[^{33}\] Analysis of variance and linear regression was performed using SPSS. A level of 0.05 was adopted for significance tests. A reduced major axis method (RMA) was used for regression analysis. RMA rather than a least-squares method (LSQ) was used because the independent variables were also associated with random error and higher and lower EFs could be under- or overestimated if LSD was applied.
Loss function (LF), defined as the quadratic sum of difference between the predicted and measured data, was used to evaluate the fitting to the model.34

Uncertainty Analysis. The overall uncertainty of the model-predicted BC emissions was analyzed using a Monte Carlo simulation with 5000 runs. A fixed coefficient of deviation (0.05) was applied for energy consumption based on a normal distribution. For EFBC, standard deviations of logEFBC derived from regression models were used as input. Median values were used for emission estimation, and a semi-interquartile range (difference between the 75th and 25th quartiles) was derived to characterize the uncertainty.

RESULTS AND DISCUSSION

Difference in EFBC between Gasoline and Diesel Vehicles. The literature reported 385 EFBC for motor vehicles (excluding motorcycles and superemitters) varied from 0.000681 to 7.20 g/kg with a median value of 0.234 g/kg, varying over 4 orders of magnitude. The frequency distribution of EFBC is presented in Figure 1A in log-scale and a typical bimodal histogram is demonstrated. The bimodal distribution can be fitted satisfactorily (p > 0.05, \( r^2 = 0.872 \)) with the two populations happening to be gasoline and diesel vehicles. A well-known fact that EFBC for diesel vehicles are significantly higher than those for gasoline ones was quantitatively confirmed.8 Therefore, the two populations were analyzed separately to evaluate other factors hereafter. Although a difference between light- and heavy-duty diesel vehicles was reported in the literature,9 the difference was not found statistically for the data collected (p = 0.946) (Figure 1B). The EFBC for gasoline and diesel vehicles were well fitted with log-normal distribution functions individually (Lilliefors test, p > 0.05). The means and standard deviations of logEFBC (log (g/kg)) were −0.17 ± 0.42 and −1.48 ± 0.58 for diesel and gasoline vehicles, respectively.

Other Factors Affecting EFBC. A number of factors other than vehicle type were tested for their effects on logEFBC using analysis of variance. Among these factors, vehicle model year and country (three categories of the United States, other developed countries, and developing countries) were found to be significant (p < 0.05) for logEFBC of both gasoline and diesel vehicles, while ambient temperature (T) was significant (p < 0.05) only for logEFBC of gasoline vehicles. It was generally recognized that EFBC decreased over time due to advances in control technology and tightening of emission regulations.35 For example, since the enforcement of EURO-I and EURO-II regulations in Thailand, EFBC for diesel vehicles had decreased over 4 orders of magnitude. Injection timing retardation technology started to apply for diesel engines during a period from 1985 to 1995. In 1993, sulfur content of diesel fuel was regulated to below 50 ppm and further restricted to less than 15 ppm in 2006.37,39 Both replacement of two-stroke by four-stroke diesel engines for and use of oxidation catalysts in transit buses occurred in 1990s.35,38 Diesel particle filters were in use since 2007.

At the beginning, the model year was also introduced as an independent parameter together with GDPc. However, no difference was found between the models with and without the model year. It was recognized that not only EFBC differed among countries at the same time but also EFBC changes over time (model year) for a given country can be explained by GDPc. Relations between logEFBC and GDPc, for both diesel and gasoline vehicles and T (for gasoline vehicles only) are shown in Figure 2. Intrinsically, both country and model year represent the status of technical progress and social-economic development in terms of BC emission. Therefore, two regression models for predicting logEFBC (log (g/kg)) using GDPc (10000 USD/cap) and T (K, gasoline vehicle only) as independent variables were derived for diesel and gasoline vehicles, respectively. The 50th and 90th percentiles of the calculated residues were 0.23 and 0.49 for diesel model and 0.30 and 0.69 for the gasoline model, respectively

\[
\log_{10} \text{EFBC} = -0.4302 \text{GDP}c + 0.7566, \\
\text{with } n = 209, \quad LF = 19.0 
\]
Although the overall countries appeared to be overadjusted by 2000 when the countries was around 20,000 USD. Equivalent standards were standard (China III) was in effect in 2007 when the middle-income and upper-middle-income economies). With only around 3,500 USD. Accordingly, GDP model year can be integrally expressed by a single variable of two inter-related factors of country development status and in EFBC can be explained by GDP, (for diesel and gasoline vehicles) and T (for gasoline vehicles only). Developed (open circles) and developing (solid triangles) countries are marked differently. Loss function (LF) was calculated to describe the goodness of fit of the regression.

![Figure 3. Relationship between the predicted and measured logEFBC for diesel (A) and gasoline (B) vehicles. The calculation was based on GDP, (for both diesel and gasoline vehicles) and T (for gasoline vehicles only). Developed (open circles) and developing (solid triangles) countries are marked differently. Loss function (LF) was calculated to describe the goodness of fit of the regression.](image)

**Effect of Technology Transfer.** Although a large variation in EFBC can be explained by GDP, EFBC for developing countries appeared to be overadjusted by GDP, leading to a systematic overestimation of logEFBC for developing countries (Figure 2). This overestimation can be explained by the fact that developing countries have learned lessons from developed countries and technical transfer helped developing countries to move relatively fast in emission controls at a similar development status. In fact, Euro III emission standards were introduced in European countries in 2000 when GDP, of these countries was around 20,000 USD. Equivalent standards were in force in the United States when the GDP, reached approximately 40,000 USD in 2004. In China, a similar standard (China III) was introduced in 2000 when the GDP, was only around 3,500 USD. Accordingly, Y was introduced to quantify the effects of experience and technology transfer (3,035 USD was used by the World Bank to distinguish lower-middle-income and upper-middle-income economies). With Y included, eqs 1 and 2 were modified as

\[
\text{logEF}_{BC} \text{(gasoline)} = -0.7055 \text{GDP}_c - 0.02080T \\
+ 5.898, \quad n = 176, \quad LF = 39.8
\]

(2)

![Figure 4. Comparison between the BC emissions predicted in this study and those reported in the literature. BC emissions in major areas in 1996 are compared on the left panel (Bond, 2004); BC emissions in China for several years are compared on the right panel (1995: by Streets; 1996: by Bond; 2000: by Cao; 2001 and 2006: by Zhang).](image)

The model calculated logEFBC are plotted against the measured ones in Figure 3. It appears that the effects of the two inter-related factors of country development status and model year can be integrally expressed by a single variable of GDP, RMA was used for the regression because higher and lower EFs could be under- or overestimated when LSD was applied (Figure S1).

![Image](image)

**Global BC Emission from Vehicles.** Based on the models developed, EFBC for and annual BC emissions from on-road vehicles from 1960 to 2006 were calculated for 221 countries/territories worldwide. The predicted annual BC emissions from various countries (medians and semi-interquartile range) are listed in supporting Excel files. The predictions were compared with those reported in the literature (Figure 4).

\[
\text{logEF}_{BC} \text{(diesel)} = -0.4302 \text{GDP}_c - 0.01105Y_{3000} \\
+ 22.52, \quad n = 209, \quad LF = 16.8
\]

(3)

\[
\text{logEF}_{BC} \text{(gasoline)} = -0.7062 \text{GDP}_c - 0.004530 \\
Y_{3000} = 0.02180T + 15.09, \quad n = 176, \quad LF = 39.6.
\]

(4)

The significantly negative slopes of Y_{3000} (p = 0.001 and 0.048 for diesel and gasoline vehicles, respectively) imply that the countries that developed later (larger Y_{3000}) benefited from learning lessons from the countries that developed earlier (smaller Y_{3000}), confirming the effect of technology transfer from developed to developing countries on EFBC reduction. Although the overall LF decreased only slightly, LF for diesel vehicles was reduced from 2.7 to 1.2 for developing countries. Moreover, mean residuals (mean differences between the measured and model predicted logEFBC) for diesel vehicles were reduced from −0.0928 to 0.0003841 and from 0.3105 to −0.004177 for developed and developing countries, respectively, indicating a considerable elimination of the systematic overestimation for developing countries.

**Model Evaluation.** If the statistics, such as the mean, of all measured EFBC are used indiscriminately for estimating emissions from all countries at all times, large uncertainties will be introduced due to large variations of all EFBC measured. If, an EFBC for that particular country and year is calculated based on our model, the variation can be reduced substantially. To quantify the effectiveness of the model in overall uncertainty reduction, variations were compared between the two methods: A) without the model, the deviations of the measured logEFBC to the mean logEFBC of either developing or developed countries were calculated (assume the average EFBC for developing and developed countries were used in emission estimation), and B) with the model, the deviations of the measured logEFBC to the predicted logEFBC were calculated. For all the data used in this study, the ranges of deviations had reduced from 2.31 (method A) to 1.42 (method B) and from 1.63 (method A) to 1.19 (method B) for diesel and gasoline vehicles, respectively, showing approximately half to 1 order of magnitude reduction in overall variation. In an arithmetic scale, it means that 87 and 64% of the variation in the reported EFBC were eliminated by the models. Although many other factors in addition to country and model year can affect the measured EFBC, they could not be quantified without enough data at this stage. Hopefully, with more data collected in the future, especially for those countries not included here, the models can be further improved.

![Image](image)
and the United States were 92 and 85 Gg in 1996, while 66 and 102 Gg were reported for the same year. Similarly, our estimated BC emissions from vehicles in China were generally higher than that in the literature.3,4,10,42

Figure 5 shows the time trends of on-road vehicle BC emissions in (A) the United States and China; (B) the four economic regions including OECD90, ASIA, REF, and ALM; and (C) global total. Model uncertainties defined as semi-interquartile ranges are also shown in (A) and (C) as the shaded areas. It appears that the emissions follow the inverted U-shaped EKCs in most countries.43,44 The emissions reached peak values in OECD90 including the United States in the middle 1970s and have been increasing continuously up to now in rapid-developing ASIA and ALM countries like China and Brazil. As a result, the relative contributions of OECD90 countries to global BC emission decreased from 73.0% in 1960 to 15.1% in 2006, while the share of ASIA countries increased from 7.27% in 1960 to 42.8% in 2006. Globally, annual BC emissions have increased from 474 Gg in 1960 to 1294 Gg in 1978, decreased to 918 Gg in 1994, and started to increase again afterward, corresponding to superimposition of two EKCs of OECD90 and ASIA/ALM countries. Although satisfactory models were developed, the future prediction was not conducted primarily because technical advances have not been kept in steady step, and new emerging technologies may lead to fast decreases in EFs over a short period in the future, leading to large uncertainties in \( E_{BC} \) prediction. A typical example is the recently introduced diesel particulate filters, the effect of which cannot be captured in the model.45 The prediction of global total emission was compared with those reported in the literature (dots in Figure 5C).11 Although similar time trends were demonstrated before late 1970, a tipping point around 1978 followed by slow decrease are predicted based on our model because relative lower \( E_{BC} \) were used even though the number of vehicles had increased continuously. Because of the same reason (higher EFs in early years), the total emissions in 1960s and 1970s predicted in this study were higher than those reported previously.

The geographical distributions of global BC emissions from motor vehicles in 1976 and 2006 are shown for 1976 and 1° × 1° resolution was used for 2006.

Figure 5. Time trends of BC emissions from motor vehicles in the United States and China (A), four country categories of OECD90, ASIA, REF, and ALM, and global total (C) from 1960 to 2006. The results are presented as medians (curves) and semi-interquartile ranges (shaded areas) in (A) and (C) according to a Monte Carlo simulation. Global annual BC emissions from motor vehicles estimated by Bond et al.11 were also shown as blue dots in (C).

Figure 6. Geographical distributions of global BC emissions from motor vehicles in 1976 (A) and 2006 (B). Country-based mean values are shown for 1976 and 1° × 1° resolution was used for 2006.

ASSOCIATED CONTENT

Supporting Information
Table S1: Vehicle fleet composition of collected EF BC for various countries. Table S2: The 385 EFBC collected from the literature with information on vehicle type, model year, measured year, marketing country, measuring method, run cycle, odometer, GDP, ambient temperature, and \( Y_{3000} \). Table S3: Coefficients of equations (fuel consumption = a × GDP + b) for predicting fuel consumption for countries not included in IEA statistics by GDP within each country categories. Figure S1: Comparison between least-squares and reduced-major-axis regression models for predicting \( \log E_{BC} \) for diesel vehicles. This material is available free of charge via the Internet at http://pubs.acs.org.

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ACKNOWLEDGMENTS

Funding for this study was provided by National Scientific Foundation of China (41130754 and 41001343), Ministry of Chinese Environmental Protection (201209018), and Beijing Municipal Government (YB20101000101). We thank Dr. Douglas Richardson for polishing the English of the manuscript.

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