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Global time trends in PAH emissions from motor vehicles

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Abstract

Emission from motor vehicles is the most important source of polycyclic aromatic hydrocarbons (PAHs) in urban areas. Emission factors of individual PAHs for motor vehicles reported in the literature varied 4 to 5 orders of magnitude, leading to high uncertainty in emission inventory. In this study, key factors affecting emission factors of PAHs (EF_{PAH}) for motor vehicles were evaluated quantitatively based on thousands of EF_{PAH} measured in 16 countries for over 50 years. The result was used to develop a global emission inventory of PAHs from motor vehicles. It was found that country and vehicle model year are the most important factors affecting EF_{PAH}, which can be quantified using a monovariate regression model with per capita gross domestic production (purchasing power parity) as a sole independent variable. On average, 29% of variation in log-transformed EF_{PAH} could be explained by the model, which was equivalent to 90% reduction in overall uncertainty on arithmetic scale. The model was used to predict EF_{PAH} and subsequently PAH emissions from motor vehicles for various countries in the world during a period from 1971 to 2030. It was estimated that the global emission reached its peak value of approximate 101 Gg in 1978 and decreased afterwards due to emission control in developed countries. The annual emission picked up again since 1990 owing to accelerated energy consumption in China and other developing countries. With more and more rigid control measures taken in the developing world, global emission of PAHs is currently passing its second peak. It was predicted that the emission would decrease from 77 Gg in 2010 to 42 Gg in 2030.

Keywords

Polycyclic aromatic hydrocarbons (PAHs); Emission factor; Vehicle emission; Global inventory; Time trend

1. Introduction

Evidence collected for polycyclic aromatic hydrocarbons (PAHs) deposition on the Greenland icesheet suggested that the global emission of PAHs has been more or less constant since the beginning of the industrial period, and PAH emission reduction in developed countries had been offset by rapid increase in energy consumption of developing world (Masclet et al., 1995). It was estimated that the annual global emission of 16 PAHs on the United States Environmental Protection Agency priority list was 520 Gg in 2004 (Zhang and Tao, 2009).

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Appendix. Supplementary material: Supplementary data related to this article can be found online at doi:10.1016/j.atmosenv.2011.01.054.

Contribution of motor vehicles to global PAH emission was less than biomass burning and wildfire (Zhang and Tao, 2009). However, unlike biomass burning and wildfire, motor vehicle emission occurs mostly in urban areas where population densities are much higher than the global average. Consequently, relative contribution of PAHs from motor vehicles to human exposure risk is much higher than its contribution to emission itself. It was estimated that inhalation intake fraction, defined as the fraction of the emission that is eventually inhaled, of benzo(a)pyrene from motor vehicles in China was 4 times greater than the mean inhalation intake fraction of all sources (Zhang et al., 2009).

For inventory development, emission rate of a PAH compound from a given source was estimated as the product of the amount of fuel consumed and the emission factor (EF) of that particular source. Thanks to the efforts of many laboratories, PAH EFs (EF_{PAH}) were measured for various activities including motor vehicle exhaust, which were used for developing PAH emission inventories (Galarneau et al., 2007; Pacyna et al., 2003; Zhang et al., 2007). In most cases, if not all, means or medians of available EF_{PAH} with acceptable quality were adopted assuming that they are the best estimates for an average condition (Xu et al., 2006; Zhang et al., 2007). Occasionally, frequency distributions of measured EF_{PAH} were used for uncertainty analysis using Monte Carlo simulation (Xu et al., 2006; Zhang et al., 2007). A thorough review on EF_{PAH} for motor vehicles was conducted in this study and it was revealed that EF_{PAH} of individual compounds reported by different laboratories varied more than 4 orders of magnitude, primarily due to the influences of many factors including country where the test performed, vehicle model year (Y_m), vehicle type, fuel type, operation mode, and ambient temperature etc. (Baek et al., 1991). Due to such a large variation, uncertainty in EF_{PAH} was the primary source of the overall uncertainty in PAH emission inventories (Xu et al., 2006).

For a better assessment on air quality and exposure risk, it is necessary to develop emission inventories with lower uncertainty. Meantime, inventories with spatial and temporal resolutions are powerful in such assessment. The best way of reducing the overall uncertainty in emission inventories is to reduce the variation in EF_{PAH}. For instance, if the total variation in EF_{PAH} could be reduced by one or two orders of magnitude, 90 or 99% of the overall uncertainty in an inventory would be removed. It was proposed that if the main factors affecting EF_{PAH} can be identified and quantified, the reduction in EF_{PAH} variation would be realized by rectifying EF_{PAH} based on these factors. An additional advantage of this approach is that the rectified EF_{PAH} can be adopted for developing spatially or/and temporally resolved emission inventories, given that the detailed information on these factors is available.

This hypothesis was tested in this study and the specific objectives were 1) to identify key factors affecting EF_{PAH} for motor vehicles; 2) to develop a quantitative model for predicting EF_{PAH} based on the main factors identified; and 3) to develop a global PAH emission inventory for motor vehicles in various countries over time.

2. Methodology

2.1. Collection of EF_{PAH} for motor vehicles

A database of EF_{PAH} for motor vehicles was established based on a thorough literature review and all the references are listed in Supplementary S1. Sixteen PAHs included in the database and evaluated in this study were naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz(a)anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DahA), indeno(l,2,3-cd)pyrene (IcdP), and benzo(g,h,i)perylene (BghiP). If a test was conducted in an extreme driving mode, using an uncommon fuel type, or using a modified vehicle, the data were excluded in this study. Grubb's test was conducted to test statistical outliers for each of the 16 PAHs after log-transformation, and no outlier were detected at a significant level of 0.05. A total of 2855 EF_{PAH} from 282 individual tests conducted in 16 countries were adopted. The vehicles used for these tests were produced from 1956 to 2005. The EF_{PAH} reported in different units were converted to mg PAH per ton of fuel combusted (mg/t). For a few reported as mass per distance traveled, vehicle fuel consumption efficiencies as functions of year and vehicle type were adopted to convert them into mg/t (Davis et al., 2010; EA, 2003; USEPA, 1995).

A number of test conditions were recorded for EF_{PAH} measurements in the literature. The often reported parameters include fuel type, vehicle type, testing year, Y_m, measurement method, country where the vehicle was marketed and operated, driving speed, catalytic converter, driving cycle, fuel consumption, starting mode, odometer reading, and particulate matter fraction etc (see S2 in Supporting Information). For those measured in a tunnel test, the Y_m was derived by subtracting the average vehicle age of the country from the testing year (Infobank, 2010; USEPA, 1995).

2.2. Modeling the influences of key factors

Among the above mentioned factors, those recorded by most EF tests are country, Y_m, and vehicle type. These factors were analyzed for their effects on EF_{PAH}. Both bivariate and monovariate regression models were conducted using ordinary least-square method. For quantifying country variable, a number of socio-economic parameters were tested by trial-and-error and it was found that gross domestic production per capita (purchasing power parity, GDP_c) is the best indicator for describing the difference in developing status among countries. It was found that GDP_c can also be used for describing temporal trends of developing status of a given country over time. Based on these results, a set of monovariate regression models were developed for EF_{PAH} of all individual PAHs based on log-transformed EF_{PAH} and GDP_c. Data of GDP_c were from the World Bank (The World Bank, 2010).

2.3. Time series of country-level PAH emission

The EF_{PAH} prediction models were applied to project PAH emissions from motor vehicles for various countries in the world from 1971 to 2030. Consumptions of gasoline and diesel by motor vehicles in individual countries from 1971 to 2006 were derived from a statistical database (IEA, 2006). Increasing rates of energy consumption in scenario A1B derived by the Intergovernmental Panel on Climate Change were adopted to estimate petroleum consumption by motor vehicles in various countries in the future, using 2006 as the base year (IPCC, 2001; Nakicenovic et al., 2000). For a comparison, the predictions were also made based on the other three IPCC energy consumption scenarios of A2, B1, and B2 (Nakicenovic et al., 2000).

2.4. Uncertainty analysis

The overall uncertainty of the model for predicting emission of PAHs from motor vehicles was estimated using Monte Carlo simulation, which was run 10,000 times with given coefficients of deviation for energy consumption (0.05 for the historical data and increased linearly from 0.05 to 0.10 from now to 2030) and the actually calculated emission factor distributions.

3. Results and discussion

3.1. Variations in EF_{PAH} for motor vehicles

Numerous tests have been conducted to measure EF_{PAH} for motor vehicles in many countries over the past 50 years. The EF_{PAH} collected in this study are summarized in Supplementary S3. For all individual PAHs, the reported EF_{PAH} vary widely. For example, 233 BaP EFs range from 0.02 to 1910 mg/t and 220 FLA EFs vary from 0.522 to 20400 mg/t, both showing striking variation of around five orders of magnitude. The coefficients of variance of EF_{PAH} for individual compounds were from 128% (ACE) to 306% (BghiP). For the 16 PAHs, the EF_{PAH} were all leptokurtically and right-skewed distributed with coefficients of skewness from 2.33 to 8.44 and the coefficients of kurtosis from 6.4 to 89.9. After log-transformation, these coefficients were reduced to values close to zero (*t*-test, $\alpha > 0.05$, see S3 in Supporting Information), suggesting that they are all log-normally distributed (see S4 in Supporting Information).

3.2. Factors affecting EF_{PAH} for motor vehicles

Of the many factors investigated, the country where EF_{PAH} were tested is one of the most critical factors affecting EF_{PAH} for vehicles simply because regulations on emission control are very different among countries. Vehicle emission is rigorously regulated in the United States and EU, while there is literally no regulation on vehicle exhaust in poor countries (ADB, 2003; Timilsina and Dulal, 2009). It is reasonable to expect that a car running in a street of Paris is usually more technically advanced than the one operated in Mexico City at the same time in the terms of emission. Such a difference can be clearly seen in the EF_{PAH} database used in this study. For example, arithmetic mean BaP EFs (EF_{BaP}) for vehicles produced from 1994 to 1996 varied widely among the United States (4.56 ± 3.19 mg/t), Sweden (6.13 ± 4.94 mg/t), Denmark (19.6 ± 25.3 mg/t), Australia (70.0 ± 64.4 mg/t), and Brazil (184 ± 73 mg/t) (Cadle et al., 1999; Chellam et al., 2005; De Abrantes et al., 2004; Durbin et al., 1998; EA, 2003; Leonidas and Zissis, 2000; Nelson et al., 2008; Riddle et al., 2007; Schauer et al., 1999; Wingfors et al., 2001). Apparently, the decreasing gradient is closely related to the socio-economic development status of these countries.

In a given country, or in a group of countries with similar development status, EF_{PAH} for a specific type of vehicles generally decreased over time due to enforcement of new regulations and development of modern control technologies. From 1992 to 2009, EU has issued five emission standards from Euro I to V for passenger cars and Euro VI was scheduled to be in force in 2014 (Timilsina and Dulal, 2009). An important step towards vehicle emission control in the United States was taken in 1970 when the Congress passed the Clean Air Act, which was further amended in 1977 and 1990 (Timilsina and Dulal, 2009). Development of most new emission control technologies were primarily driven by these regulations. The first-generation catalytic converter introduced in mid 1970s helped to cut car emission substantially (USEPA, 1999). After the three-way catalysts with on-board computer and oxygen sensor hit the market in 1980s, more evident reduction in car emission was achieved (USEPA, 1999). Although PAH emission was not regulated directly, the new technologies aiming at other pollutants including particulates and nitrogen oxide helped to trim down PAH emission “unintentionally”. For the same reason, vehicle emission was also gradually reduced in developing countries over years, although the progress was hysteretic (ADB, 2003). For example, China III emission standard, which is similar to Euro III, already went into effect in China in 2007 (Timilsina and Dulal, 2009).

Taking the United States and European countries as examples, the time dependence of EF_{PAH} is illustrated by plotting EF_{BaP} against Y_m in Fig. 1, demonstrating significantly negative correlations ($p < 0.005$). In the United States, EF_{BaP} for light-duty gasoline vehicles decreased approximately two orders of magnitude over 30 years from 300 mg/t in early 1970s to 1.4mg/t in 2000, which was equivalent to a half-life of 3.9 years. In European countries, EF_{BaP} for all types of vehicles decreased from 113 to 3.0 mg/t with a half-life of 5.7 years over the past 3 decades. A similar trend was observed for other countries including Brazil, Australia, and Mexico.

There are lines of evidence suggesting significant difference in EF_{PAH} among different types of vehicles. For vehicles produced in the United States in late 1990s, EFs of PHE, FLA, and PYR were 3.01, 1.02, and 2.01 mg/t for light-duty gasoline vehicles and were 91.4, 47.6, and 67.1 mg/t for heavy-duty diesel vehicles, respectively (Riddle et al., 2007). It was also noted that the EF_{PAH} measured using different methods could be very different. For example, it was reported that EF_{PAH} derived from real world test were significantly higher than those from dynamometer tests (Kristensson et al., 2004), and EF_{PAH} observed in a tunnel test were considerably higher than those measured in a roadside test (Wingfors et al., 2001). Based on the result of an investigation on the influences of fuel type and driving conditions on PAH emission, it was revealed that the emission increased with either higher PAH content in the fuel or higher cruising speed (Westerholm et al., 1992; Westerholm and Li, 1994). It was also found that PAH emission over transient driving cycles was higher than that over steady-state driving cycles (Kado et al., 2005). Partition of PAHs between gaseous and particulate phases is temperature dependent (Grieshop et al., 2006), leading to different EF_{PAH} at different ambient temperatures (Cadle et al., 1999). Other factors affecting EF_{PAH} include vehicle age and load, lubricant oil (Ravindra et al., 2008), cold starting (Paturel et al., 1996), as well as the errors associated with sample collection and analysis.

3.3. Prediction of EF_{PAH} for motor vehicles

Although a dozen of factors were found to have significant effects on EF_{PAH}, country where vehicles operated and Y_m are the most important. For this reason, records on these two factors are available for almost all EF_{PAH} measurements in the database. The factor of “country” is an attribute reflecting the socio-economic-technical development status. A number of relevant quantitative variables including GDP_c, energy consumption, population density, and income were evaluated for their relationship with EF_{PAH}. Among them, GDP_c was found to be the best one in terms of predicting log-transformed EF_{PAH}. Therefore, bivariate linear regression models were developed for predicting EF_{PAH} (log(mg/t)) using GDP_c (1000USD) and Y_m (AD) as two independent variables for the 16 PAHs individually at the first place:

$$\log(EF_{PAH})=a \times GDP_c+b \times Y_m+C,$$

where a (log(mg/t)/(1000USD)), b (log(mg/t)), and C (log(mg/t)) are regression coefficients of the bivariate regression models and the calculated results of these coefficients, together with p values and coefficients of determination, can be found in Supplementary S5. Although Y_m is also a critical factor affecting EF_{PAH} (Fig. 1), the results of the regression modeling revealed that this variable was not significant in most cases (mean p values for the 16 PAHs were 0.02 and 0.27 for GDP_c and Y_m, respectively). Moreover, it was found that Y_m was significantly correlated with GDP_c for individual countries ($p < 0.05$ for the Unite States, European countries, and Australia), indicating non-orthogonality of the two variables. Intrinsically, both factors of country and Y_m represent the status of technical evolution and social-economic development, although the former represents geographical difference while the latter signifies temporal change. As such, the EF_{PAH} measured in developed countries were generally lower than those in developing countries at the same period, while EF_{PAH} reported for a given country, either developed or developing, generally decreased over time. In fact, both variables were found to be correlated with GDP_c, which was then used as an integrated independent variable for a monovariate regression model for predicting EF_{PAH}. A set of monovariate regression models were developed for predicting EF_{PAH} (mg/t) for individual PAHs based on GDP_c (1000 USD):

$$\log(EF_{PAH})=k \times GDP_c+C,$$

where k (log(mg/t)/(1000USD)) and C (log(mg/t)) are slope and interception, respectively. Physically, k is a decreasing rate constant of $\log(EF_{PAH})$ as GDP_c increases, while 10^C (mg/t) represents the emission factor of a “prototype vehicle” without emission control. It appears that the relationship between EF_{PAH} and GDP_c follows a typical single exponential function with negative k values, which is universally used to model decay processes. The results of the modeling are presented in Fig. 2 using ACY, PYR, CHR, and BaP as examples and similar results for all other PAHs studied are provided in Supplementary S6.

It is evident that the overall uncertainty in EF_{PAH}, subsequently, in emission estimation can be substantially reduced after taking country and model year into account. According to the models developed, r^2 varied from 0.14 for ACE to 0.42 for DahA with a mean value of 0.29 for the 16 PAHs, indicating that almost 30% of the variation in $\log(EF_{PAH})$ can be explained. It was estimated that 29% variation reduction in log-scale for EF_{PAH} with variations of 3–5 orders of magnitude equivalents to over 90% of the reduction arithmetically in overall variation. No doubt, more than an order of magnitude reduction in variation of EF_{PAH} can greatly improve the quality of the emission estimation.

Although significant difference in PAH emission between light-duty gasoline and heavy-duty diesel vehicles was reported (Riddle et al., 2007). The models were not substantially improved by separating the two types of vehicles in this study. In fact, when the two types of vehicles were modeled separately, the results were similar to each other with only a single exception of NAP (see S7 in Supporting Information). Therefore, a unified model was recommended for both gasoline and diesel vehicles and further improvement may become possible in the future when more data are available. A number of other factors including method for EF_{PAH} measurement, ambient temperature, and driving mode were also investigated in this study by checking the interrelationship between these factors and the residues of the monovariate regression models. Unfortunately, the sample size reported for these parameters were limited and were not enough for supporting further improvement of the model. Still, there is potential for further improvement of the model if only more data can be collected.

3.4. Model validation

Based on the model calculated EF_{PAH} and petroleum consumption by motor vehicles, either historically recorded or predicted for years to come (IPCC, 2001; IEA, 2006; Nakicenovic et al., 2000), annual emission of PAHs from motor vehicles for all countries around the world were calculated for a period from 1971 to 2030. The models were validated by comparing our results with those reported in the literature for a number of countries. Fig. 3 shows several examples of the comparison. Good agreement was found between our results and those reported for the United States, Canada, United Kingdom, and Netherlands (EEA, 2010; Environment Canada, 2010; NAEI, 2010; USEPA, 2010) (see S8 in Supporting Information). Unfortunately, our results do not agree with those of some developed European countries such as France, Germany, Italy, and Sweden (EEA, 2010) (see S8 in Supporting Information). In general, in case of the inconsistency, continuous decrease in annual emission of PAHs over time was predicted by our model for these European countries, while an increasing trend was reported in the literature (France in Fig. 3 is a typical example). It is very likely that although the numbers of motor vehicles have increased over these years, even quicker decrease in EF_{PAH} has lead to decrease in annual emission in all developed countries. For example, the measured EF_{PAH} in Sweden decreased more than one order of magnitude over the last fifteen years, in a pace much faster than the increase rate of vehicle number (Bergvall and Westerholm, 2009; Westerholm et al., 1996, 2001; Wingfors et al., 2001). Another piece of evidence was that the ambient air PAH levels in major cities of western European countries decreased in past decades (Menichini et al., 2006; Schauer et al., 2003; Valerio et al., 2009). For example, total concentrations of several PAHs in ambient air in traffic-oriented sites in two Italian cities including Genoa and Siracusa decreased significantly (Valerio et al., 2009). For major European countries including United Kingdom, Sweden, Switzerland, Netherlands, Greece, Germany, France, and Belgium where most EF_{BaP} were measured, the model predicted half-life of BaP (5.8 year) agrees excellently with that derived directly from the measurements (5.7 year). For this reason, we recommend that the methodology used by the European Environmental Agency for PAH emission estimation should be reviewed and updated.

3.5. Difference in EF_{PAH} among PAH compounds

Although EF_{PAH} of different PAHs for motor vehicles varies widely, the dependence on GDP_c (eventually on country and vehicle model year) follows a similar pattern. Moreover, it was found that the k values of the regression models for individual PAHs were log-transformed vapor pressure $\log(P_s)$ dependent (see S9 in Supporting Information). Positive correlation between k and $\log(P_s)$ was significant ($p < 0.10$), suggesting that during the course of technical development, the efficiencies of the emission reduction for lower P_s PAHs were higher than those for higher P_s PAHs, likely due to the fact that the particulate matter, which the majority of lower P_s PAHs are bound to, is one of the main targeted pollutants in emission control. Still, more data is needed in the future to confirm this trend.

3.6. Time trend of PAH emission from motor vehicles in the world

Annual emissions of PAHs from motor vehicles for all countries around the world from 1971 to 2030 are listed in Supplementary S10. The prediction beyond 2006 was based on IPCC A1B scenario. Globally, the total annual emissions of 16 PAHs from motor vehicles were 82.7 (56.1–122.9 as semi-interquartile range from Monte Carlo simulation), 94.1 (64.5–137.8), 71.7 (50.1–102.8), 67.5 (47.0–97.3), and 76.5 (49.7–116.0) Gg in 1971, 1980, 1990, 2000, and 2010 and will be 69.2 (39.4–119.6) and 41.6 (21.0–81.1) Gg in 2020 and 2030, respectively. Time trend of global emission from 1971 to 2030 is presented in Fig. 4 (top left panel). In addition, the total emissions of 16 PAHs in the United States, Germany, Russia, China, and India are also presented in Fig. 4. The emissions in the United States and Germany decreased from 31.7 to 3.20 Gg in 1971 to 0.836 and 0.269 Gg in 2010, respectively. Similar trends can be found for the entire developed world. These countries were the major contributors in 1970s and 1980s leading to the first peak in global emission during this period. On the other hand, PAH emission in China and India increased from 1.648 (1.132–2.394) and 0.890 (0.628–1.272) Gg in 1971 to 21.5 (12.9–34.8) and 6.07 (3.85–9.44) Gg in 2010. Fast increase in PAH emissions during last 10–20 years can also be seen in other developing countries in economic transit periods. Globally, although total emission reduced approximately 30% from 1980 to 1990 after emission control measures were introduced in developed countries, continuous decrease in these countries has been offset by the increase in rapidly developing countries. Fortunately, the global vehicle emission of PAHs is currently passing over the second peak and will decrease in the future primarily due to the predicted emission decrease in China and other developing countries. In China, for example, although the number of vehicles increased at annual rate of 21%, average emission of each vehicle also decreased approximately 18% each year. In fact, China III emission standard equivalent to Euro III was introduced in 2007, and China IV emission standard equivalent to Euro IV will be enforced in 2011 (Timilsina and Dulal, 2009). As a result, the emission from China is approaching its turn point currently and the relative contribution of China to the global motor vehicle PAH emission will decrease from 28% in 2010 to 21 and 10% in 2020 and 2030, respectively, if only the current trend of controlling effort would continue. Time trend pattern of EF_{PAH} of India is similar to that of China except that the peak will not be reached till 2020 (The peak between 1995 and 2001 was likely due to error in petroleum consumption data). For comparison, the prediction beyond 2006 was also made based on other three IPCC energy scenarios of A2, B1, and B2 (Nakicenovic et al., 2000) and the results are shown in Supplementary S11.

Since the emission from other major sources including wildfire, biomass burning, and consumer product usage will not change as quickly as those from motor vehicles, the relative contribution of motor vehicles to total PAH emission from all sources is expected to decrease, which would lead to a decrease in inhalation exposure of urban residences to PAHs.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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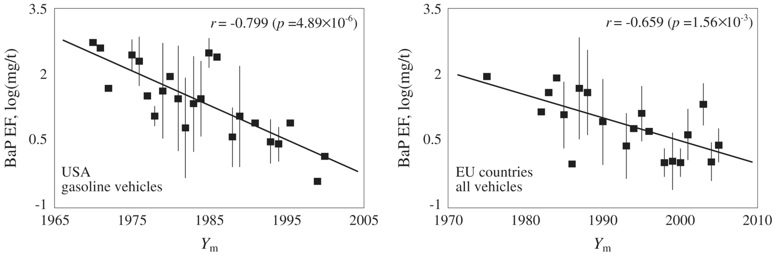

Fig. 1.

Relationship between the measured EF_{BaP} and Y_m for A) light-duty gasoline vehicles in the United States from 1970 to 2000 (left panel) and B) all types of vehicles in European countries from 1975 to 2005 (right panel). Annual means and standard deviations of EF_{BaP} are presented in log-scale.

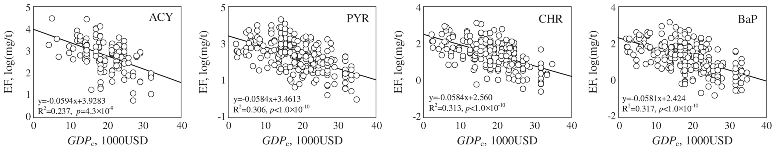

Fig. 2.

Linear dependence of $\log(EF_{PAH})$on GDP_c. The results for ACY, PYR, CHR, and BaP are presented as four representative compounds.

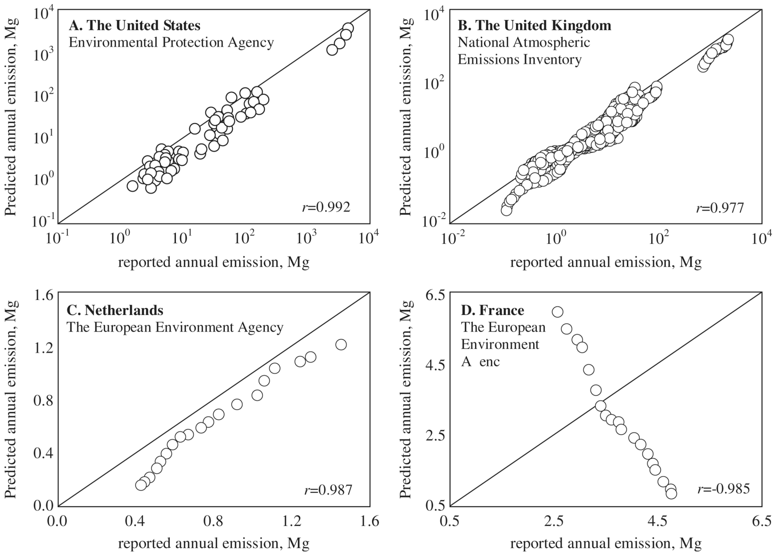

Fig. 3.

Comparison of annual PAH emissions for motor vehicles between our model estimation and the literature reported values. A. emissions of individual PAH compounds in the United States (USEPA) in 1999, 2002, 2005, and 2008; B. emissions of individual PAH compounds in the United Kingdom from 1990 to 2007; C and D. emissions of BbF, BkF, BaP, and IcdP in Netherlands and France from 1990 to 2008, *r* is the Pearson's coefficient.

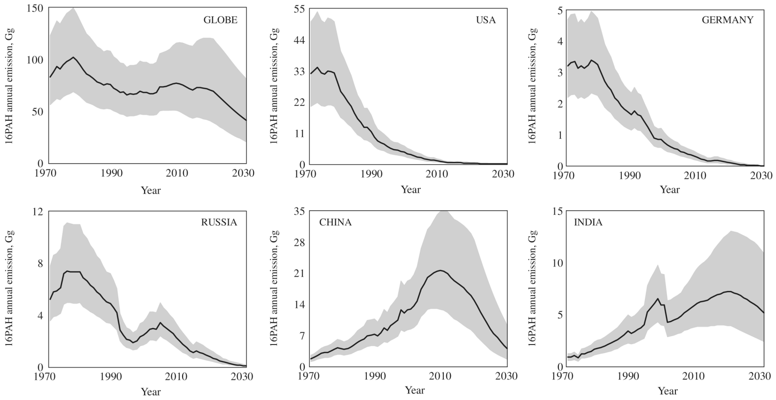

Fig. 4.

Time trends of motor vehicle emissions of 16 PAHs during a period from 1971 to 2030 for the world and several representative countries including the United States, Germany, China, India, and Russian. The predicted results are presented as medians and semi-interquartile ranges of Monte Carlo simulation and the latter are used to describe the overall uncertainties of the prediction.