Atmospheric Environment 101 (2015) 166–176

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Modeling airborne benzo(a)pyrene concentrations in the Czech Republic

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HIGHLIGHTS

- Models for estimation of daily and monthly ambient B[a]P levels were developed.
- The concentrations seasonality was addressed by combined CART–MLR models.
- The models reproduced very accurately monthly mean ambient B[a]P concentrations.
- Spatial across-site extrapolations revealed reliable models performance.
- Temporal extrapolations revealed comparable errors to the spatial extrapolations.

ARTICLE INFO

Article history:
Received 2 September 2014
Received in revised form
12 November 2014
Accepted 14 November 2014
Available online 15 November 2014

Keywords:
PAHs
B[a]P
Multivariate linear regression
Classification trees
Air pollution monitoring

ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are complex hazardous organic compounds that are introduced into the atmosphere as by-products of partial combustion processes. For common atmospheric conditions, the large molecular weight PAHs, such as benzo(a)pyrene (B[a]P), are found in the particulate phase and are believed to account for a considerable amount of the fine particulate matter toxic potential. Nonetheless, unlike meteorological variables and criteria pollutants, PAHs are very rarely monitored on a routine basis in most parts of the world. We present methodology for development and evaluation of a model for estimation of daily and monthly ambient B[a]P concentrations. The model utilizes a very large ambient B[a]P database from three sites in the Czech Republic. The difficulties faced when dealing with ambient PAH data are discussed. Model performance was evaluated by a complete internal-, external-, and temporal cross validations. The models reproduced very accurately monthly mean ambient B[a]P concentrations and provided acceptable daily mean B[a]P concentrations. Spatial extrapolations resulted in small deterioration of the models' performance. The temporal backward extrapolation revealed comparable errors to the spatial extrapolations in spite of the dramatic emissions reduction in the early years of the study period.

1. Introduction

Monitoring of ambient pollutants is required for air resources management and for exploring relationships between air pollution and health outcomes. For example, environmental epidemiology studies normally use air quality monitoring data to derive exposure metrics, and may suffer from considerable uncertainties in sparsely monitored regions. Criteria air pollutants (NO_x, NO, NO_2, CO, SO_2, O_3, PM_{10} and $PM_{2.5}$) are monitored on a wide spatiotemporal scale global-wise. Yet, many other pollutants are not monitored regularly at most places. This situation is not expected to change dramatically in the coming years due to technological and mainly financial limitations. In such cases, it is oftentimes required to rely on indirect means for estimating ambient concentrations of harmful pollutants.

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http://dx.doi.org/10.1016/j.atmosenv.2014.11.031
1352-2310/

Polycyclic aromatic hydrocarbons (PAHs) are a class of complex organic compounds with two or more benzene rings. Anthropogenic sources of PAHs include incomplete combustion of organic fuels (residual oil, wood, coal, gasoline and diesel). Over the years, PAHs have received increased attention, since some of them are highly carcinogenic or mutagenic. For example, the International Agency for Research on Cancer classifies benzo(a)pyrene (B[a]P) in Group 1, i.e. an agent that is carcinogenic to humans (IARC, 2014). Under certain atmospheric conditions and due to their low vapor pressure, the large molecular weight PAHs are believed to account for a considerable amount of the fine particulate matter toxic potential (Dejmek et al., 2000; Binkova and Sram, 2003; Ohura et al., 2004; Hertz-Picciotto et al., 2007; Rubes et al., 2007; Soucy et al., 2007; Sram et al., 2011, 2013). Nonetheless, although PAHs are hazardous substances (in particular, particle bound PAHs; Jia et al., 2011), spatiotemporal data on ambient PAHs levels and reliable long-term records for studying their chronic effects are limited (Srogi, 2007). Namely, unlike meteorological variables and criteria pollutants, PAHs are very rarely monitored on a routine basis in most parts of the world. Consequently, most studies on human exposure to ambient PAHs are based on relatively short measurement campaigns. Oftentimes, B[a]P (and B[a]P equivalent) concentrations are used as a marker of exposure to the total mixture of ambient PAHs (Matthias et al., 2009).

PAHs, and B[a]P in particular, are examples of pollutants that are regulated without sufficient environmental information, due to lack of data on their spatiotemporal concentrations. Since the PAHs monitoring density is not expected to increase dramatically in the coming years, indirect means for estimating ambient PAH concentrations are sought. In general, air quality models can be divided into two types: mechanistic models and empirical models. Mechanistic models use mathematical equations that mimic the atmospheric processes that govern pollutant dispersion, reaction and deposition. Empirical models are data driven models that relay on statistical relationships between explanatory and dependent variables. Unlike mechanistic models, empirical models do not require emissions inventories or a detailed and accurate description of the complex physicochemical processes involved, since these processes are captured by the data (i.e. all the information about the phenomena is contained within the records). However, a very large database is normally required to be able to develop a reliable empirical model. To date, only a handful of studies addressed methods for estimating ambient B[a]P concentrations. Matthias et al. (2009) developed a mechanistic model for calculating B[a]P concentrations over Europe on a 54 × 54 km^2 grid. The model was evaluated against measurements taken at 8 background sites. Whereas the concentration seasonal pattern was captured, a concentration underestimation of ~50% was obtained with model results showing much lower variability than the observations. The discrepancies were attributed to uncertainties in the PAHs emissions inventory and degradation rates, resulting from the highly variable combustion efficiency and fuel quality (Bostrom et al., 2002). In contrast with the mechanistic approach, Lobscheid et al. (2007) and Callen et al. (2010) presented multivariate linear regression models for estimating daily mean logarithmic transformed B[a]P concentrations. Whereas model results were obtained for a relatively large geographical scale, suitable for epidemiological studies, the models were developed only for days in which the measured concentrations were higher than the instrument detection limit, thus introducing bias to the prediction.

This work reports the development of statistical models for estimating ambient daily and monthly mean B[a]P concentrations at three locations in the Czech Republic. We limited the explanatory variables to criteria pollutants and to common meteorological variables, which are frequently measured and enable model evaluation and application at different locations and times. Each model consists of two sub modules: a classification and regression tree model for estimating low B[a]P concentrations and a multivariate linear regression model (MLRM) for estimating B[a]P concentrations on days in which its levels are expected to be above the detection limit. The models' performance was examined by means of a complete internal-, external-, and temporal cross validation.

2. Methods

2.1. Study area

Data from four monitoring stations in the Czech Republic were available for this study. The Teplice (population of ~51,000) site (TP) is a highly industrialized city in northern Bohemia, 90 km northwest of Prague, with reported high air pollution levels due to emissions from local industry, power plants, strip-mining of coal and associated industries, residential space heating, and motor vehicles. Under severe meteorological conditions, such as wintertime inversions, pollutant emissions tend to be trapped within the valleys of the northwestern Czech Republic, where Teplice resides (Pinto et al., 1998). Over the study period (1995–2006), the major air pollution sources have been closed or their emissions diminished considerably (Benes et al., 2006). The Prachatice (population of ~12,000) site (PT) is located 130 km south of Prague in a rural and relatively sparsely populated area (Sram et al., 1996). The area is considered to be less polluted than the TP area (Pinto et al., 1998) yet, as will be discussed below, a major PAHs source impacted the PT site making it unsuitable for model development within this study. The Prague Smichov (PRG-SM) monitoring station (Prague population ~1,187,000) is located in an urban commercial area with high traffic density. The Prague Libus (PRG-LB) monitoring station is considered to be the background station for Prague as it is characterized by residential and suburban areas (Binkova et al., 2003).

2.2. Data

The database available for this study included monitored concentrations of NO_x, NO, NO_2, SO_2, CO, O_3, PM_{10} and $PM_{2.5}$ and 13 PAHs as well as ambient temperature records. In general, the reporting frequency of SO_2, NO_x, NO, NO_2, CO, O_3 and the temperature (daily means) is much higher than of PM_{10}, $PM_{2.5}$ and PAHs, with the number of PAH measurements differing considerably between the years. The B[a]P database we used included ~6000 measurements, about an order of magnitude larger than the databases used in previous studies (e.g. Lobscheid et al., 2007). PAHs concentrations were measured 2–3 times a week using Versatile Air Pollutant Samplers (VAPs; URG Corp., USA) following the sampling procedure described by Pinto et al. (1998). In brief, fine particles (<2.5 μm) were captured on a 47 mm quartz filter and analyzed by high-performance liquid chromatography for various PAH concentrations. The B[a]P measurement detection limit was 0.2 ng/m^3, with all measurements below 0.2 ng/m^3 logged as 0.1 ng/m^3. The number of measurements of each pollutant in each year at the four monitoring sites is presented in the Supplementary Material. The monitoring datasets used in this study covered the years 1995–2006 in the TP and PT monitoring sites and the years 2000–2006 in the PRG-SM and PRG-LB monitoring stations. Ambient temperature records at PRG-LB site were not available until the end of 2004. Since the absence of these data was crucial for model development and as the temperatures in the two sites in Prague were found to be very similar and highly correlated (0.99<, data not shown), temperature measurements from PRG-SM were used at PRG-LB.

Processing each of the PAHs independently greatly complicates the modeling work, since different locations are often characterized by distinct PAH species profile. Moreover, since the toxicity of different PAHs vary considerably the common approach is to lump all the PAHs together (total PAH concentration, tPAH), weighting their individual concentrations according to their B[a]P toxic equivalence. The B[a]P toxic equivalent factor, TEF (Tsai et al., 2004; Chen and Liao, 2006), enables comparing PAH concentrations at different locations using a uniform scale. Moreover, from a health risk perspective, the total PAHs carcinogenic potential (in terms of B[a]P equivalent toxicity) is oftentimes more important than the concentrations of individual PAHs. As will be shown, B[a]P concentrations were correlated with the tPAH concentration (both the gaseous and particulate phases) and with the equivalent carcinogenic potential of the tPAH. Hence, we used B[a]P as a marker of exposure to all the PAHs, enabling the development of a model that can be spatially extrapolated.

2.3. Classification and regression tree model

A binary classification tree has been developed for dividing the explanatory variables data into two subgroups based on the observed B[a]P concentrations. The threshold value was set as the B[a]P measurement detection limit. The output of this model is a set of rules for dividing the explanatory variables into one group that can be used for quantitative estimation of B[a]P concentrations, and another group that represent conditions when B[a]P concentrations are estimated to be below the threshold value and a preset low value of 0.1 ng/m^3 is assigned. In general, binary tree classifiers are constructed by repeated splits of subsets of the dataset into two descendent subsets, beginning with the whole dataset at the first generation. In practice, the model was developed such that each tree bifurcation divides the database into two distinct subgroups with the aim of decreasing the overall within-group heterogeneity (Breiman et al., 1984). We used the Gini index as a measure of the impurity (heterogeneity), securing that at each consecutive node the two data subsets are maximally distinct. The Gini index represents the weighted average relative difference across all subgroups.

2.4. Multivariate linear regression

Multivariate linear regression models were developed for estimating ambient B[a]P concentrations on days in which the concentrations are expected to be above the threshold value. The regression takes the following form,

$$Y_i = \beta_o + \beta_1 X_1 + \beta_2 X_2 + \ldots + \beta_n X_n + \varepsilon, \quad (1)$$

where Y_i is the predicted daily average B[a]P concentration, β_0 is an intercept, $\beta_1 \ldots \beta_n$ are coefficients associated with the explanatory variables $X_1 \ldots X_n$ and ε is the residual error. For such a model to be valid the residuals should be normally distributed and independent of the explanatory variables. Based on data availability, the potential predictors were criteria pollutant concentrations and the ambient temperature. Two alternatives for reducing the number of model variables were examined: stepwise regression and constrained stepwise regression. Stepwise regression is a common method for reducing the number of variables in a model to a minimum. However, this method may not have a single solution and its result depends on the chronology of individual steps. Furthermore, if applied independently for a number of datasets (e.g. representing different sites) different predictors may be selected as optimal model variables. This poses severe limitations on the model spatial transferability, i.e. application in different sites. To ensure that all the local models use identical variables, we used constrained stepwise regression. In this approach, all the models are developed simultaneously in a stepwise manner but at each step the same variables are selected in all the models in an optimal way.

2.5. Model evaluation

The performance of the combined models developed in this study was evaluated at three levels: (1) an internal cross validation – data from each site were divided into two subsets. The learning subset was used for model parameterization and the test subset was used for the evaluation process. (2) An external cross validation – model parameterization was done using data from one site whereas model evaluation was done using data from a different site. (3) A spatiotemporal evaluation – model parameterization was done using data from 2000 to 2006 from three sites and model evaluation was done using data from TP in the early years, 1995–1999. The models' performance was evaluated against daily B[a]P concentration observations and for monthly average B[a]P levels, using monthly mean daily predictions and observations. Both qualitative (visual inspection, descriptive statistics of the magnitude and trend of model over/under prediction) and quantitative measures were used for model evaluation. The quantitative model performance measures used were the Pearson product–moment correlation coefficient (r) (Chang and Hanna, 2004), the coefficient of determination (R^2), the root mean square error (*RMSE*) (Borrego et al., 2008), the mean absolute difference (*MAD*) and the fraction of the predictions within a factor of two relative to the observations (*FAC2*) (Chang and Hanna, 2004). A summary of the various measures and the differences between them, promoting the use of a basket of such measures, has recently been published by Simon et al. (2012).

3. Results

3.1. Air pollution trends

Fig. 1 depicts annual mean concentrations of SO_2, NO_x, O_3 and $PM_{2.5}$ at the four Czech monitoring sites during the study period. A clear decrease in the yearly mean SO_2 and $PM_{2.5}$ concentrations is seen in the PT and TP monitoring sites until 1999. From 2000 to 2006 the yearly mean concentrations are stable, with one clear peak in 2003 $PM_{2.5}$ concentrations attributed by the Czech Hydrometeorological Institute to the very dry meteorological conditions that occurred during that year (CHMU, 2004). Fig. 1 depicts that the PRG-SM and TP monitoring sites exhibit the highest NO_x concentrations and the lowest O_3 concentrations, contrary to PT. The high NO_x and low O_3 pattern seen in PRG-SM and TP is common to areas where background ozone levels are partially titrated by locally emitted NO_x (Comrie, 1997). The opposite NO_x/O_3 trend seen in PT suggests that the PT monitoring site measured background O_3 concentrations and is not affected by local emissions.

Annual mean concentrations of total PAHs (tPAHs) and of B[a]P are presented in Fig. 2. Since the measurement frequency varied throughout the year, with a higher number of measurements and higher concentrations in the winter, a simple arithmetic mean of the observations would result in overestimated annual mean concentrations. Hence, annual mean concentrations were calculated based on monthly means. Concentrations of tPAH and B[a]P in TP and PT were similar until 1999 although based on the understanding gained from Fig. 1 the two sites could be expected to reveal different trends. The high PAH concentrations at PT prior to 1998 resulted from a significant point source in close proximity to the monitoring site (Dejmek et al., 2000). Thus, the PT monitoring

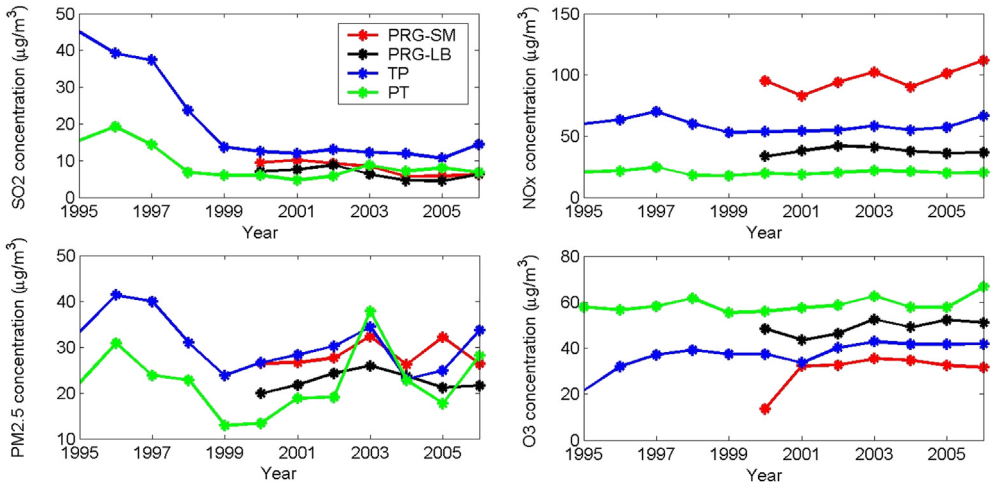

Fig. 1. Annual mean concentrations of SO_2 (upper left), NO_x (upper right), $PM_{2.5}$ (lower left), and O_3 (lower right) in the TP (blue), PT (green), PRG-SM (red) and PRG-LB (black) monitoring sites. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

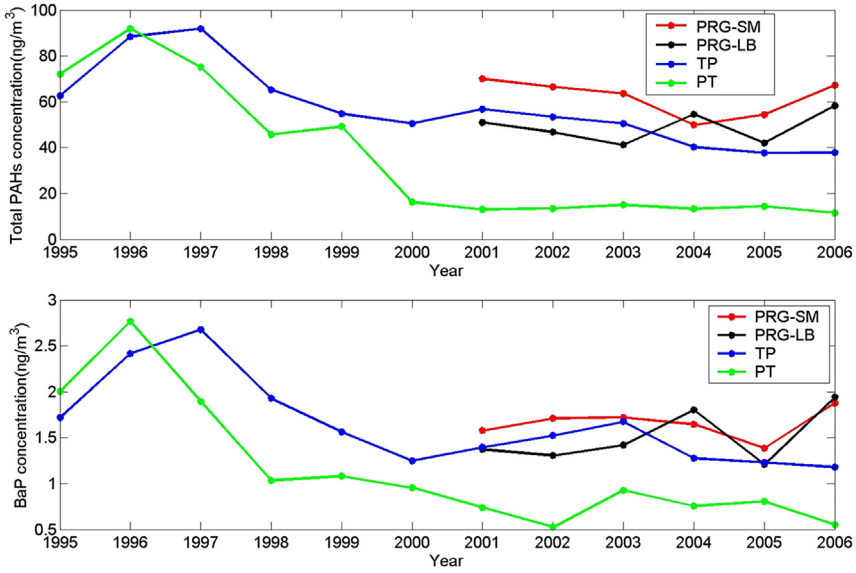

Fig. 2. Annual mean concentrations of tPAHs (upper plate) and B[a]P (lower plate). Annual mean concentrations for 2000 in PRG-LB and PRG-SM are not presented due to missing data in January–April.

station seems to represent distinct conditions also with respect to PAH concentrations than the other sites.

The B[a]P and tPAH trends (Fig. 2) are highly correlated and support the common notion that B[a]P is a good marker of exposure to other PAHs. Moreover, both the B[a]P and tPAH concentrations are correlated with the carcinogenic potential in terms of B[a]P equivalent concentrations (Fig. 3). In all the sites, B[a]P accounted for ~60% of the total carcinogenic potential, with the weighted sum of the six compounds: benzo(a)pyrene, dibenzo(ah)anthracene, benzo(b)fluoranthene, benzo(a)anthracene, indeno(1,2,3-cd)pyrene and benzo(k)fluoranthene accounting for 94%–97% of the total carcinogenic potential. Once again, the PT PAH data seem to be in discordance with the data from the other sites. Fig. 4 presents mean B[a]P concentrations in the summer (JJA) and winter (DJF) of 1995–2006, with winter B[a]P concentrations higher by an order of magnitude than summer concentrations. In particular, winter mean B[a]P concentrations in the years 2000–2006 in TP, PRG-SM and PRG-LB were mostly >3 ng/m^3 whereas summer mean concentrations were mostly <0.2 ng/m^3, i.e. below the B[a]P detection limit.

In summary, based on descriptive statistics and on pollutant patterns and trends three points relevant for model development need to be emphasized, (a) B[a]P concentrations could be used to proxy exposure to the carcinogenic potential of the total PAHs combined, (b) models for predicting B[a]P concentrations, which are not fitted to local conditions, could be developed only for TP, PRG-SM, and PRG-LB, and (c) the models should account separately for winter and summer.

3.2. MLRM development

Criteria pollutants (SO_2, NO_x, NO, NO_2, CO, O_3, $PM_{2.5}$, PM_{10}), the season (binary variable) and the ambient temperature were candidate explanatory variables. All variables (except the season) were log-transformed, to ensure that model results comply with the requirement that the residuals are normally distributed with

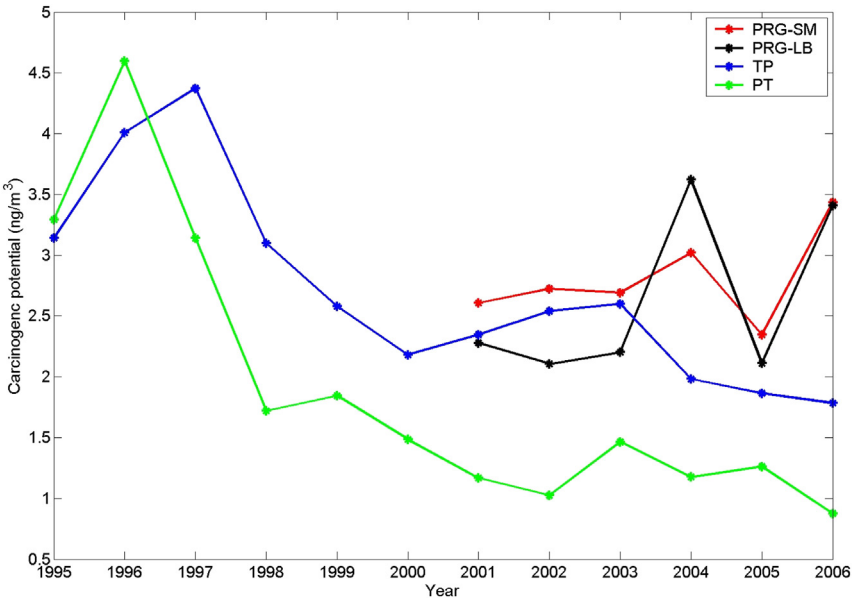

Fig. 3. Carcinogenic potential in terms of B[a]P equivalent concentration of the TEF-weighted tPAHs.

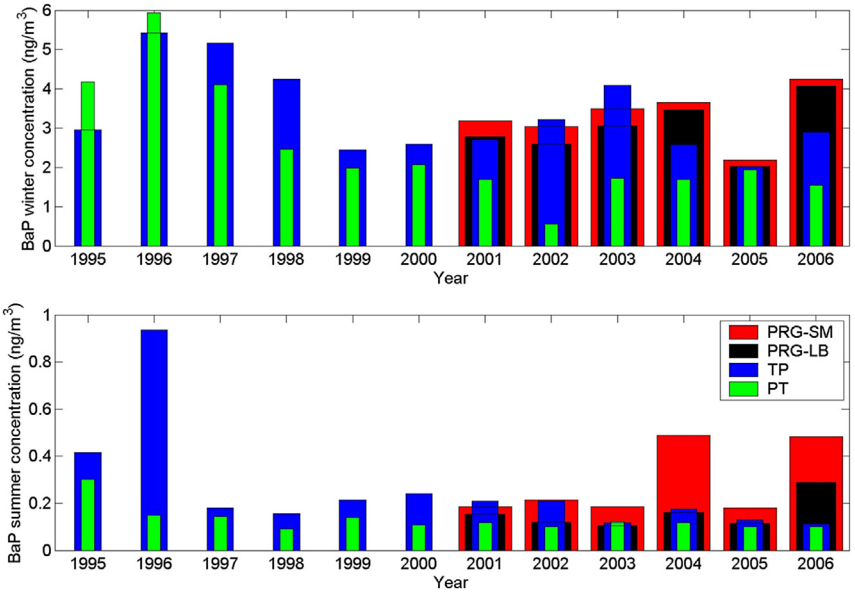

Fig. 4. Winter (December, January, February; upper plate) and summer (June, July, August; lower plate) mean B[a]P concentrations in 1995–2006.

zero means. Only data from days in which the daily B[a]P measurements were above the threshold value (the measurement detection limit) were used, otherwise assumptions related to the development of multivariate linear regression models (MLRM) were violated. Data from 2000 to 2006 from the TP, PRG-SM and PRG-LB monitoring sites were used for developing two models at each site, one with the optimal set of variables and one based on the constrained stepwise regression approach. The models were evaluated by a complete internal (i.e. site specific) cross validation procedure. Evaluation of the models for estimating daily (Table 1) and monthly (Table 2) B[a]P concentrations revealed that the models developed using the constrained stepwise regression performed as good as those developed using standard stepwise regression. Since uniformity of model variables across the different sites is preferable and was set as a goal in this work, model results reported henceforth are based on the following variables: NO_x, NO_2, temperature, $PM_{2.5}$ and season (winter: November–March, summer: April–October), designated as the constrained MLRM variables. As seen in Tables 1 and 2, the R^2 of the site-specific models ranged between 55–58% and 79–88% for the daily and monthly predictions, respectively. Table 3 depicts the parameters of the models in the three sites, based on data from 2000 to 2006.

3.3. Development of classification trees

Data from 2000 to 2006 in the three monitoring sites were also used for the development and evaluation (complete internal cross validation) of the classification trees. To be consistent with the MLRM and to minimize the overall number of variables in the combined model, the input variables to the classification trees were

Table 1
Performance of the MLRMs for estimating daily mean B[a]P concentrations. Results of the two best models (stepwise regression and constrained stepwise regression) at each site, based on 2000–2006 data when observed B[a]P concentrations were larger than the threshold value. (SR – stepwise regression, CSR – constrained stepwise regression).

Station	PRG-SM		PRG-LB		TP	
Model	SR	CSR	SR	CSR	SR	CSR
Variables	CO, O_3, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	O_3, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season
Measured mean (ng/m^3)	0.98	0.96	1.00	1.00	0.87	0.87
Estimated mean (ng/m^3)	0.83	0.78	0.81	0.81	0.68	0.68
R^2	0.56	0.55	0.58	0.58	0.58	0.58
RMSE (ng/m^3)	0.66	0.65	0.75	0.76	0.64	0.64

Table 2
Like Table 1 but for the MLRMs that estimate monthly mean B[a]P concentrations.

Station	PRG-SM		PRG-LB		TP	
Model	SR	CSR	SR	CSR	SR	CSR
Variables	CO, O_3, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	O_3, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season	NO_x, NO_2, temp., $PM_{2.5}$, season
Measured mean (ng/m^3)	0.87	0.84	0.87	0.86	0.70	0.70
Estimated mean (ng/m^3)	0.78	0.75	0.87	0.87	0.59	0.59
R^2	0.88	0.87	0.80	0.79	0.88	0.86
RMSE (ng/m^3)	0.33	0.33	0.42	0.44	0.31	0.31

Table 3
Model parameters of the daily mean B[a]P estimation models at the three sites based on data from 2000 to 2006 (mean and 95% CI).

	PRG-SM	PRG-LB	TP
Intercept	92.55 (66.56, 118.54)	114.86 (82.42, 147.3)	89.43 (65.92, 112.94)
ln(NO_x) (μg/m^3)	0.71 (0.39, 1.02)	0.74 (0.26, 1.2)	0.84 (0.59, 1.1)
ln(NO_2) (μg/m^3)	−0.90 (−1.44, −0.35)	−0.40 (−1.04, 0.23)	−0.79 (−1.25, −0.32)
ln(Temperature) (K)	−16.41 (−21.15, −11.68)	−20.57 (−26.44, −14.7)	−16.12 (−20.37, −11.87)
ln($PM_{2.5}$) (μg/m^3)	0.54 (0.35, 0.73)	0.32 (0.09, 0.54)	0.42 (0.28, 0.55)
Season (categorical)	−1.42 (−2.07, −0.77)	−1.37 (−2.11, −0.64)	−0.38 (−0.94, 0.18)

the constrained MLRM variables. The season always came out as the most important variable, accounting for the largest decrease in data heterogeneity. In general, most of the winter measurements were allocated to a distinct group for which the daily averaged measured B[a]P concentrations (which were not a model variable) were above the threshold value. The data in this group were used for developing the MLRM. The rules of the classification tree that correspond to conditions when the daily B[a]P concentrations were estimated to be below the threshold value, i.e. for data points in the second group, were used for predicting these cases and assigning a preset value of 0.1 ng/m^3 to the ambient B[a]P concentrations. The performance of the classification trees is presented in Table 4.

3.4. Internal cross validation

The station-specific MLRM and classification tree were combined and evaluated together using a complete cross validation (i.e. internal validation) procedure. Performance measures of the models that estimate the daily mean ambient B[a]P concentrations in 2000–2006 are presented in Table 5. Similar results were obtained at the three sites, with the models underestimating the high winter concentrations in 2004. Nonetheless, in general the model results follow nicely the seasonal pattern. In PRG-SM, the mean absolute difference was about half the measured mean, and the correlation coefficient was relatively low. This was attributed to 14 cases in which the differences between measured and estimated concentrations were very large. When these outliers were removed the correlation increase dramatically and the RMSE (a measure highly affected by outliers) decrease by ~30%. Similar dramatic improvement in model performance measures following removal of a very small fraction of outliers (<3%) was obtained also for the

Table 4
Performance of the classification tree models.

	PRG-SM	PRG-LB	TP
Total number of measurements	496	485	641
Number of measurements with B[a]P > D.L.	396	334	479
Number of correct model estimates that B[a]P < D.L.	53	137	108
Number of wrong model estimates that B[a]P < D.L	44	57	42
Number of correct model estimates that B[a]P > D.L	352	277	437
Number of wrong model estimates that B[a]P > D.L.	47	14	54
Overall success (%)	81.7	85.4	85.0

Table 5
Performance of the site-specific combined classification tree-MLRM models for estimating daily mean B[a]P concentrations in 2000–2006.

	PRG-SM		PRG-LB		TP	
	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	496	482	485	473	641	620
Measured mean (ng/m^3)	1.54	1.34	1.26	1.08	1.53	1.39
Estimated mean (ng/m^3)	1.31	1.23	1.03	1.00	1.36	1.29
MAD (ng/m^3)	0.78	0.60	0.64	0.50	0.70	0.58
R^2	0.58	0.69	0.58	0.65	0.59	0.68
RMSE (ng/m^3)	1.55	1.00	1.36	0.89	1.16	0.88
FAC2 (%)	63.9	65.6	71.1	72.9	66.4	67.9

models developed for PRG-LB and TP. Hence, two values for each performance measure are presented for each of the models in Table 5, one based on all the estimated B[a]P concentrations and another based on estimates from which outliers were omitted. The latter correspond to all the cases where the estimated B[a]P concentrations satisfied the condition.

$$B[a]P_{estimated} < \overline{B[a]P}_{measured} + 2 \cdot STD_{B[a]P_{measured}}. \quad (2)$$

As seen, the FAC2 is less affected by removing the outliers.

The monthly mean B[a]P estimations were better than the daily mean B[a]P estimations, with high correlations between the measured and estimated B[a]P concentrations found in the three sites (Fig. 5, Table 6). Since the monthly mean B[a]P concentrations are averages of the daily estimated concentrations, the effect of outliers on the monthly means is smaller. The estimated monthly mean B[a]P concentrations show high R^2 (~0.9) with the corresponding observations and more than 85% of the estimates falling within a factor of two from the observations. The normalized mean absolute difference (the ratio of *MAD* to the measured mean) is mostly ~20%.

3.5. External cross validation

The spatial performance of the models was evaluated by calculating model parameters (i.e. parameterization) at one site, using data from the years 2000–2006, and applying the model for estimating ambient B[a]P concentrations in the years 2000–2006 at the other two sites. Table 7 presents the performance measures of the models that estimate daily mean ambient B[a]P concentrations. The ratio of the mean absolute error (*MAD*) to the measured mean ranged between 0.48–0.61 (mean 0.54) and the R^2 ranged between 0.53–0.58 (all data). Clearly, when outliers (<3.3% of the data) were removed the models' performance improved (R^2: 0.61–0.69). Once more, the quality of the monthly mean estimates was better than that of the daily estimates (Table 8, all data) and removing the outliers further improved it. For example, Fig. 6 depicts measured vs. estimated monthly mean B[a]P concentrations in the PRG-SM site using models that were parameterized in the PRG-LB and TP sites.

Table 6
Like Table 5 but for estimating monthly mean B[a]P concentrations.

	PRG-SM		PRG-LB		TP	
	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	51	51	51	51	65	65
Measured mean (ng/m^3)	1.36	1.27	1.08	1.01	1.46	1.32
Estimated mean (ng/m^3)	1.19	1.16	0.95	0.94	1.26	1.21
MAD (ng/m^3)	0.31	0.27	0.30	0.25	0.32	0.24
R^2	0.90	0.92	0.85	0.89	0.86	0.91
RMSE (ng/m^3)	0.47	0.38	0.48	0.38	0.54	0.34
FAC2 (%)	86.3	88.2	88.2	90.2	90.8	92.3

3.6. Spatiotemporal cross validation

The performance of the models when estimating daily mean B[a]P concentrations in TP in the years 1995–1999 based on parameterization using data from PRG-SM, PRG-LB or TP from the years 2000–2006 are presented in Table 9. Again, model performance improved when the outliers were removed. Table 10 presents the corresponding values for the monthly mean B[a]P estimates. The monthly predictions are consistently better than the daily predictions although in most cases they underestimate the 1995–1999 TP monthly means (Fig. 7). The performance of the models under the spatiotemporal cross validation tests were comparable to those obtained in the external cross validation tests.

4. Discussion

The order of magnitude summer-to-winter difference in ambient B[a]P concentrations is in agreement with previous findings (Ravindra et al., 2006; Sisovic et al., 2008; Li et al., 2009). This suggests that exposure to PAHs during exposure windows shorter than a year, e.g. during pregnancy, could be biased towards the higher winter concentrations if naïve annual mean concentrations

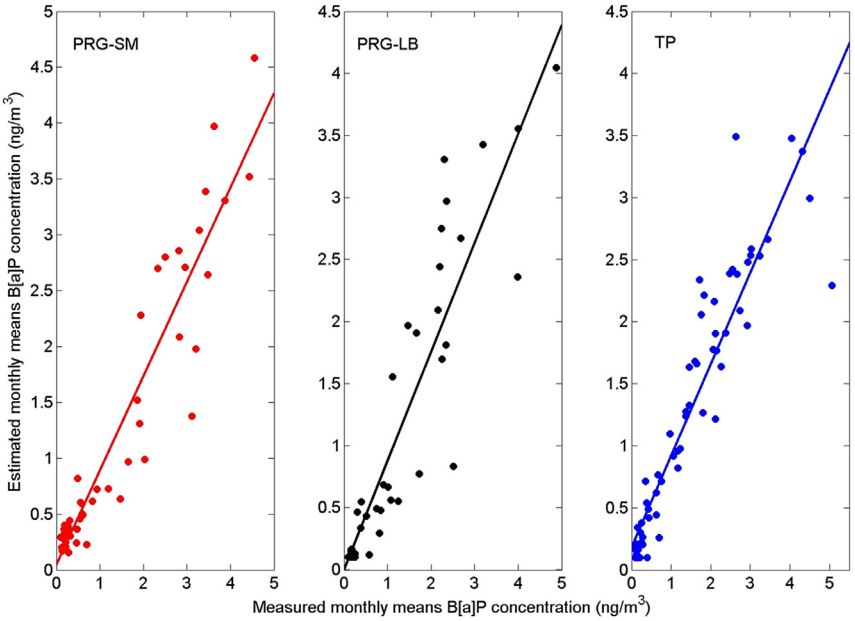

Fig. 5. Measured vs. estimated monthly mean ambient B[a]P concentrations at PRG-SM, PRG-LB and TP monitoring sites in the years 2000–2006. The Models' performance is detailed in Table 6.

Table 7
Across-site model performance when estimating daily mean B[a]P concentrations in 2000–2006 (external cross validation).

Site of estimated B[a]P	PRG-SM				PRG-LB				TP			
B[a]P data used for model parameterization	PRG-LB		TP		PRG-SM		TP		PRG-SM		PRG-LB	
	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	603	583	603	584	590	573	590	572	764	746	764	742
Measured mean (ng/m^3)	1.71	1.51	1.71	1.48	1.48	1.27	1.48	1.25	1.47	1.40	1.47	1.38
Estimated mean (ng/m^3)	1.90	1.65	1.39	1.32	1.08	1.05	0.89	0.85	1.60	1.46	1.90	1.63
MAD (ng/m^3)	0.99	0.76	0.82	0.64	0.74	0.57	0.80	0.62	0.76	0.66	0.90	0.70
R^2	0.53	0.66	0.55	0.69	0.56	0.66	0.55	0.61	0.58	0.66	0.54	0.66
RMSE (ng/m^3)	1.93	1.25	1.57	1.02	1.48	0.97	1.63	1.08	1.24	0.99	1.81	1.12
FAC2 (%)	60.7	62.4	61.0	62.8	65.6	67.4	59.3	60.8	61.1	62.0	63.6	65.4

Table 8
Like Table 7 but for monthly mean B[a]P concentrations.

Site of estimated B[a]P	PRG-SM				PRG-LB				TP			
B[a]P data used for model parameterization	PRG-LB		TP		PRG-SM		TP		PRG-SM		PRG-LB	
	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	63	63	63	63	63	63	63	63	77	77	77	77
Measured mean (ng/m^3)	1.46	1.38	1.46	1.35	1.27	1.16	1.27	1.16	1.41	1.35	1.41	1.34
Estimated mean (ng/m^3)	1.59	1.47	1.21	1.20	0.93	0.93	0.78	0.77	1.49	1.39	1.76	1.54
MAD (ng/m^3)	0.48	0.38	0.37	0.29	0.44	0.35	0.55	0.45	0.35	0.29	0.57	0.42
R^2	0.90	0.93	0.91	0.94	0.78	0.83	0.79	0.80	0.82	0.87	0.80	0.86
RMSE (ng/m^3)	0.77	0.54	0.56	0.40	0.77	0.58	0.94	0.74	0.56	0.43	0.94	0.60
FAC2 (%)	77.8	79.4	81.0	84.1	76.2	79.4	77.8	81.0	88.3	88.3	85.7	89.6

are used as the exposure metric. Moreover, apart from seasonal related emissions and dispersion disparities the season in which the exposure takes place may have another crucial impact. Namely, the potential carcinogenicity is distributed in a non-uniform fashion along the year among both the gaseous and particulate PAH species (e.g. Fig. 4), with the PAH species profile showing seasonal dependence. Hence, PAHs and B[a]P concentration estimates must account for this seasonality. Yet, models that were developed for estimating ambient B[a]P did not account, to date, for this aspect. In this study B[a]P concentrations lower than the measurement detection limit were estimated using a classification tree model that has been developed to detect conditions that accompany such cases. This unique approach relies on the availability of a rich database of ambient B[a]P concentrations in the Czech Republic, which enabled the development of MLRM for estimating B[a]P concentrations when conditions allowed it. Reliable estimation of the low ambient B[a]P concentrations in the summer is important, since exposure to low ambient B[a]P

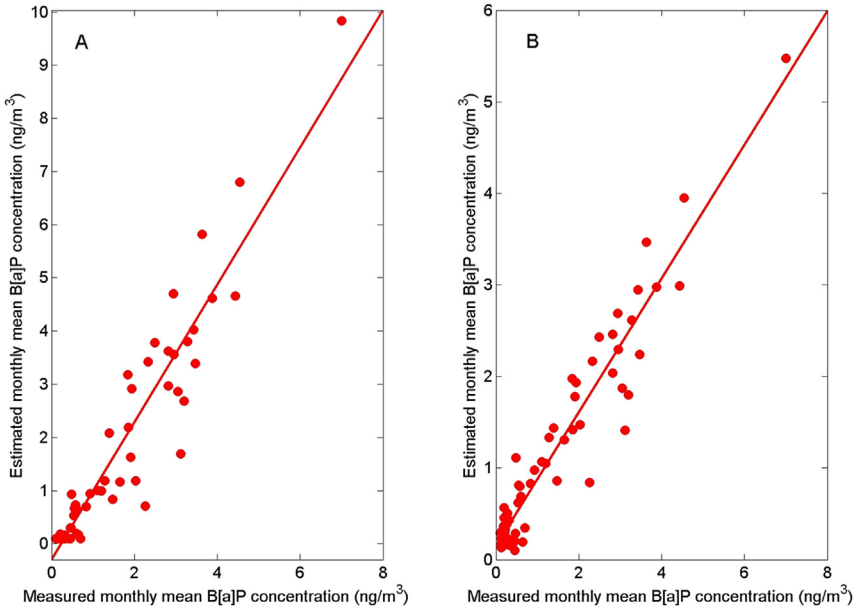

Fig. 6. Estimated vs. measured monthly mean B[a]P concentrations at PRG-SM in the years 2000–2006. A – model parameters based on 2000–2006 data from PRG-LB. B – model parameters based on 2000–2006 data from TP. The models' performance is detailed in Table 8.

Table 9
Model performance when estimating daily mean B[a]P concentrations in TP in the years 1995–1999 (spatiotemporal cross validation).

B[a]P data used for model parameterization	PRG-SM 2000–2006		PRG-LB 2000–2006		TP 2000–2006	
Site and time of estimated B[a]P	TP 1995–1999		TP 1995–1999		TP 1995–1999	
	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	840	818	840	816	840	816
Measured mean (ng/m^3)	2.92	2.62	2.92	2.65	2.92	2.60
Estimated mean (ng/m^3)	2.32	2.28	2.87	2.65	1.95	1.90
MAD (ng/m^3)	1.38	1.15	1.46	1.18	1.47	1.21
R^2	0.53	0.65	0.48	0.64	0.53	0.66
RMSE (ng/m^3)	2.42	1.73	2.63	1.80	2.59	1.84
FAC2 (%)	64.2	65.8	64.9	66.4	61.0	62.8

concentrations was linked to adverse reproductive effects (Choi et al., 2006).

The MLRM developed in this study use criteria pollutants, temperature and season (categorical variable) as predictor variables. Although accounting for additional explanatory variables, such as the wind speed and direction (Lobscheid et al., 2007; Callen et al., 2010) could possibly improve the model performance, such data were not available at all the monitoring sites for all the study period. Thus, including these variables would have resulted in models that could not be spatially extrapolated across sites. Moreover, meteorological variables may not account for the strength of the PAHs emission sources (apart from the absence/presence of space heating), since they proxy mainly dispersion and degradation processes. Consequently, models that use mostly meteorological variables may be suitable for estimating ambient B[a]P concentrations only when the emissions are stable over time. In contrast, in this work we used concentrations of criteria pollutants as explanatory variables, assuming that their ambient concentrations shadow the varying PAHs source strengths. Whereas this assumption seems to work reasonably well when applying the models to areas for which they have not been trained (i.e. the external evaluation procedure), the results of the spatiotemporal evaluation were somewhat inferior, i.e. the effect of the dramatic change in PAHs source strengths during the early study period on ambient B[a]P concentrations was only partially captured by the model.

Monthly mean B[a]P concentrations prediction in this work is based on averaging estimates obtained at the highest (daily) temporal data resolution available for us. Clearly, an alternative approach could have been to first take the mean of all the explanatory variables and then use these averages to model the monthly B[a]P mean. In spite of the MLRM being a linear model, these approaches are not equivalent and their prediction could differ considerably, depending on the fraction of missing values, etc. (Yuval and Broday, 2014). It is therefore highly recommended in such cases to use data at the highest temporal resolution available and average out only the final predictions to obtain predictions on a coarser time scale.

Comparing the performance of the models developed in this study with that of previously reported models can be done only based on daily B[a]P concentration estimates and only for cases with B[a]P concentrations above the threshold value. Previously reported coefficients of determination (R^2) ranged between 0.57–0.72 (Lobscheid et al., 2007) and 0.79–0.88 (Callen et al., 2010) whereas here we report raw R^2 values of 0.55–0.58 (Table 1). However, when the combined classification tree-multivariate linear regression models were used (i.e. including days with low B[a]P concentrations) R^2 ranged between 0.65 and 0.69 (without outliers; Table 5). Moreover, the fraction of explained variance (R^2) of the monthly mean B[a]P estimates was even higher, ranging between 0.85–0.90 (all data) and 0.89–0.92 (without outliers; Table 6). In fact, this should not be underestimated, since monthly mean exposure to PAHs was found to be associated with various health effects, in particular pregnancy outcomes (Dejmek et al., 2000; Perera et al., 2003, 2006; 2009, 2012). It is therefore noteworthy that Lobscheid's et al. (2007) and Callen's et al. (2010) models, which were developed only for days in which the measured concentrations were higher than the detection limit, introduce bias to the estimation of monthly mean exposures to B[a]P.

Lobscheid et al. (2007) reported that when their model was parameterized in one air basin and later applied to other air basins the R^2 ranged between 0.47 and 0.5. Similarly, Callen et al. (2010) parameterized their model for Monzon, Spain (based on only 31 measurements) and estimated B[a]P concentrations in Zaragoza, reporting model cross validation of $R^2 = 0.76$. The models developed in this study are characterized by R^2 for the external cross validation (i.e. model application across sites) that ranged between 0.53–0.58 (all data) and 0.61–0.69 (without outliers; Table 7). All the performance measures were higher when estimating monthly mean B[a]P concentrations (Table 8). Clearly, the accuracy of the estimation in places other than the one for which the model has been parameterized is lower, yet the spatial extrapolation of the models to air basins for which they have not been trained (model transferability) is in general supported by our results.

Successful temporal validation could support the use of the models for time periods outside the learning dataset. In spite of the large decrease in ambient B[a]P concentrations in the years 1995–1999 and the change in the relative strength of different B[a]P sources, model predictions for the early time period were almost as good as those obtained for the spatial extrapolation evaluation tests (Tables 9 and 10). In agreement with our previous results, daily mean B[a]P concentration estimates in TP in the years 1995–1999 were not as good as the monthly mean estimates. These results support our assumption that models for estimating B[a]P concentrations using criteria pollutants as explanatory variables could be responsive to changing emission conditions.

5. Summary and conclusions

Using a classification tree as a tool for identifying days on which B[a]P concentrations are expected to be low (below the measurement detection limit or any other threshold) improved our predictions and provide reliable estimates of ambient B[a]P concentrations in the summer. In particular, the combined regression tree-MLRM models enable the estimation of daily and monthly mean B[a]P concentrations throughout the year without violating the assumptions on which the MLRM is based. The large database of ambient B[a]P concentrations in the Czech Republic enabled a detailed study of the combined classification tree-MLRM modeling approach, and to address quantitatively many drawbacks normally associated with such a modeling. Model performance suggests that

Table 10
Like Table 9 but for monthly mean B[a]P concentrations.

B[a]P data used for model parameterization	PRG-SM 2000–2006		PRG-LB 2000–2006		TP 2000–2006	
Site and time of estimated B[a]P	TP 1995–1999		TP 1995–1999		TP 1995–1999	
	All data	Without outliers	All data	Without outliers	All data	Without outliers
Number of measurements	60	60	60	60	60	60
Measured mean (ng/m^3)	2.07	1.90	2.07	1.93	2.07	1.90
Estimated mean (ng/m^3)	1.60	1.58	1.90	1.79	1.41	1.40
MAD (ng/m^3)	0.63	0.51	0.61	0.49	0.76	0.62
R^2	0.83	0.86	0.81	0.85	0.86	0.87
RMSE (ng/m^3)	0.94	0.72	0.90	0.71	1.13	0.90
FAC2 (%)	78.3	83.3	80.0	83.3	80.0	85.0

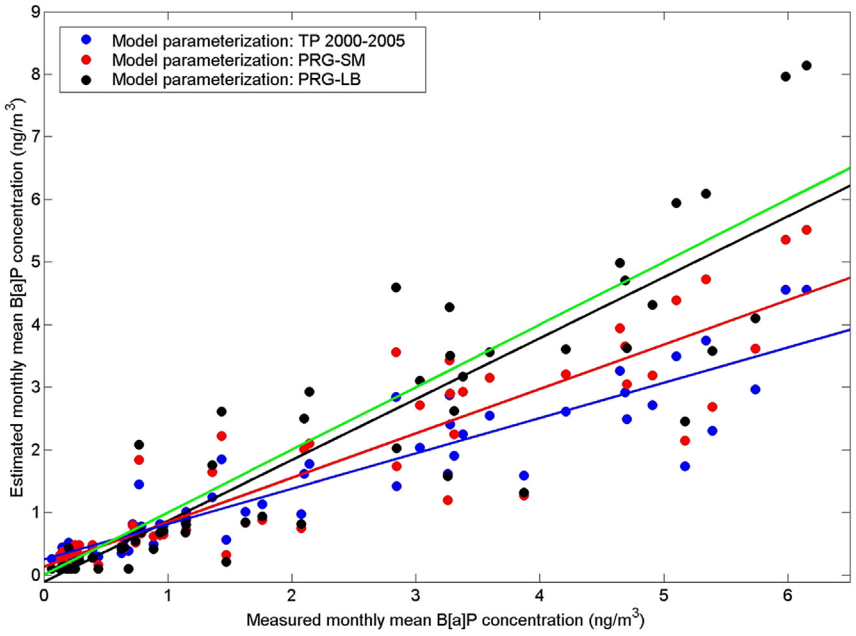

Fig. 7. Estimated vs. measured monthly mean B[a]P concentrations in TP in the years 1995–1999 based on models that were parameterized using 2000–2006 data from TP (blue), PRG-SM (red), and PRG-LB (black). The green line represents the 1:1 line, the other lines are the best linear fits to their respective data points. The models' performance is detailed in Table 10. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the models are very reliable for estimating monthly mean ambient B[a]P concentrations. Namely, both spatial extrapolation and spatiotemporal extrapolations of the models resulted in only a slight decrease in model performance. These findings have two important implications: (1) estimation of past ambient B[a]P concentrations may be possible, and (2) estimates of monthly mean B[a]P concentrations are more reliable than daily means estimates, and may be useful in retrospective environmental health studies.

Acknowledgment

This work was supported by the ENVIRISK consortium agreement SSPE-CT-2005, Contract no. 044232, under the Sixth Framework Program for R&D of the Research Directorate General of the European Commission, the CITISENSE grant agreement n° 308524.F under the European Union Seventh Framework Program (FP7/2007-2013), and by the Technion Center of Excellence in Exposure Science and Environmental Health (TCEEH).

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.11.031.

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