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Comparison of Black Smoke and $PM_{2.5}$ Levels in Indoor and Outdoor Environments of Four European Cities

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Recent studies on separated particle-size fractions highlight the health significance of particulate matter smaller than 2.5 μm ($PM_{2.5}$), but gravimetric methods do not identify specific particle sources. Diesel exhaust particles (DEP) contain elemental carbon (EC), the dominant light-absorbing substance in the atmosphere. Black smoke (BS) is a measure for light absorption of PM and, thus, an alternative way to estimating EC concentrations, which may serve as a proxy for diesel exhaust emissions. We analyzed $PM_{2.5}$ and BS data collected within the EXPOLIS study (Air Pollution Exposure Distribution within Adult Urban Populations in Europe) in Athens, Basel, Helsinki, and Prague. 186 indoor/outdoor filter pairs were sampled and analyzed. $PM_{2.5}$ and BS levels were lowest in Helsinki, moderate in Basel, and remarkably higher in Athens and Prague. In each city, Spearman correlation coefficients of indoor versus outdoor were higher for BS (range $r_{Spearman}$: 0.57−0.86) than for $PM_{2.5}$ (0.05−0.69). In a BS linear regression model (all data), outdoor levels explained clearly more of indoor variation (86%) than in the corresponding $PM_{2.5}$ model (59%). In conclusion, ambient BS seizes a health-relevant fraction of fine particles to which people are exposed indoors and outdoors and exposure to which can be assessed by monitoring outdoor concentrations. BS measured on $PM_{2.5}$ filters can be recommended as a valid and cheap additional indicator in studies on combustion-related air pollution and health.

Introduction

The effects of airborne particulate matter (PM) on human health have been examined in numerous epidemiologic studies (*1*), several of which highlight the health significance of fine particles smaller than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) (*2*−*6*). Soot particles, consisting of an elemental carbon (EC) core and thousands of adsorbed substances, are an important constituent of $PM_{2.5}$ in urban settings worldwide. In earlier times, coal combustion was a large source for soot particle emissions; however, nowadays in Western Europe and the U.S., between 67% and 90% of the atmosphere's content of EC is estimated to be produced by diesel-powered vehicles (*7*−*10*). Complete combustion of diesel fuel, as it can be almost achieved in industrial burners and domestic heating, produces water and carbon dioxide, but use of diesel in motor vehicles results in incomplete combustion and the formation of carbonaceous particles (*11*). Therefore, EC particle concentrations could be used as a surrogate for traffic-related air pollution in urban areas. Nevertheless, it must be considered that the contribution of other primary combustion sources such as coal-burning heating or industry can vary significantly between cities (*7*, *12*, *13*).

Diesel exhaust particles (DEP) completely fall within the size range of $PM_{2.5}$ with submicron mass-median diameters. Exposure to diesel exhaust has been observed to cause changes in lung function and a number of other respiratory symptoms. Diesel exhaust is probably carcinogenic to humans and might be an important factor contributing to the allergy pandemic (*11*).

The common analytical methods to detect EC are expensive and destructive for the sample material. Because EC (or graphitic or black carbon) is the dominant light absorbing substance in the atmosphere (*9*, *14*, *15*), measuring light absorption or reflectance of PM, collected on filter media, is an alternative way to estimate EC concentrations in the atmosphere. Reflectometry provides a simple and cheap, yet sensitive, method to estimate carbonaceous combustion products, like DEP or soot particles from other sources, on particle sample filters.

Methods comparing the blackness of sample filters to assess smoke concentrations have been used since the beginning of the 20th century (*16*). In 1964, a standard calibration curve to convert reflectance into mass concentration ($\mu g/m^3$) was defined by the OECD, known as the black smoke (BS) method (*17*). At that time, suspended PM in the atmosphere was dominated by carbonaceous combustion products, mainly from coal burning. Since then, the use and type of fuel has changed, and coal heating has been reduced. A number of studies (*16*, *18*) showed that a universal relation between BS and PM no longer prevailed; therefore, gravimetric methods to determine PM were preferred. In recent years, disadvantages of gravimetric methods became evident; although studies on separated size fractions of PM could clarify some associations between health effects and particles (*2*−*6*, *19*, *20*), gravimetric methods do not allow for the main components of PM to be distinguished by characteristics such as, for example, elementary composition, primary or secondary formation, emission sources, and so forth. However, this information is crucial for establishing clean air regulations.

Considering EC as a potential indicator for traffic-related particles, the goal of our work was to analyze the relation between indoor and outdoor BS and to compare it with the equivalent associations for $PM_{2.5}$. As we spend about 90% of

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TABLE 1. Sample Characteristics

	Athens N = 43		Basel N = 41		Helsinki N = 82		Prague N = 20	
	obs	%	obs	%	obs	%	obs	%
season								
winter (Oct−March)	30	69.8	19	46.3	39	47.6	9	45.0
summer	13	30.2	22	53.7	43	52.4	11	55.0
home location								
suburban, small[a]	18	41.9	17	41.5	32	39.0	20	100.0
downtown	5	11.6	11	26.8	16	19.5		
suburban, high[b]	20	46.5	8	19.5	32	39.0		
industrial zone			2	4.9	2	2.4		
other			3	7.3				
traffic density[c]								
low	32	74.4	25	61.0	55	67.1	2	10.0
high	11	25.6	16	39.0	27	32.9	18	90.0
type of heating								
district	9	21.4	13	31.7	55	67.9	12	60.0
central	29	69.1	24	58.5	21	25.9	3	15.0
gas			2	4.9	3	3.7	5	25.0
electric	4	9.5	2	4.9	2	2.5		
gas appliances[d]								
yes	3	7.0	11	26.8	2	2.4	12	60.0
smoke[e]								
yes	15	34.9	7	17.5	2	2.4	8	40.0
	mean	**sd**	**mean**	**sd**	**mean**	**sd**	**mean**	**sd**
use of gas appliances[f]	0.3	1.2	2.1	8.2	0.1	0.3	6.6	14.6
cigarettes per day[g]	14.3	10.2	5.0	3.8	2.5	3.5	10.5	5.8
	winter	**summer**	**winter**	**summer**	**winter**	**summer**	**winter**	**summer**
open window[h]	4.8	23.1	8.9	27.2	4.3	25.1	1.3	17.9

[a] Self-contained houses or multistorey buildings within green spaces. [b] Multistorey buildings in densely built area. [c] "High traffic density": "traffic volume (TV) on the nearby street" = "heavy/continuous" or "heavy vehicle traffic volume (HTV) on the nearby street" = "all of the time/often". "Low traffic density" = "medium/light" "TV on the nearby street" and "rarely/never" "HTV on the nearby street". [d] Households reported "gas cooking" or "single gas stove heating". [e] Households reported smoking. [f] Hours of "gas cooking" and "single gas stove heating" during 48 h measurement period. [g] Nonsmokers excluded. [h] Hours during 48 h measuring period.

the time indoors (*21*), indoor concentrations will determine factual exposure to the pollutant. Therefore, associations between outdoor levels of air pollution and health outcomes may be plausible only if health relevant constituents of ambient air pollution efficiently penetrate indoors.

To assess the relevance of indoor sources, which can add remarkable amounts to indoor particle concentrations, we used multiple linear regression models. Further, correlations between BS and $PM_{2.5}$ indoors and outdoors and several stratified analysis were applied to describe and compare the characteristics of the two exposures. The suitability of BS as a supplementary indicator in exposure assessment will be discussed.

Materials and Methods

In our work, we analyzed $PM_{2.5}$ data and reflectance (absorption coefficient/meter (abs coeff/m)) from indoor and outdoor filters sampled within the framework of the European EXPOLIS study (Air Pollution Exposure Distribution within Adult Urban Populations in Europe) (*22*). The analyzed filter samples ($N = 186$ pairs of participants with both valid indoor and outdoor data) were collected from October 1996 to March 1998 at the homes of the study participants in Athens (Greece; $N = 43$), Basel (Switzerland; $N = 41$), Helsinki (Finland; $N = 82$), and Prague (Czech Republic; $N = 20$). Descriptive statistics of the study population are shown in Table 1. $PM_{2.5}$ measurements were performed in the study centers using identical equipment and following common standard operation procedures (*23*). Indoor and outdoor air at each participants home was filtered over two consecutive nights (two nights on one filter), from the time when the participant would usually return from work to the time the participant would usually leave home for work (approximately 2×15 h). Exposure samples of work time are not analyzed in this paper. Details on the EXPOLIS design, the study population, and the applied methodologies have been published elsewhere (*22−25*).

After the weighing to determine particle mass, the filters were sent to the EXPOLIS center Basel for reflectance analysis. The blackness of the $PM_{2.5}$ filter deposit was measured using a reflectometer (EEL model 43; Diffusion Systems Ltd., London, U.K.), which measures the reflection of the light incidence in percent. Blank filters were used to set reflectance to 100%. To compensate for inhomogeneities, the reflectance of each sampled filter was measured on five different spots of each filter and then averaged. The absorption coefficient was calculated using the following formula (ISO 9835 (*26*)): absorption coefficient per meter (abs coeff/m) = $0.5A \ln(R_0/R)/V$, where A = loaded filter area; R_0 = reflectance of blank filters (in percent); R = reflectance of sampled filters (in percent); and V = sampled volume of air (in m^3).

The average reflectance of the five measured spots on each filter was used to calculate the absorption coefficient, which was then multiplied by 10^5 to make the readings more comprehensible.

Reflectometry applied in the current work was done following the standard operation procedure (SOP) from the ULTRA study (*27*), which is based on the international standard ISO 9835 (*26*). It does not correspond exactly to the black smoke method defined by the OECD protocol (*17*), because of the cutoff size of 2.5 μm and the different type of filters (Gelman Teflo 47 mm, 2 μm pore size) used. Also,

TABLE 2. Air Pollution Levels and Ratios between Indoors and Outdoors, Summer and Winter, and Smoking and Nonsmoking, Respectively

center	*N*	mean ± sd indoor	mean ± sd outdoor	median of indoor/outdoor ratios (paired)[a]	winter/summer ratio of means (unpaired)[b] indoor	winter/summer ratio of means (unpaired)[b] outdoor	smoke[c]/no-smoke ratio of means indoors (unpaired)[b]
BS (abs coeff/m)							
Athens	43	2.92 ± 2.50	3.30 ± 2.56	0.87	1.52	1.75	1.28[b] (15)
Basel	41	1.37 ± 0.58	1.39 ± 0.51	0.92	1.24[b]	1.23	1.30[b] (7)
Helsinki	82	0.78 ± 0.46	0.97 ± 0.44	0.79	1.00	1.05	1.23 (2)
Prague	20	2.74 ± 0.81	2.98 ± 1.30	0.96	1.21	1.37	0.94[b] (8)
$PM_{2.5}$ (μg/m^3)							
Athens	43	35.6 ± 29.4	37.3 ± 27.4	0.90	1.47	1.58[b]	1.41 (15)
Basel	41	21.0 ± 16.7	19.3 ± 11.5	0.98	1.58[b]	1.28	1.89[b] (7)
Helsinki	82	9.5 ± 6.1	10.5 ± 7.1	0.91	0.84	1.10	1.91 (2)
Prague	20	34.4 ± 28.7	27.3 ± 10.4	1.04	1.13	1.35[b]	2.09[b] (8)

[a] No mean ratio of indoor outdoor pairs significantly different from 1 at 5% level in paired *t* test. Only median of indoor outdoor ratios shown. [b] Ratios of means significantly different from 1 at 5% level in unpaired *t* test. [c] *N* of households with tobacco smoke exposure in parentheses.

reflectance was not converted into units of mass concentrations. Nevertheless, in this work, the expression black smoke (BS) is used synonymously to reflectance, measured as light absorption coefficient per meter.

To ensure the reliability of the used reflectance analysis method, we repeated measurements of 32 randomly selected filters five times each, five spots in each measurement. All of these repetitions were completed within 1 week, 1 year after the complete EXPOLIS filter set had been measured for reflectance. The reliability of the method was determined using reflectance readings directly. Mean difference between the original EXPOLIS measurements and the repeated measurements was calculated to judge the reliability of the method. Detailed information on the reliability of the applied method is available in a thesis (*28*).

For further data analysis, reflectance of the sample filter was expressed as absorption coefficient per meter of the sample filter. Descriptive statistics for indoor and outdoor levels of BS and $PM_{2.5}$ were calculated for the entire sample and stratified by study centers, season, and smoking status (smoke = households reporting smoking).

Spearman correlation was used to describe associations between indoor and outdoor levels as well as between BS and $PM_{2.5}$ concentrations. To assess the impact of different sources on indoor concentrations, multiple linear regression models were developed. Indoor concentrations of BS and $PM_{2.5}$ were chosen as dependent variables. Measured outdoor concentrations and questionnaire data on traffic density at home (self-reported), tobacco smoke, use of gas appliances, home location, heating system, duration of open window, season, vacuum cleaning, and further indoor characteristics were considered as potential predictors. The models for the total data were adjusted for center. The descriptive statistics of the study population for all variables with relevant frequencies are shown in Table 1.

Feasibility of linear regression was tested with the usual diagnostic tools. To prevent inaccurate standard errors due to observed nonnormal distribution and heteroscedasticity of residuals, coefficients were calculated with robust standard errors (*29*). Criteria for inclusion of variables in the multiple linear regression models were an increase of adjusted R^2 or change of crude estimate in either of the two models. The variable "season" was not included in the models because of collinearity with the measured outdoor concentration. The traffic variables were not used either. They did not improve the models, probably because of their inaccuracy or collinearity with the outdoor variable. The variable "home location", on the other hand, increased R^2 and was, therefore, used for the models, although there might be some collinearity with the outdoor variable, too. Duration of "gas cooking" and "single gas stove heating" were combined to the variable "use of gas appliances". For the models, the data of a pipe smoker was excluded, because, as a single observation, it could not be predicted by a specific pipe variable.

TABLE 3. Spearman Correlation Coefficients for Complete Data Sets, Summer Data, and Winter Data

center	in vs out correlation BS	in vs out correlation $PM_{2.5}$	BS vs $PM_{2.5}$ correlation indoor	BS vs $PM_{2.5}$ correlation outdoor
total data				
Athens	0.86	0.67	0.83	0.90
Basel	0.84	0.58	0.78	0.74
Helsinki	0.83	0.69	0.74	0.66
Prague	0.57	0.05	0.16	0.87
summer				
Athens	0.87	0.90	0.70	0.86
Basel	0.86	0.78	0.87	0.69
Helsinki	0.86	0.78	0.73	0.63
Prague	0.15	0.33	−0.15	0.38
winter				
Athens	0.88	0.55	0.86	0.90
Basel	0.82	0.42	0.72	0.85
Helsinki	0.74	0.62	0.73	0.68
Prague	0.73	−0.22	0.55	0.93

Data were analyzed using STATA Statistical Software 6.0 (*30*) on Windows 95 and Windows NT.

Results

In the reliability study, the mean differences between the five repeated measurements and the original EXPOLIS values of the 32 filters were within the range of −0.03 to 0.76 (average 0.21) reflectance units. Expressed as relative difference the range was between −0.05% and 1.3% with an average of 0.65% (results not shown). More details on the reliability of the applied method are available in a thesis (*28*).

Descriptive statistics for BS and $PM_{2.5}$ are summarized in Table 2. Fine particle concentrations were found to be lowest in Helsinki, moderate in Basel, and remarkably higher in Athens and Prague. Ratios between indoor and outdoor concentrations tend to be higher for $PM_{2.5}$ than for BS. Overall, particle concentrations were higher in winter than in summer, although not in Helsinki with generally low PM levels. Ratios between smoking and nonsmoking indoors were higher for $PM_{2.5}$ than for BS.

In Table 3, Spearman correlation coefficients between indoor and outdoor levels as well as between BS and $PM_{2.5}$

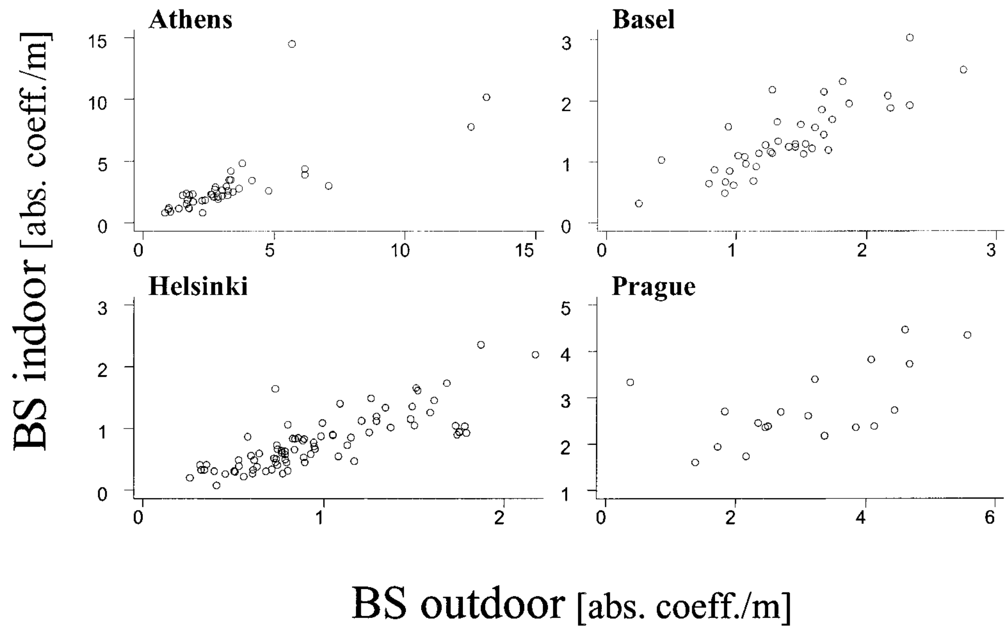

FIGURE 1. Indoor against outdoor concentrations of BS in the four EXPOLIS study centers Athens, Basel, Helsinki, and Prague (note: different scale ranges for each center and axis).

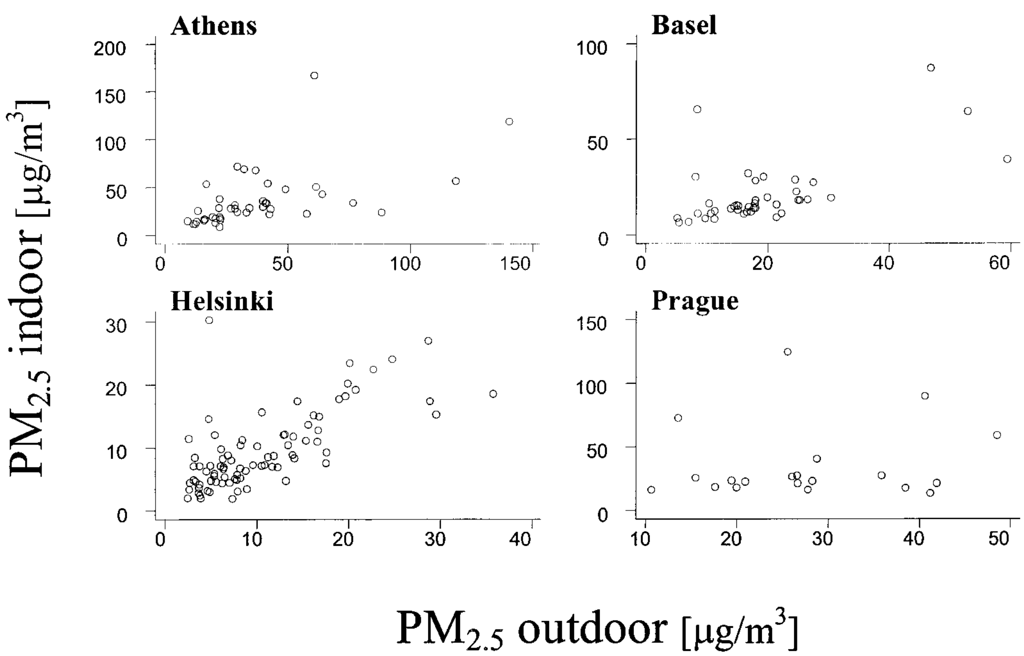

FIGURE 2. Indoor against outdoor concentrations of $PM_{2.5}$ in the four EXPOLIS study centers Athens, Basel, Helsinki, and Prague (note: different scale ranges for each center and axis).

concentrations for the four cities are listed for all data and stratified by summer and winter. Overall, Spearman correlation coefficients between indoors and outdoors were higher for BS than for $PM_{2.5}$. No significant correlation was found between indoor and outdoor concentrations of $PM_{2.5}$ in Prague. Spearman correlation between BS and $PM_{2.5}$ outdoors (Table 3) was very high in Athens and Prague ($r =$ 0.90 and 0.87, respectively) and somewhat lower in Basel and Helsinki ($r = 0.74$ and 0.66, respectively). Correlation between BS and $PM_{2.5}$ indoors was similar to outdoors, except for Prague where it was very low. The scatter plots in Figures 1 and 2 show indoor against outdoor concentrations of BS and $PM_{2.5}$, respectively, for the four study centers.

Univariate and multiple linear regression models to predict indoor concentrations of BS (Table 4a) and $PM_{2.5}$ (Table 4b) showed common patterns valid throughout the four centers as well as some center-specific phenomena. In univariate regression (Table 4a, crude estimates), BS outdoor levels explained 83%, 66%, 62%, and 31% of the indoor variation in Athens, Basel, Helsinki, and Prague, respectively. For $PM_{2.5}$, the corresponding R^2 values were clearly lower (40%, 36%, 48%, and 0.0%, respectively). Throughout the four centers in both univariate and multivariate regressions, slopes were steeper for BS than for $PM_{2.5}$ outdoor concentrations. Accordingly, including additional variables improved $PM_{2.5}$ models, increasing R^2 by 7%, 18%, 3%, and 71% in Athens, Basel, Helsinki, and Prague, respectively, while gain of explained indoor variation in multiple BS models was rather small or even negative (1%, −1%, 5%, and 8%, respectively). Still, the multivariate models for BS explained more of the variance of indoor concentrations (84%, 65%, 67%, and 39%, respectively) than the corresponding models for $PM_{2.5}$ (47%,

TABLE 4. Linear Regression Models for the Impacts on BS and $PM_{2.5}$ Indoor Concentrations[a]

(a) Linear Regression Models for the Impacts of BS Outdoor Concentration, Cigarettes, Use of Gas Appliances, and Home Location on BS Indoor Concentration (abs coeff/m)

	black smoke indoor (abs coeff/m)				
	Athens[b]	Basel	Helsinki	Prague	total data[b,c]
crude estimate (*N*)	42	41	82	20	185
BS outdoor (abs coeff/m)	0.62**	0.94**	0.82**	0.37*	0.70**
	(0.49−0.76)	(0.71−1.17)	(0.64−1.01)	(0.01−0.73)	(0.61−0.78)
constant	0.62**	0.07	-0.02	1.64**	0.29**
	(0.24−1.01)	(−0.26 to 0.40)	(−0.18 to 0.14)	(0.45−2.84)	(0.15−0.44)
adjusted R^2	0.83	0.66	0.62	0.31	0.83
multiple regression (*N*)	42	40	82	20	184
BS outdoor (abs coeff/m)	0.61**	0.86**	0.79**	0.20	0.61**
	(0.48−0.75)	(0.57−1.15)	(0.60−0.98)	(−0.22 to 0.63)	(0.49−0.73)
per cigarette/day	0.02*	0.03		−0.03	0.02
	(0.00−0.04)	(−0.01 to 0.08)		(−0.09 to 0.02)	(0.00−0.03)
per hour use of gas appliances		0.01**		0.03	0.00
		(0.01−0.02)		(−0.00 to 0.06)	(−0.01 to 0.02)
home location[d]					
downtown	0.11	−0.03	0.21*		0.18*
	(−0.27 to 0.49)	(−0.26 to 0.20)	(0.00−0.42)		(0.03−0.32)
suburban, high	0.39	0.16	0.16**		0.25**
	(−0.04 to 0.82)	(−0.28 to 0.60)	(0.06−0.27)		(0.09−0.40)
constant	0.35	0.12	−0.11	2.12**	0.46*
	(−0.08 to 0.79)	(−0.25 to 0.48)	(−0.27 to 0.06)	(0.75−3.50)	(0.10−0.82)
adjusted R^2	0.84	0.65	0.67	0.40	0.86

(b) Linear Regression Models for the Impacts of $PM_{2.5}$ Outdoor Concentration, Cigarettes, Use of Gas Appliances, and Home Location on $PM_{2.5}$ Indoor Concentration ($\mu g/m^3$)

	$PM_{2.5}$ indoor ($\mu g/m^3$)				
	Athens[b]	Basel	Helsinki	Prague	total data[b,c]
crude estimate (*N*)	42	41	82	20	185
$PM_{2.5}$ outdoor ($\mu g/m^3$)	0.50**	0.89**	0.60**	0.33	0.68**
	(0.20−0.79)	(0.27−1.50)	(0.44−0.76)	(−0.87 to 1.53)	(0.50−0.85)
constant	14.20**	3.94	3.21**	26.15	6.36**
	(4.09−24.30)	(−7.54 to 15.43)	(1.26−5.17)	(−9.26 to 61.55)	(3.16−9.55)
adjusted R^2	0.40	0.36	0.48	−0.04	0.41
multiple regression (*N*)	42	40	82	20	184
$PM_{2.5}$ outdoor ($\mu g/m^3$)	0.48**	0.74*	0.56**	−0.36	0.51**
	(0.19−0.76)	(0.12−1.36)	(0.36−0.77)	(−0.98 to 0.24)	(0.30−0.74)
per cigarette/day	0.38	3.22*		2.29*	0.94*
	(−0.11 to 0.87)	(0.34−6.10)		(0.11−4.48)	(0.09−1.78)
per hour use of gas appliances		0.23**		1.10**	0.70**
		(0.07−0.39)		(0.36−1.84)	(0.20−1.21)
home location[d]					
downtown	7.96*	0.34	3.11		3.12
	(0.09−15.84)	(−7.66 to 8.34)	(−1.31 to 7.54)		(−1.16 to 7.39)
suburban high	13.98*	6.62	1.87*		6.78**
	(3.29−24.67)	(−5.35 to 18.58)	(0.30−3.45)		(2.72−10.85)
constant	5.98	2.38	2.16*	28.42**	5.58
	(−3.28 to 15.24)	(−7.85 to 12.62)	(0.22−4.11)	(10.67−46.17)	(−2.10 to 13.25)
adjusted R^2	0.47	0.54	0.51	0.69	0.59

[a] Robust 95% confidence intervals in parentheses. (*) Significant at 5% level. (**) Significant at 1% level. [b] One influential point (pipe smoker) excluded in Athens and total data. [c] Multiple regression model for total data adjusted for "center". [d] Reference value: "suburban area with small buildings"; prague: all $N =$ "downtown".

54%, 51%, and 71%, respectively), with the exception of Prague. In Prague, impact of outdoor concentration on indoor concentration was small. Variables for indoor sources (use of gas appliances, cigarettes) were strong predictors for $PM_{2.5}$ indoor concentration, explaining 71% of the variation, but not for BS ($R^2 = 8\%$). In Helsinki, indoor variables did not predict indoor concentrations of BS. The location of the households, however, was associated stronger with BS than with $PM_{2.5}$. In Athens, on the other hand, home location variables differed significantly only for $PM_{2.5}$. In Basel and Prague, use of gas appliances had a significant impact on indoor concentrations, but the effects in Basel are very small. In Prague however, the effect of gas burning on $PM_{2.5}$ indoor concentration is strong.

Discussion

Indoor concentrations of BS, measured as light absorption of $PM_{2.5}$ filters, could be shown to be higher correlated with outdoor levels and apparently less influenced by indoor particle sources than indoor concentrations of $PM_{2.5}$.

The distribution of particulate air pollution across Europe showed the expected pattern, with low concentrations in Helsinki and much higher levels in Athens and Prague. Particle levels in Basel were in a medium range.

The models to predict indoor concentrations revealed consistent associations throughout the four centers, although differences between centers were remarkable. In the models including the four cities, besides the outdoor concentrations, the main predictors for $PM_{2.5}$ are the number of cigarettes smoked per day and the duration of the use of gas appliances, while, for BS levels indoors, the determining factor is the BS outdoor concentration. For all centers, the β coefficients for the impact of BS outdoor on BS indoor concentrations are higher as compared to the equivalents for $PM_{2.5}$.

However, the models for Prague differed from the others, showing generally lower impacts from ambient particle concentrations. The low impact from outdoors is consistent with low ventilation rates observed in Prague (Table 1) but, because of the small sample size, not conclusive. In this city, the multivariate models explain more of the variation for $PM_{2.5}$ than for BS. In the other cities, the explained variation (R^2) of univariate and multivariate indoor BS models is larger than for $PM_{2.5}$. Apart from Athens, cigarettes had a stronger impact on $PM_{2.5}$ concentrations as compared to BS. Among participants in Prague, heating by gas burning heaters, that are installed one per flat or one per floor and, therefore, produce indoor emissions, was common and may explain the observation of a strong impact by the use of gas appliances on $PM_{2.5}$ indoor concentration. The use of gas appliances in Basel shows small significant coefficients for both BS and $PM_{2.5}$. While, for $PM_{2.5}$, these results are consistent with findings of other studies (*31*−*33*), it remains questionable whether the observed contribution of gas appliances to BS is a real effect.

Comparing indoor with outdoor levels, ventilation has to be taken into account. Ventilation, measured as hours with an opened window during the measuring period of 48 h, varied substantially between the cities and the seasons. We compared correlations between indoors and outdoors, stratified by winter and summer, representing periods with low and high ventilation rates, respectively (see Table 1). One would expect lower correlation coefficients when the ventilation rate is lower. Indeed, comparing summer and winter correlations, this is strongly the case for $PM_{2.5}$ (Table 3). Indoor and outdoor BS, however, show only in Basel and Helsinki moderately weaker associations in winter. In Prague, correlation between indoor and outdoor BS is clearly better during winter months, although exclusion of one influential point would raise the Spearman coefficient for the summer to 0.43. These findings indicate that penetration of soot particles is less affected by different ventilation rates than is $PM_{2.5}$, which is in line with findings of other studies on indoor penetration of different particle-size fractions (*34*).

With respect to future applications, correlations between BS and $PM_{2.5}$ outdoors are of particular interest, as measurements will usually be done at fixed sites, monitoring outdoor air. High correlations between BS and $PM_{2.5}$ outdoors indicate that, in Athens and Prague, major parts of $PM_{2.5}$ originate from the same sources as BS, whereas the contribution to $PM_{2.5}$ of nonblack particles, such as sea dust, crustal, or secondary sulfate and nitrate particles, seems to be larger in Basel and Helsinki, where lower correlations have been observed. Stratified analysis by season should allow for conclusions about the particle sources. For example, the lower correlation between BS and $PM_{2.5}$ in Prague during summer is pointing toward the important role of carbonaceous particle emissions by (coal) heating in winter, whereas the more or less constant correlation throughout the year between the two measures in the other cities might indicate the predominance of particles emitted by traffic. In Helsinki, a smaller proportion of $PM_{2.5}$ seems to originate from the BS sources, as correlation between BS and $PM_{2.5}$ is lower compared to the other centers. Besides local primary combustion, nonblack particles from long distance transport, soil dust, and, especially in winter, from sea salt and deicing probably contribute significantly to $PM_{2.5}$ (*35*). Furthermore, the generally low levels and small ranges of pollutants in Helsinki may partially explain these findings.

Certain limitations are set to the interpretation of our data. The questionnaire variable on tobacco smoke, the main source for indoor particles, is not very precise; thus, its contribution to indoor concentrations may be underestimated. The tobacco variable used reflects mean cigarette consumption in the households, as declared by the participants, but not necessarily the amount of cigarettes or other tobacco products consumed during the measuring period. Traffic density was estimated by self-reported traffic on the street nearest home. It neither produced useful quantitative nor relative data for the model and, therefore, was dropped. Furthermore, our data is restricted to weekdays only. Comparing the models of the four cities, several center-specific characteristics have to be taken into account. First, the range of measured concentrations varies between centers, making comparisons of correlations more difficult. Also, the number of observations differs from center to center, yielding less stable estimates for Prague with only 20 observations as compared to, for example, Helsinki with 82. In Prague, all participants lived in the center of the city.

Various studies could show the validity of reflectance measurements on filters as a proxy for the EC content of particulate matter in filtered air (*35*−*38*). In comparison to thermo-analytical methods to detect EC, the BS method is nondestructive, far less expensive, and can be measured from the same filters which have been collected for gravimetric and elemental analysis. The results from the repeated measurements ascertained the reliability of the applied BS method; however, further standardization could increase its accuracy (*28*).

Our results underpin the hypothesis that BS indoors is predominantly originating from outdoor air pollution, whereas $PM_{2.5}$ can have significant additional sources indoors. Further, BS particles seem to penetrate into buildings more easily than $PM_{2.5}$, which is more depending on active ventilation. Therefore, BS measurements of ambient outdoor air can be considered a useful estimate of BS concentrations in indoor environments too.

Spatial variability within cities, however, is expected to be somewhat larger for BS than for $PM_{2.5}$ (with higher BS levels at traffic-exposed sites). This should be taken into account when selecting fixed monitoring sites (*39*, *40*).

Overall, the current work could show that indoor concentrations of BS highly correlate with outdoor concentrations, measured as reflectance of $PM_{2.5}$ filters. This allows for the consideration of BS as a supplementary indicator of particulate outdoor air pollution. BS seizes a health-relevant fraction of fine particles to which people are exposed indoors and outdoors and exposure to which can be assessed by monitoring outdoor concentrations. In conclusion, reflectance measurements of $PM_{2.5}$ filters can be recommended as a valid and cheap additional measure of EC to be used, for example, in studies on traffic-related air pollution and health.

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