Journal of the Air & Waste Management Association
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/uawm20

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Published online: 27 Dec 2011.

To cite this article: Martin Röösli , Charlotte Braun-Fährlander , Nino Künzli , Lucy Oglesby , Gaston Theis , Markus Camenzind , Patrick Mathys & Johannes Staehelin (2000) Spatial Variability of Different Fractions of Particulate Matter within an Urban Environment and between Urban and Rural Sites, Journal of the Air & Waste Management Association, 50:7, 1115-1124, DOI: 10.1080/10473289.2000.10464161

To link to this article: http://dx.doi.org/10.1080/10473289.2000.10464161

TECHNICAL PAPER

ISSN 1047-3289 *J. Air & Waste Manage. Assoc.* **50**:1115-1124

Spatial Variability of Different Fractions of Particulate Matter within an Urban Environment and between Urban and Rural Sites

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ABSTRACT

The spatial variability of different fractions of particulate matter (PM) was investigated in the city of Basel, Switzerland, based on measurements performed throughout 1997 with a mobile monitoring station at six sites and permanently recorded measurements from a fixed site. Additionally, PM_{10} measurements from the following year, which were concurrently recorded at two urban and two rural sites, were compared.

Generally, the spatial variability of PM_4, PM_{10}, and total suspended particulates (TSP) within this Swiss urban environment (area = 36 km^2) was rather limited. With the exception of one site in a street canyon next to a traffic light, traffic density had only a weak tendency to increase the levels of PM. Mean PM_{10} concentration at six sites with different traffic densities was in the range of less than ±10% of the mean urban PM_{10} level. However, comparing the mean PM levels on workdays to that on weekends indicated that the impact of human activities, including traffic, on ambient PM levels may be considerable.

Differences in the daily PM_{10} concentrations between urban and more elevated rural sites were strongly influenced by the stability of the atmosphere. In summer, when no persistent surface inversions exist, differences between urban and rural sites were rather small. It can therefore be concluded that spatial variability of annual mean PM concentration between urban and rural sites in the Basel area may more likely be caused by varying altitude than by distance to the city center.

IMPLICATIONS

It has become popular in recent years to use the concentration of PM as an indicator of air pollution exposure in epidemiologic studies. Since many of these studies assess the exposure of subjects based on one measurement per city, the accuracy of this technique will markedly affect the result of cross-sectional studies. High spatial variability of PM could result in a large non-differential misclassification of exposure that would lead to a smaller recognized health effect of air pollution.

The remarkable spatial homogeneity of long-term mean PM levels clearly reduces the error of assigning data from one fixed monitoring site to all study subjects living in Basel, as was done in recent cross-sectional health studies in Switzerland (Swiss Study on Air Pollution and Lung Diseases in Adluts, Swiss Study on Childhood Allergy and Respiratory Systems). In fact, all participants lived in urban Basel, rendering PM_4, PM_{10}, and even TSP useful city-wide surrogates for long-term exposure to outdoor air pollution.

INTRODUCTION

In the past decade, a body of epidemiologic evidence has emerged which demonstrates a range of health effects due to both long-term and short-term exposure to particulates at concentrations commonly occurring in ambient air. In epidemiologic studies investigating long-term effects of PM on health, exposure is usually assigned to a group or population level based on fixed-site monitoring data.[1-5] Several Swiss studies comparing the health of children[2] and adults[1,3] living in different areas of the country also based the

exposure assessment on one fixed-site monitor in each study area. However, since these studies were cross-sectional in design, little is known about the small-scale spatial variability of PM within the study areas, although it may affect the observed results. A higher spatial variability leads to a higher expected misclassification error associated with exposure, generally biasing the regression coefficient of the association between air pollution and health towards the null.

Several studies in the United States and Great Britain found long-term mean $PM_{2.5}$ and PM_{10} concentrations to be uniformly distributed within an urban environment (scale = 1–20 km), whereas a higher within-city variability was observed for the coarse fraction ($2.5 < d < 10\ \mu m$).[6-9] However, two recent studies reported within-city spatial variability for both $PM_{2.5}$ and PM_{10} in Canada and California,[10-11] but neither of these studies explicitly compared urban sites with different exposures to traffic.

A recent study in the Netherlands comparing daytime $PM_{2.5}$ and PM_{10} concentrations at urban background sites and at sites heavily exposed to traffic showed that PM levels were approximately 30% higher at the sites exposed to traffic, mainly due to increased concentrations of black smoke. In addition, the elemental concentrations of Fe and Si in the PM_{10} fraction were significantly higher at the traffic-exposed sites.[12] In Switzerland, measurements of vertical and horizontal PM_{10} concentration profiles orthogonal to a road in the city of Zurich showed that mean ground-level measurements of PM_{10} directly at the roadside were $30\ \mu g/m^3$. Roadside measurements 20 m above ground and ground-level measurements at a distance of 80 m from the road were approximately $4\ \mu g/m^3$ (13%) lower.[13]

However, until now, no systematic assessment of the spatial variability in a Swiss urban environment and its association with traffic density has been carried out. Results of studies from the United States and the Netherlands may be difficult to transfer to the Swiss situation because of existing differences in topography. In particular, it is not known whether the surrounding hills of Basel may affect the small-scale and meso-scale (20–30 km) spatial variability of PM.

Thus, in the present study, concentrations of different fractions of particulate matter (PM_4, PM_{10}, TSP, and coarse fractions) were measured during 1997 at six locations within the city of Basel that differed in traffic density. Between April 1998 and April 1999, additional PM_{10} measurements were concurrently performed at two urban and two rural sites in the Basel area.

The main purpose of the present study was to evaluate whether it is appropriate to use one monitoring site per city for PM exposure assessment in a cross-sectional health study. We quantified the spatial variability of different fractions of PM in an urban environment and between urban and rural locations. Moreover, we were interested in factors that might influence this spatial variability.

METHODS

Air Pollutant Measurements

For the present analyses, two sets of data were used, both of which were recorded in the Basel area. Basel is a city of about 200,000 inhabitants located in the northwestern part of Switzerland at 250 m above sea level, and it is surrounded by suburbs and more elevated rural areas.

The first data set included measurements performed between January 8 and December 22, 1997, as part of the Basel Risk Assessment Study of Ambient Air Pollutants (BRISKA). Twenty-four hour mean concentrations of TSP, PM_{10}, and PM_4 were recorded at six temporary sites within the city of Basel by means of a mobile monitoring station rotating every two weeks. (Due to technical problems, PM_4 was not measured until March 1.) This resulted in a 13-day sampling period from each site in each season (generally a total of 52 measurements per site). The six temporary sites differed in traffic density and were assumed to be representative of the traffic exposure of the Basel population (see Table 1). Two sites (1 and 2) were situated in typical residential areas with low road traffic density, one site (3) was located on a square in the very center of Basel, approximately 150 m away from a road with moderate traffic, and three sites (4–6) were located close to roads with a high traffic volume (16,300–29,500 passenger cars daily; 1130–4920 heavy-duty vehicles daily). The maximum distance between these six sites was 3.3 km. PM_{10} was also measured permanently at a fixed monitoring station of the Air Quality Management Agency of Basel (site C), to represent the urban background. In addition, permanent measurements of PM_4, PM_{10}, and TSP recorded throughout 1997 at a fixed-site monitoring station located at the outskirts of Basel (NABEL station)[14] were used as reference for the present analyses.

The second set of data was recorded from April 1, 1998, to April 30, 1999, at two rural and two urban sites (see Table 1). In addition to urban site C (urban background), a monitoring station was located in a street canyon with heavy traffic close to a traffic light (site D). Rural sites A and B were 25 and 20 km to the southeast of the city at altitudes of 900 and 600 m, respectively (Basel = 250 m). At all four stations, simultaneous measurements of 24-hr mean concentrations of PM_{10} were performed.

All PM concentrations (PM_4, PM_{10}, and TSP) were measured with High Volume Samplers (HVS) (Digitel DHA 80). The samples were collected on quartz filters (QF 20 Schleicher & Schuell), changed automatically at 12:00 a.m. local time (GMT + 1hr, wintertime). The air was sampled 2.40 m above ground. Particle concentrations were determined using pre- and post-sampling filter weights, taking into account

Table 1. Overview of the database and the characteristics of the measurement sites.

Date	PM	Sampling Period	Type	Site Name	Passenger Cars/ 24hr	Heavy Duty Vehicles/24hr	Comments
1/8/97– 12/22/97	PM_4, PM_{10}, TSP	permanent temporary sites, 13 days per season at each site using mobile monitoring station	Urban	NABEL	no traffic		outskirts of Basel (reference site)
				1	5000	260	residential
				2	5200	370	residential
				3	6200	500	center of Basel
				4	16,300	3410	high truck quota
				5	27,100	1130	sloped lane
				6	29,500	4920	cross-way
4/1/98– 4/30/99	PM_{10}	permanent	Urban	D	17,000	890	street canyon, traffic light
				C	11,500	950	PM_{10} also measured during 1997
			Rural	B	no traffic		Altitude = 600 m, 20 km from Basel
				A	no traffic		Altitude = 900 m, 25 km from Basel

Note: Traffic counts originate from the traffic inventory of the city of Basel.[17]

the sampled air volume. The filters were equilibrated under controlled temperature (15–24 °C) and relative humidity (47–60%) 48 hr prior to weighing, in order to ensure consistent values for particle mass.

As a quality control measurement, an additional 21 blank filters were placed in the HVS device at the monitoring site for 14 days but were not exposed. Weight differences between pre- and post-sampling weighing of these blank filters were, on average, associated with a concentration of –0.04 μg/m³ (standard error = 0.80 μg/m³).

The air-flow volume of the HVS was calibrated at 500 L/min at all monitoring sites. The flow was kept constant using a light barrier. The volume was corrected for temperature. The applied procedure is described in ref 15.

Data quality control of the measurements revealed that PM_4 had actually been recorded instead of $PM_{2.5}$. This was due to errors in the calculation of the geometry of the device's head, which led to the larger cut-off. To assess how comparable the PM_4 measurements were to the more common measurement of $PM_{2.5}$, the EPA-WINS Impactor used for the Air Pollution Exposure Distribution among Adult Urban Populations in Europe (EXPOLIS) Basel study[16] was repeatedly installed at the fixed-site NABEL monitoring station throughout 1997. The measurements indicated that PM_4 levels are highly correlated with $PM_{2.5}$ levels, though on average PM_4 levels are 11% higher (25.2 vs 22.3 μg/m³). The coarse fractions were determined by calculating the differences between the three size fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$, and $PM_{(TSP-4)}$).

Statistical Analyses

The measurements at sites 1–6 in Basel, performed during 1997 by the mobile monitoring station, could not be directly used to calculate annual means and to assess the spatial variability, as they had not been performed concurrently. Comparing raw PM levels between the sites could have yielded differences in the concentrations of PM, simply owing to differing meteorological conditions prevailing during the sampling period, or to temporal varying source strengths such as traffic density, or both. Two methods were used to deal with such influences on the calculation of mean values for each temporary site. First, the 52 measurements from each temporary site were compared with the concurrent measurements from the permanent NABEL site. The Pearson correlation coefficients and the mean ratios between the measurements from each temporary site and the corresponding measurements from the NABEL site were calculated. As the distribution of ratios is asymmetrical, mean values were obtained by taking the logarithm of the ratios and then exponentiating that value.

Second, to adjust for meteorological conditions, a multiple regression model was designed for each site and for each particulate size fraction. The daily PM measurements from the temporary station were regressed against the respective daily PM measurements from the NABEL station and against meteorological parameters. The following mathematical expression describes the model:

$$C_{temp.site} = a + b \cdot C_{NABEL} + \sum_{i=1}^{n} k_i \cdot m_i \qquad (1)$$

where C = concentration of PM_4, PM_{10}, TSP, or coarse fractions at the six temporary sites (temp. site) and at the reference site (NABEL), m_i = meteorological parameters, and a, b, and k_i are estimated coefficients.

Meteorological parameters that were significant in at least one model were considered as covariates of the final model that was used for all sites. The following meteorological parameters were included in the preliminary models: season (binary: Oct–Mar = 1), daily means of temperature

and humidity, daily sum of global radiation, daily minimum and maximum of the temperature gradient between ground level and a tower measurement 243 m above ground, and the daily vector sum of all wind-direction measurements taken every 10 min.

In general, each model was built with 52 pairs of daily PM measurements (temporary and NABEL) and meteorological parameters. Based on these models, daily PM concentrations for the remaining days were predicted. Annual and seasonal means were then calculated on the basis of all observed and predicted PM values. Because of the missing PM_4 values in January and February, models for PM_{10} and TSP were calculated for both the January–December and March–December periods.

The models were validated with the PM_{10} measurements from site C, which had been measured permanently throughout the year. The measurement periods, 1–6, were defined to correspond to the four time intervals during which the mobile monitoring station was located at the respective sites 1–6. Each measurement period consisted of 52 days with PM measurements (13 days in each season). First, the mean PM_{10} concentration at site C was calculated for each measurement period, yielding 22.7 $\mu g/m^3$, 24.3 $\mu g/m^3$, 27.8 $\mu g/m^3$, 30.0 $\mu g/m^3$, 23.3 $\mu g/m^3$, and 32.2 $\mu g/m^3$, respectively, for measurement periods 1–6. The annual mean at site C computed from the daily measurements throughout the year was 27.9 $\mu g/m^3$. Thus, estimates of annual means based on averages of the available PM measurements during one measurement period would give rather variable results compared to the "true" mean of 27.9 $\mu g/m^3$.

Second, the PM_{10} readings of site C in each measurement period were used as dependent variables for the regression models described above (i.e., including the PM_{10} values of the reference station and meteorological parameters as covariates). Based on these models, PM_{10} concentrations for the remaining days were predicted and annual means calculated. The model-based annual means for site C could then be compared to the measured annual mean PM_{10} concentration at site C. The six annual means derived from the regression models were 26.9 $\mu g/m^3$, 27.8 $\mu g/m^3$, 26.4 $\mu g/m^3$, 27.6 $\mu g/m^3$, 28.4 $\mu g/m^3$, and 30.4 $\mu g/m^3$, respectively, for measurement periods 1–6. These results indicate that the annual means computed from regression models are indeed close to the measured mean concentration of 27.9 $\mu g/m^3$ at site C, although some inaccuracy could not be eliminated.

In order to compare the spatial variability of the different size fractions of PM across the six temporary sites, coefficients of spatial variation were calculated. For each size fraction, the overall mean of the six sites and the corresponding standard deviation were computed using the measurements from March to December to avoid undue influence from the missing PM_4 measurements in January and February. The coefficient of spatial variation (cv) for each particulate size fraction was obtained by dividing the standard deviation by the overall mean.

To evaluate the hypothesis that proximity to traffic might influence the local PM measurements, the annual mean PM concentrations of each of the six temporary sites were plotted against the respective daily mean number of passenger cars and heavy-duty vehicles, and linear regression models were calculated to estimate the regression coefficients of this association. The information on traffic density was provided by the traffic inventory of the canton Basel-Stadt.[17] This inventory contains information on average numbers of cars and trucks passing a given address each hour and is updated by traffic counts every 3–4 years. Again, these analyses were based on the measurements from March to December, 1997.

The differences between mean PM concentrations on workdays and weekends may reflect the influence of regional anthropogenic sources, since many human activities such as industrial processes, construction work, or road traffic are reduced on weekends. Thus, the mean concentration for each PM fraction was calculated for each day of the week. In addition, the ratio between mean concentrations on workdays and weekends and the corresponding two-sided t-test were computed. These analyses were based on the data provided by the fixed monitoring station (NABEL) recorded between March and December, 1997.

The second data set consisting of simultaneously performed measurements at four fixed monitoring sites (two urban and two rural) was used to assess differences in annual mean PM_{10} concentrations between urban and rural sites. In addition, monthly mean values at the two rural and the two urban sites were compared to assess whether seasonal factors such as the number of days with thermal inversion may influence urban–rural differences. Temperature gradients between the urban and rural sites were computed by calculating the differences between the daily maximum temperature at the urban (NABEL) station and rural station A. A positive gradient is indicative of an inversion layer.

Data Analyses

Data were analyzed using S-Plus 4.5 Professional Release 2 for Windows.

RESULTS

Spatial Variability of Different Fractions of Particulate Matter within an Urban Environment

Overall mean PM concentrations across the six temporary stations for the measurements performed between March and December were 33.1 $\mu g/m^3$ (TSP), 24.7

μg/m³ (PM_{10}), and 19.9 μg/m³ (PM_4). The annual mean TSP and PM_{10} concentrations (January–December) were 3.8 and 2.4 μg/m³ higher, respectively, than mean values for the period from March to December, due to an episode of high air pollution in January (PM_4 was not measured in January and February). The ratio of the mean PM_4 versus PM_{10} concentrations (March–December) was 81%, and the ratio was 75% for the mean PM_{10} versus TSP concentrations.

Comparisons of the 52 measurements from each temporary site with the concurrent measurements from the NABEL site are shown in Table 2. In each size fraction, the mean ratios of the concurrent measurements were generally lowest at the residential sites (1 and 2). Only $PM_{(10-4)}$ did not show this pattern. For the coarse fractions, a wide range of ratios was found between the six sites, whereas for PM_4, PM_{10}, and TSP a smaller range was found, indicating more spatial uniformity. The correlation coefficients (R^2) of the measurement were highest for PM_{10} but were also mostly above 0.9 for PM_4 and TSP. The R^2 of all coarse fractions ($PM_{(TSP-10)}$, $PM_{(TSP-4)}$, $PM_{(10-4)}$) was below 0.8 at all six sites. The R^2 of the measurements was always lower than the corresponding R^2 from the meteorology-based regression models, even taking into account the lower number of degrees of freedom in the regression models. The standard errors of the regression coefficient of the NABEL measurements indicate that the t-value was lower for the coarse fraction models than for the PM_4, PM_{10}, and TSP models.

Table 2. Mean ratio (ratio measured) and R^2 (R^2 measured) between concurrently measured PM concentrations at the six temporary sites and the permanent NABEL site, as well as R^2 (R^2 model) and regression coefficients β ±standard error (β ±SE model) of the regression models. The latter included the measurements from the temporary sites as dependent variables and the permanent NABEL measurements and meteorological parameters as explanatory variables.

		Residential 1 [μg/m³]	Residential 2 [μg/m³]	Center 3 [μg/m³]	Traffic 4 [μg/m³]	Traffic 5 [μg/m³]	Traffic 6 [μg/m³]
PM_{10}	ratio (measured)	0.90	0.94	0.99	0.97	0.96	1.04
(Jan–Dec)	R^2 (measured)	0.97	0.97	0.97	0.98	0.96	0.98
	R^2 (model)	0.99	0.99	0.99	0.99	0.99	0.99
	β (±SE) model	0.94 (±0.02)	0.97 (±0.03)	0.88 (±0.03)	0.90 (±0.03)	1.03 (±0.03)	1.03 (±0.04)
TSP	ratio (measured)	1.06	1.02	1.18	1.19	1.12	1.34
(Jan–Dec)	R^2 (measured)	0.84	0.94	0.95	0.93	0.93	0.92
	R^2 (model)	0.93	0.97	0.97	0.94	0.95	0.94
	β (±SE) model	0.93 (±0.06)	1.20 (±0.06)	1.05 (±0.07)	1.20 (±0.07)	1.14 (±0.06)	1.06 (±0.07)
PM_4	ratio (measured)	0.75	0.82	0.92	0.88	0.99	0.98
(Mar–Dec)	R^2 (measured)	0.95	0.78	0.98	0.91	0.97	0.98
	R^2 (model)	0.98	0.93	0.99	0.98	0.99	0.99
	β(±SE) model	0.85 (±0.04)	0.91 (±0.07)	0.93 (±0.03)	0.97 (±0.04)	1.06 (±0.03)	0.94 (±0.05)
PM_{10}	ratio (measured)	0.88	0.93	0.99	0.99	0.96	1.03
(Mar–Dec)	R^2 (measured)	0.97	0.88	0.97	0.93	0.96	0.98
	R^2 (model)	0.99	0.96	0.99	0.97	0.99	0.99
	β (±SE) model	0.91 (±0.03)	0.92 (±0.06)	0.88 (±0.03)	0.98 (±0.05)	1.03 (±0.03)	1.00 (±0.05)
TSP	ratio (measured)	1.00	0.97	1.18	1.20	1.12	1.32
(Mar–Dec)	R^2 (measured)	0.94	0.85	0.95	0.79	0.93	0.92
	R^2 (model)	0.97	0.96	0.97	0.86	0.95	0.95
	β(±SE) model	0.92 (±0.05)	1.00 (±0.07)	1.05 (±0.07)	1.14 (±0.16)	1.14 (±0.06)	1.06 (±0.10)
$PM_{(TSP-10)}$	ratio (measured)	1.92	1.01	3.52	2.66	4.25	3.51
(Mar–Dec)	R^2 (measured)	0.73	0.56	0.43	0.44	0.46	0.63
	R^2 (model)	0.86	0.87	0.53	0.69	0.63	0.83
	β(±SE) model	0.85 (±0.26)	1.09 (±0.21)	0.65 (±0.46)	1.28 (±0.39)	1.35 (±0.36)	1.93 (±0.35)
$PM_{(TSP-4)}$	ratio (measured)	1.61	1.40	2.04	1.93	1.40	2.24
(Mar–Dec)	R^2 (measured)	0.68	0.60	0.66	0.57	0.61	0.63
	R^2 (model)	0.87	0.84	0.72	0.76	0.74	0.80
	β(±SE) model	0.87 (±0.16)	0.90 (±0.16)	1.56 (±0.35)	1.33 (±0.29)	1.33 (±0.21)	1.57 (±0.23)
$PM_{(10-4)}$	ratio (measured)	1.35	1.45	1.26	1.43	0.80	1.37
(Mar–Dec)	R^2 (measured)	0.30	0.31	0.44	0.66	0.11	0.76
	R^2 (model)	0.59	0.90	0.66	0.79	0.44	0.92
	β (±SE) model	0.75 (±0.16)	0.71 (±0.15)	0.50 (±0.11)	0.80 (±0.12)	0.55 (±0.20)	1.04 (±0.13)

Note: β is the coefficient of the permanent NABEL measurements. Model coefficients of the meteorological parameters are not shown, as they had a considerably weaker influence than the measurements from the NABEL site.

Table 3 shows the mean concentrations of the different fractions of particulate matter at the six temporary urban sites, which were calculated based on the regression models. Again, lowest mean concentrations of PM_4, PM_{10}, and TSP occurred at the residential sites (1 and 2) and had a weak tendency to increase with increasing road traffic density. The biggest differences between the residential sites and the more traffic-exposed sites were detected for $PM_{(TSP-10)}$ and $PM_{(TSP-4)}$, but not for $PM_{(10-4)}$. The calculated coefficients of spatial variation, as well as the ratio of the highest to the lowest mean concentration at the six sites, were clearly higher for the coarse fractions than for PM_4, PM_{10}, and TSP. The lowest spatial variability was found for the PM_{10} fraction.

To evaluate whether there was an association between the traffic density at a given site and the respective concentrations of the different size fractions of PM, linear regression models were calculated. The results are given in Table 4, and Figure 1 illustrates the association. Overall, only a very weak association between local PM concentrations and traffic density was observed, and it reached statistical significance only for PM_4 and passenger cars.

Figure 2 depicts the mean concentrations of the different PM size fractions for each day of the week. The concentrations of all size fractions were lowest on Sunday, increased from Monday to Wednesday, and decreased again on Friday and Saturday. The ratios between the mean concentrations on workdays and weekends increased with increasing particle size. The concentration of $PM_{(TSP-10)}$ was 51% higher on workdays than on weekends, and $PM_{(10-4)}$ was 32% higher. On workdays, TSP, PM_{10}, and PM_4 were elevated by 20, 17, and 14%, respectively. These differences were statistically significant for PM_{10} ($p = 0.045$), TSP ($p = 0.012$), $PM_{(10-4)}$ ($p = 0.0007$), and $PM_{(TSP-10)}$ ($p = 0.0028$). For PM_4, the difference was of borderline significance ($p = 0.107$).

Spatial Variability of PM_{10} between Urban and Rural Sites

To assess differences in PM levels between the rural and urban sites, the PM_{10} measurements monitored from April 1, 1998, to April 30, 1999, were used. During this period, the PM_{10} levels were generally lower than in 1997 due to different meteorological conditions. In 1997, the annual mean PM_{10} concentration at urban site C was 27.9 $\mu g/m^3$, whereas the average PM_{10} level between April 1998 and March 1999 was only 22.4 $\mu g/m^3$. Figure 3 gives the boxplots for the measurements at the four sites from April 1998 to March 1999. The highest PM_{10} concentrations were measured at urban site D, situated in a street canyon with high traffic density. The annual mean value at this site was more than one-third higher than the PM_{10} concentration at urban site C, which represented urban background exposure. The annual PM_{10} levels at rural sites A and B were 48% and 31% below the levels of site C, respectively.

Figure 4 shows the monthly PM_{10} concentrations at the four monitoring sites. During the summer months

Table 3. Calculated mean concentration of different fractions of particulate matter for the periods of January–December and March–December as well as coefficient of spatial variability and the ratio of the highest and lowest concentration of each PM fraction.

	Residential 1 [$\mu g/m^3$]	Residential 2 [$\mu g/m^3$]	Centre 3 [$\mu g/m^3$]	Traffic 4 [$\mu g/m^3$]	Traffic 5 [$\mu g/m^3$]	Traffic 6 [$\mu g/m^3$]	cv [%]	PM Ratio highest/lowest
PM_{10} (Jan–Dec)	27.6	28.4	30.1	28.6	29.6	32.0	5.2	1.16
(CI)	(26.1; 29.2)	(26.4; 30.4)	(27.9; 32.3)	(26.1; 31.0)	(27.7; 31.5)	(28.7; 35.2)		
TSP (Jan–Dec)	34.5	36.4	41.5	40.8	38.9	45.4	9.9	1.32
(CI)	(30.1; 38.8)	(32.6; 40.1)	(36.7; 46.3)	(33.7; 47.8)	(35.2; 42.7)	(39.2; 51.5)		
PM_4 (Mar–Dec)	17.4	18.0	20.5	19.7	21.4	22.1	9.4	1.27
(CI)	(15.3; 19.5)	(16.0; 19.9)	(18.6; 22.5)	(18.2; 21.1)	(19.6; 23.1)	(18.7; 25.4)		
PM_{10} (Mar–Dec)	23.0	23.7	26.0	24.8	24.3	28.0	7.2	1.21
(CI)	(21.2; 24.9)	(21.7; 25.7)	(23.8; 28.2)	(23.0; 26.7)	(22.4; 26.2)	(23.8; 32.1)		
TSP (Mar–Dec)	30.6	29.4	36.9	34.0	34.2	42.1	13.3	1.43
(CI)	(27.3; 34.0)	(27.0; 31.8)	(32.1; 41.8)	(26.3; 41.7)	(30.4; 37.9)	(34.5; 49.7)		
$PM_{(TSP-10)}$ (Mar–Dec)	7.3	5.1	10.4	8.2	9.5	14.1	33.8	2.78
(CI)	(4.7; 9.8)	(3.0; 7.1)	(5.4; 15.4)	(1.2; 15.3)	(5.5; 13.5)	(9.9; 18.3)		
$PM_{(TSP-4)}$ (Mar–Dec)	12.9	10.8	16.1	13.6	12.6	19.9	22.4	1.84
(CI)	(10.1; 15.8)	(8.7; 12.9)	(10.8; 21.3)	(6.5; 20.7)	(9.1; 16.1)	(14.8; 24.9)		
$PM_{(10-4)}$ (Mar–Dec)	5.4	5.4	5.6	4.9	2.6	6.3	25.2	2.40
(CI)	(3.8; 7.0)	(4.4; 6.5)	(4.0; 7.2)	(3.7; 6.2)	(1.0; 4.2)	(4.4; 8.1)		

Note: cv = standard deviation/mean; CI = confidence interval.

Table 4. Results from the linear regression analyses of mean PM concentration at the six urban sites and mean number of cars and trucks passing the site per day.

	Passenger Car Model				Truck Model			
	Intercept	β	(CI of β)	R^2	Intercept	β	(CI of β)	R^2
PM_4	17.8	1.38	(0.10; 2.65)	0.69	18.8	0.59	(-0.46; 1.63)	0.38
PM_{10}	23.6	0.91	(-0.91; 2.73)	0.33	23.7	0.69	(-0.15; 1.54)	0.56
TSP	30.5	2.73	(-1.48; 6.95)	0.45	31.4	1.76	(-0.42; 3.94)	0.56
$PM_{(TSP-10)}$	6.2	1.95	(-0.71; 4.62)	0.51	7.1	1.11	(-0.47; 2.68)	0.49
$PM_{(TSP-4)}$	12.2	1.45	(-1.97; 4.87)	0.26	12.2	1.18	(-0.43; 2.79)	0.51
$PM_{(10-4)}$	5.6	-0.39	(-1.86; 1.09)	0.12	4.7	0.18	(-0.69; 1.05)	0.08

Note: β refers to an increase of 10,000 passenger cars and 1000 trucks.

(April–October), the variability of the monthly PM_{10} levels was comparable at all four stations. From November to March, however, the monthly mean PM_{10} concentration increased more at the two urban sites than at the rural stations. An analysis of the meteorological conditions revealed days with positive temperature gradients from December to February between the more elevated rural site A (900 m) and the urban NABEL site (250 m). This is characteristic for the presence of an inversion layer and was positively associated with increased levels of PM_{10} at the urban site (see Figure 5). During the warm months, on the other hand, vertical temperature gradients were rarely positive.

DISCUSSION

In general, ambient levels of PM_4, PM_{10}, and TSP were distributed rather homogeneously within the urban area of Basel. A higher spatial variability was found for the coarse fractions ($PM_{(TSP-10)}$, $PM_{(TSP-4)}$, $PM_{(10-4)}$). These findings are in line with previous findings[6-9,16] and did not depend on the method used to analyze the spatial variability. However, using multiple regression models to calculate mean

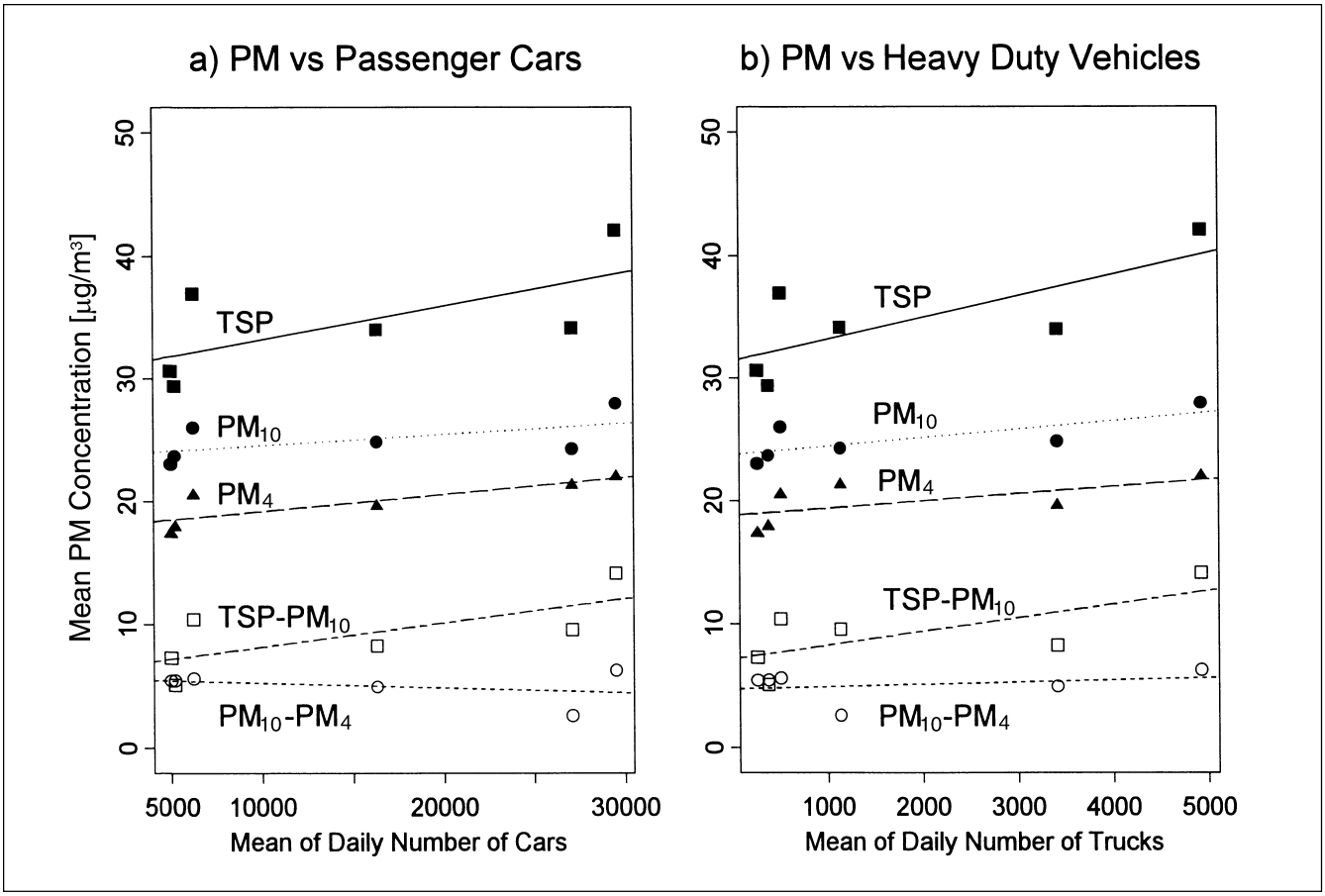

Figure 1. Mean calculated PM concentration of the different size fractions of PM for March 1 to December 31, 1997, plotted against the mean daily counts of passenger cars (a) and heavy-duty vehicles (b).

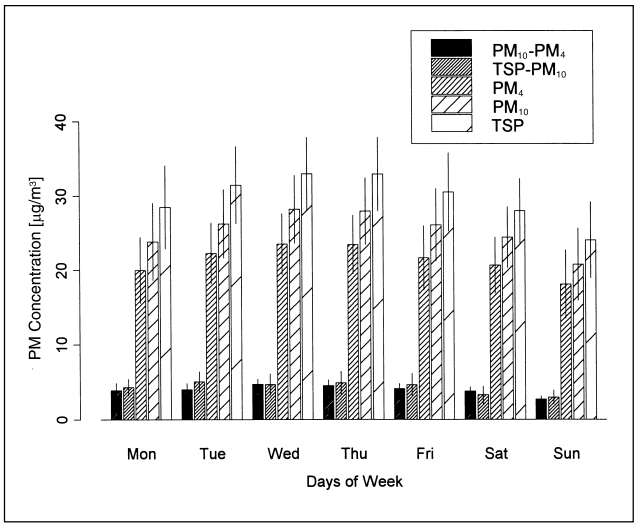

Figure 2. Mean concentrations (± confidence interval) of PM_4, PM_{10}, TSP, and coarse fractions ($PM_{(10-4)}$, $PM_{(TSP-10)}$) by day of the week. Measurements are from the fixed monitoring site (NABEL) for March–December 1997.

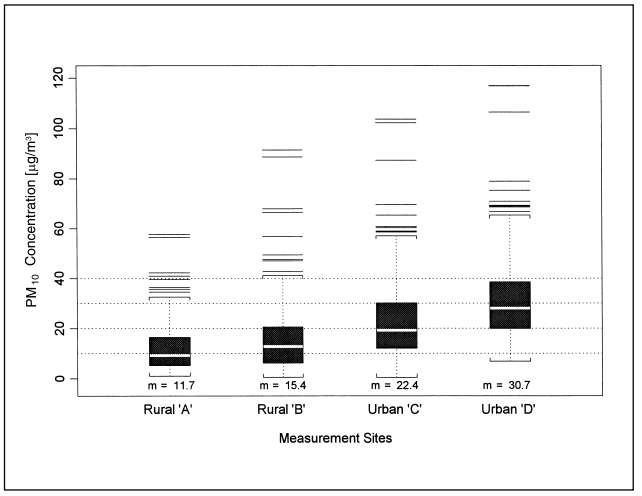

Figure 3. Boxplots of PM_{10} measurements at two urban and two rural sites from April 1, 1998, to March 31, 1999. The box contains 50% of all measurements; the white line represents the median value and *m* equals the arithmetic mean value.

values yielded slightly higher R^2 values as compared to the crude analyses. Comparing only ratios without taking meteorological conditions into account may bias the result, because the ratios systematically varied with varying pollution levels, which in turn are a function of the meteorological conditions.

The slightly higher spatial variability of PM_4 as compared to PM_{10} was an unexpected finding. It is possible that this result occurred by chance. Alternatively, one might speculate that the size distribution between PM_4 and PM_{10} at the two residential sites as compared to the more traffic-exposed sites was slightly changed on average due to known processes[18,19,20] such as sedimentation,

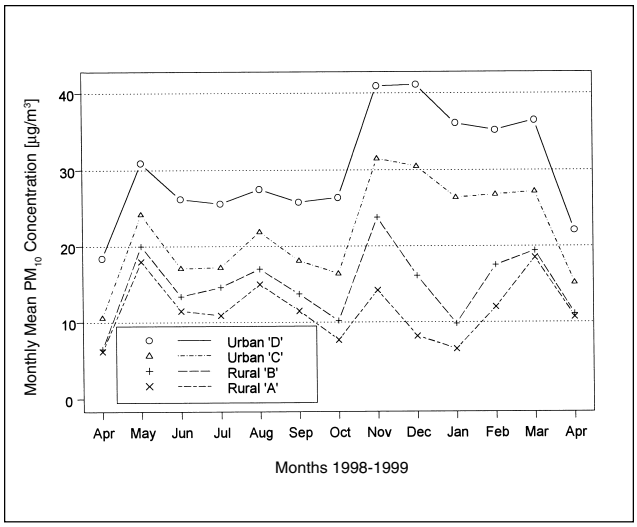

Figure 4. Monthly mean values of PM_{10} at two urban and two rural sites.

coagulation, and condensation of water vapor on the particle surface. These processes are effective in the ultrafine particles (< 50 nm) on a small spatial scale.[21,22] Whether they influence the size distribution of PM_4 and PM_{10} in our study area cannot be determined with certainty with our data.

Only a weak and mostly insignificant association was observed between the volume of road traffic at the six sites and the concentration of PM. The models are, however, limited due to the relatively small range of observed traffic and the fact that only six sites were analyzed. Moreover, other factors, such as building density and distance to the road plane, were not absolutely identical among the six sites. The PM concentration at site 3 was higher than expected from traffic counts alone, suggesting that factors other than traffic influence the long-term concentration of PM. On the other hand, PM_{10} measurements from site D demonstrate that, at some locations, considerable small-scale spatial gradients in the long-term levels of PM may occur within Basel due to a specific traffic situation. Site D was located in a street canyon close to a traffic light.

The differences in the mean PM concentrations on different days of the week, however, suggest that human activities that are reduced on weekends, such as road traffic, construction work, and industrial processes, can markedly influence the levels of PM in Basel, even if they generally were not found to have a particular effect on the small-scale spatial variability.

The differences between urban and rural sites were determined by the existence of an inversion layer. A strong impact of persistent surface inversion layers on the levels of PM has also been found in the Swiss alpine region.[23] In Basel, the accumulation of pollutants in the air on days with persistent surface inversion due to reduced dilution

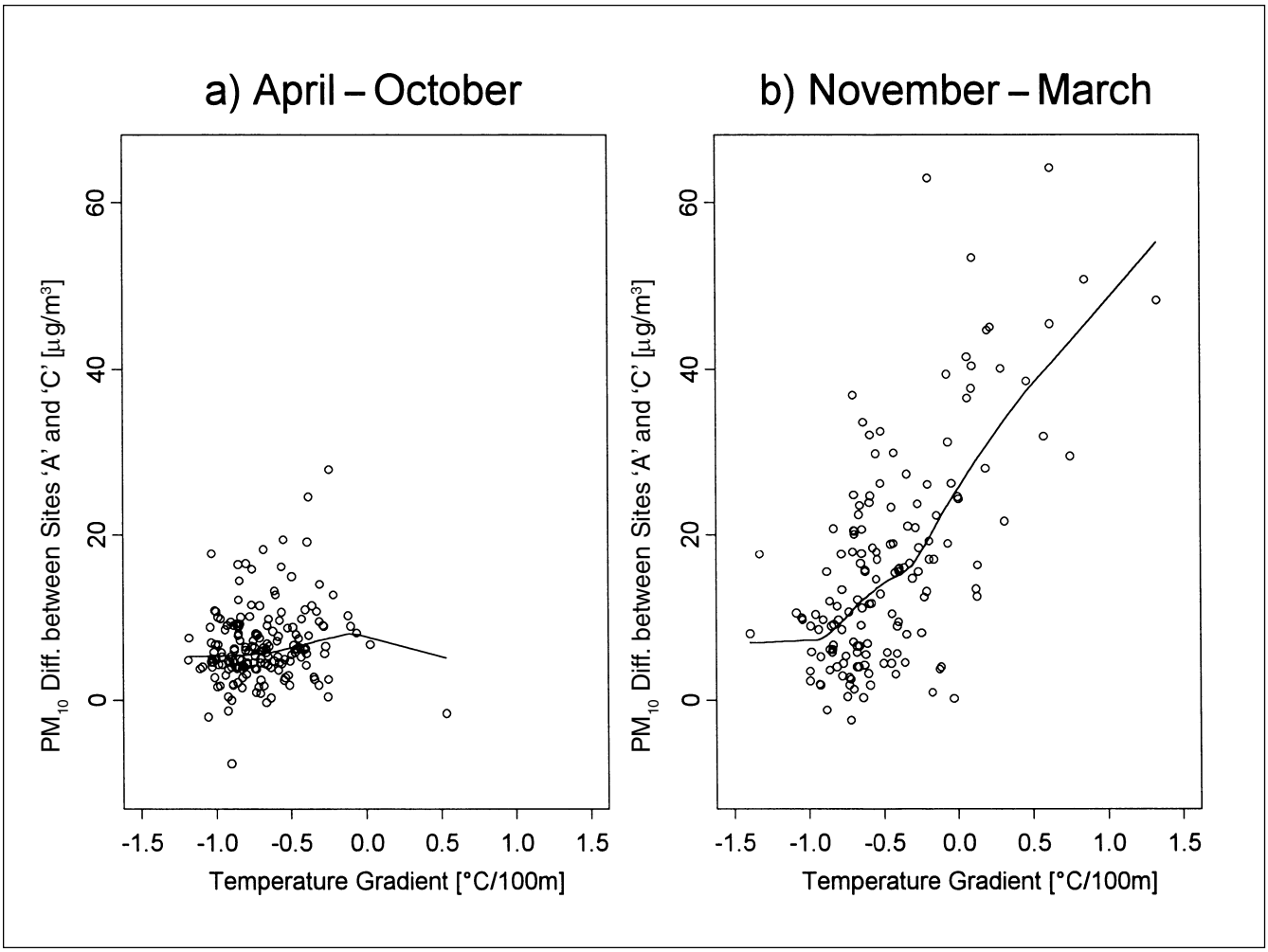

Figure 5. Smooth plot of the association of vertical temperature gradient and daily PM_{10} difference between urban site C and rural site A for April–October 1997 (a) and November–March (b).

can be demonstrated by the mean PM_{10} levels on the 21 days that had a positive temperature gradient between rural site A and Basel (November–March). The mean PM_{10} concentrations were 56 (D), 48 (C), 27 (B), and 12 µg/m^3 (A). The average urban value (C) for this period was more than twice as high as the annual mean PM_{10} level at this site, whereas the PM_{10} concentration at site A was equal to its annual mean value. It is known that, in general, the altitude of the upper boundary of an inversion layer is lowest during December and January, and of the 21 days with a positive temperature gradient, 15 occurred during December 1998 and January 1999. In November and March, persistent surface inversions are not unusual, but the boundary is often higher than 900 m, resulting in increased monthly PM_{10} levels at the elevated rural sites as well. With our data, it was not possible to identify unequivocal days with inversion above 900 m, due to missing information on the vertical temperature gradient. In summer, persistent surface inversions rarely occur. Thus, from April to October, only two days with a positive temperature gradient between sites A and C were observed, and they may also have been caused by factors other than inversions. Therefore, it can be concluded that differences between PM concentrations at urban and rural sites may be more likely caused by varying altitude than by the distance from the city center.

The remarkable spatial homogeneity in long-term mean PM levels clearly reduces the error of assigning data from one fixed monitoring site to all study subjects living in Basel, as was done in the former Swiss health study SAPALDIA. In fact, all participants lived in urban Basel, rendering PM_4, PM_{10}, and even TSP useful city-wide surrogates for long-term exposure to outdoor air pollution.

ACKNOWLEDGMENTS

The authors thank Catherine Wyler and Franziska Zimmerli for their careful proofreading of the manuscript. Thanks also to Hansruedi Moser from the Air Quality Management Agency of Basel for helpful discussions. This study was funded by the foundation MGU (Man–Society–Environment)

of the University of Basel and by the BUWAL (Swiss Federal Agency of Environment, Forest, and Landscape), Bern. We would also like to acknowledge the Air Quality Management Agency of Basel for providing meteorological and air pollution data.

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