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Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area

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Abstract

Numerous epidemiological studies have demonstrated the association between particle mass (PM) concentration in outside air and the occurrence of health related problems and/or diseases. However, much less is known about indoor PM concentrations and associated health risks. In particular, data are needed on air quality in schools, since children are assumed to be more vulnerable to health hazards and spend a large part of their time in classrooms.

On this background, we evaluated indoor air quality in 64 schools in the city of Munich and a neighbouring district outside the city boundary. In winter 2004–2005 in 92 classrooms, and in summer 2005 in 75 classrooms, data on indoor air climate parameters (temperature, relative humidity), carbon dioxide (CO_2) and various dust particle fractions (PM_{10}, $PM_{2.5}$) were collected; for the latter both gravimetrical and continuous measurements by laser aerosol spectrometer (LAS) were implemented. In the summer period, the particle number concentration (PNC), was determined using a scanning mobility particle sizer (SMPS). Additionally, data on room and building characteristics were collected by use of a standardized form. Only data collected during teaching hours were considered in analysis. For continuously measured parameters the daily median was used to describe the exposure level in a classroom.

The median indoor CO_2 concentration in a classroom was 1603 ppm in winter and 405 ppm in summer. With LAS in winter, median PM concentrations of 19.8 $\mu g\,m^{-3}$ ($PM_{2.5}$) and 91.5 $\mu g\,m^{-3}$ (PM_{10}) were observed, in summer PM concentrations were significantly reduced (median $PM_{2.5} = 12.7\,\mu g\,m^{-3}$, median $PM_{10} = 64.9\,\mu g\,m^{-3}$). $PM_{2.5}$ concentrations determined by the gravimetric method were in general higher (median in winter: 36.7 $\mu g\,m^{-3}$, median in summer: 20.2 $\mu g\,m^{-3}$) but correlated strongly with the LAS-measured results. In explorative analysis, we identified a significant increase of LAS-measured $PM_{2.5}$ by 1.7 $\mu g\,m^{-3}$ per increase in humidity by 10%, by 0.5 $\mu g\,m^{-3}$ per increase in CO_2 indoor concentration by 100 ppm, and a decrease by 2.8 $\mu g\,m^{-3}$ in 5–7th grade classes and by 7.3 $\mu g\,m^{-3}$ in class 8–11 compared to 1–4th class. During the winter period, the associations were stronger regarding class level, reverse regarding humidity (a decrease by 6.4 $\mu g\,m^{-3}$ per increase in 10% humidity) and absent regarding CO_2 indoor concentration. The median PNC measured in 36 classrooms ranged between 2622 and 12,145 particles cm^{-3} (median: 5660 particles cm^{-3}).

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The results clearly show that exposure to particulate matter in school is high. The increased PM concentrations in winter and their correlation with high CO_2 concentrations indicate that inadequate ventilation plays a major role in the establishment of poor indoor air quality. Additionally, the increased PM concentration in low level classes and in rooms with high number of pupils suggest that the physical activity of pupils, which is assumed to be more pronounced in younger children, contributes to a constant process of resuspension of sedimented particles. Further investigations are necessary to increase knowledge on predictors of PM concentration, to assess the toxic potential of indoor particles and to develop and test strategies how to ensure improved indoor air quality in schools.

Keywords: Indoor; Schools; Classrooms; Particulate matter; Carbon dioxide; Particle number concentration; Ultrafine particles

1. Introduction

Numerous epidemiological studies have been carried out during the last decades which demonstrated the correlation between the pollution of outside air with toxic substances and the occurrence of health related problems and/or diseases. On the background of these findings, particularly in recent years the focus of research has clearly shifted towards particulate matter, notably fine particles—e.g. $PM_{2.5}$—and ultra fine particles with diameters less than 100 nm (EPA (US-Environmental Protection Agency), 2004; WHO (World Health Organization), 2004).

Stationary outdoor particle mass (PM) concentration is a frequently used indicator of PM exposure in large epidemiologic cohort studies and has been shown to be associated with health risks on the population level. Stationary outdoor PM concentration, however, is only of limited use for the assessment of individual exposure level and thus estimates of relative risk derived from these studies might be biased by measurement errors. Individual exposure to PM occurs in diverse microenvironments, where particles may originate from a wide variety of sources. In the indoor environment, in which people spend most of their time, both indoor and outdoor sources contribute to PM levels. Indoor PM is affected by ambient concentrations, air exchange rates, penetration factors, as well as deposition and resuspension mechanisms. In this complex microenvironment, activities such as cooking, cleaning, walking, and particularly smoking cause the formation of PM in indoor air.

Although numerous measurements of PM in the outside air have been conducted, only few data on indoor air pollution are available. Investigations into PM are even more rare for schools and related indoor facilities than for other microenvironments. As school-aged children spend approximately 30% of their daytime in school and may be regarded as particularly vulnerable to potential health hazards, more precise data on exposure to air pollution in this setting is urgently required.

Therefore, the aim of the present study was to determine the quality of indoor air in different seasons in a large number of schools in Bavaria, in particular the concentration of particulate matter. Furthermore, we aimed for the comparison of results on PM concentrations obtained by two different established techniques and for the identification of potential determinants of PM concentration in exporative analysis.

2. Methods

2.1. Study description and sampling sites

Sixty-four primary and secondary schools located in the northern part of the city of Munich and in a neighbouring rural district took part in the study, thus allowing data collection from city, rural and small town areas. In the winter measurement period (December 2004 to March 2005) 92 classrooms, and in the summer measurement period (May to July 2005) 75 classrooms, respectively, were included. Fifty-eight classrooms were measured at both occasions.

In each classroom measurements were done during one school day (about 5 h), each day a different school was monitored. The sampling and measuring position in the classroom was opposite to the black board, about one meter above floor level, the level at which the pupils would normally inhale. This was chosen as a ''typical'' location inside the room, it being away from the door, thus avoiding disturbances resulting from air currents.

Information on the general conditions in the classroom were collected by a standardized form, which had been tested in a brief pre-test period. This

included the condition of the buildings, the size of the classroom, the weather and, where necessary, potentially significant influences and disruptive factors. Teachers were asked to document the number of subjects in the room and the ventilation conditions prevailing during class as well as during the breaks.

2.1.1. Sampling methods

Filter-based measurements of $PM_{2.5}$ in the classrooms were conducted with a medium volume sampler using a flowcontrolled pump working with a constant flow of $2.3\,m^3\,h^{-1}$ (Derenda, Teltow, Germany). The sample inlet was a $PM_{2.5}$ sampler, having a 50% collection efficiency for particles with a 2.5 μm aerodynamic diameter. Sample volumes were calculated from elapsed time indicators and flow measurements before sampling with calibrated rotameters. A Munktell 47 mm binder free glass fibre filter with a pore size of 2 μm was used. Filter assembly and disassembly were performed with great care so as to avoid damage, contamination or the dislodging of particles before final weighing was completed. The filters were stored and conditioned for 48 h in a room with controlled temperature and humidity before they were weighed (recommendation of DIN EN 12341) to prevent biases. Each filter was weighed four times prior to sampling and four times afterwards with a microbalance (Mettler-Toledo AX26DR, Greifensee, Switzerland) with 2 μg reading accuracy.

Continuous measurements of PM (e.g. PM_{10}, PM_4, $PM_{2.5}$) were also done using an optical laser aerosol spectrometer (LAS) (Dust monitor 1.108, Grimm Technologies, Inc., Ainring, Germany). This spectrometer works by constantly drawing the air sample via a volume controlled pump ($1.2\,l\,min^{-1}$.) through a flat beam of laser light. All scattered signals generating while the particles cross this beam are detected with a high-speed photo diode, analysed by an integrated pulse height analyser and counted. The LAS measures particle concentrations in 16 nominal size bins from about 0.1 to 2.5 μm. For our purposes the continuous measurements were stored minute-by-minute on a data logger.

A TSI model 3034 scanning mobility particle sizer (SMPS) (TSI Inc., Shoreview, MN, USA) was used to measure particle number concentrations (PNC) for a discrete size distribution of aerosols within a range of 10–487 nm. The SMPS consists of an electrostatic classifier and a CPC. With an inlet flow of $1\,l\,min^{-1}$ electrically charged particles pass from the neutralizer into the differential mobility analyser (DMA). The DMA contains two concentric metal cylinders with an electric field in-between. Here, the particles are separated depending on their electrical mobility and counted with a condensation particle counter (CPC) after passing through a butanol saturated atmosphere, which forces the particles to grow to detectable size. Number concentrations are given for 54 channels (32 channels per decade), particle surface area and volume are estimated based on the number concentration and assuming a spherical shape of particles. All estimates were standardized with the geometric width of the size channel (dlogDp). Measurements were done in 3-min intervals.

Indoor carbon dioxide was measured using a continuously monitoring infrared sensor (Testo 445, Germany). The instrument was programmed for a 1 min data logging interval. Calibration of the CO_2 sensors included linearity checks at four concentrations (0, 350, 1000, 2000 ppm) during the weeks prior to sampling. Indoor humidity and temperature were measured simultaneously with a separate sensor connected to the Testo instrument. The same equipment was used for the measurement of outdoor carbon dioxide, temperature and humidity, which was implemented in the morning and afternoon for 5 min in 15 s intervals, at approximately 1 m vertically above ground.

2.2. Statistical methods

Continuously measured particle concentrations were summarized to a daily median for each classroom. Only data collected in time periods when classrooms were occupied were used in analysis. After description of the distribution of PM measurements, the correlation of parameters of PM with indoor temperature and humidity, number of occupants, and classroom size (surface area, volume, volume per occupant), was evaluated with the spearman rank correlation coefficient. Differences in PM by floor covering (smooth vs. carpet) and class level (three categories: 1–4, 5–7, 8–11) were tested with the Wilcoxon rank sum and Kruskal–Wallis test, respectively. Associations between factors significantly correlated with PM were then estimated in bivariate linear regression, assuming a linear relationship. Differences between summer and winter measurements were calculated for classrooms measured at both occasions and the *t*-statistic

used to determine statistical significance. The intraclass correlation coefficient was derived by analysis of variance and used to determine the correlation of classroom measurements within the same school. For the comparison between gravimetrically and continuously obtained $PM_{2.5}$ measurements, the whole observation period on one day, including times of non-attendance, were considered in the analysis.

3. Results

The condition of the school buildings and the furnishings and fittings of the classrooms were extremely variable. The size of the classrooms ranged between 47–98 m^2 (median: 68 m^2) and the volume between 160–437 m^3 (median: 222 m^3). During the period of occupancy (ca. five school hours daily) the classrooms were occupied by 9–35 subjects (median: 24). The attendance (number of pupils) in winter did virtually not differ from attendance in summer. As usual for school buildings in Germany, none of them had an air conditioning system, but there was a mechanical ventilation equipment in two schools only. In 85 of the classrooms investigated in winter and in 69 of classrooms investigated in summer the floors were covered by smooth surface material (wood and linoleum). Carpeting was observed in both measurement periods in 7 rooms only.

During the winter, the median inside room temperature fluctuated between 18 and 25 °C (median: 22 °C) and the relative humidity between 22% and 60% (median: 38%) (Table 1). In the second (summer) period the temperatures ranged between 21 and 29 °C (median: 24 °C) and the relative humidity between 32% and 70% (median: 51%). The CO_2 levels in the classrooms ranged between 598 and 4172 ppm (median: 1608 ppm) in winter and between 480 and 1875 ppm (median: 785 ppm) in summer. The corresponding outside air concentrations during the measuring periods varied between 386 and 472 ppm (winter) and 341 to 485 ppm (summer).

According to the ASHRAE Standard 62-1989 indoor CO_2 levels should not exceed 1000 ppm to ensure satisfactory comfort. Values exceeding this threshold indicate insufficient fresh air and are associated with a higher frequency of health complaints (ASHRAE (American Society of Heating and Refrigerating and Air-conditioning Engineers), 1989). While during the winter period in 92% of the classrooms the CO_2 daily medians did not meet the ASHRAE Standard, the percentage of classrooms with increased CO_2 concentration fell to 28% in summer. This reduction is even more marked for CO_2 readings over 1500 ppm. In winter, in 60% of classes the daily median CO_2 concentration exceeded 1500 ppm, while in summer this threshold was reached by only 9%. In classrooms

Table 1
Distribution of classroom-specific median temperature, relative humidity and carbon dioxide in indoor and outdoor air

	N	Minimum	10th Percentile	Median	90th Percentile	Maximum	Mean
Winter indoor							
Temperature (°C)	91	18	20	22	23	25	22
Relative humidity (%)	91	22	28	38	49	60	38
CO_2 (ppm)	90	598	1009	1608	2724	4172	1759
Winter outdoor							
Temperature (°C)	40	−8	−6	−0.6	5	12	−0.3
Relative humidity (%)	40	42	46	66	81	82	65
CO_2 (ppm)	40	381	385	405	467	490	414
Summer indoor							
Temperature (°C)	75	21	22	24	27	29	25
Relative humidity (%)	75	32	41	51	62	70	51
CO_2 (ppm)	75	480	570	785	1459	1875	890
Summer outdoor							
Temperature (°C)	76	10	12	18	24	26	19
Relative humidity (%)	76	49	52	65	85	94	66
CO_2 (ppm)	38	338	355	383	448	509	391

measured at both occasions, CO_2 concentrations were on average reduced by 864 ppm (95% confidence interval: 670–1028 ppm) in summer compared to winter.

3.1. Comparison between gravimetrically and continuously measured $PM_{2.5}$

The results of PM measurements are given in Table 2. Looking at all schools, the $PM_{2.5}$ concentrations obtained with the filter-based gravimetric technique were in general higher than those obtained with the real-time monitored LAS and ranged between 4.3 and 73.1 $\mu g\,m^{-3}$ (median: 37.0 $\mu g\,m^{-3}$) in winter and between 9.8 and 55.1 $\mu g\,m^{-3}$ (median: 22.1 $\mu g\,m^{-3}$) in summer.

In 89 classrooms—40 in the winter and 49 in the summer period—$PM_{2.5}$ comparative measurements were conducted during the school days using both measurement techniques. In both observation periods, a statistically significant non-parametric correlation between results obtained by these two methods were observed ($r = 0.68$, $p < 0.001$ for winter and $r = 0.72$, $p < 0.001$ for summer). In both measurement periods LAS produced in general lower findings; only in 2% of the summer measurements and in 21% of the winter measurements readings obtained with LAS were higher than those obtained by gravimetry. Compared to the filter-based method, the LAS resulted in a reduction of the median $PM_{2.5}$ by 77% (winter) and 63% (summer). Looking at both summer and winter period combined and applying a bivariate linear regression model, the proportion of variance in gravimetrically obtained $PM_{2.5}$ values explained by LAS-obtained $PM_{2.5}$ values was 63%. After exclusion of two outliers (defined as: ratio PM gravimetrically to continuously >5) this proportion increased to 71% (see Fig. 3).

3.2. PM mass concentrations

The results of the PM measurements are given in Table 2. For all parameters relating to mass, higher concentrations were observed in winter than in summer. In winter, the results of the real-time monitored $PM_{2.5}$ and PM_{10} (using LAS) fluctuated between 2.7 and 80.8 $\mu g\,m^{-3}$ (median: 19.8 $\mu g\,m^{-3}$) and 16.3 and 313 $\mu g\,m^{-3}$ (median: 91.5 $\mu g\,m^{-3}$), respectively, during class teaching time. In summer, measurements ranged between 4.6 and 34.8 $\mu g\,m^{-3}$ (median: 12.7 $\mu g\,m^{-3}$) for $PM_{2,5}$ and 18.3 and 178 $\mu g\,m^{-3}$ (median: 64.9 $\mu g\,m^{-3}$) for PM_{10}.

As Fig. 1 shows, PM concentrations within schools were strongly correlated and this correlation was particularly high for small PM fractions. For example, the intraclass correlation of classrooms within the same school with respect to PM_1 was 0.90. Thus, for PM_1 90% of the variability between PM measurements were caused by differences between schools and only 10% by differences between classrooms within schools. In winter, intraclass correlations were even slightly higher than in summer (0.93 for PM_1, 0.88 for $PM_{2.5}$, 0.71 for PM_4 and 0.64 for PM_{10}).

In 58 classrooms data on PM were collected in both winter and summer. Among these classrooms, a reduction in PM_{10} and $PM_{2.5}$ in the summer

Table 2
Distribution of classroom-specific particulate matter concentrations in indoor air of schools using daily medians

	N	Minimum	10th Percentile	Median	90th Percentile	Maximum	Mean
Winter							
$PM_{2.5}$ (gravimetrically)	42	4.3	21.5	36.7	62.9	73.1	38.9
$PM_{2.5}$ (LAS)[a]	79	2.7	8.8	19.8	44.9	80.8	23.0
PM_4 (LAS)[a]	79	12.3	27.2	63.4	120.0	243.8	71.9
PM_{10} (LAS)[a]	79	16.3	43.2	91.5	168.8	313.2	105.0
Summer							
$PM_{2.5}$ (gravimetrically)	49	9.8	14.6	20.2	30.9	55.1	22.1
$PM_{2.5}$ (LAS)[a]	74	4.6	34.8	12.7	20.9	34.8	13.5
PM_4 (LAS)[a]	74	15.5	20.0	42.9	76.6	121.5	44.8
PM_{10} (LAS)[a]	74	18.3	31.8	64.9	124.1	178.4	71.7
Particle number concentration ($N\,cm^{-3}$)[a]	36	2,622	3,873	5,660	10,566	12,145	6,509

[a]Computed using median daily values; LAS: laser aerosol spectrometer (real-time monitoring).

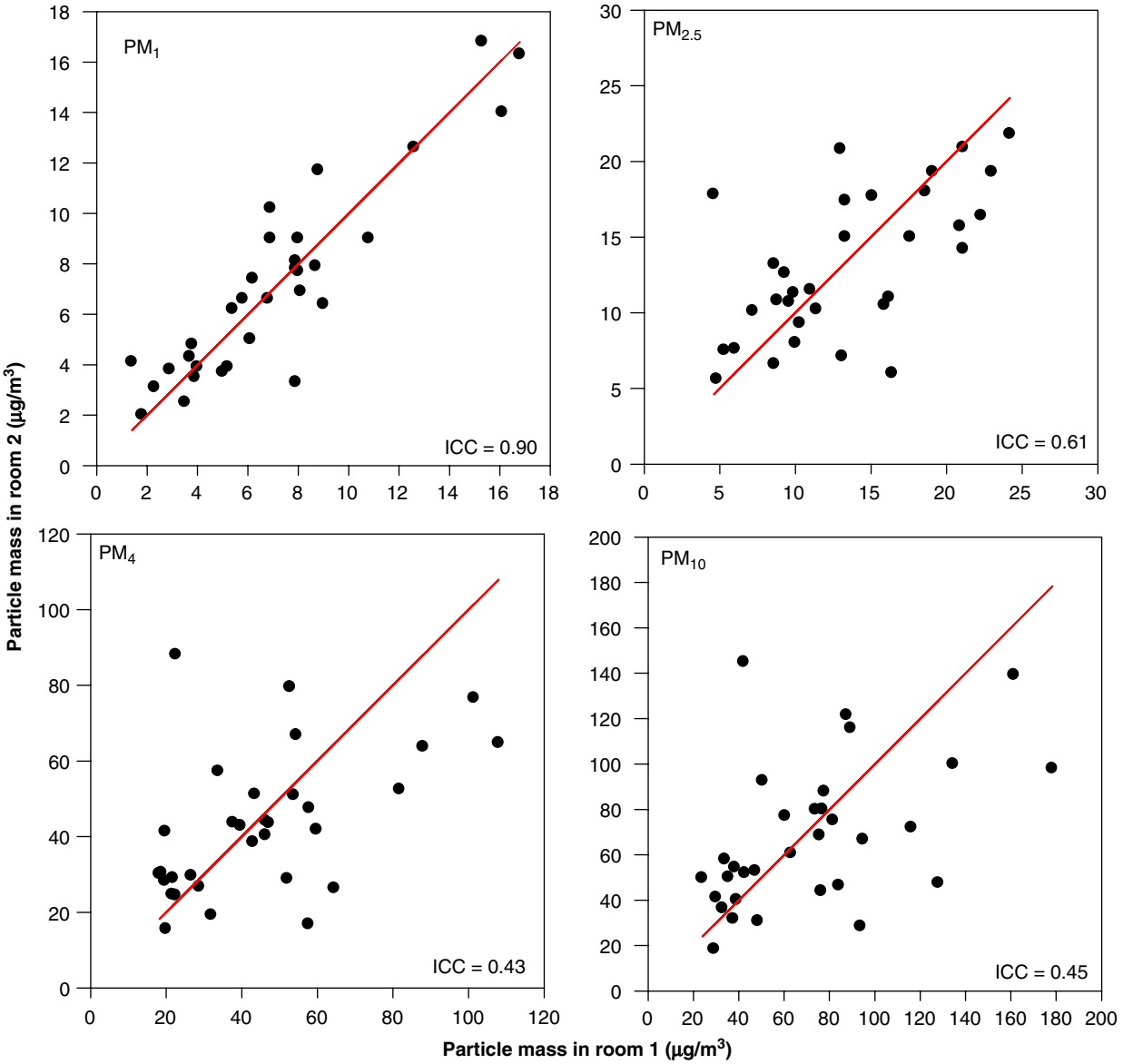

Fig. 1. Particle mass concentration in classrooms belonging to the same school, summer.

period was observed in 74% and 78% cases, respectively. The median reduction potentials of all classrooms were 36% (PM_{10}) and 35% ($PM_{2.5}$). On average, PM concentrations were decreased by $10.4\,\mu g\,m^{-3}$ ($PM_{2.5}$) and $31.8\,\mu g\,m^{-3}$ (PM_{10}) in summer compared to winter. These differences were highly statistical significant.

A significant negative correlation between humidity and real-time monitored $PM_{2.5}$ concentration was observed in winter, while a significant positive correlation between these two factors was found in summer and no correlation between humidity and PM_{10} and gravimetrically measured $PM_{2.5}$ was observed (Table 3). A significant positive correlation between temperature and gravimetrically measured $PM_{2.5}$ was observed in summer only. In winter, but not in summer, PM_{10} and gravimetrically measured $PM_{2.5}$ are significantly correlated with parameters of room size and number of occupants. An increase in volume per subject by $1\,m^3$, for example, was associated with a decrease in median PM_{10} concentration by $6.3\,\mu g\,m^{-3}$ and in

Table 3
Correlation between room characteristics and PM concentrations in classrooms in the winter and summer measurement period

	Winter period			Summer period		
	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
	LAS	LAS	Gravimetry	LAS	LAS	Gravimetry
Correlation (p-value)						
Humidity (%)	−0.32 (0.004)*	−0.09 (0.42)	−0.14 (0.38)	0.24 (0.04)*	−0.06 (0.63)	−0.04 (0.79)
Indoor Temperature (°C)	0.05 (0.67)	0.12 (0.30)	0.03 (0.85)	0.19 (0.10)	−0.02 (0.89)	0.34 (0.02)*
Number of occupants	0.13 (0.24)	0.37 (0.0007)*	0.20 (0.21)	0.11 (0.34)	0.20 (0.09)	−0.02 (0.90)
Floor surface (m^2)	−0.08 (0.46)	−0.30 (0.007)*	−0.44 (0.003)*	0.01 (0.94)	−0.08 (0.48)	0.17 (0.25)
Room volume (m^3)	−0.16 (0.17)	−0.25 (0.02)*	−0.38 (0.01)*	0.14 (0.22)	0.15 (0.22)	0.20 (0.16)
Volume/occupant (m^3)	−0.19 (0.09)	−0.41 (0.0002)*	−0.39 (0.01)*	0.02 (0.84)	−0.02 (0.87)	0.12 (0.41)
CO_2 indoor concentration	0.12 (0.28)	0.27 (0.02)*	0.28 (0.08)	0.25 (0.03)*	0.57 (<0.0001)*	0.14 (0.35)
χ^2 *(p-value)*						
Floor covering (carpet vs. smooth)	1.44 (0.23)	1.32 (0.25)	0.24 (0.62)	0.01 (0.96)	0.30 (0.59)	2.10 (0.15)
Class level (3 categories)	21.80 (<0.0001)*	34.06 (<0.0001)*	11.14 (0.004)*	22.7 (<0.0001)*	25.1 (<0.0001)*	5.33 (0.07)

*p-value < 0.05.

gravimetrically measured $PM_{2.5}$ concentration by 2.0 μg m^{-3} (Table 4). No differences were found between the PM content of air in classrooms with smooth—wood or linoleum—floors and rooms with carpeted floor. It should, nevertheless, be borne in mind here that only seven of the classrooms included in the investigation had a carpeted floor, which results in low power for the detection of significant correlations.

However, a statistically significant influence of class level on the PM content was apparent in both measuring periods. In primary schools (class level 1–4) higher PM values were found than in classrooms of secondary schools. The median PM_{10} concentrations in primary schools ranged from 67 to 313 μg m^{-3} (median: 139 μg m^{-3}) in winter and from 48 to 178 μg m^{-3} (median: 89 μg m^{-3}) in summer. In contrast, in the classes level 8–11 PM_{10} values were reduced from 27 to 124 μg m^{-3} (median: 72 μg m^{-3}) (winter) and to 18 to 93 μg m^{-3} (median: 43 μg m^{-3}) (summer). Apart from differences in classroom fittings, flooring etc., the more intense physical activity of the younger children could be an explanation of this correlation.

There were two classrooms included with mechanical airing systems. In these classrooms (grade 7 and 8), in winter CO_2 concentrations were 701 and 598 ppm, LAS measured $PM_{2.5}$ concentrations 2.4 and 4.9μg m^{-3}, LAS measured PM_{10} concentrations 27.4 and 22.8 μg m^{-3}. In summer, only one of these classrooms was included and the respective results were 628 ppm CO_2, 6.0 μg m^{-3} $PM_{2.5}$ and 28.4 μg m^{-3} PM_{10}.

3.3. PM number concentration, particle surface and volume

PNC measured with the SMPS in 36 classrooms in summer are summarized in Table 2. The median number concentrations in the classrooms ranged from 2622 to 12,145 particles cm^{-3} (Median: 5660 cm^{-3}). As an example, Fig. 2 shows the PNC by particle size in seven classrooms during one school day (for each channel, the approximately 80 individual measurements were summarized to a median).

4. Discussion

In this study, for the first time, results on various particle related parameters in indoor air are presented for a large number of schools. Our results clearly show that exposure to particulate matter in school is high. The wide range of PM concentrations indicate the large potential for reduction and the need for identification of factors responsible for this variability. We observed a strong seasonal variability, with air quality being particularly poor in winter. Further parameters correlated with increased concentrations of PM were small room size, high number of occupants, high CO_2 concentrations and low class level. The findings may help

Table 4
Determinants of PM concentrations, results of the bivariate linear regression

	Winter period		Summer period	
	β[a] (95% CI)[b]	R^{2c}	β (95% CI)	R^2
$PM_{2.5}$, LAS				
Humidity (10 %)	−6.41 (−10.66, −2.16)	0.1049	1.67 (0.17, 3.17)	0.0639
CO_2 indoor concentration (100 ppm)	—	—	0.49 (0.12, 0.85)	0.0898
Class level		0.2750		0.2893
1–4	Reference		Reference	
5–7	−13.45 (−20.40, −6.50)		−2.79 (−5.63, 0.06)	
8–11	−18.22 (−25.24, −11.20)		−7.34 (−10.14, −4.54)	
PM_{10}, LAS				
Number of occupants	4.41 (1.93, 6.89)	0.1401	—	—
Floor surface (10 m^2)	−14.14 (−27.55, −0.74)	0.0542	—	—
Room volume (100 m^3)	−21.08 (−46.46, 4.29)	0.0343	—	—
Volume/occupant (m^3)	−6.29 (−9.41, −3.17)	0.1731	—	—
CO_2 indoor concentration (100 ppm)	2.30 (0.46, 4.14)	0.0754	5.76 (3.67, 7.86)	0.2945
Class level		0.3936		0.3108
1–4	Reference		Reference	
5–7	−55.69 (−78.64, −32.73)		−24.53 (−42.79; −6.27)	
8–11	−79.56 (−102.73, −56.39)		−50.65 (−68.63, −32.68)	
$PM_{2.5}$, gravimetry				
Indoor temperature (°C)	—	—	0.53 (−0.82, 1.88)	0.0135
Floor surface (10 m^2)	−7.02 (−12.39, −1.66)	0.1490	—	—
Room volume (100 m^3)	−7.96 (−18.16, 2.24)	0.0586	—	—
Volume/occupant (m^3)	−2.04 (−3.89, −0.19)	0.1106	—	—
Class level		0.2555	—	—
1–4	Reference			
5–7	−5.83 (−16.63, 4.96)			
8–11	−20.14 (−31.29, −9.00)			

[a]Regression coefficient from linear regression, change in PM per change in unit determinant.
[b]Confidence interval.
[c]Percent variance explained.

in explaining the mechanisms how high PM concentrations develop and further which strategies may be effective to improve air quality in classrooms.

Our results are in line and extend findings of previous studies on PM levels in indoor air of public buildings and factors correlated with poor indoor air quality. Only limited data on indoor PM concentration in public buildings are at present available. In Germany, investigations have been undertaken in Berlin and Baden-Wuerttemberg. Results reported from Berlin on PM_4 concentration are of similar magnitude than our findings. In the 73 nursery schools which were investigated from November 2000 to March 2001, a median PM_4 concentration of 52.6 $\mu g\,m^{-3}$ (range: 13–128.4 $\mu g\,m^{-3}$) was detected (Fromme et al., 2005) and similarly in an investigation of 33 classrooms a median PM_4 concentrations of 60 $\mu g\,m^{-3}$ (range: 24–106 $\mu g\,m^{-3}$) was found.

In the Baden-Wuerttemberg study, in which 54 classrooms were measured in winter and spring, low concentrations of $PM_{2.5}$ were reported (median $PM_{2.5} = 15\,\mu g\,m^{-3}$, range: 5–40 $\mu g\,m^{-3}$) (Link et al., 2004). However, these classrooms were measured for one entire week and thus data obtained during long time periods when no subjects were in the rooms—i.e. at night—were included in analysis. If analysis would be restricted to time periods, when pupils attended class, one would expect increased median PM levels.

A number of mostly small investigations on PM concentrations, which included only very few classrooms, have been conducted in the Netherlands, the US and China (Roorda-Knape et al., 1998; Janssen et al., 1997, 1999, 2001; Keeler et al., 2002; Scheff

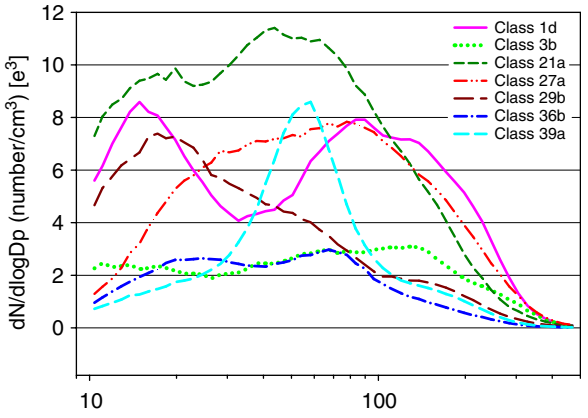

Fig. 2. Concentration of particle number by particle size in seven classrooms.

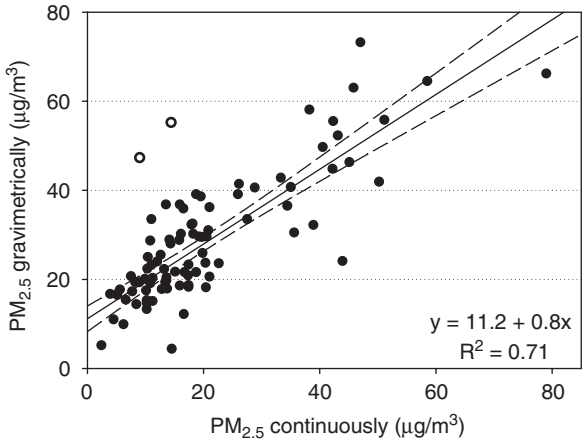

Fig. 3. Correlation between $PM_{2.5}$ measured continuously and gravimetrically (lines represent regression line and 95% confidence interval; $N = 87$ pairs and two outliers marked; R^2: 0.63 with outliers).

et al., 2000; Ligman et al., 1999; Shaughnessy et al., 2002; Turner et al., 2002; Lee and Chang, 2000). Due to the high variability between classrooms these studies are only of limited value for the estimation of typical exposure levels. Only in two studies more than ten classrooms were examined and here mean PM_{10} levels of 51 to 166 $\mu g\,m^{-3}$ (Roorda-Knape et al., 1998) and a mean $PM_{2.5}$ concentration of 23.0 $\mu g\,m^{-3}$ (Janssen et al., 2001) were observed, which agree with the concentrations we found. Relatively small amounts of PM_{10} with mean summer and winter indoor concentrations of 46.7 and 39.1 $\mu g\,m^{-3}$, respectively, were found in 40 classrooms in Korea (Son et al., 2006). The values described were near by the outdoor concentrations.

When comparing PM concentrations in different studies, the measurement method should be taken into account, since gravimetric assessment generates higher results than LAS-assessment. However, while absolute values differ strongly, the rank order of values is comparable with both methods, as indicated by the high correlation in our study (Fig. 3). Thus, categorization of classrooms in those with high and those with low PM levels should lead to comparable results with both methods.

One reason for the difference in PM concentration measured with LAS technique and gravimetry may be, that, while PM is weighted directly in gravimetry, in LAS technique PM is estimated only based on the number or particles counted and additionally based on assumptions of the shape of the particles and their density. These assumptions may not hold true and thus may cause an underestimation of PM, especially with respect to particles with a more complex shape. Furthermore, to minimize potential sampling artefacts we aimed for maximal standardization of measurement procedures and conducted a clear quality management.

Indoor sources such as tobacco smoke and other factors such as heating can in general contribute to indoor PM levels, but are unlikely to be of relevance in the classrooms we examined. For example, since smoking usually is not allowed in public school buildings and in our study there was no direct connection (e.g. a ventilation system) between teachers rooms (where smoking may be allowed) and the classrooms examined, tobacco smoke can be ruled out as an indoor source of PM. This assumption was confirmed by the simultaneously measured VOC in the classrooms. Heating or for example candle burning can also be excluded as a significant indoor source because all schools use a central heating system and we did not register any other burning inside the classrooms.

The influence of season on PM concentrations we observed has been reported before from the US (Keeler et al., 2002). This difference is most likely due to the different ventilation practice in summer and winter. Due to increased ventilation in spring, summer and autumn indoor PM levels may strongly depend on outdoor levels, while in winter the classroom PM may be more strongly influenced by indoor activities.

Floor coverings in schools have been continually controversially discussed in Germany. They can serve as a reservoir for dirt and microorganisms and be a potential source of allergens and therefore a

health risk. Whilst in our study no significant differences between PM values in classrooms with carpets and those with hard surface floorings were reported, Shaughnessy et al. (2002) observed significantly higher $PM_{2.5}$ levels in carpeted classrooms ($17\,\mu g\,m^{-3}$) than in those with hard surface floors ($10\,\mu g\,m^{-3}$). Because data in the aforementioned study were only available for five rooms, confounding by other covariables cannot be excluded. Also, in a further study, no marked differences in PM_{10} levels were observed between two school gym rooms, one carpeted and one non-carpeted (Turner et al., 2002). The effect of cleaning routine on indoor PM concentrations has not been evaluated in previous studies but might be important.

Our results suggest two potential mechanisms for the development of increased PM concentrations in schools. On the one hand, the continually high CO_2 values we observed particularly in winter indicate an insufficient ventilation routine in schools. This lack of ventilation may inhibit the transport and removal of, especially, larger particles from room interiors to the outdoor. On the other hand, classrooms today are less frequently and less thoroughly cleaned (a mere twice weekly wipe over the flooring, perhaps) so that the sedimented dust particles are only partly removed from indoor spaces. This in turn leads, in combination with a large number of pupils in relation to room area and volume, to a continued resuspension of particles from the room surface. The impact of ventilation is emphasized by the fact, that in the two classrooms with a mechanical ventilation system the air quality appears to be improved.

There is yet another factor to be mentioned: the physical activity of the pupils. This influence was confirmed by the increased particle concentrations during the school day when pupils are in the room compared to sampling times when no pupils are present (data not shown). The increased concentrations of particulate matter in classrooms with younger children, who develop much more indoor activity than older ones, support this theory.

We did not present results on the difference in PM by location of school, since location was strongly correlated with school type. Secondary schools were found mainly in suburbs and city areas, while the schools recruited in the rural areas were mainly primary schools. Since class level is a strong predictor of PM concentration (see Table 3), the presentation of results by location could lead to a somewhat misleading impression.

The examination of two classrooms per school enabled us to partition the total variance in variance explained by school and variance explained by classroom within the school. We could show that particularly for small particles the concentration depends strongly on factors related to the school, as indicated by the high intraclass correlation coefficient. Predominantly school specific are factors such as the school building itself, the outdoor concentrations, the furniture, cleaning procedures and room size. Classroom-specific differences such as the difference in actual number of pupils and their activity as well as differences in airing behaviour appear to be important for the concentration of larger particles, but less important for the concentration of particles of small size.

We were not able to collect outdoor PM concentrations parallel to indoor levels. However, PM_{10} outdoor levels in Munich are available from air pollution measurement sites at roads with both low and high traffic loads. At these measurement sites in winter, a mean PM_{10} concentration of $38\,\mu g\,m^{-3}$ and in summer a mean PM_{10} concentration of $26\,\mu g\,m^{-3}$ was measured. These outdoor estimates are much lower than the concentrations we did observe in schools.

In the Netherlands one study including 12 school sites near motorways, the mean PM_{10} levels during school time varied strongly, not only between schools but also within the same school buildings (Roorda-Knape et al., 1998). The authors concluded that the indoor concentrations were neither correlated with the proximity of the schools to motorways nor with traffic intensity. In a similar study, in the years 1994 and 1995 Janssen et al. (1997) found higher PM_{10} values in the indoor air of four schools in Wageningen and Amsterdam than in the corresponding outdoor air, with 8 h averaged findings ranging from 81 to $157\,\mu g\,m^{-3}$. The same study group reported a mean $PM_{2.5}$ of $19.9\,\mu g\,m^{-3}$ for one classroom in the city of Wageningen (range: 14.1 to $35.2\,\mu g\,m^{-3}$), and a high correlation between indoor and ambient fine particulate matter (Janssen et al., 1999). In 1997/1998 Janssen et al. (2001) conducted a more extensive Dutch study, including 24 schools and a sampling period from April 1997 to May 1998. They found indoor $PM_{2.5}$ values between 7.7 and $52.8\,\mu g\,m^{-3}$ (mean: $23.0\,\mu g\,m^{-3}$) and increased $PM_{2.5}$ concentrations associated with increased lorry traffic density and decreased distance to motorways.

Measurements of PNC in schools have so far been described in one study only. Compared to our

study, in which PNCs of 2622 to 12,145 particles cm^{-3} were found in 36 classrooms in spring/summer, the PNC ranged between 2390 and 75,500 particles cm^{-3} (city) and 1720 and 49,100 particles cm^{-3} (rural area) (Link et al., 2004). The median PNCs were 8310 (city) and 4070 particles cm^{-3} (rural area), whilst in our study a median of 5660 particles cm^{-3} was observed.

Other studies on number concentrations of particles in schools are available, but did not report concentration of fine particles $< 1\,\mu m$ (Kinshella et al., 2001; Blondeau et al., 2005; Poupard et al., 2005). Also, no systematic studies on PNC in German dwellings and only few on PNC in outdoor air in Germany exist. Measurements in Erfurt, for instance, have shown that the daily number of ultra fine particles (10–100 nm) recorded at a city monitoring site reaches maxima of 21,000 particles cm^{-3} in the daytime and 5000 particles cm^{-3} at night and is higher in winter than in summer (Kreyling et al., 2003; Pitz et al., 2003). In Australia, concentrations of submicron particles (7–808 nm) in apartments were on average 18,200 particles cm^{-3} (during indoor activities) and 12,400 particles cm^{-3} (no indoor activities) (Morawska et al., 2003).

However, the number of fine and ultra fine particles measured in classrooms was in the same range or lower as the results from residences or outdoor monitoring sites and show little variation during teaching hours. This is probably due to the absence of typical indoor sources of ultra fine particles, such as cooking and cigarette smoke, which could temporarily produce high concentrations.

A major strength of our investigation was the restriction of measurements to actual teaching hours. In studies including time periods, in which no pupils are in class, PM concentrations might be underestimated. Thus, for example, in an investigation in the US, after the modification of the sampling procedure from 24 to 8-h measurements the mean PM_{10} values were twice as high as before (Yip et al., 2004). Our examination, which covered teaching hours only, will thus give a more realistic estimate of the PM concentrations pupils are exposed to at school.

While we measured the quantity of particulate mass in indoor air, we did not evaluate the quality, i.e. the chemical composition of particulate matter. Chemical composition of indoor PM may differ from that of outdoor PM and thus may cause different effects. There are quite a few studies dealing with the chemical composition of PM (for example elemental carbon, sulfate content) from residential indoor locations, and these chemical contents can be used to trace their various sources (ambient, personal and indoor). Data on toxicity, however are at present available only for pure substances and outside air particles. One in vitro study in Boston has examined 14 paired indoor and outdoor samples ($PM_{2.5}$) more closely for proinflammatory response, with special reference to rat alveolar macrophages (Long et al., 2001). Even after adjustment for the endotoxin concentration of each sample, the indoor-generated particles revealed a greater inflammatory effect (production of tumour necrosis factor) than the samples of ambient origin. In another study, Monn and Becker (1999) determined the cytotoxicity and proinflammatory activity of water soluble components of fine and coarse particles in blood monocytes. Toxicity and cytokine production were induced by outdoor coarse particles but not by outdoor $PM_{2.5}$ and particles collected indoors. But, according to the authors, a possible role of gram-negative bacteria and/or endotoxins could not be excluded. The mutagenic activity (examined by Salmonella microsuspension assay), however, appears to be similar for indoor and outdoor dust samples (Kado et al., 1994).

The importance to differentiate between ambient and nonambient components of total personal particle exposure in epidemiologic studies has been discussed by Ebelt et al. (2005). However, while the authors gave a summary estimate for the health effect of ambient exposure (particles from outdoor origin) they did not present a corresponding estimate for exposure to nonambient particles of indoor origin.

To develop a valid risk assessment and effective risk management strategies, the following questions have to be the basis of further research: (I) is the chemical composition of particulate matter inside school buildings different from that in outside air? (II) How large is the toxic potential of these indoor respirable particles, i.e., compared to typical outside PM? (III) What sources and mechanisms are responsible for the high PM concentration and how can the exposure of school children be minimized?

Furthermore, strategies to minimize the exposure must be developed and tested for effectiveness in intervention studies to eventually be able to reduce health risks of children.

5. Conclusion

Our results as well as findings from earlier studies clearly show that exposure to particulate matter in school is high. In our explorative analysis we were able to identify parameters correlated with increased concentrations of PM such as high CO_2 concentrations and low class level. Only in winter additionally small room size and high number of occupants were associated with increased PM. Further research is needed to confirm these findings and identify additional determinants of PM concentration, and evaluate how these findings can be translated into preventive action.

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