

Mass concentration and elemental composition of PM₁₀ in classrooms

Nicole A H Janssen, Gerard Hoek, Bert Brunekreef, Hendrik Harssema

Abstract

Objectives—To investigate the sources of high concentrations of particles of <10 µm diameter (PM₁₀) in classrooms, observed in a previous study on childhood exposure to PM₁₀, and to study the correlation between classroom and outdoor concentrations of mass and elements of PM₁₀.

Methods—Measurements of PM₁₀ were conducted in two schools and outdoors in Amsterdam, the Netherlands. Averaging time was 24 hours for the outdoor measurements and both 8 hours (school time) and 24 hours for the classroom measurements. Analysis by x ray fluorescence was used to measure the elemental composition of 55 samples from the 11 days when measurements were conducted simultaneously in both classrooms and outdoors.

Results—For most elements, classroom concentrations were considerably higher than outdoor concentrations, especially during school hours. The highest classroom/outdoor ratios were found for the elements from soils Si, Ca, and Ti. The only measured elements that were not increased were S, Br, Pb, and Cl, which are dominated by non-crustal sources. For S, Br, and Pb, which are generally associated with particles <1 µm, significant correlations between classroom and outdoor concentrations and between the two classrooms were found. The other elements generally had low correlations.

Conclusions—The results show that the high PM₁₀ concentrations found in our classrooms are probably due to resuspension of coarse particles or suspension of soil material. Due to these excess coarse particles, the correlation between classroom and outdoor concentrations is lower for elements associated with coarse particles than for elements associated with fine particles. As the general composition of PM₁₀ in classrooms differs from the composition of PM₁₀ in ambient air, the high PM₁₀ mass concentrations in classrooms can probably not be directly compared with ambient air quality guidelines.

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Keywords: particles; PM₁₀; elemental composition; classrooms; children

Several studies describe high personal exposures to particles <10 µm diameter (PM₁₀) compared with outdoor concentrations.¹⁻⁴ In a study on childhood exposure to PM₁₀ in the Netherlands, personal exposures were on average three times

higher than outdoor concentrations.³ An important part of this difference could be explained by high PM₁₀ concentrations in classrooms, which occurred especially during school time. In the three schools where PM₁₀ measurements were conducted, concentrations during school hours were two to five times higher than outdoor concentrations and about twice as high as 24 hour average classroom concentrations. Measurements of PM₁₀ conducted in 11 other primary schools in the Netherlands also showed highly increased classroom concentrations during school hours.⁵

In the particle total exposure assessment methodology (PTEAM) study,^{1,2} daytime personal PM₁₀ concentrations were about 60% higher than outdoor and indoor concentrations. During night time—when people were sleeping for about 2/3 of the time—personal PM₁₀ concentrations were more similar to indoor and outdoor concentrations. Resuspension of household dust caused by human activity was suggested as one of the causes of the increased daytime personal exposures. Thatcher and Layton⁶ studied the effect of resuspension by measuring different ranges of particle size before and after several resuspension activities. Normal activity by four people or continuous walking and sitting by one person resulted in a twofold to fourfold increase of particles in the 5-10 µm size range. Resuspension of particles, caused by the presence and activity of about 30 children in the relatively small volume of a classroom, can therefore be considered a likely cause of the high PM₁₀ concentrations found in our classrooms. Furthermore, several studies suggest that resuspension of particles occurs predominantly in the coarse fraction of PM₁₀.^{2,6,7} Consistent with this, measurements of PM_{2.5} in a classroom did not show significant differences between classroom and outdoor concentrations.⁸

To further investigate the increased PM₁₀ concentrations found in classrooms, we measured the elemental composition of PM₁₀ samples collected simultaneously in two classrooms and outdoors. Furthermore, the correlations between ambient and classroom concentrations of PM₁₀ mass and elements were evaluated. Elemental composition of particles can be used to interpret what sources are important. Also, specific elements can be associated with fine or coarse particles.

Methods

STUDY DESIGN

Repeated measurements of PM₁₀ were conducted in two primary schools in Amsterdam.³ In these classrooms, 24 hour average and 8 hour averages during school time were

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Table 1 Detection limits (DLs), % above DL, and % above uncertainty limits (UL) for elements measurable on at least 50% of outdoor and classroom filters

	Mean field blank (ng/m ³)		DL (ng/m ³)		% > DL	% > UL
	School hours*	24 h†	School hours*	24 h†		
S	3.8	1.3	36.3	12.1	100	100
Pb	-2.4	-0.8	3.6	1.2	100	100
Br	-4.6	-1.5	3.7	1.2	98	87
Cl	-28.1	-9.4	15.8	5.3	100	100
Cr	-0.4	-0.1	3.0	1.0	96	89
Zn	2.4	0.8	4.1	1.4	100	100
Cu	-0.5	-0.2	5.7	1.9	100	100
Mn	-3.0	-1.0	5.0	1.7	100	98
K	42.3	14.1	102.9	34.3	100	100
Fe	9.3	3.1	20.6	6.9	100	100
Si	-92.6	-30.9	26.0	8.7	100	98
Ca	7.9	2.6	16.0	5.3	100	100
Ti	1.4	0.5	8.1	2.7	100	89
Sr	1.2	0.4	2.9	1.0	100	98

*Based on a sampled volume of 4.8 m³.

†Based on a sampled volume of 14.4 m³.

Table 2 Median mass and elemental concentrations of classroom and outdoor PM₁₀ (µg/m³ for mass, ng/m³ for elements; n=11)

	School 1			School 2			
	School hours	24 h	Non-school hours*	School hours	24 h	Non-school hours*	Outdoor 24 h
Mass	164.2	73.3	34.7	77.8	44.2	28.5	28.5
S	1173	756	482	878	769	662	862
Pb	43	31	18	71	61	49	26
Br	14	7	6	13	8	6	11
Cl	1759	1571	1537	2893	1553	1288	1499
Cr	19	8	2	17	9	6	2
Zn	165	77	46	142	90	61	24
Cu	34	22	17	19	13	10	6
Mn	66	27	14	16	9	6	8
K	1439	648	338	536	319	189	140
Fe	2135	934	500	614	442	341	209
Si	10264	4224	1853	3992	1978	1060	194
Ca	5919	2540	1075	3322	1919	1222	232
Ti	524	271	152	235	153	114	10
Sr	29	12	5	15	7	4	2

*Estimated classroom concentration during non-school hours; estimated = $(C_{24h} \times t_{24h} - C_{\text{school hours}} \times t_{\text{school hours}}) / (t_{24h} - t_{\text{school hours}})$

measured. Outdoor measurements were only 24 hour averages. Analysis was by x ray fluorescence on filters from all days that measurements were conducted in both schools simultaneously, and outdoor values were similarly measured. This involved 55 samples from 11 days, collected between 23 January and 7 March 1995.

SAMPLING SITES

The study was conducted in Amsterdam, which has about 720 000 inhabitants and is situated about 25 km east of the North Sea. In the inner city of Amsterdam, air pollution levels are influenced primarily by emissions from motorised traffic and long distance transport. The industrial area of Amsterdam is relatively small and is in the west and north of Amsterdam.

Classroom measurements were conducted in two schools in the inner city of Amsterdam. School 1 was built in 1954 and school 2 in 1926. In school 2, the stairwell had been repainted just before the start of the study. Distance between the schools was 1 km. School 1 was situated about 150 m south of a road with a traffic intensity of 17 000 vehicles a day and school 2 about 75 m east of a road with a traffic intensity of 14 000 vehicles a day. In each school, measurements were conducted in the

classroom of children who participated in the study on childhood exposure to PM₁₀.³ Both classrooms had uncarpeted floors and were situated on the first floor. Measurements were conducted at 1.5 m height, and away from the door and the blackboard to avoid disturbances by air currents and dust sources.

Outdoor measurements were conducted in a park in the city centre, about 150 m away from the nearest road and away from other local sources of particles. The outdoor monitoring site was 3 km from school 1 and 2 km from school 2. Given the small spatial variation of PM₁₀ concentrations within The Netherlands,^{9 10} PM₁₀ concentrations measured at the outdoor monitoring site were considered to be representative of PM₁₀ concentrations throughout the city.

SAMPLING METHODS

Measurements of PM₁₀ were conducted with a Harvard Impactor (ADE, Harrison, Maine, USA)¹¹ operating at 10 l/min, with Anderson 37 mm 2 µm pore size Teflon filters. For the classroom measurements, two averaging times were used: 24 hour measurements at the same time as the outdoor measurements (1500–1500) and 8 hour measurements during school time (0800–1600). Details about the sampling and weighing procedures and detection limits have been published elsewhere.³ All measurements were above the detection limit.

ANALYSIS OF THE ELEMENTAL COMPOSITION

The elemental composition of the samples was analysed by energy-dispersive x ray fluorescence at the United States Environmental Protection Agency in Research Triangle Park (North Carolina, USA). For each element the uncertainty per sample was calculated based on several factors including the concentration of the element in the sample and the propagated uncertainty calculated for sampling and analysis variables, such as the calibration uncertainty and the system stability.¹² This uncertainty means that there is a 68% chance that the true concentration lies within the reported concentration $\pm 1 \times$ the uncertainty. The uncertainty limit was calculated as three times the uncertainty. Uncertainty limits thus changed from sample to sample for each element. Only elements with concentrations higher than the uncertainty limit on at least 50% of both outdoor and indoor filters were included in the data analysis.

As well as uncertainty limits, detection limits, defined as three times the SD of field blanks divided by the nominal sample volume, were calculated. Three field blanks were analysed. Mean field blank values were subtracted from all sample values.

Results

Fourteen elements were measurable on at least 50% of both the outdoor and indoor filters. Mean field blank concentrations, detection limits, and percentage above the detection and uncertainty limits of those 14 elements are presented in table 1. Elements are grouped to their main ambient sources: S, Pb, and Br (anthropogenic), Cl (marine), Cr, Zn, Cu, Mn, K, Fe

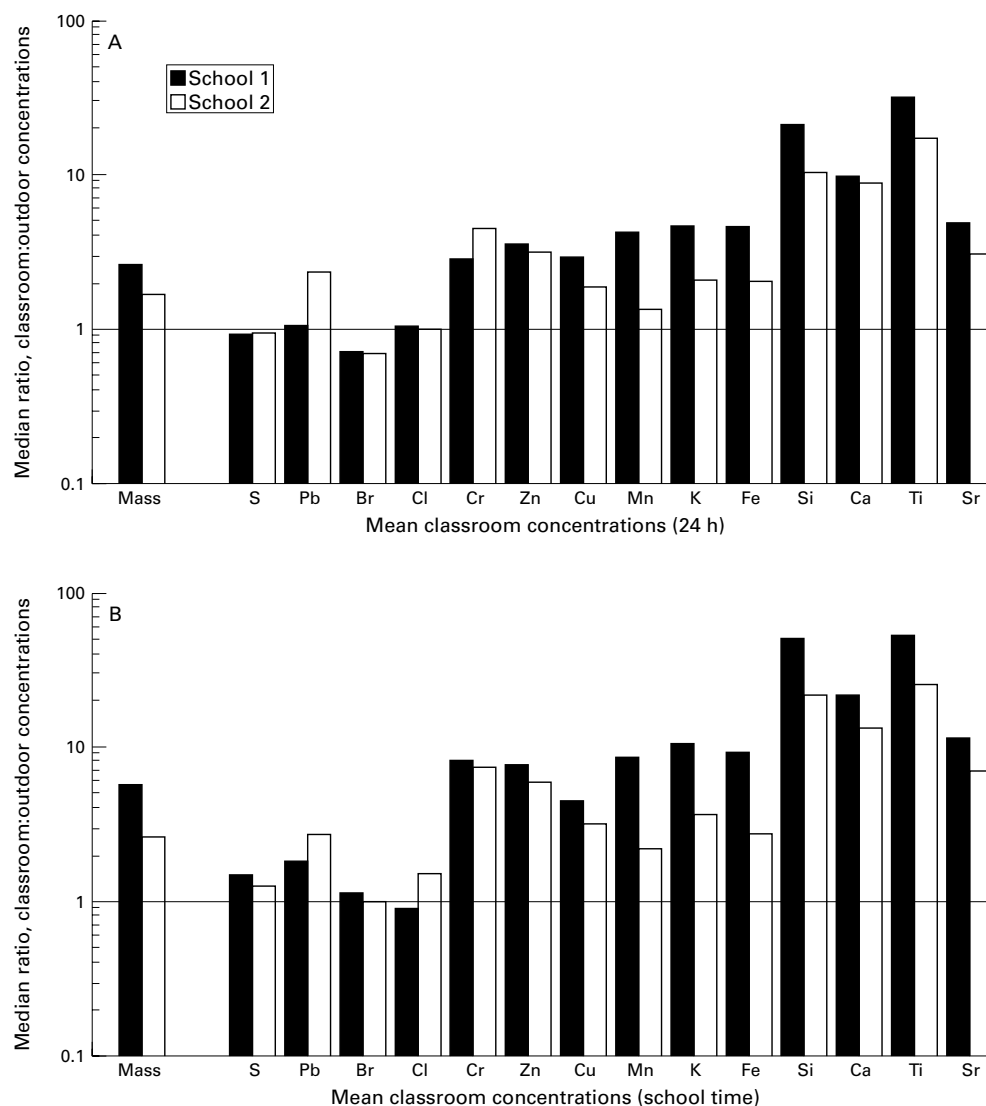


Figure 1 Median ratio between classroom and outdoor concentrations for (A) 24 hour mean and (B) school time classroom concentrations.

(both crustal and anthropogenic) and Si, Ca, Ti, Sr (crustal).¹³⁻¹⁶ Values that were above the uncertainty limit were always also higher than the detection limit. Mean field blank values can differ from zero, both negatively and positively,

due to subtle differences between the batch of filters used to set the background and the batch of filters used for sampling. For most elements, correcting for the mean field blank values did not result in a substantial change in the concentrations. For Br, however, the correction resulted in a >60% increase in the 8 hour average concentrations and about a 20% increase in the 24 hour average concentrations.

Mass and elemental concentrations are presented in table 2. Ratios between classroom and outdoor concentrations are shown in figure 1. Medians are presented because concentrations and ratios were not normally or log normally distributed for all elements. For most elements, classroom concentrations were considerably higher than outdoor concentrations. This was especially the case during school time. The highest classroom/outdoor ratios were found for the soil elements Si, Ca, and Ti, with median classroom/outdoor ratios up to 50 for school time classroom Si and Ti concentrations in school 1. Only for S, Br, and Cl, 24 hour average classroom concentrations were lower than or comparable with outdoor concentrations. For

Table 3 Spearman correlation between classroom concentrations and 24 h mean outdoor concentrations (n=11)

	24 h Mean classroom concentrations			8 h Mean classroom concentrations during school hours		
	Outdoor-school 1	Outdoor-school 2	School 1-school 2	Outdoor-school 1	Outdoor-school 2	School 1-school 2
Mass	0.63*	0.43	-0.02	0.36	0.32	-0.05
S	0.84**	0.95**	0.76**	0.58†	0.54†	0.64*
Pb	0.95**	0.52	0.55†	0.66*	0.19	0.30
Br	0.75**	0.85**	0.74**	0.23	0.53†	0.67*
Cl	0.46	0.95**	0.39	0.27	0.64*	0.64*
Cr	0.45	-0.31	-0.15	0.39	0.00	-0.29
Zn	0.81**	0.34	0.12	0.52	0.32	0.43
Cu	-0.02	0.58†	0.35	0.40	0.05	0.04
Mn	0.63*	0.61*	0.45	0.56†	0.42	0.70*
K	0.72*	0.14	0.12	0.65*	-0.01	0.08
Fe	0.63*	0.51	0.38	0.50	0.07	0.50
Si	0.63*	0.05	-0.08	0.72*	-0.19	0.07
Ca	0.63*	-0.43	-0.34	0.40	-0.41	0.14
Ti	0.49	0.29	-0.18	0.51	-0.27	-0.09
Sr	0.26	0.32	-0.06	0.29	0.15	-0.22

†p<0.10; *p<0.05; **p<0.01.

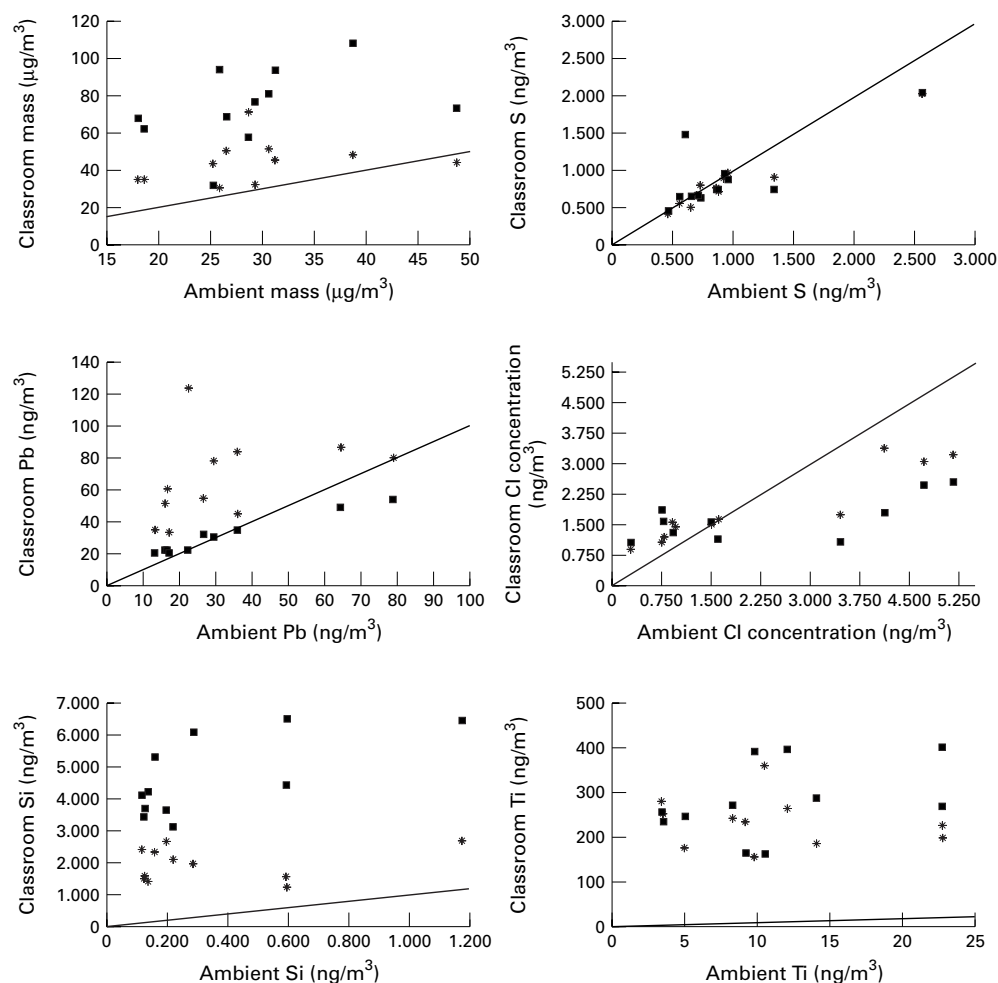


Figure 2 Relation between 24 hour mean outdoor and classroom concentrations (■=school 1; *=school 2; —=1:1 line).

Pb, 24 hour average classroom concentrations were similar to outdoor concentrations in school 1, whereas much higher classroom concentrations were found in school 2.

Spearman correlations between classroom and outdoor concentrations, and between the two classrooms are presented in table 3. Figure 2 shows the relations between 24 hour average classroom and outdoor concentrations for some elements. Because outdoor measurements were only conducted with a 24 hour averaging time, the comparison between classroom and outdoor concentrations should focus on the 24 hour average classroom concentrations. Classroom mass concentrations were only moderately correlated with outdoor concentrations. A significant correlation was only found between outdoor and 24 hour average classroom concentrations in school 1. For several crustal elements, 24 hour average concentrations in school 1 were also significantly correlated with outdoor concentrations, but other correlation coefficients were generally low and not significant. For S and Br, however, all concentrations that were measured with the same averaging time (between outdoor and 24 hour average classroom concentrations, and between the two classrooms) were significantly correlated. For Pb, significant correlations were found for school 1 but not for school 2.

Data on wind direction per hour, obtained from the measuring site Schiphol (Amsterdam Airport) of the Royal Dutch Meteorological Institute, showed that on 8 of the 11 days of measurements, the wind was directed from the sea (south west to north west; 210° – 330°) for over 66% of the time. On such days, PM_{10} concentrations in The Netherlands are relatively low.¹⁰ On the other 3 days, the wind was directed mainly from the south east to south west (120° – 240°). Mean temperature on the days of measurements was 6°C (range 2°C – 9°C), thus no salting of roads occurred.

Discussion

This study has shown that mass concentrations and most elemental concentrations of PM_{10} in classrooms were considerably higher than outdoor concentrations, especially during school hours. The highest classroom/outdoor ratios were found for the soil elements Si, Ca, and Ti. The only elements that showed 24 hour average classroom concentrations lower than or equal to the corresponding outdoor concentrations were S, Br, and Pb (at school 1 only), which are dominated by combustion sources, and Cl, which is dominated by marine aerosol. Given the repainting that was conducted in school 2 just before the start of the study, the

high Pb concentrations in school 2 were possibly caused by removal of old lead based paint.

No information is available about elemental concentrations in schools. Several studies in homes and offices have generally found lower indoor/outdoor ratios for mass and crustal elements than the ones found in our classrooms.^{7 17 18} In the PTEAM study,^{1 2} mass and elemental concentrations of PM₁₀ in the 178 homes were generally lower or similar to outdoor concentrations, both during daytime and during night time. During night time, this was also the case for personal exposure measurements. For daytime personal samples, however, the pattern of results was more similar to our study: mass concentrations (mean 150 µg/m³) were considerably higher than outdoor and indoor concentrations (mean 95 µg/m³), and, except for S, all elements measured were also increased by 50%–100% in the personal concentrations compared with the indoor concentrations. Nevertheless, these increases are small compared with the ones found in our classrooms, especially for Si, Ca, and Ti.

In the classrooms, no particle emitting sources such as smoking, and woodburning and gas stoves were present. The high classroom concentrations are also not likely to be caused by local traffic: the highest concentrations were found in school 1, which was situated at greater distance and much less often downwind of the nearest busy road than school 2. Furthermore, the highest classroom concentrations were found during school hours, when the classrooms were occupied by about 30 children. Hourly PM₁₀ data of the same period, obtained from the Amsterdam site of the National Air Quality Monitoring Network, did not show significant differences between school time and 24 hour average outdoor concentrations (29.2 µg/m³ and 29.6 µg/m³ respectively). The differences between school time classroom measurements and outdoor concentrations can therefore not be explained by diurnal variation in ambient concentrations. The most probable cause of the increased classroom concentrations, therefore, is resuspension of settled dust or suspension of soil material brought in by the children's shoes. Use of chalk for writing on the blackboard could also be a dust source. Chalk consists mainly of calcium carbonate and will therefore predominantly affect Ca concentrations. As the classroom/outdoor ratios for Si and Ti were higher than for Ca, chalk dust was probably not the most important source. Several studies found that human activity can cause increased indoor particle concentrations.^{2 6 7} These studies, moreover, suggest that human activity contributes primarily to resuspension of coarse particles.^{2 6 7} In our study, the highest classroom/outdoor ratios were found for soil related elements and the lowest ratios were found for elements dominated by combustion sources. As soil elements are generally associated with coarse particles and elements from combustion sources are generally associated with particles <1 µm,^{19 20} this is consistent with the general finding that resuspension activities mainly affect coarse particle concentrations.

The largest classroom/outdoor ratios were found for Si, Ca, and Ti. These elements showed large differences in their relative contribution to classroom and outdoor mass concentrations. In total, Si and Ti contributed about 6% to the schooltime classroom mass concentrations, compared with <1% to the outdoor mass concentrations. As these elements can be considered as the most specific soil elements, this suggests that the increased classroom concentrations are not only caused by resuspension of settled dust, but also by suspension of soil material.

The highest correlations between classroom and outdoor concentrations and between classrooms were found for S, Br, and Pb (school 1). These elements have in common that they all had 24 hour average classroom concentrations lower than or similar to outdoor concentrations and originate primarily from combustion sources, which are generally associated with particles <1 µm. For Cl, classroom/outdoor ratios were also low, but concentrations were less well correlated than for S, Br, and Pb. Chlorine differs from these elements in that it has marine aerosol as its main source, which is generally associated with coarse particles. Elements that were increased in the classroom samples, generally showed low correlations. In the PTEAM study,² S was the only element that was not increased in the personal samples compared with indoor samples. Also, S was the only element for which personal and indoor concentrations were highly correlated with outdoor concentrations ($R^2=0.8-0.9$). For mass and all other elements, personal, and outdoor concentrations were less well correlated ($R^2=0.1-0.4$). Several studies on personal, indoor, and outdoor sulphate have also found high correlations, with correlation coefficients between personal and outdoor concentrations or between indoor and outdoor concentrations ranging from 0.81 to 0.99.²¹⁻²⁴ In one of these studies,²¹ mass concentrations of respirable suspended particles were also measured. The correlation between personal and outdoor respirable suspended particle concentrations ($R=0.69$) was lower than between personal and outdoor sulphate ($R=0.81$). In a study on childhood exposure to fine particles,⁸ classroom concentrations were similar to and highly correlated ($R>0.90$) with ambient concentrations. When personal or indoor concentrations were significantly influenced by resuspension of predominantly coarse particles, these excess coarse particles reduce the correlation between personal or indoor concentrations and outdoor coarse particle concentrations, whereas correlations for fine particles were less influenced.

Recently, it has been suggested that fine particles (PM_{2.5}) more than coarse particles (PM₁₀ minus PM_{2.5}) are specifically responsible for the associations found between air pollution by PM and mortality.²⁵ Also, it has been hypothesised that the (metal) composition of the particles affects the associated health response.²⁶ In this case, indoor PM₁₀ measurements in environments with a lot of activity would not provide a good measure of the relevant exposure. Furthermore, as the general compo-

sition of PM₁₀ in classrooms differs from the composition of PM₁₀ in ambient air, the high PM₁₀ mass concentrations in classrooms cannot be directly compared with ambient air quality guidelines.

Conclusion

This study has shown that the high PM₁₀ mass concentrations in classrooms are probably due to resuspension of coarse particles or suspension of soil material, caused by the activity of the children. Due to these excess coarse particles, the correlation between classroom and outdoor concentrations is lower for elements associated with coarse particles than for elements that are generally associated with fine particles. As the general composition of PM₁₀ in classrooms differs from the composition of PM₁₀ in ambient air, the high PM₁₀ mass concentrations in classrooms can probably not be directly compared with ambient air quality guidelines.

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