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Personal exposures and indoor, residential outdoor, and urban background levels of fine particle trace elements in the general population

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Personal exposures and indoor, residential outdoor, and urban background levels of PM_{2.5} and PM₁ were measured simultaneously in Göteborg, Sweden. A total of 270 24-hour samples from 30 subjects were analyzed for elemental concentrations using X-ray fluorescence (XRF) spectroscopy. Personal exposures to PM_{2.5} were significantly higher for Cl, Ca, Ti, and Fe compared with the other sites. For most elements, residential outdoor levels were significantly higher than urban background levels. Correlations between personal exposure and stationary measurements were moderate to high for Zn, Br, and Pb ($r_s = 0.47\text{--}0.81$), while Ca and Cu showed low correlations. The penetration indoors from outdoors was 0.7, as calculated from S and Pb ratios. For the pairs of parallel PM₁ and PM_{2.5} measurements, only Ca and Fe levels were significantly lower for PM₁ at all sites. Significant correlations were found between urban background mass concentrations and personal exposure levels for elements attributed to combustion processes (S, V, and Pb) and resuspended dust (Ti, Fe, and Zn), indicating that both sources could be relevant to health effects related to urban background mass. Air mass origin strongly affected the measured urban background concentrations of some elements (S, Cl, V, Ni, Br, and Pb). These findings were also seen for personal exposure (S, Cl, V, and Pb) and indoor levels (S, Cl, V, Ni, and Pb). No differences were seen for crustal elements. Air mass origin should be taken into account in the description and interpretation of time series studies of air pollution and health.

²⁵ Introduction

The general population is exposed to a number of air pollutants of natural and anthropogenic origin. The mixture is generally complex and pollutant levels vary greatly, both $<10\ \mu\text{m}$ (PM₁₀) have been the traditional standard, but measurement of PM₁₀ has over the years been complemented with monitoring of fine particles with a diameter $<2.5\ \mu\text{m}$ (PM_{2.5}). The submicron fraction of PM_{2.5}, PM₁, has not been monitored or comprehensively studied. Instead, aerosol scientists often use different types of multistage or cascade impactors in determining size distributions of various particulate pollutants¹. The fact that the majority of combustion-derived particles are of submicron size makes PM₁ an important parameter to study in urban environments.

Numerous epidemiological studies have found associations between concentrations of ambient fine particulate matter (PM_{2.5}) and adverse health effects²⁻⁵. Some studies have shown that ambient concentrations do not always reflect the personal exposure⁶⁻⁸. In recent years, several studies

locally and over time. The traditional way to measure air pollution is at a centrally located monitoring station that monitors exceedances of air quality standards. For measuring particulate matter, particles with an aerodynamic diameter

comparing personal exposure and indoor and ambient levels of fine particle mass concentrations have been performed by researchers in Europe and North America. These studies have mostly focused on PM_{2.5} and sometimes also on black smoke. Only a few studies have characterized the chemical composition of the fine particulate matter in personal exposures^{6, 9-12}. Additional information obtained from chemical analysis may make it possible to estimate the contribution from different sources and their influence on personal exposure¹³. The penetration of ambient air indoors can be calculated using elements with no indoor sources (e.g., S or Pb) and thus, assessment of the influence of ambient air on indoor air can be improved.

The aims of the present study were to characterize the personal exposure to trace elements in fine particulates (PM_{2.5} and PM₁), to describe the relations between personal, indoor, residential outdoor, and urban background (PM_{2.5}) concentrations, and to assess the influence of different air masses on the measured concentrations.

Materials and Methods

Study design and subjects

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The study took place in Göteborg, Sweden, during two spring periods and two autumn periods in the years 2002 and 2003 (April 2 – June 7 and September 26 – November 6, 2002, and March 27 – June 12 and October 7–30, 2003). Personal, indoor, and residential outdoor measurements of PM_{2.5} and PM₁ in the homes of the subjects, as well as PM_{2.5} at a stationary outdoor urban background station were performed.

The subjects were recruited from two groups. In the first group, out of 30 adults (aged 20–50 years) randomly selected from the population living in Göteborg, 24 agreed to participate in the study. Four were excluded due to lack of possibilities of performing residential outdoor measurements. The second group consisted of ten volunteers among the staff of our Department in Göteborg. Repeated measurements were performed in ten of the 20 randomly selected subjects and in ten staff members. The latter group was recruited from our staff since two sets of sampling equipment for personal measurements were used. The handling of the double setup was complex, but these subjects were easier to instruct. The study therefore included 30 subjects, eight men and 22 women. There were three smokers among the 20 randomly selected participants, and none among the ten volunteers.

All subjects completed a questionnaire about age, occupation, and type of home and heating system, as well as daily activities. The daily activities diary included questions regarding smoking, exposure to environmental tobacco smoke, time spent at home, at work, indoors and outdoors, time spent in cars and buses, and occupational exposure. One subject in the randomly selected study group was found to have significant workplace exposure and was therefore excluded from the analysis of personal exposure. The study was approved by the Ethics Committee at Göteborg University.

Monitoring

Identical sets of equipment were used for personal exposure and indoor and residential outdoor sampling, namely, a GK2.05 (KTL) cyclone for PM_{2.5} sampling and a Triplex SCC1.062 cyclone for PM₁ sampling. Both cyclones were connected to BGI 400S personal sampling pumps with flow rates of 4 and 3.5 l min⁻¹, respectively. For PM_{2.5} measurements at the urban background station, an EPA-WINS impactor (PQ100 EPA-WINS Basel PM_{2.5} Sampler) with a flow rate of 16.7 l min⁻¹ was used. The sampling time was 24 hours. We used 37 mm Teflon filters (Pall Teflo, R2PJ037) in the cyclones and 47 mm Teflon filters (Pall Teflo, R2PJ047) in the WINS impactor.

Parallel measurements of both PM_{2.5} and PM₁ were performed indoors and outside the residences. Personal measurements were performed in two ways. The randomly selected individuals were equipped with a small shoulder bag with one PM_{2.5} cyclone and a pump attached to it. The cyclone was attached to the shoulder strap and placed near the breathing zone. The ten subjects from our staff were equipped with two sets of sampling equipment, near the breathing zone, one on each side. During the first measurement, one PM_{2.5} cyclone and one PM₁ cyclone was used for determination of the size distribution of particles in the personal exposure. During the second measurement, two PM_{2.5} cyclones were

used to study the precision of the measurements. All measurements (personal exposure, and indoor, outdoor, and urban background) for each subject started simultaneously. In total, 50 sampling periods (on 47 different days) yielded altogether 270 filters. Normally, only one subject was investigated each day. Information about the weather (temperature, wind speed, and direction, rain, etc.) was provided by the Environmental Agency in Göteborg.

Analytical techniques

All filters were weighed before and after exposure using a CAHN C-30 microbalance placed in a temperature- and humidity-controlled room¹⁴. An energy-dispersive X-ray fluorescence (EDXRF) spectrometer at the Department of Chemistry, Atmospheric Science, Göteborg University¹⁵, was used to analyze the elemental composition of all filter samples. The EDXRF spectra were processed and quantified using the Quantitative X-ray Analysis System (QXAS) and the Analysis of X-ray spectra by Iterative Least-square fitting (AXIL)^{16, 17}. All samples were analyzed using a livetime of 1,000 seconds, a tube voltage of 55 kV, a tube current of 25 mA, and a Mo secondary target. More information about calibration and quality control has been presented elsewhere¹⁴. For some filters with low concentrations, a more narrow and fine-tuned spectrum fit was obtained to improve the data recovery of the lighter elements (up to V). The mean analytical precision was 5%, as calculated from repeated analysis (N = 5) of two randomly selected filters, one having a low and the other, a high mass loading. In total, 65 field blanks were analyzed and concentrations were below the limit of detection (LoD) for all elements except Fe and Zn, but their concentrations were low and did not change the results.

Air mass back trajectories

Air mass back trajectories were computed using the NOAA ARL HYSPLIT Model¹⁸ to investigate the effect of long distance transported (LDT) pollution. For each sampling day, 96-hour air mass back trajectories were computed at startup time and 6, 12, 18, and 24 hours later. The trajectories were divided into four classes, representing different source areas, namely, continental areas (central and Eastern Europe), marine areas, the Nordic countries Finland, Norway, and Sweden, and the UK, or remained undetermined (for trajectories that shifted classes during the sampling day). The classification was made according to the criterion that all trajectories during a sampling period must have a major path belonging to the same class.

Statistical analysis

Statistical calculations were performed with the SAS System for Windows, version 9.1¹⁹. Correlations between elemental concentrations in different microenvironments were assessed using the Spearman rank correlation coefficient (r_s) and differences between pairs of personal, indoor, outdoor, and ambient levels using the Wilcoxon signed rank test. For unpaired observations, Wilcoxon's rank sum test was used. Statistical significance refers to $p < 0.05$ in two-tailed tests. If the elemental concentration was below the LoD, the LoD

Table 1 Results of the urban background PM_{2.5} measurements of trace elements (all concentrations in ng m⁻³).

Element	First measurement (N = 28)			
	Mean	Median	#>LoD ^a	Range
S	620	320	21	95–1900
Cl	97	54	17	25–460
K	55	50	28	32–130
Ca	21	17	28	6.6–62
Ti	2.1	1.9	15	1.3–3.8
V	3.4	2.4	16	1.0–13
Mn	1.6	1.4	17	0.67–3.8
Fe	36	33	28	7.1–100
Ni	1.6	1.2	21	0.33–5.7
Cu	2.1	1.4	27	0.33–11
Zn	14	11	28	2.8–38
Br	1.7	1.4	28	0.47–4.3
Pb	3.3	2.1	28	0.94–11

^a LoD = limit of detection.

divided by the square root of two was used in the calculations²⁰.

Results

The results for personal, indoor, residential outdoor, and urban background measurements are summarized in Tables 1 and 2. The elements Cr, Rb, and Sr have been omitted due to low detection frequency (<33%), as has Mn except for the urban background measurements. In general, mass concentrations were low. The median value for the personal PM_{2.5} exposure was 8.4 µg m⁻³ and for indoor, residential outdoor, and urban background PM_{2.5} measurements, 8.6, 6.4, and 5.6 µg m⁻³, respectively (not shown in the Tables). For PM₁, the median levels were 5.4, 6.2, and 5.2 µg m⁻³ for personal, indoor, and outdoor levels, respectively. More

15 details regarding mass concentrations will be presented elsewhere.

There was moderate variability for most elements in duplicate personal samples, with coefficients of variation (CVs) including the analytical CV ranging from 16% to 53%. The lowest CVs were found for K and Pb, and the highest, for Zn and Fe. The variability for Ni was very high (CV 204%), probably due to contamination during sampling.

For PM_{2.5}, the personal exposures were significantly higher than both outdoors and urban background measurements for the elements Cl, K, Ca, Ti, Fe, and Cu (Table 3). The median individual ratios between personal exposure and outdoor locations for Ca, Fe, and Cu varied from 2 to 4. Compared with the urban background station, the concentrations of all elements except S, Br, and Pb were higher for personal exposure. Personal exposure was also higher than indoor levels for Cl, Ca, Ti, Fe, and Br, but lower than outdoor Pb (median individual ratio 0.75). Residential outdoor levels were significantly higher than the corresponding indoor levels for Br and Pb, but lower for Ti and Cu. The residential outdoor levels were significantly higher than the urban background levels for all elements except for Fe, Cu, and Pb.

In Fig. 1, Spearman correlations between personal PM_{2.5} exposure and indoor, residential outdoor, and urban background PM_{2.5} levels are shown for some elements. The associations for Zn, Br, and Pb were all relatively high ($r_s = 0.47$ – 0.81), while Ca and Cu showed low or no associations between personal exposure and the other sites. For Fe, only the personal exposure-outdoor relationship ($r_s = 0.54$), and for K (not shown in the Figure), only the personal exposure-indoor relationship ($r_s = 0.70$) was statistically significant.

Table 2 The PM_{2.5} and PM₁ measurements of the personal exposure and indoor and residential outdoor levels of trace elements (concentrations in ng m⁻³).

Element	Personal PM _{2.5} (N = 29)				Indoor PM _{2.5} (N = 30)				Residential outdoor PM _{2.5} (N = 29)			
	Mean	Median	#>LoD	Range	Mean	Median	#>LoD	Range	Mean	Median	#>LoD	Range
S	–	<470	12	270–1400	–	<450	11	240–1500	640	460	17	190–1800
Cl	270	170	21	61–920	220	140	17	61–770	200	140	21	57–840
K	140	96	29	39–690	140	90	30	48–710	82	78	28	32–200
Ca	110	80	29	27–670	69	53	30	15–450	34	28	24	4.6–85
Ti	11	9.5	25	3.7–27	8.0	7.1	18	2.9–20	6.3	5.2	17	3.3–21
V	4.7	4.0	15	2.7–9.4	4.5	3.9	17	2.6–9.5	5.5	3.9	19	2.1–14
Fe	68	69	29	23–150	39	36	30	12–100	45	31	29	8.8–200
Ni	4.2	2.6	20	0.89–46	4.6	1.4	17	0.67–63	–	<1.6	13	0.65–5.5
Cu	10	6.6	28	1.1–81	9.8	3.9	29	0.97–91	2.6	1.3	18	0.65–17
Zn	21	16	29	6.6–70	17	14	30	6.4–40	22	15	29	5.5–85
Br	2.0	1.3	23	0.91–14	1.8	1.3	22	0.67–12	2.0	1.3	25	0.91–5.1
Pb	2.9	2.6	21	0.92–8.3	3.2	2.7	22	0.91–8.3	4.6	2.6	27	0.90–20

Element	Personal PM ₁ (N = 10)				Indoor PM ₁ (N = 30)				Residential outdoor PM ₁ (N = 29)			
	Mean	Median	#>LoD	Range	Mean	Median	#>LoD	Range	Mean	Median	#>LoD	Range
S	–	<470	1	240–1200	–	<440	3	240–1400	–	<450	5	240–2000
Cl	–	<110	2	54–160	–	<130	5	64–250	–	<110	5	44–170
K	80	82	10	50–130	130	74	28	31–640	76	68	28	34–170
Ca	32	23	9	8.4–87	22	18	20	7.6–73	–	<12	13	5.1–78
Ti	6.5	6.3	5	3.7–11	–	<5.3	7	4.2–12	–	<5.0	8	2.2–9.5
V	–	<4.2	3	2.8–8.9	–	<4.4	14	2.5–11	5.6	4.7	16	2.2–14
Fe	28	25	10	7.6–68	17	13	30	3.4–77	23	14	29	3.7–140
Ni	8.2	1.2	5	0.83–58	3.4	1.5	16	0.81–28	3.3	1.4	15	0.73–28
Cu	5.0	4.4	10	1.6–14	6.4	2.8	22	0.81–55	–	<1.1	13	0.73–12
Zn	15	14	10	7.6–37	14	12	30	4.6–37	15	14	29	5.2–30
Br	1.6	1.5	7	0.83–4.4	1.8	1.2	15	1.0–12	1.5	1.4	21	0.78–4.3
Pb	3.6	2.8	7	1.1–11	3.0	2.2	17	0.84–9.4	4.1	1.5	24	1.0–17

Note: Where the median value is below the limit of detection (LoD) for the element in question, the value is notated with a < sign.

Table 3 Difference in mean PM_{2.5} concentration between personal exposure (P) and indoor (I), residential outdoor (O), and urban background (UB) levels (all differences in ng m⁻³), together with the corresponding p-values calculated using Wilcoxon's signed-rank test.

Element	P-I (N = 29)		P-O (N = 28)		P-UB (N = 27)		I-O (N = 29)		O-UB (N = 27)	
	Mean difference	p-value	Mean difference	p-value	Mean difference	p-value	Mean difference	p-value	Mean difference	p-value
S	^a	^a	-79	0.293	-33	0.870	-130	0.167	64	0.028
Cl	40	0.043	72	0.013	170	<0.001	30	0.282	91	<0.001
K	0.18	0.924	64	<0.001	92	<0.001	59	0.077	28	<0.001
Ca	36	<0.001	70	<0.001	87	<0.001	32	0.001	14	<0.001
Ti	2.5	0.032	4.2	<0.001	8.7	<0.001	1.4	0.070	4.2	<0.001
V	0.22	0.992	-0.83	0.184	1.2	<0.001	-1.1	0.108	2.4	<0.001
Fe	29	<0.001	23	<0.001	33	<0.001	-6.3	0.681	9.8	0.071
Ni	-0.55	0.220	2	0.162	2.7	0.002	2.5	0.841	0.73	0.003
Cu	0.21	0.292	7.8	<0.001	5.8	<0.001	7.3	0.002	0.52	0.511
Zn	4.20	0.181	0.11	0.657	7.7	0.003	-5.6	0.051	7.5	0.004
Br	0.28	0.044	0.09	0.411	0.34	0.590	-0.27	0.044	0.35	0.021
Pb	-0.18	0.941	-1.2	0.013	-0.21	0.397	-1.5	0.010	1.3	0.059

^a Number of samples above LoD<50%. Statistically significant differences are marked in bold.

The outdoor-urban background associations were high for S, V, Br, and Pb ($r_s > 0.7$) and moderate for Cl, Fe, Cu, and Zn ($0.5 < r_s < 0.7$). The penetration indoors from outdoors was calculated as the ratio for both S and Pb. The median penetration was 0.72 for S and 0.69 for Pb, with 95% confidence intervals of 0.5–1 for both.

For the pairs of parallel PM_{2.5} and PM₁ measurements, personal, indoor, and outdoor Ca and Fe levels were significantly higher for PM_{2.5} than for PM₁ (Table 4). The value for Cu was higher with the personal and indoor PM_{2.5} measurements, while values for Zn and Br were higher for outdoor PM_{2.5} (Table 4). The median ratios between PM₁ and PM_{2.5} for Ca and Fe were in the ranges of 0.31–0.40 and 0.38–0.41, respectively. The PM₁/PM_{2.5} ratios for Cu were 0.76 indoors and 0.58 for personal measurements. For outdoor measurements, the ratios for both Zn and Br were 0.8. The correlations between the parallel PM_{2.5} and PM₁ samples for personal exposure, indoor, and residential outdoor levels are shown for some elements in Fig. 2. All correlations were high for Pb ($r_s \geq 0.75$) and moderate for K ($r_s = 0.58$ –0.71), while for Zn and Br ($r_s = 0.86$), they were high only outdoors (K and Br not shown in the Figure). Weak correlations were found for

Table 4 Difference in mean concentration of trace elements between PM_{2.5} and PM₁ for personal exposure, indoor, and residential outdoor levels (all differences in ng m⁻³), together with the corresponding p-values calculated using Wilcoxon's signed-rank test.

Element	Personal (N = 10)		Indoor (N = 30)		Residential outdoor (N = 29)	
	Mean diff.	p-value	Mean diff.	p-value	Mean diff.	p-value
S	^a	^a	^a	^a	42	0.391
Cl	98	0.064	70	0.681	44	0.519
K	6.7	0.375	15	0.005	6.1	0.108
Ca	35	0.019	47	<0.001	18	<0.001
Ti	1.8	0.160	1.9	0.078	0.79	0.292
V	0.087	0.922	-0.78	0.086	-0.26	0.254
Fe	33	0.037	22	<0.001	22	<0.001
Ni	-4.8	0.625	1.2	0.818	-1.2	0.547
Cu	3.1	0.049	3.4	<0.001	0.48	0.263
Zn	3.7	0.105	2.9	0.068	6.1	0.002
Br	0.016	0.846	0.07	0.723	0.53	<0.001
Pb	-0.69	0.570	0.21	0.119	0.58	0.060

Statistically significant differences are marked in bold.

^a Number of samples above LoD<50%..

Ca and moderate correlations for Fe indoors and outdoors.

Correlations between mass and elemental concentrations at 25 different locations are presented for some elements in Table 5. For Ca, Fe, and Zn, the correlations with PM mass followed the same trend, with significant correlations for residential outdoor levels and personal exposure and weak correlations indoors and at the urban background station. For K and Br, the 30 correlations were relatively similar at all places. On the other hand, Pb only showed significant correlations outdoors and in urban background levels. In addition, S was highly correlated with urban background PM_{2.5} mass concentrations ($r_s = 0.82$) (not shown in the Table).

35 Statistically significant correlations between urban background mass and elemental concentrations of the personal exposure were found for Ti ($r_s = 0.40$, not shown in Table 5), Fe, Zn, and Pb. In addition, correlations for S and V were significant ($r_s = 0.70$ for both), but since a large fraction of 40 these last two elements were below the LoD, the results should be interpreted with caution. No correlations were found for Cl, K, Ca, and Cu.

The origin of the sampled air strongly affected the measured concentrations of some of the elements. When 45 comparing marine with continental air, the *urban background* PM_{2.5} levels of S, Br, and Pb were significantly higher for continental air masses, while the Cl level was higher for marine air masses (Fig. 3 on page 8). This was also the case with Cl and Pb in the residential outdoor measurements. Other 50 significant urban background differences between air masses are presented in Figure 3. For the crustal elements Ca, Ti, Mn, Fe, Cu, and Zn, no differences between any of the air masses were observed.

Some differences between air masses were even large 55 enough to be statistically significant in the personal exposure and/or indoor levels. In the *personal exposure* measurements, significantly higher levels of Pb were found for continental air masses compared with both marine and Nordic air, while significantly higher V levels were seen for continental and UK 60 air masses compared with Nordic air. Air masses originating from the UK showed significantly higher levels of S compared with marine air. In addition, Cl levels of marine air were significantly higher compared with continental air.

Indoors, higher levels were seen for continental S and Pb 65 compared with Nordic air, while levels of Cl were higher in

Table 5 Spearman correlations between PM mass and the corresponding concentrations of some elements in the urban background (UB), residential outdoor (O), indoor (I), and personal exposure (P) samples, as well as correlations between PM_{2.5} mass at the UB site and the elemental concentrations for personal exposure. Values for PM₁ are presented in parenthesis where applicable.

Element	UB Mass vs. elements	O Mass vs. elements	I Mass vs. elements	P Mass vs. elements	UB mass vs. personal elements
K	0.46	0.65 (0.36)	0.51 (0.40)	0.54 (0.32)	0.16
Ca	0.10	0.61 (-)	0.20 (0.18)	0.76 (0.86)	0.31
Fe	0.30	0.80 (0.64)	0.36 (0.53)	0.60 (0.78)	0.51
Zn	0.43	0.72 (0.55)	0.38 (0.10)	0.49 (0.52)	0.54
Br	0.75	0.45 (0.76)	0.60 (-)	0.60 (0.76)	^a
Pb	0.50	0.83 (0.55)	0.16 (-)	0.21 (0.18)	0.40

Note that PM₁ was not measured at the urban background station.

Significant values are marked in bold. Statistics are not shown if number of samples above LoD < 65%.

^a Not evaluated since the range in personal exposure was too narrow.

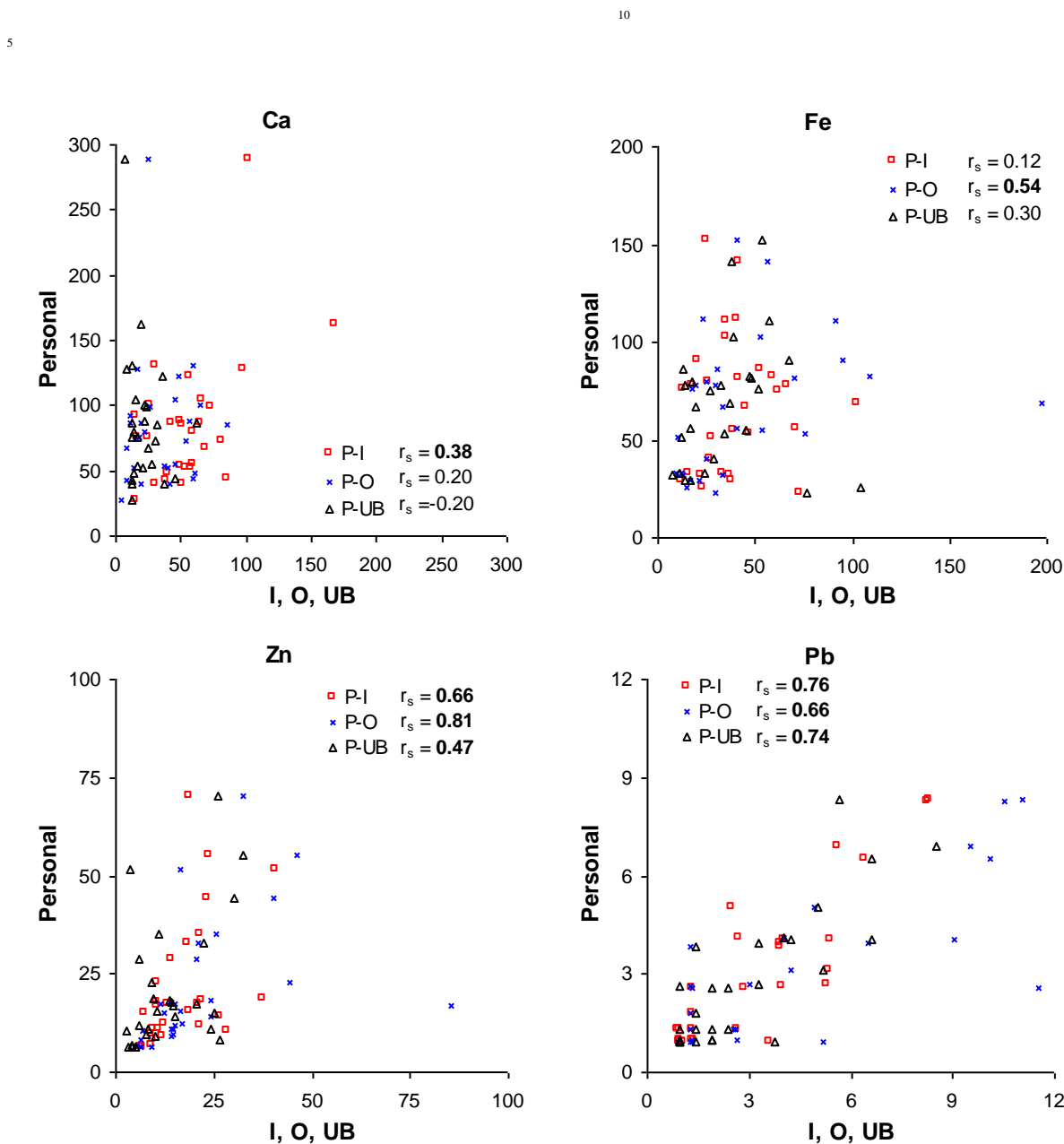


Fig. 1 Concentrations of Ca, Fe, Zn, and Pb (in ng m⁻³) in personal PM_{2.5} exposure (P) versus indoor (I), residential outdoor (O), and urban background (UB) levels. Significant correlations are marked in bold. Note: For Ca, one high personal exposure (670 ng m⁻³) and its corresponding indoor, outdoor, and urban background values (450, 28, and 9 ng m⁻³, respectively) are not shown in the graph.

marine air than in continental air. In addition, air masses coming from the UK had significantly higher levels of V and Ni compared with Nordic air masses.

Discussion

5 Personal exposure versus indoor, residential outdoor, and urban background concentrations

The urban background station had lower levels compared with residential outdoor levels for all elements and the differences were significant for all except Fe and Pb (Table 3). The elevated rooftop placement of the urban background station in the campus area of the Faculty of Medicine situated in central Göteborg makes measurements taken at this site decidedly less affected by traffic, local industries, and pollution from the harbor compared with measurements outside some subjects' homes.

The reason why personal exposure concentrations were higher than outdoor concentrations (outdoor and urban background levels) for many elements is that indoor sources had a significant impact on personal exposure. Only elements such as S, Br, and Pb, with mainly outdoor sources, did not show any differences. Differences between personal exposure and indoor levels followed the same pattern as did the personal exposure-outdoor concentration and personal exposure-urban background differences for Cl, Ca, Ti, and Fe. Elements with only outdoor sources, such as V and Pb, did not show any significant differences between personal exposure and indoor levels, and neither did elements with likely contributing indoor sources, such as K, Ni, Cu, and Zn.

Only few other studies have presented data on elemental concentrations for both personal exposure, and indoor and outdoor levels of fine particles. These were performed in Amsterdam, The Netherlands, and Helsinki, Finland⁹, in New

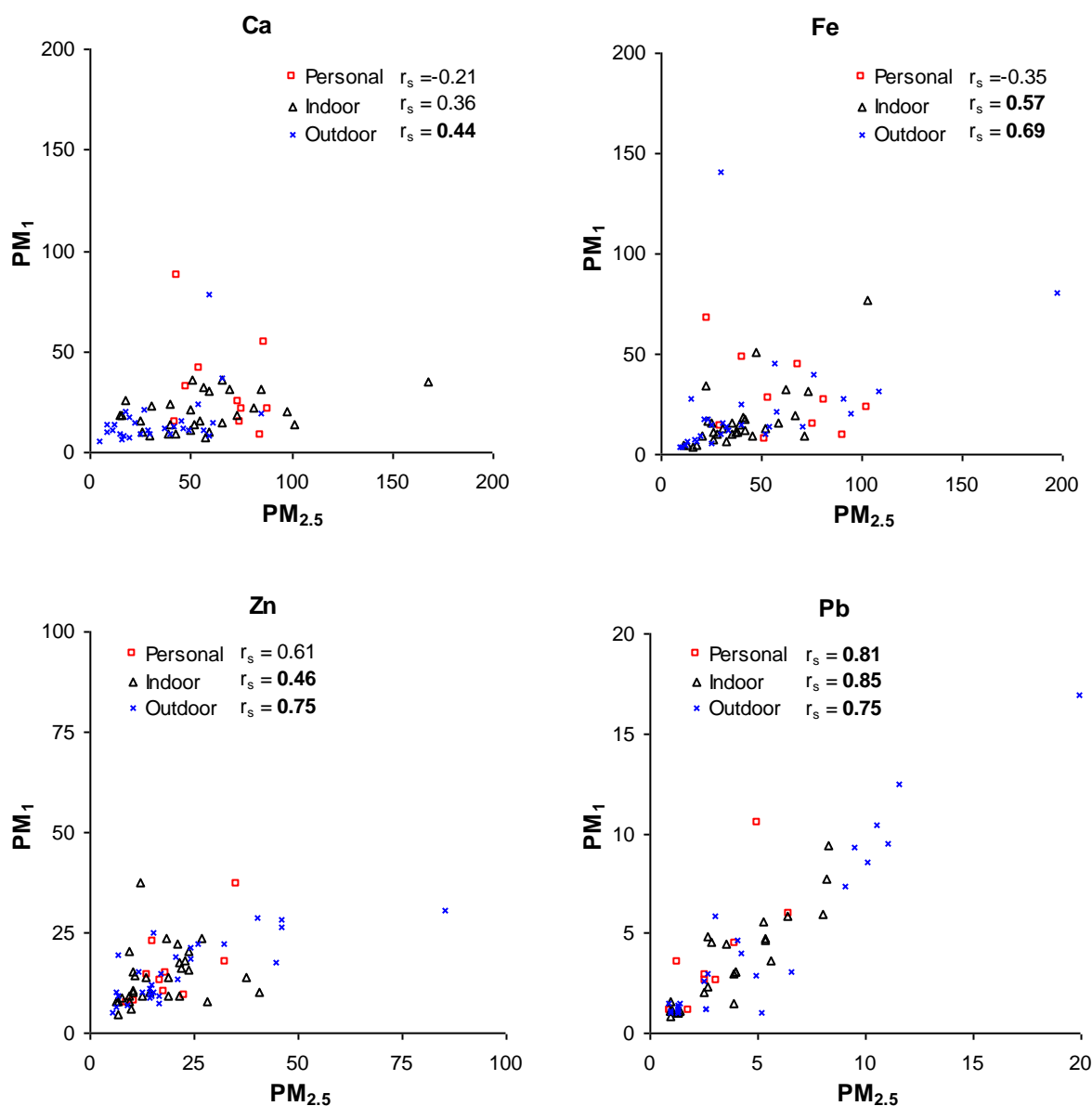


Fig. 2 Concentrations of Ca, Fe, Zn, and Pb in PM₁ versus PM_{2.5} (in ng m⁻³) at different locations. Significant correlations are marked in bold.

York, US¹⁰, in Oxford, UK¹¹, in Seattle, US¹², and in Hagfors, Sweden¹⁴. Compared with the present study, Amsterdam had higher levels of PM mass and S at all sites, while the levels of Cl and K at all sites as well as personal Ca were higher in Göteborg. In Helsinki, the levels were generally similar, except for S, which was higher in Helsinki, and Cl and K, which were higher in Göteborg. New York had much higher concentrations of PM mass, S, Pb, and all transition metals except Ti and V for all sites. In Oxford, UK, outdoor concentrations of S, Cl, and Br were higher than in Göteborg, while the other elements had comparable concentrations. Personal exposure and indoor levels in Oxford were higher for all elements except for Ti and V, which were higher in Göteborg. In Seattle, on the other hand, concentrations were similar or somewhat lower (Cl, K, Ti, V, Ni, and Cu) for all places. In Hagfors, Sweden, a small community investigated during the winter season, domestic wood burning for space heating is very common and levels of elements associated with wood combustion (K and Zn) and S (from LDT pollution) were higher, while Ti, V, and outdoor Cl were lower compared with the present study. This reflects the fact that population density, geographic location, predominating wind direction, and local activities have an important impact on element concentrations.

The penetration indoors from outdoors was calculated for both S and Pb since neither element has any indoor sources, and the I/O ratios of 0.72 and 0.69, respectively, were almost equal. Other studies have reported I/O ratios of similar magnitude for S or sulfate, ranging from 0.6 to 0.9²¹⁻²⁴, with somewhat lower values being found in winter than in summer. However, during winter in Sweden, a lower penetration factor (0.5) was found¹⁴.

PM₁ versus PM_{2.5}

The consistent feature of significantly higher concentrations of Ca and Fe in PM_{2.5} compared with PM₁ for personal exposure and indoor and outdoor levels was expected since for both elements, coarse particles (e.g., windblown, soil-derived dust and abrasion) are major sources. The low-end tail of the coarse fraction mode of these elements contributes to the supermicron part of the total PM_{2.5}¹. Similarly, personal and indoor Cu levels were affected by coarse fraction indoor sources. Zn and Br, on the other hand, showed significant differences outdoors, but not indoors or in personal exposure. This was probably due to coarse particle sources outdoors (mainly tire wear for Zn and sea salt particles for Br). For all other elements, no significant differences were found, suggesting that these elements exist mainly in the accumulation mode and are consequently collected by both the PM₁ and the PM_{2.5} cyclones. Comparing the PM₁ versus PM_{2.5} correlations outdoors, indoors, and in personal exposure is complicated since both indoor and personal exposure samples are made up of a mixture of indoor sources and penetration indoors from outdoor sources. In addition, the personal exposure is affected by direct exposure outdoors and also, by the personal cloud around a person. The elements with the highest correlations between PM₁ and PM_{2.5} were elements with combustion processes as the main submicron sources (e.g., K and Pb), while elements with multiple (indoor

and outdoor) sources (e.g., Cl and Ca), and also including the coarse mode, were mostly non-correlated.

Correlations between elements and particulate matter mass

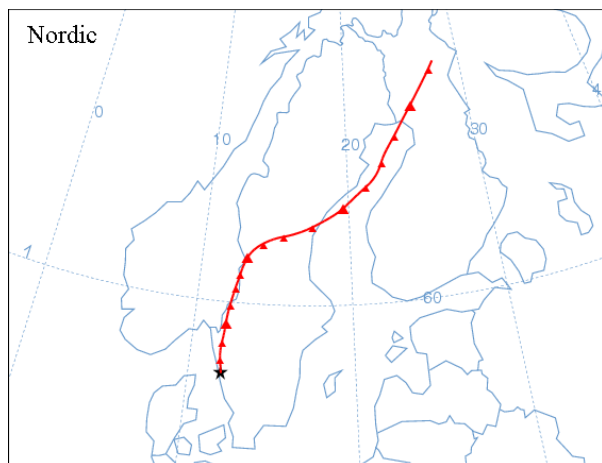
The common practice of measuring only the PM mass of indoor, outdoor, and personal exposure provides little information on the sources of the collected particles. Chemical characteristics (e.g., transition metals, soot, sulfates, nitrates, and volatile organic compounds) give information about the sources (e.g., traffic, wood smoke, and marine air) of the particles. It is also important to understand how concentrations of different chemical species vary with PM mass. In the present study, the overall highest PM versus element correlations were found for the residential outdoor measurements, followed by the personal exposure levels (Table 5). The correlations for the urban background station were similar to the ones presented earlier for Göteborg²⁵.

There was no correlation between PM mass and elements of crustal origin (Ca and Fe) at the urban background site, while the correlation was high for the residential outdoor measurements. This can be explained by the fact that windblown soil and road dust are more easily collected near the ground than at a rooftop site. The good correlation with PM mass for elements from combustion sources (K and Pb) for both residential outdoor and urban background filters is due to the fact that the dominating part of the combustion aerosol comes from polluted LDT air masses and is more evenly distributed across the city. On the other hand, Pb was not correlated to indoor or personal exposure PM mass. The probable reason for this is that Pb is not related to indoor activities and that the outdoor contribution to indoor mass concentrations is small. Meng et al.²⁶ showed that ambient sources could explain about 56% of indoor mass concentrations.

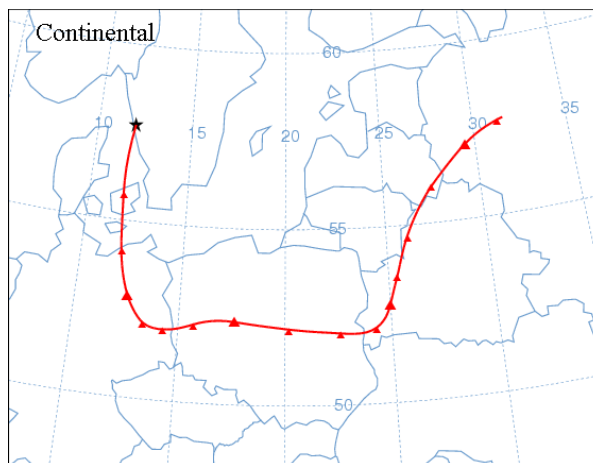
Use of PM mass at urban background stations has been the most common way to monitor exposure to particulate air pollution. There is a link between elevated concentrations of urban background PM mass and both short- and long-term health effects^{4, 5, 27, 28}. In the present study, significant correlations were found between urban background PM mass and personal exposure to the elements S, V, and Pb (attributed to combustion processes and LDT pollution) as well as to Ti, Fe, and Zn (local sources such as resuspended dust and road/tire wear). This suggests that both LDT and local sources could be relevant for health effects from particulate matter. In a previous study, a high correlation between urban background PM mass and personal S was found as well¹⁴. Further studies are needed to investigate whether similar conclusions can be drawn in other cities.

The effect of air mass origin on elemental concentrations

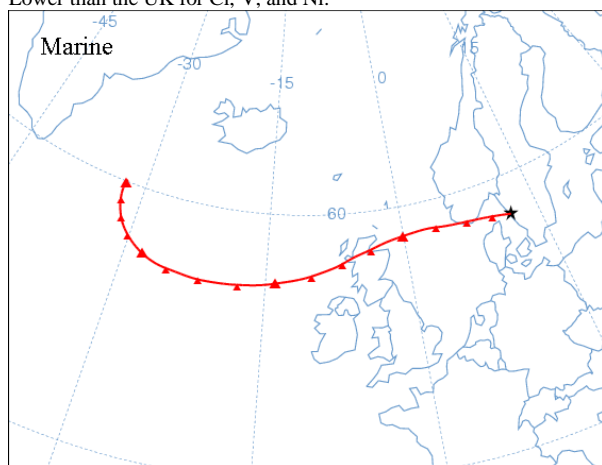
The four trajectory classes represent different levels and mixtures of trace elements. Nordic air is typically clean, containing low PM_{2.5} mass concentrations and low trace element levels. Marine air has high levels of sea salt particles (represented by high Cl levels) and elevated concentrations of V and Ni (from ship emissions), but low levels of other elements. As expected, the continental air in the present study had higher PM_{2.5} mass than the other classes, with high levels



N = 8 days.
 S 120 ng m^{-3} , Cl 37 ng m^{-3} , V 1.0 ng m^{-3} , Br 1.4 ng m^{-3} , and Pb 2.1 ng m^{-3} .
 Lower than continental for S, V, Ni, Br, and Pb.
 Lower than marine for Cl, V, and Ni.
 Lower than the UK for Cl, V, and Ni.



N = 15 days.
 S 960 ng m^{-3} , Cl 46 ng m^{-3} , V 2.8 ng m^{-3} , Br 2.4 ng m^{-3} , and Pb 3.8 ng m^{-3} .
 Higher than marine for S, Br, and Pb, and lower for Cl.
 Lower than the UK for V.



N = 6 days.
 S 230 ng m^{-3} , Cl 290 ng m^{-3} , V 3.8 ng m^{-3} , Br 0.94 ng m^{-3} , and Pb 1.4 ng m^{-3} .
 Lower than the UK for S, and higher for Cl.



N = 11 days.
 S $1,000 \text{ ng m}^{-3}$, Cl 62 ng m^{-3} , V 6.1 ng m^{-3} , Br 1.4 ng m^{-3} , and Pb 1.9 ng m^{-3} .

Fig. 3 Examples of the four trajectory classes and median concentrations of some key elements at the urban background station. Significant differences between air mass trajectories are presented.

of S, V, Ni, Br, and Pb, all of which can be attributed to industry and combustion processes. Air masses passing over the UK had a mixture of marine and continental characteristics, with high levels of S, V, and Ni and somewhat elevated levels of Cl, Br, and Pb. This is consistent with previous studies from Sweden^{14, 29}.

When comparing the different air masses that reached Göteborg, some of the differences between trajectory classes were large enough to have a statistically significant impact also on indoor and personal exposure levels. This was the case with S, V, Ni, and Pb, which were associated with combustion processes, and Cl, from sea salt particles. As could be expected, there were no differences between elements of crustal origin. We found no other studies presenting data on the impact of air mass origin on personal exposure. A better understanding of how regional transport processes (and seasonal pattern) can influence personal exposure could be important for the design and interpretation of time series analyses of PM levels and short-term effects on health.

Conclusions

Personal exposures to $\text{PM}_{2.5}$ were significantly higher for Cl, Ca, Ti, and Fe compared with the other sites, and for most elements, residential outdoor levels were significantly higher than urban background levels.

Personal exposure to elements attributed to combustion processes (S, V, and Pb) and resuspended dust (Ti, Fe, and Zn) were significantly correlated to urban background total $\text{PM}_{2.5}$ mass concentrations. This indicates that both sources could be relevant to health effects related to urban background mass.

Air mass origin strongly affected the measured urban background concentrations of some elements (S, Cl, V, Ni, Br, and Pb). These findings were also observed for personal exposure (S, Cl, V, and Pb) and indoor levels (S, Cl, V, Ni, and Pb). Air mass origin should therefore be taken into account in the description and interpretation of time series studies of air pollution and health.

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