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Sources of fine particulate matter in personal exposures and residential indoor, residential outdoor and workplace microenvironments in the Helsinki phase of the EXPOLIS study

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Objectives This study assessed the source contributions to the mass concentrations of fine particles (PM_{2.5}) in personal exposures and in residential indoor, residential outdoor, and workplace indoor microenvironments of the nonsmoking adult population unexposed to environmental tobacco smoke in Helsinki, Finland.

Methods The elemental composition of 48-hour personal exposure and residential indoor, residential outdoor, and workplace indoor PM_{2.5} was analyzed by energy-dispersive X-ray fluorescence spectrometry for 76 participants not exposed to environmental tobacco smoke and 102 participating residences with no smoking in Helsinki as a part of the EXPOLIS study. Subsequently, a principal component analysis was used to identify the emission sources of PM_{2.5}-bound elements and black smoke in each microenvironment, and this information was used to identify the corresponding sources in personal exposures. Finally, source reconstruction was done to determine the relative contributions of each source type to the total PM_{2.5} mass concentrations.

Results Inorganic secondary particles, primary combustion, and soil were the dominant source types for the PM_{2.5} mass concentration in all the microenvironments and personal exposures. The ratio of the residential indoor-to-outdoor PM_{2.5} concentration was close to unity, but the corresponding elemental ratios and source contributions varied. Resuspension of soil dust tracked indoors was a much larger contributor to residential and workplace indoor PM_{2.5} than soil dust to residential outdoor PM_{2.5}. Source contributions to personal PM_{2.5} exposures were best approximated by data from residential and workplace indoor microenvironments.

Conclusions Population exposure assessment of PM_{2.5}, based on outdoor fixed-site monitoring, overestimates exposures to outdoor sources like traffic and long-range transport and does not account for the contribution of significant indoor sources.

Key terms elemental composition, fine particles, long-range transport, principal component analysis, soil, source apportionment, source reconstruction, traffic.

As a consequence of the need to regulate particulate emissions from the most apparent sources, such as industry and traffic, current regulations and measurements of particulate matter with an aerodynamic diameter of <2.5 µm (PM_{2.5}) have focused on ambient-air pollution. Ambient-air measurements are often made in large monitoring programs, but they may not reflect personal exposure to and sources of PM_{2.5} in the population.

People spend most of their time indoors, and therefore a large part of their personal daily exposure to pollutants from indoor and outdoor origins occur in places they usually consider unpolluted, such as homes, offices, and automotive cabins (1). Although many studies have identified sources of particulate matter with an aerodynamic diameter of <10 µm (PM₁₀) in ambient air (2, 3), the relative contributions to total PM_{2.5} in air are not

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equally well known. A large proportion of the PM₁₀ mass concentration is comprised of coarse particles (PM_{10-2.5}) formed by mechanical or wind abrasion. In contrast, the PM_{2.5} fraction contains a much larger proportion of fine particles originating from primary emissions, secondary gas-to-particle reactions, and coagulation processes in the atmosphere.

In the particulate total exposure assessment methodology (PTEAM) study carried out by the Environmental Protection Agency (EPA) in the United States (4, 5, 6), PM_{2.5} concentrations and their sources were determined inside and outside the homes of 178 randomly selected nonsmoking subjects in Riverside, California. The median residential outdoor PM_{2.5} concentration was slightly higher (36 µg/m³) than the median residential indoor PM_{2.5} concentration (34 µg/m³) in daytime, but this difference was considerably larger (35 versus 26 µg/m³) at night. Smoking, cooking, dusting, and vacuuming were found to be the dominant indoor sources. Subsequently, house volume and air exchange rate were both found to be weak, but statistically significant determinants of the PM_{2.5} concentration in residential indoor air. In homes with smoking, particles of ambient origin, smoking, cooking, and other sources contributed 60%, 30%, 3% and 7 %, respectively, to the indoor PM_{2.5} concentration. The results of two other large-scale exposure studies (more than 150 homes), the Harvard six-city study and the New York State ERDA study, showed that the two most important indoor sources of PM_{2.5} were smoking and cooking (7). It must be noted that none of these studies identified all the indoor sources, and thus a more-detailed source apportionment of indoor sources of PM_{2.5} is needed.

The present study was conducted in Helsinki, Finland, as part of the EXPOLIS project, which is a study of air pollution exposure in European cities (8). Our principal aim was to identify sources of PM_{2.5} in personal exposures with the help of a principal component analysis (PCA) of the elemental compositions of PM_{2.5} in residential outdoor, residential indoor, and workplace indoor microenvironments. In addition, we examined the interaction of sources in these microenvironments. Finally, source reconstruction (9, 10) was applied to assess the contribution of different sources to personal and microenvironmental PM_{2.5} mass concentrations.

Material and methods

EXPOLIS (Helsinki) was a representative population-based study on air pollution exposure among adults. A core sample of 2523 subjects (25–55 years of age) was randomly selected from the population census in the Helsinki metropolitan area (total population ~1 million).

They were mailed a short screening questionnaire (75% response rate) concerning their home environment, occupation, socioeconomic status, and willingness to participate in the study. Responses to more-detailed questionnaires and 48-hour time activity diaries were obtained from 201 monitored participants selected randomly from those willing to participate in the study. Microenvironmental (residential indoor, residential outdoor, workplace indoor) and personal exposure concentrations of PM_{2.5} were monitored for these participants between October 1996 and December 1997.

Environmental tobacco smoke had the highest source contribution to PM_{2.5} exposure among the participants who reported such exposure (11). In an effort to avoid confounding of the source apportionment due to elements (like cadmium) common to tobacco smoke and emissions from combustion processes (4, 6, 12, 13), a subset of participants not exposed to environmental tobacco smoke was included in the principal component analysis. Exposure to environmental tobacco smoke was defined conservatively as any reported contact with tobacco smoke at any time, in any microenvironment, during the 48-hour monitoring period. This definition included active smokers, those living with smoking partners, and those working with people who smoked inside the workplace at any time during the monitoring period. These criteria left 102 residences in which no one smoked indoors and in which 76 participants not exposed to environmental tobacco smoke lived and could be included in the source apportionment analysis.

Collection of samples

A continuous program of consecutive 48-hour monitoring periods was employed in which 2–3 participants were monitored from Monday to Wednesday and another 2–3 from Wednesday to Friday each week. Thus each participant was monitored once. Samples were collected inside and outside the residence and inside the workplace of each participant during the times when he or she was expected to be in that microenvironment during the 48-hour monitoring period. In addition, each participant carried, or kept within arm reach at all times, an aluminum briefcase that contained a personal PM_{2.5} sampling apparatus. The full details have been given in two other reports (8, 14).

Personal samples of PM_{2.5} exposure were collected on two Teflon filters, one during the workday, including commuting, and the other during leisure time, spent usually in and around the residence. Personal PM_{2.5} cyclones (KTL-Cyclone GK2.05, at 4 l/min by BGI Inc, Waltham, MA, USA) and low-flow impactors (EPA Well Impactor Ninety-Six, at 16.7 l/min by BGI Inc) were used as personal exposure monitors and microenvironmental monitors, respectively. Filters (Gelman

Teflo R2PJ037 for personal exposure monitors and R2PJ047 for microenvironmental monitors) were weighed using a Mettler MT5 microbalance (Mettler-Toledo AG, Greifensee, Switzerland) in a weighing room with a relative humidity of 8–48% during the study period. Detailed descriptions of the PM_{2.5} methodology, detection limits, quality assurance, and quality control have been given by Koistinen et al (15) and Hänninen et al (16).

Element and black smoke analysis

An elemental analysis of the PM_{2.5} filters was performed by energy-dispersive X-ray fluorescence (ED-XRF) spectrometry using an X-Lab 2000 (SPECTRO Analytical Instruments, Germany 1998) in the Institute for Mineralogy and Petrography, University of Basel (Switzerland). The X-ray fluorescence device was calibrated for a quantitative elemental analysis with seven commercially available standards. A more-detailed description of the element analysis of the PM_{2.5} samples has been given in Mathys et al (17). The relative standard deviation for the element analysis of duplicate PM_{2.5} samples ranged from 3.4% (calcium) to 4.8% (bromine) (18).

The blackness of the PM_{2.5} on the filters was measured with a reflectometer (EEL model 43, Diffusion Systems Ltd, London, UK). A reflectometer emits light onto the filter and detects the light reflected back from the particles. In this work, reflectance was analyzed according to the method defined in international standard ISO 9835 (19). The protocol of the Organization for Economic Cooperation and Development (20) was followed in the analysis with the exception of that for the particle cut-size and filter material, which were 2.5 µm and Teflon (Gelman Teflo 47mm, 2 µm pore size), respectively, in our work. The expression “black smoke” is used as a synonym for the absorption coefficient defined by the procedures presented in the preceding description.

Source identification

Receptor modeling fundamentals have been reviewed comprehensively by Gordon (21), Cooper & Watson (22) and Henry et al (23). Briefly, receptor models can be any of the following types: chemical mass balance methods or multivariate, microscopic and source-receptor hybrids. Many multivariate methods are based on a factor analysis of individual pollutants or element components to identify underlying patterns that explain common variations among a set of variables, which represent physical properties causing changes in the trace metal concentrations. In our study, principal component analysis was applied to identify sources of microenvironmental and personal PM_{2.5} exposure. A factor analysis

differs from a principal component analysis in that the former is based on the correlation between variables while the latter deals with variances. The linear recombination of eigenvectors in the correlation matrix of element concentrations with a Varimax rotation produced source vectors in the principal component analysis (23). Thus the rotated factors represented major sources or meteorological effects that explained common variations in the element concentrations of the microenvironmental and personal PM_{2.5} samples among the participants of our study.

The Statistical Package for Social Sciences (SPSS Inc, Chicago, IL, USA) version 9.0 for Windows was used for the analysis (Varimax rotation) with natural log-transformed data on element concentration for all the microenvironments. Element concentrations below the detection limit were replaced by half the detection limit. Subsequently, elements with a mean concentration lower than three times the standard deviation of duplicate samples or the maximum detection limit (each sample had its own detection limit) were excluded from the analysis of that particular microenvironment. Principal components with eigenvalues larger than 1 entered the source identification analysis, and absolute coefficients smaller than 0.40 were omitted from the tables to facilitate the identification of PM_{2.5} sources.

Source reconstruction

Specific mass contributions of sources identified in the principal component analysis were calculated using source reconstruction (9, 10, 24). Element concentrations were combined with source fingerprint data from the literature on the chemical composition of particles from each source, and this information was used to compute mass contributions.

Typical elements in soil [aluminum (Al), silicon (Si), calcium (Ca), iron (Fe), titanium (Ti), potassium (K)] were calculated to their common oxides using the approaches and assumptions of Malm et al (10) and Brook et al (9) to estimate the total mass of the soil particles according to the following equation: soil = 2.20 Al + 2.49 Si + 1.63 Ca + 1.58 Fe + 1.94 Ti + 1.41 K.

The contribution of seasalt to the total chlorine concentration was calculated from measured chlorine. According to Ojanen et al (25), 89% of chlorine in urban outdoor air in Helsinki originates from seasalt. The total mass of compounds (eg, chlorine, sodium, sulfate ion, magnesium, calcium, potassium, and bromine) in seasalt were calculated from the seasalt-to-chlorine ratio, which could be estimated from the seasalt-composition reported by Seinfeld & Pandis (26). It must be noted that a considerable amount of the chlorine in seasalt could have been lost from the PM_{2.5} mass due to reactions with nitric acid and sulfuric acid. Thus the

calculation of seasalt using chlorine (Cl) as the reference element produces a minimum estimate for the total seasalt mass. The contribution of seasalt to the ambient PM_{2.5} concentration was estimated according to the following equation: seasalt = 0.89 × 1.816 × Cl.

Inorganic secondary particles (later secondary particles) were calculated as the sum of sulfate, nitrate, and ammonium. Sulfate was calculated from the measured sulfur, and nitrate and ammonium were calculated from the ammonium-to-sulfate and nitrate-to-sulfate ratios taken from the air quality data collected by the Helsinki Metropolitan Area Council from April 1996 to June 1997 (25), a period partially overlapping the period of field work in our study. A large proportion of the ammonium nitrate could have evaporated from the Teflon filters, and, if so, the mass concentrations of nitrate, ammonium, and total PM_{2.5} could have been underestimated. It must be noted that organic secondary particles were also formed in the atmosphere, but they were not included in the category of secondary particles.

The remaining PM_{2.5} mass consisted of primary combustion particles, nonvolatile primary and secondary organic particles, and particles from tire wear, water, and the like (in short, combustion and other particulate matter, which is referred to elsewhere in this paper as CoPM). The CoPM was calculated by subtracting the sum of the contributions from the other major sources identified in the principal component analysis from the total PM_{2.5} mass concentration.

Results

Principal component analysis

The dominant factors for PM_{2.5} mass in each microenvironment and personal exposure were identified, and the contributions of the variations in each factor to the variation in the total element mass were assessed. Tables 1–4 show the rotated component matrices (Varimax rotation) for the microenvironmental and personal PM_{2.5} samples. The values in the tables represent correlations of specific element concentrations with source vectors in a multivariate space, which allows the identification of sources of PM_{2.5} constituents. Not surprisingly, the four factors were often associated with the same elements in different microenvironments and in personal exposure samples. For instance, the first two factors were associated with the same elements in residential outdoor and workplace indoor microenvironments, but the factors seemed to appear in reverse order in residential indoor microenvironments. Several elements (bromine, iron, magnesium, sodium, nickel, and lead)

of the personal exposure samples did not meet the quality assurance goals, probably because the sample volumes were too small. Therefore, they were excluded from the analysis, and the exclusion resulted in a lower resolution of the sources.

Factor 1 of the residential outdoor microenvironments was highly correlated with bromine, aluminum, potassium, and sulfur, and less strongly with black smoke. A similar factor was identified as factor 2 of the residential indoor microenvironments (bromine, sulfur, lead, and black smoke), as factor 1 of the indoor workplace environments (aluminum, calcium, potassium, black smoke, magnesium, bromine and sulfur), and as factor 2 of the personal exposures (black smoke, potassium,

Table 1. Rotated component matrix in the principal component analysis of the PM_{2.5}-bound elements and black smoke in residential outdoor samples (N=101). See table 5 for the definition of the source. (PM_{2.5} = particulate matter with an aerodynamic diameter of < 2.5 µm, * = coefficients -0.4 < x < 0.4)

Element	Factor 1	Factor 2	Factor 3	Factor 4
Aluminum	0.799	*	*	*
Bromine	0.807	*	*	*
Chlorine	*	*	0.888	*
Iron	*	0.852	*	*
Potassium	0.728	0.471	*	*
Magnesium	*	*	0.669	*
Sodium	*	*	0.781	*
Lead	*	0.677	*	*
Sulfur	0.720	*	*	-0.476
Silicon	*	*	*	0.909
Zinc	*	0.915	*	*
Black smoke	0.545	0.563	*	*
Source	A	B	C	E

Table 2. Rotated component matrix in the principal component analysis of the PM_{2.5}-bound elements and black smoke in indoor samples for residences without environmental tobacco smoke (N=102). See table 5 for the definition of the source. (PM_{2.5} = particulate matter with an aerodynamic diameter of < 2.5 µm, * = coefficients -0.4 < x < 0.4)

Element	Factor 1	Factor 2	Factor 3	Factor 4
Aluminum	*	*	0.852	*
Bromine	*	0.819	*	*
Calcium	*	*	0.771	*
Chlorine	-0.446	*	*	0.648
Iron	0.696	*	*	*
Potassium	0.504	*	*	0.591
Phosphorus	*	*	*	0.813
Lead	*	0.678	*	*
Sulfur	*	0.749	*	*
Silicon	*	*	0.79	*
Zinc	0.813	*	*	*
Black smoke	0.695	0.436	*	*
Source	B	A	E	D

Table 3. Rotated component matrix in the principal component analysis of the PM_{2.5}-bound elements and black smoke in indoor samples for workplaces without environmental tobacco smoke (N=69). See table 5 for the definition of the source. (PM_{2.5} = particulate with an aerodynamic diameter of < 2.5 µm, * = coefficients -0.4 < x < 0.4)

Element	Factor 1	Factor 2	Factor 3	Factor 4
Aluminum	0.779	*	*	*
Bromine	0.535	*	*	*
Calcium	0.652	*	*	0.410
Chlorine	*	*	0.645	*
Copper	*	0.739	*	*
Iron	*	0.765	*	*
Potassium	0.635	*	*	*
Magnesium	0.601	*	*	*
Nickel	*	0.637	*	*
Phosphorus	*	*	0.773	*
Lead	*	*	*	-0.787
Sulfur	0.459	*	-0.753	*
Silicon	*	*	0.450	0.679
Zinc	*	0.850	*	*
Black smoke	0.615	0.489	*	*
Source	A	B	D	E

Table 4. Rotated component matrix in the principal component analysis of PM_{2.5}-bound elements and black smoke in 48-hour personal exposure samples without environmental tobacco smoke (N=76). See table 5 for the definition of the source. (PM_{2.5} = particulate matter with an aerodynamic diameter of < 2.5 µm, * = coefficients -0.4 < x < 0.4)

Element	Factor 1	Factor 2	Factor 3	Factor 4
Aluminum	0.841	*	*	*
Calcium	0.797	*	*	*
Chlorine	*	*	0.660	*
Copper	*	*	*	0.820
Potassium	*	0.768	*	*
Manganese	0.452	0.552	*	*
Phosphorus	*	*	0.851	*
Sulfur	*	0.710	-0.543	*
Silicon	0.825	*	*	*
Zinc	*	*	*	0.718
Black smoke	*	0.855	*	*
Source	E	A	D	B

sulfur, and magnesium). Obviously, there was a common source, denoted as "A", for these factors (tables 1–4).

Factor 2 of the residential outdoor microenvironments was highly correlated with zinc, iron, and lead, and it had a weaker correlation with black smoke and potassium. A similar factor was identified as factor 1 of the residential indoor microenvironments (zinc, iron, black smoke, potassium with a negative correlation with chlorine), as factor 2 of the workplace indoor microenvironments (zinc, iron, copper, nickel, and black smoke), and as factor 4 of personal exposure (copper, zinc). This source was denoted as "B" (tables 1–4).

Chlorine, sodium, and magnesium were highly correlated with factor 3 of the residential outdoor microenvironments (denoted "C"). In the other microenvironments and personal exposures, chlorine was associated with a different factor: factor 4 of the residential indoor microenvironments (phosphorus, chlorine, and potassium), factor 3 of the workplace indoor microenvironments (phosphorus, chlorine, and silicon with a negative correlation with sulfur), and factor 3 of personal exposures (phosphorus, chlorine and a negative correlation with sulfur). The respective phosphorus–chlorine source was denoted as "D" (tables 1–4).

Factor 4 of the residential outdoor microenvironments was strongly correlated with silicon and had a weak negative correlation with sulfur. A factor with silicon was identified as factor 3 of the residential indoor microenvironments (aluminum, silicon, and calcium), as factor 4 of the indoor workplace environments (silicon, calcium, and a negative correlation with lead), and as factor 1 of personal exposures (aluminum, calcium, and silicon). The respective source was denoted as "E" (tables 1–4).

Table 5 summarizes all the factors for all the microenvironments. This summary helps to identify and locate the main sources of PM_{2.5} constituents and their respective contributions to the exposure variations. It shows semi-quantitatively the contribution of each emission source to the total element or black smoke variation in each microenvironment. The total variances explained by the four factors for residential outdoor, residential indoor, and workplace indoor microenvironments and personal exposures were 75%, 73%, 65%, and 71%, respectively. These results show that the variances could be largely explained by these four sources. On the other hand, still a significant portion of the percentage of explained variance could not be explained by these sources, and this phenomenon should be investigated in more detail in forthcoming studies. Source A represented long-range transported secondary and primary particles, source B covered other combustion sources (mainly traffic in Helsinki), source C referred to sea and de-icing salt in outdoor air, source D represented indoor aerosol from cleaning agents and possibly cooking, and source E was soil mineral dust from indoor and outdoor sources.

Indoor-to-outdoor ratios

Table 6 shows the median elemental indoor-to-outdoor ratios for residences free of environmental tobacco smoke. The respective median ratios of the total PM_{2.5} mass concentrations and black smoke are given for comparison. The median ratios for lead, sulfur, bromine, and black smoke were approximately 0.6–0.7 for winter and spring, and generally 0.9–1 for summer and 0.7–0.9 for fall. Interestingly, these elements were associated with

Table 5. Comparison of the main sources of PM_{2.5}-bound elements and black smoke identified in a principal component analysis for residential outdoor (RO), residential indoor (RI), and workplace indoor (WI) microenvironments and 48-hour personal exposures (P48). (A = long-range transported secondary and primary PM, B = other combustion sources (mainly traffic in Helsinki), C = seasalt, D = indoor aerosol from cleaning agents and possibly cooking, E = soil mineral dust from indoor and outdoor sources, + = loading > 0.4 in a particular microenvironment or personal exposure, o = loading in 3 out of the 4 microenvironments or exposure, += loading in all the microenvironments and exposure, - = loading < 0.4 in a particular microenvironment or personal exposure, · = elemental data not included in the analysis or factor not present, PM_{2.5} = particulate matter with an aerodynamic diameter of < 2.5 µm)

Element	Source A				Source B				Source C				Source D				Source E			
	RO	RI	WI	P48																
Aluminum	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	+
Bromine	o	o	o	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Calcium	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	o	o	o
Chlorine	-	-	-	-	-	+	-	-	+	-	-	-	-	o	o	o	-	-	-	-
Copper	-	-	-	-	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-
Iron	-	-	-	-	o	o	o	-	-	-	-	-	-	-	-	-	-	-	-	-
Potassium	o	-	o	o	+	+	-	-	-	-	-	-	-	+	-	-	-	-	-	-
Magnesium	-	-	+	-	-	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-
Manganese	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+
Sodium	-	-	-	-	-	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-
Nickel	-	-	-	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
Phosphorus	-	-	-	-	-	-	-	-	-	-	-	-	-	o	o	o	-	-	-	-
Lead	-	+	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+
Sulfur	+	+	+	+	-	-	-	-	-	-	-	-	-	-	+	+	+	+	-	-
Silicon	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	+	+	+	+
Zinc	-	-	-	-	+	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-
Black smoke	+	+	+	+	o	o	o	-	-	-	-	-	-	-	-	-	-	-	-	-
Variation in micro-environment explained by the source (%)	24	19	21	21	23	21	19	12	16	-	-	-	-	14	15	16	12	19	10	22

sources A and B in the principal component analysis, and therefore indicated long-range transport and local combustion sources. The penetration of these elements indoors closely mirrored the length of time that windows were kept open in the residences during different seasons. According to the data collected by questionnaires, windows were open almost all the time during the 48-hour study period in summer, but for only about an hour in winter. The indoor-to-outdoor ratios for zinc and iron were remarkably low (0.5–0.6) for winter and clearly higher (mostly 0.7–1) for spring, summer, and fall; this result also indicates mainly outdoor sources. None of the median indoor-to-outdoor ratios for the aforementioned elements was over 1 for any season. The median ratios for the remaining elements (potassium, chlorine, and aluminum), all with significant indoor sources, exceeded 1 nearly throughout the year, the ratios being highest in fall, followed by summer.

Source reconstruction

Figure 1 shows the contribution of each identified source to the microenvironmental and personal PM_{2.5} concentrations calculated by mass reconstruction. These sources

Table 6. Median seasonal indoor-to-outdoor ratios of the PM_{2.5}-bound elements (N = 92) and black smoke (N = 78) and the total PM_{2.5} mass concentration (N = 92) for the residences of participants not exposed to environmental tobacco smoke. (PM_{2.5} = particulate matter with an aerodynamic diameter of < 2.5 µm)

	Winter	Spring	Summer	Fall	Total
Lead	0.67	0.56	0.86	0.69	0.67
Sulfur	0.60	0.63	0.90	0.75	0.69
Bromine	0.57	0.72	0.98	0.89	0.77
Black smoke	0.65	0.67	0.91	0.88	0.79
Zinc	0.58	0.75	0.66	0.75	0.68
Iron	0.52	0.96	0.90	0.95	0.83
Potassium	0.95	1.05	1.01	1.08	1.05
Chlorine	1.01	1.24	1.37	1.74	1.24
Aluminum	1.19	1.08	1.41	2.20	1.27
PM _{2.5}	0.77	1.03	0.95	0.92	0.92

were not exactly equal to sources A–E, which were identified by the principal component analysis. For example, source A (long-range transport) of the analysis contained both primary combustion particles and secondary particles, which are two separate sources in figures 1 and 2. Figure 2 shows the clear dependency of source A on easterly winds.

The indoor-to-outdoor ratios higher than 1 (table 6) suggested an indoor source for chlorine in addition to

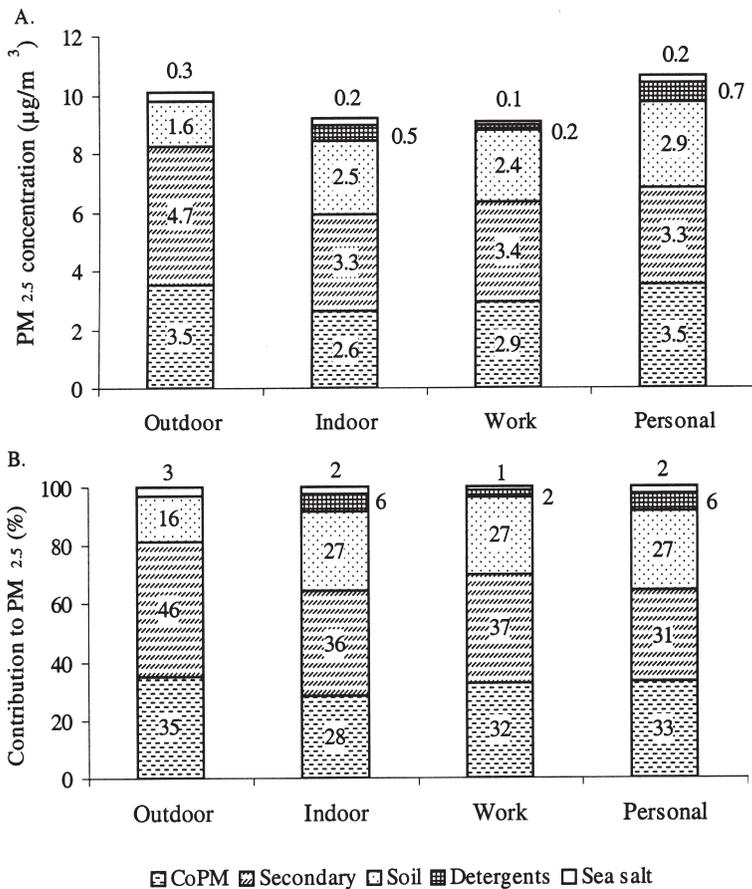


Figure 1. Absolute (A) and percentage (B) contribution of sources to the PM_{2.5} concentrations in residential outdoor, residential indoor, and indoor workplace microenvironments and in personal 48-hour exposures of the participants not exposed to environmental tobacco smoke. (PM_{2.5} = particulate matter with an aerodynamic diameter of <2.5 µm)

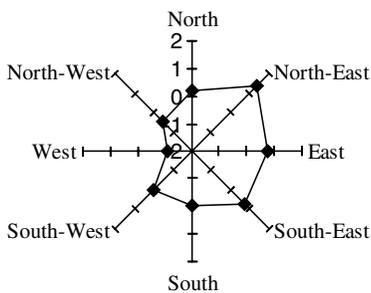


Figure 2. The wind rose for the average factor scores of long-range transport in residential outdoor microenvironments (factor 1).

seasalt. For the residential indoor and workplace indoor microenvironments and personal exposures, marine chlorine was separated from the indoor source. Using the indoor-to-outdoor ratio for summer as reference (for the best estimate of marine and indoor sources without interference from air filtration), we multiplied the chlorine concentrations by a factor of 0.73 (1/1.37) to get an estimate of marine chlorine in these microenvironments for source reconstruction.

In the principal component analysis, an additional phosphorus-chlorine source was identified in the residential indoor and workplace indoor microenvironments and in the personal exposures to PM_{2.5}. It was probably

from cleaning products or detergents. For simplicity, the contribution of this source was calculated as phosphoric anhydride (a common oxide for phosphorus, exact identification of the compound not possible). The total (unevaporated) PM_{2.5} mass concentrations in residential outdoor, residential indoor, and workplace indoor microenvironments and personal 48-hour exposures of the participants not exposed to environmental tobacco smoke were 10 µg/m³, 9.2 µg/m³, 9.1 µg/m³, and 10.6 µg/m³, respectively (figure 1).

Discussion

In our study, we confined the principal component analysis to microenvironments in which environmental tobacco smoke was not present in order to focus on the other factors contributing to the PM_{2.5} concentrations. It should be remembered that, for those exposed to environmental tobacco smoke, the dominant source of PM_{2.5} was tobacco smoke.

The indoor and outdoor microenvironmental PM_{2.5} concentrations were measured at fixed sites, inside and outside of residences and at the workplace, which were the most relevant environments for the participants'

personal exposures. While all the subjects spent an overwhelming majority of their time in these microenvironments, some of them may have acquired a quantitatively or qualitatively significant portion of their exposure elsewhere (eg, in transport between home and work). The personal exposure samples reflected the combination of all PM_{2.5} sources encountered by the participants in all the microenvironments entered during the 48-hour monitoring period.

The central role of black smoke in the principal component analysis indicated that sources A and B were connected to combustion processes, while the three other sources (C, D, and E) were unrelated to combustion.

Identification of sources of fine particulate matter

The strongest source (A) represented long-range transported, mainly secondary, aerosols loaded by sulfur, but there were also primary combustion particles loaded by black smoke from wood burning (potassium) and traffic (table 5). It showed a clear dependency on easterly winds (figure 2), and this dependency could hardly be caused by differences in traffic patterns within Helsinki, as the measurements were made outside of the participant's residences, which were randomly spread over the Helsinki metropolitan area. In addition, the dependency on wind direction did not result from a small number of samples with high concentrations. The plausible sources of these long-range transported pollutants could be Saint Petersburg in Russia, 300 kilometers east of Helsinki, and the large power plants fired by oil shale in Narva, Estonia, about 100 kilometers southeast of Helsinki. In 1996, the Narva region was responsible for 78%, 72%, and 68% of the total particulate matter, sulfur dioxide, and nitrogen oxide emissions in Estonia. [See the Estonian environmental home page, 1997, of the Estonian Ministry of Environment: available at <http://www.envir.ee/ehp/ohku96.htm> (accessed 27 February 2002).]

Source B represented local combustion emissions, mostly from traffic, because lead, iron, zinc, and black smoke are traditionally linked with gasoline and diesel-powered engine emissions (table 5). It is not surprising that particulate emissions from diesel and gasoline combustion appeared in the same factor, as these vehicle types are highly associated with each other when compared with other sources. The only other significant local combustion sources in Helsinki are large power plants generating district heat with high-efficiency flue gas cleaning and high stacks. Source B was likely to represent all combustion emissions within the Helsinki metropolitan area, as the factor scores were independent of wind direction. In the workplace indoor microenvironments, also copper and nickel were associated with source B, which was probably due to additional sources from traffic, which were occupational or more

predominant during daytime hours. These elements were not associated with source B in residential outdoor or indoor microenvironments, but copper was associated with source B also in personal exposures. Because iron and lead were not included in the principal component analysis of the personal PM_{2.5} samples, the contribution of traffic emissions to source B may have become somewhat underestimated in the personal exposures.

Source C could be distinguished only in outdoor air. Its association with chlorine, sodium, and magnesium clearly identified salt from sea spray aerosols in the Baltic Sea, de-icing of roads, or both (table 5) (25, 27). Willison et al (28) have reported that salt in samples of particulate matter at least 100 kilometers from the coastline in England originated mainly from the sea and contributed 10–20% and 2–3% to the mass concentrations of coarse and fine particulate matter, respectively. Conditions in Finland, however, differ from those in England during winter months, because water in the Baltic Sea around Helsinki has a lower salt content than the Atlantic Ocean and the sea is frozen over from approximately the end of December through the end of March. Moreover, much more salt is spread on roads in Finland than in England during the winter.

Source D, loaded with phosphorus and chlorine, could only be seen in residential indoor and workplace indoor microenvironments and in personal exposures (table 5). These elements are related to biological organic material, especially to washing powders, cleaning agents and disinfectants (29). The similarity of source D in the residential indoor and workplace indoor microenvironments and in personal exposures suggested the same cleaning product source. Phosphorus was not found in residential outdoor air, and the indoor-to-outdoor ratios for chlorine in homes exceeded 1 during all seasons and therefore indicated a strong indoor source for this factor. Surprisingly, phosphorus levels were 10 times higher in residential indoor microenvironments in the fall than in other seasons (data not shown), while the use of cleaning agents and detergents is not expected to vary so much on a population level. The association of chlorine with source D implied cleaning product sources, as the chlorine concentrations were higher in residential and workplace indoor microenvironments than in residential outdoor air. The median personal exposure concentrations of phosphorus were 5–10 times higher than the residential indoor concentrations in the winter, spring and summer, but the two concentrations were similar in the fall (data not shown). This finding suggests that phosphorus originated mainly from a source strongly related to human behavior, such as cleaning products, disinfectants, or personal hygiene products.

Silicon was clearly the marker of source E, which represented mineral dusts from outdoor (wind, traffic)

and indoor (floor dust) sources. Silicon is a characteristic tracer for the high silica soil present in the Helsinki metropolitan area (25, 30, 31), and therefore this factor is likely to be related to resuspended soil particles from either human activities (traffic) or wind. In the residential and indoor workplace microenvironments and personal exposures, source E was also associated with calcium, a finding indicating indoor resuspension of tracked-in soil dust and probably also dust from calcium-rich building materials. For an unknown reason iron did not appear in source E as expected.

Indoor-to-outdoor ratios

The seasonal indoor-to-outdoor ratios for the total PM_{2.5} mass concentration were close to unity, except for winter. However, this result seemed to be an oversimplification, as the elemental indoor-to-outdoor ratios varied considerably, depending on the season (table 6). These seasonal variations indicated differences in source emissions or their penetration into the indoor environments via ventilation. As expected from the questionnaire data on seasonal differences in open windows, the indoor-to-outdoor ratios for elements closely associated with source A "long-range transport" (sulfur, bromine, and black smoke) were approximately 0.6–0.7 for winter and spring and approached 1 for summer. Interestingly, a different pattern was observed for the ratios for elements closely associated with source B "traffic" in indoor and outdoor microenvironments (zinc, iron). These elements showed a remarkable similarity in their ratios for spring, summer and fall, but the ratios were much lower for winter. The corresponding ratios for the other elements (aluminum, chlorine, and potassium) were close to or exceeded 1 (sometimes considerably so) for all the seasons and therefore pointed to emission contributions from indoor sources. One significant indoor source may well have arisen through the resuspension of dust and soil particles tracked in from outdoors and deposited on floors and other indoor surfaces.

Thus the residential building structures in northern latitudes tend to restrict, especially in winter, the penetration of particles from outdoor sources (mainly distant and local combustion processes) into indoor environments. This finding agrees with the conclusions of Sarnat et al (32) and Long et al (33) in the cities of Baltimore and Boston in the United States. However, the restricted penetration of elements from outdoor sources was overshadowed by the simultaneously increased emissions of soil and dust particles from indoor sources.

Source reconstruction

The main contributors to PM_{2.5} in residential and work microenvironments were inorganic secondary particles

and CoPM (figure 1). Their relative contributions in an urban background station (Vallila) in Helsinki were 43% and 46%, respectively, in a previous study by Ojanen et al (25), whose findings agree well with the results of our current study on residential outdoor microenvironments (46% and 35%, respectively). Similar source contributions to fine particles in ambient air have been reported by Schauer et al (34) for downtown Los Angeles in southern California. In their study, secondary particles, diesel and gasoline powered automobiles, soil, industry, and wood burning contributed 40%, 39%, 10%, 6%, and 5% to the PM_{2.5} mass concentration, respectively. Moreover, vehicle emissions and secondary aerosols have been the dominant source categories for PM_{2.5} in Birmingham in the United Kingdom, as reported by Harrison et al (35). ApSimon et al (36) modeled the atmospheric transport of primary PM_{2.5} across Europe and obtained 2 µg/m³ for the Helsinki area. If this is a valid estimate for the current study period (1996–1997), 43% of the CoPM in PM_{2.5} in residential outdoor air would have come from local sources. Our results also agree with those based on virtual impactor data reported by Pakkanen et al (37) from Helsinki. They found that 46% of the PM_{2.3} originated from local sources, mostly traffic (25). Similarly, traffic was an important source for CoPM, identified by a principal component analysis in our study. The seasalt contribution to outdoor PM_{2.5} in our study also agreed with the results of Ojanen et al (25), being 3% in both studies.

The contribution of soil to the PM_{2.5} mass concentration in residential outdoor microenvironments (16%) was twice as high as that (8%) reported by Ojanen et al (25). The higher soil contributions in our study were probably due to sampling close to people's residences instead of sampling at a central fixed site. The highest soil contributions to outdoor PM_{2.5} occurred in the spring due to the "spring dust episodes" (38, 39). These episodes take place typically during a short period when snow and ice have melted and left behind large quantities of finely ground sand, which is spread on the roads in winter and resuspended by wind and traffic.

The high soil contribution to the PM_{2.5} concentration in residential and indoor workplace microenvironments, combined with the relatively high indoor-to-outdoor ratio for the soil tracer aluminum, indicated the presence of a substantial indoor source, as discussed earlier (table 6, figure 1). The source contributions to workplaces were similar to those of the residential indoor microenvironments, as the majority of participants worked in offices and services. The only exception was that the average contribution of CoPM was somewhat higher in workplaces, probably due to a higher and closer traffic contribution in the workplaces during the daytime measurements, while the residential indoor microenvironments were monitored in the evenings and at night.

Additional occupational sources were present in some manufacturing workplaces, but the number of participants working in such an environment was too low to affect the results at the population level (14). It should be noted, however, that certain occupational sources (ie, vehicle emissions in garages, metal industry processes) could have made very significant contributions to the PM_{2.5} exposures of workers.

Similar sources of PM_{2.5} in different microenvironments and the appearance of these sources in personal exposures were expected, as the personal exposure samples represented the summed exposures in all the microenvironments in which the participants spent their time. Particles from outdoor sources penetrate indoors (7, 33, 40), and therefore these sources are also seen in residential and workplace indoor microenvironments. In addition, many of the same consumer and cleaning products are used in both residences and workplaces.

Most strikingly, the contributions of both CoPM and long-range transport to the PM_{2.5} concentrations of residential and workplace indoor exposures and personal exposures were up to 35% lower than in residential outdoor air. Thus the estimation of population exposures to PM_{2.5} of outdoor origins on the basis of mere outdoor data would have resulted in an overestimation of these sources in Helsinki. The contributions of CoPM and secondary particles to personal PM_{2.5} exposures were highest in summer, probably due to increased penetration indoors and longer times spent outdoors (data not shown).

Concluding remarks

Our study of an adult urban population not exposed to tobacco smoke produced several important findings. First, inorganic secondary particles, particles from combustion processes, and soil dust dominated the source types for the PM_{2.5} mass concentration in all the microenvironments and personal exposures. Second, the indoor-to-outdoor ratios of elemental concentrations in the PM_{2.5} depended on the source and varied between seasons due to widely differing ventilation characteristics in residences in the northern latitudes, although the indoor-to-outdoor ratios for the total PM_{2.5} mass concentration were close to unity. Third, the analysis of the indoor-to-outdoor ratios, combined with a principal component analysis and source reconstruction, indicated the presence of substantial residential indoor and workplace indoor sources from the resuspension of soil dust particles tracked into indoor environments and probably cleaning associated with the use of phosphorus- and chlorine-containing products. Fourth, the contributions of different sources to personal PM_{2.5} exposures were the most closely approximated by data from residential and workplace indoor microenvironments. Finally, the population exposure assessment of PM_{2.5} based on outdoor

fixed-site monitoring overestimated exposures to outdoor sources like traffic and long-range transport, and did not account for the contribution of significant indoor sources. Thus the concentration-response functions with outdoor fixed-site data may underestimate the true association between personal exposures to PM_{2.5} of outdoor origins and respective health effects.

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