DISTRIBUTION OF AIRBORNE PARTICLES FROM MULTI-EMISSION SOURCE

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Abstract. The purpose of this work was to study the distribution of airborne particles in the surroundings of an iron and steel factory in southern Finland. Several sources of particulate emissions are lying side by side, causing heavy dust loading to the environment. This complicated multi-pollutant situation was studied mainly by SEM/EDX methodology. Particles accumulated on Scots pine bark were identified and quantitatively measured according to their element content, size and shape. As a result, distribution maps of particulate elements were drawn and the amount of different particle types along the study lines was plotted. Particulate emissions from the industrial or energy production processes were not the main dust source. Most emissions were produced from the clinker crusher. Numerous stockpiles of the industrial wastes and raw materials also gave rise to particulate emissions as a result of wind erosion. It was concluded that SEM/EDX methodology is a useful tool for studying the distribution of particulate pollutants.

Keywords: air pollutants; bark particles, biomonitoring, SEM/EDX, steel industry

1. Introduction

The distribution and accumulation of particulate pollutants are presently an object of intensive research (Kouimtzis and Samara, 1995; Hinds, 1999). Airborne dust causes serious health effects and other problems in urban and industrial environments. In many cases several sources simultaneously produce different emissions including different kinds of particles. Therefore it is not easy to study the role of each pollutant, and there are not many good methods to identify and quantitatively measure different particulate pollutants and to locate their origin and distribution in the environment. It is generally known that steel plant particulate emissions are mainly deposited near the plant (Pilegaard, 1979; Vestergaard *et al*., 1986; Ettala *et al*., 1986; Mukherjee and Nuorteva, 1994; Türkan *et al*., 1995). The investigations have usually been performed by measuring the distribution of iron and other metal elements in the surroundings of the plants. Similar studies have been done also around the steel plant investigated in this work (reviewed by Monni and Mäkinen, 1995), including environmental impacts on pine forests and epiphytic lichens (Holmberg and Pihlström, 1992), on soil and pine bark (Fritze, 1991), on element

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content of forest mosses (Rinne and Mäkinen, 1988) and lichens (Holmberg and Pihlström, 1992), as well as soil microbial response (Fritze, 1991; Bååth *et al.*, 1992; Fritze and Bååth, 1993) and response of epiphytic microflora of pine needles (Pasanen and Fritze, 1992).

This work was performed under multi-pollutant industrial conditions, characterized by several different particle emission sources. The aim of the research was to identify different types of particles and to measure their amount as well as to study their origin and distribution. New methodological applications (Haapala, 1998) were used and their fitness for this kind of research was tested.

2. Materials and Methods

The research was performed in the surroundings of the iron and steel plant Koverhar (Rautaruukki Ltd) in Hankoniemi, on the coast of southern Finland. Scots pine (*Pinus sylvestris* [L.]) forests on sandy and relatively even ground are characteristic for the study area, which is bordered on the east by a shallow, brackish arm of the Gulf of Finland.

Total steel production at the plant in 1998 was 550 000 tons. Total particle emissions from industrial processes and energy production were 250 tons, containing 8.2 tons of Zn and 1.8 tons of Pb. Annual production of blast furnace clinker was 115 000 tons and converter clinker 106 000 tons (in 1997). These clinkers give rise to considerable dust emissions since they are used in a clinker crusher, which produces great quantities of dust without any filtering. Numerous stockpiles are stored on the industrial fields, including those of clinker, different ash materials, waste from the desulfurization process (gypsum), as well as piles of raw materials such as iron pellets, Ca-minerals, and coke. Handling and transportation of those materials as well as wind erosion produce airborne particles. In wintertime the snow is covered by a thin layer of grey dust, visible over an area of about one square kilometer. The ground and trunks of trees are colored in many places by red (iron-bearing) or gray dust.

The fieldwork was performed during the winter of 1998. Pine bark samples were collected from 23 plots (Figure 1). The samples were located on four lines, reaching (1) 2800 m to N-NE from the plant, (2) 1250 m to (SE), (3) 1600 m to NW, and (4) 800 m to W. The shoreline of the Gulf lies about 800 east from the plant going in south-north direction. Small pieces (about 3 cm^2) of bark were cut in each plot from three contiguous trees on the side facing the factory at about a 1.5 m height.

The samples were air-dried in paper bags, then mounted on sample stubs and coated with gold by a Jeol Fine Coat Ion Sputter JFC-1100 coating unit. The preparations were examined with a Jeol JSM-840 scanning electron microscope (SEM) and a Jeol Semaphore image-recording system. Analyses were performed directly in the SEM by the PGT Imix EDX microanalyzer. The voltage for the en-

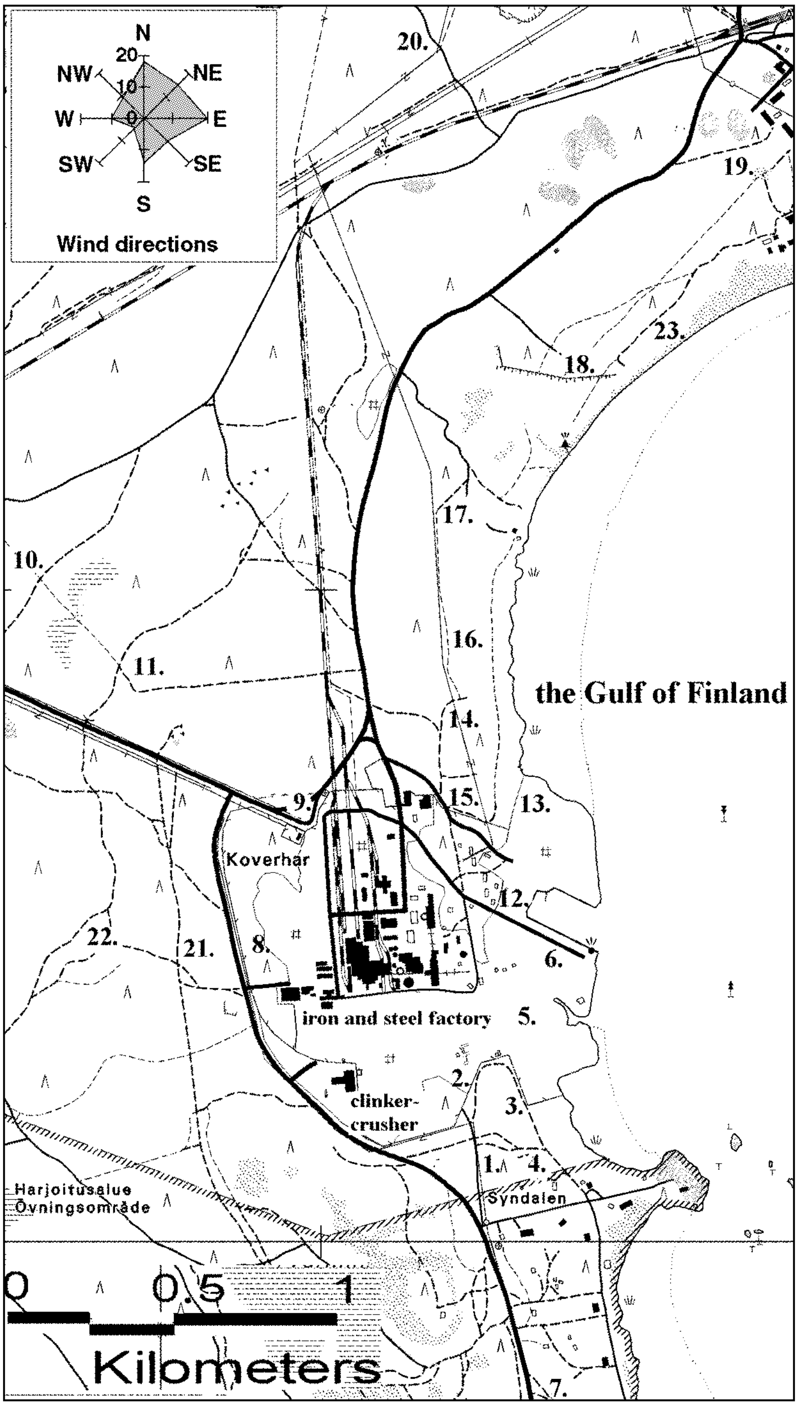

Figure 1. Sample spots and wind directions in the study area.

ergy dispersive X-ray (EDX) analysis was 15 kV, and the time of X-ray collection was 60 s.

To measure the total element content on the bark surface, two randomly selected points of four bark pieces (eight points for each sample plot) were studied by SEM/EDX with a 50-fold magnification (see Haapala 1998) scanning the whole SEM-area for X-ray collection. Concentrations of Ca, Mg, Si, Al, K, Fe, and Na were measured as mean values of 8 points. Ca-concentrations are given as grams per square meter of pine bark (Figure 2) after standardization with reference preparations. For the concentrations of other elements, arbitrary 'EDX units' are used, since the reference materials were not developed. For the identification of different particle types and for the measurement of their shares, 10 randomly selected single particles of each four bark pieces (40 particles for each sample plot) were recorded according to their size, shape and element content with the SEM/EDX methods used by Koponen (1994), Juhanoja and Meinander (1994) and Meinander (1994). These measurements made possible the source apportionment of the particles.

For the identification, original particles from different emission sources were used as a reference material ('fingerprints') after being studied by the same methodology. The element composition of single particles was also compared with the information attained from literature and from the laboratory of the Koverhar steel plant, where the chemical composition of waste and raw materials was analyzed. Using all this information, we divided the particles into 10 different particle types. At the same time, pollutant distribution was measured by the moss-bag method (SFS 1994), but the results of these studies are only briefly reported here.

The Kruskal-Wallis one-way analysis of variance method was used to study the differences in element content between the sample plots. The correlation of different elements in the particles was calculated by the Spearman Rank method. To help the identification of different particle types, principal component analysis (eigenvalues) of each sample plot was performed. Statistix version 1.0 was used for the statistical calculations; Surfer 5.00 program was used to produce distribution maps for particulate emissions; and MS Excel 5.0 was used for plotting the different particle types in sample plots.

3. Results and Discussion

The study area is heavily polluted by dust particles. The dust deposition during two months in February–March 1998 was investigated by the moss-bag method (SFS, 1994). In the two most polluted plots, the ash content of the moss-bags was 73 and 43%, which was almost totally made up of airborne particulates. Iron accumulation in the moss-bags was maximally 49 000 mg kg^{-1} month^{-1} and increase in Ca-concentration 30 000 mg kg^{-1} month^{-1}, respectively. Many bags were covered by a thick dust layer. Particle deposition is strongly dominated by local emissions, the accumulated element concentrations being much higher than in many similar

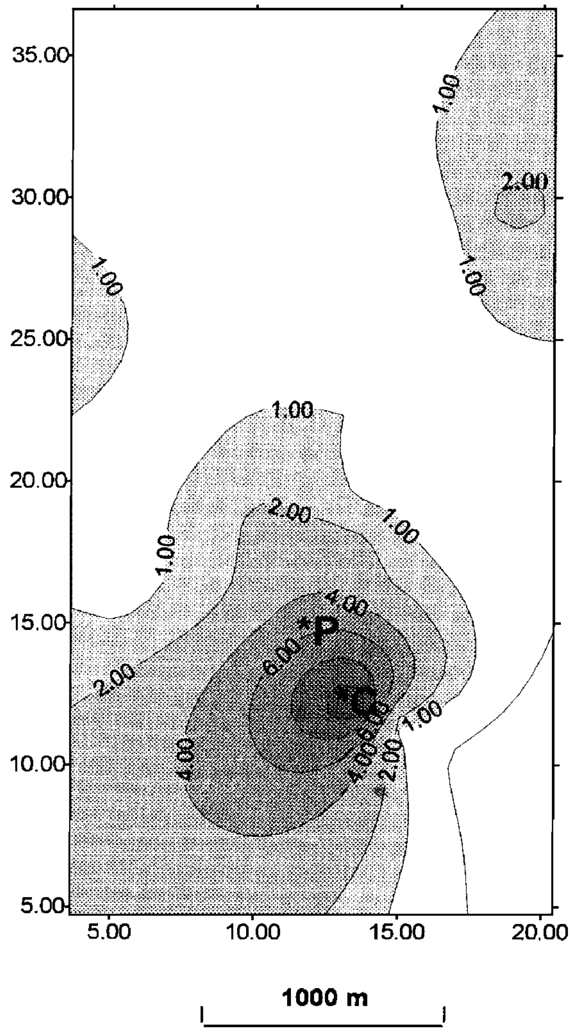

Figure 2. Distribution map for calcium in the surroundings of the steel plant. Contents on pine bark surface as 'EDX units': 1 EDX unit = 109 mg m^{-2}, P = steel plant, C = clinker-crusher.

investigations (Vestergaard *et al.*, 1986; Mukherjee and Nuorteva, 1994; Türkan *et al.*, 1995; Vasconcelos and Tavares, 1998). Local dust emissions have an alkalizing impact on the environment. Soil pH of the acid sandy podzol has risen from 4.1 to 6.6 during years 1961–1989 (Fritze, 1991).

3.1. Distribution of particulate elements

Distribution maps of element content on pine bark surface are presented in Figures 2, 3 and 4. For some elements the distribution curves are extended in a southwesterly direction from the steel plant. This can be seen for calcium in Figure 2,

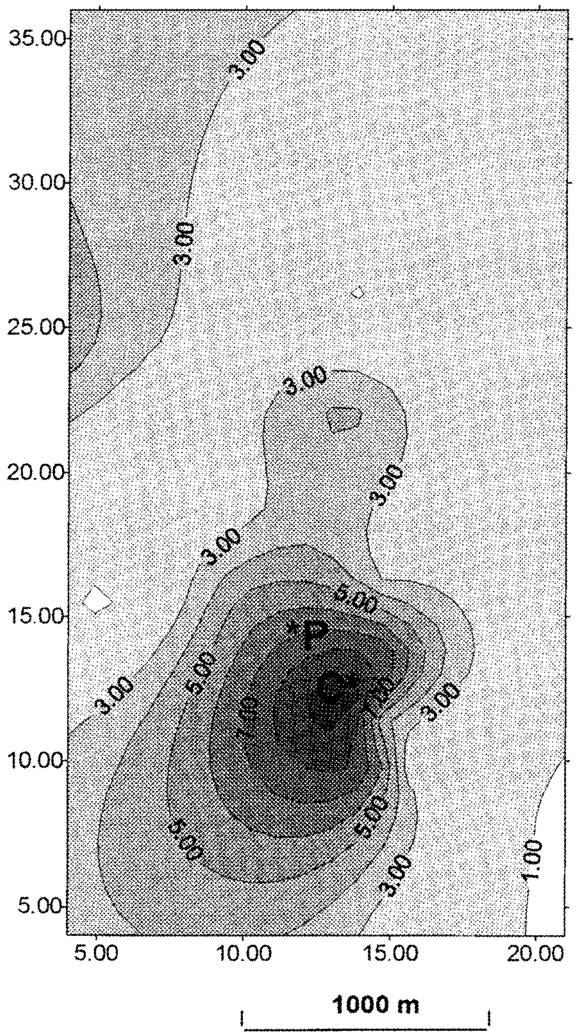

Figure 3. Distribution map for silicon in the surroundings of the steel plant (for further explanations see Figure 2).

but it was similarly clear for two other abundant pollutant elements: silicon and aluminum.

This result was unexpected, because the winds most often blow in the opposite direction (Ilmatieteen laitos, 1996; Karlsson, 1997; Pokki, 1998) and only rarely to the southwest. We suppose that the position of the clinker crusher as well as local topography give rise to this kind of pollutant distribution. The clinker crusher is superior to all other sources in producing Si-Al-Ca-containing dust. It is located on the southwestern side in the factory area. Due to big steel plant buildings and other

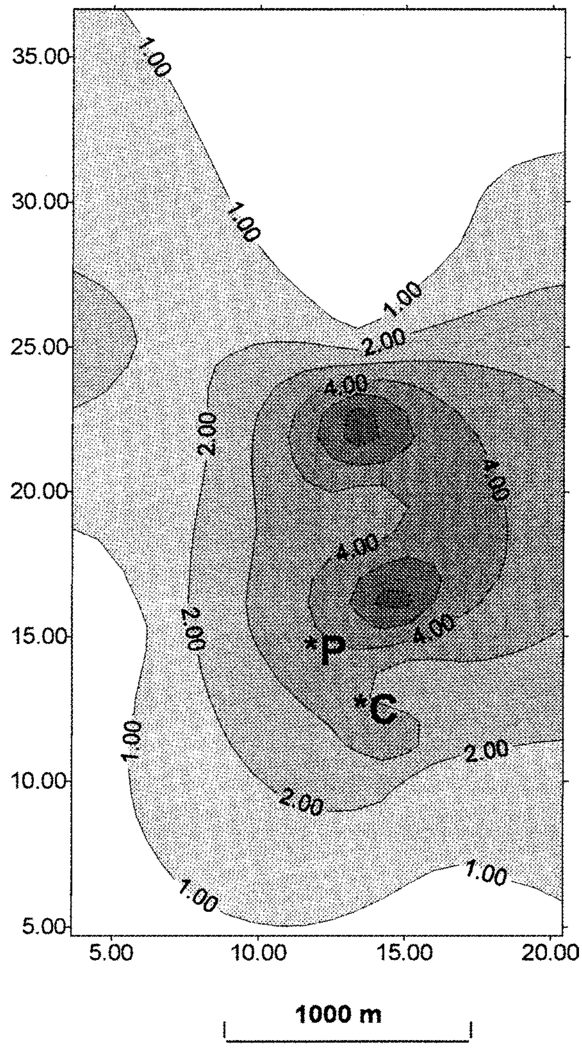

Figure 4. Distribution map for iron in the surroundings of the steel plant (for further explanations see Figure 2).

topographic hindrances like stockpiles, this dust cannot easily go to the northeast or any direction except southwest.

However, a smaller extension of distribution to the northeast was detected in the case of Si (Figure 3) and Al. These elements are included in many pollutants that are emitted from other sources than the clinker crusher and may thus be distributed in a different way. The Ca/Si-ratio was 1.22 near the crusher and 0.23 at about 940 m northeast of the steel plant and 1260 m northeast of the crusher, which means that silicon is proportionally distributed more in that direction. In fact, silicon showed three dispersion extensions, as did magnesium, which is also a lesser component of many emissions. It must also be remembered that the particle size may vary among different emissions and has an impact on the distribution distance.

A different distribution pattern was observed for iron. The curves of the iron map (Figure 4) are strongly extended to the east and east-northeast. This is near the main wind direction. Iron-containing particles are emitted from many sources, but mainly from the end of the conveyer along which iron pellets are transported from the harbor to the factory. The trees and ground near the conveyer termination are strongly colored red by iron dust. Iron-containing particles are also emitted directly from the processes and from some stockpiles. On the other hand, the dust from the clinker crusher contains very little iron. The conveyer termination lies on the northeastern side of the factory. Additionally, a stockpile of converter filter dust (containing 50% iron) is situated 600 m northeast from the plant, thus giving rise to the red-brown colour of its nearest surroundings.

Sulfur showed a unique distribution pattern. As a rule, only small amounts of sulfur were measured on the pine bark surfaces. However, a lot of sulfur was found on pine bark in sample plot 5, near the waste pile of the desulfurization process (limestone-gypsum). It was evident that gypsum-containing particles are blown by the wind from this pile and they are deposited nearby. Sulfur-containing emissions are also produced from the power plant as a sulfur dioxide, but it is in a gas form and not deposited much in the research area.

3.2. Identification of particles

3.2.1. *Correlation of Elements*

Correlation analysis was used to distinguish different particle types. Correlation of elements in 40 particles on the surface of pine bark were calculated separately for every sample plot. It was supposed that the elements which had a significant correlation probably existed in the same particles and vice versa. However, elements existing together in one type may exist separately in other particle types. Therefore the interpretation of the correlation analysis was not easy. It was supplemented by the principal component analysis in which particle data of different samples plots was calculated to find special coexistence and separate-existence of elements.

Sample plot 20 was selected for background comparison. The correlation table (Table I) shows that Si, Al, K and Mg have significant correlations with each other. Their coexistence probably indicates soil mineral particles. Also, Ca and Fe should be included in soil particles, but they did not have significant correlations with the Si-Al-K-Mg-group. It was concluded that Ca and Fe existed not only in soil mineral particles, but also in different particles originating from the steel plant.

Principal component analysis supported this observation. The first vector (Table II) was characterized by Al, K, Mg and Si; the second one by Ca and S, indicating $CaSO_3$/$CaSO_4$ particles; and the third one by Fe, possibly connected with some other elements. However, only a few particles were identified by their high Fe

TABLE I

Correlations (*Spearman Rank*) of elements in particles on the surface of pine bark in sample plot 20 (n = 40). *P<0.05, **P«0.01, ***P≤0.001

	Al	Ca	Fe	K	Mg	S
Ca	0.060					
Fe	0.069	0.021				
K	0.57**	0.19	0.037			
Mg	0.48**	–0.019	0.16	0.42**		
S	–0.084	0.23	0.00	0.055	–0.37*	
Si	0.57***	–0.17	–0.23	0.38*	0.37*	–0.28

TABLE II

Principal component analyses of particulate element data on the surface of pine bark in sample plot 20

Vector	Eigenvalue	Variance (%)	Cumulative variance (%)
1	2.64	37.7	37.7
2	1.39	19.8	57.5
3	1.02	14.6	72.1

Factor	Vector 1	Vector 2	Vector 3
Al	0.55	0.077	0.26
Ca	0.075	0.57	–0.024
Fe	–0.23	–0.27	0.79
K	0.43	0.31	0.32
Mg	0.51	–0.19	0.18
S	–0.14	0.65	0.073
Si	0.42	–0.22	–0.41

or Ca/S content (Figure 5). Soil mineral particles made up the major part of all particulate matter in sample plot 20.

In the vicinity of the steel plant the situation was different. An almost similar correlation between Si-Al-K-Mg was observed in sample plot 5. The total amount of Si and other soil-indicating elements was much higher here than in sample plot

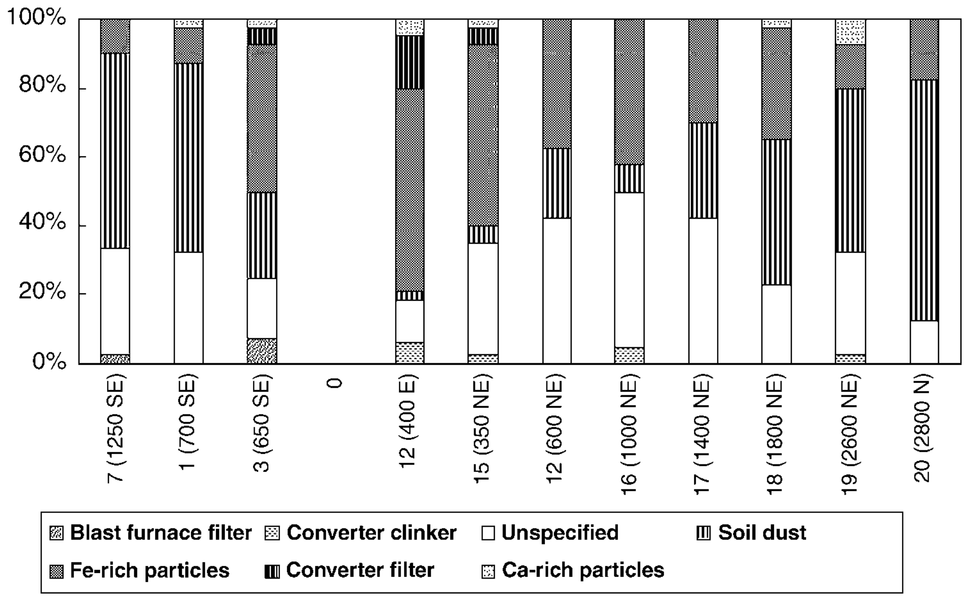

Figure 5. The share of different particle types on a 4 km long study line. X-axis titles include numbers of sample plots and, in brackets, distance and direction from the steel plant.

20, but nevertheless the amount of the soil's mineral particles was very low (Figure 6), since most of the Si-Al-K-Mg-containing particles were of anthropogenic origin. On the other hand, Ca had a very high correlation coefficient with sulfur. This was caused by the many $CaSO_3$/$CaSO_4$ particles on the surface of the pine bark. They originated from the nearby stockpile of desulfurization waste. Similarly, the correlation of elements and principal component analyses of other sample plots gave interesting information about coexistence and separate-existence of elements. However, the identification of the particles was finally done by comparing the element content of each particle.

3.2.2. *Particle Types*

Ten particle types were classified for the identification. A 'fingerprint' SEM/EDX spectrum for each particle type was produced using the original dust as reference material (see fingerprint for type 7 in Figure 7).

The particle types were:

1. Iron-rich particles. Several particle types contained a high Fe concentration, but this type included only such particles that did not have high content of other common elements, like silicon and calcium.
2. Soil dust particles were difficult to separate from some other types due to similar elemental composition, namely Si (highest content), Al (second highest),

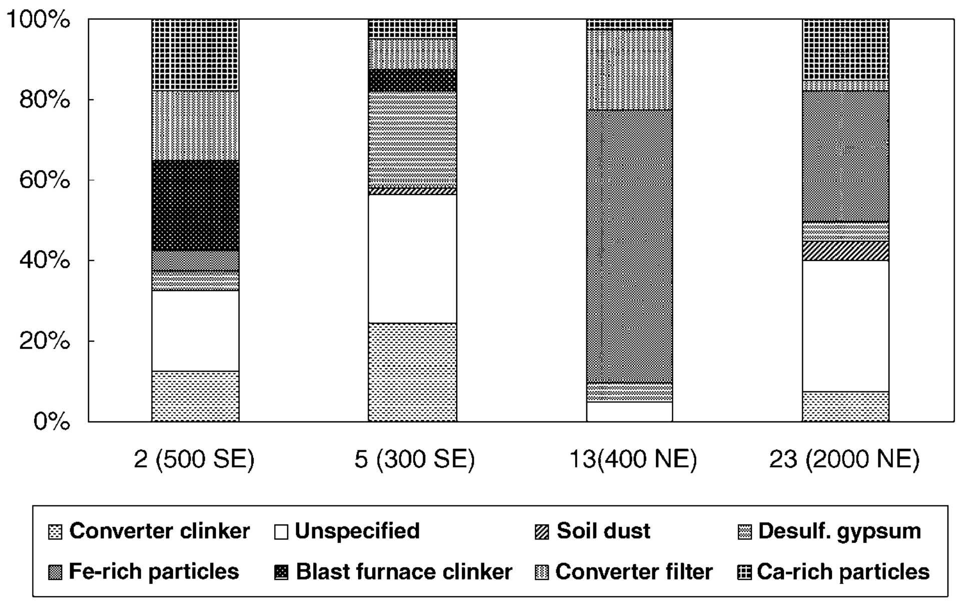

Figure 6. The share of different particle types in sample plots of special interest. X-axis titles include numbers of sample plots and, in brackets, distance and direction from the steel plant.

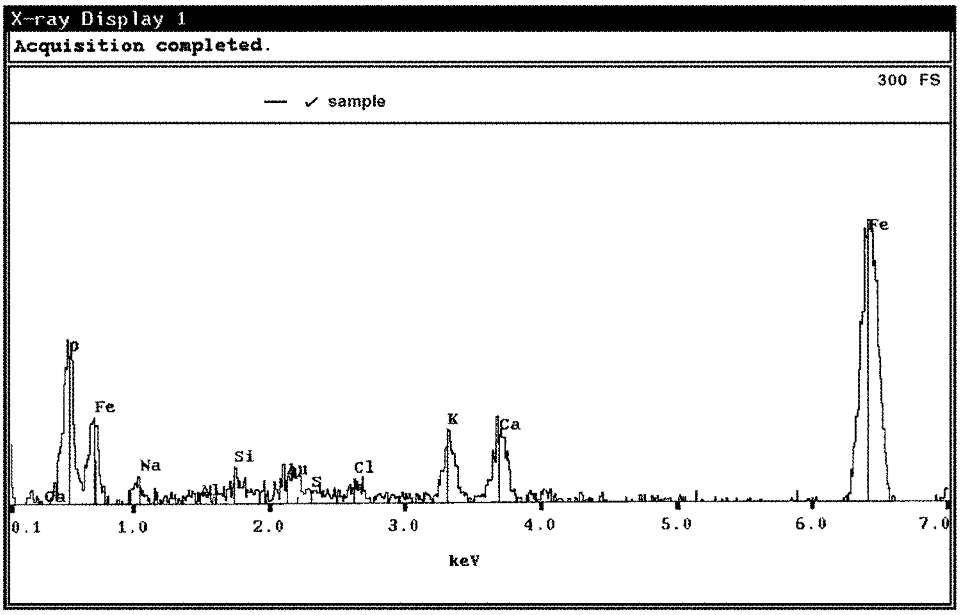

Figure 7. SEM/EDX spectrum ('fingerprint') of converter filter dust particle.

K, Mg, Ca, and Fe. However, particles originating from steel plant processes or wastes had increased concentrations of Ca, Fe, or Mg, unlike soil dust particles.

3. Unspecified local emissions. This type included particles which could not be identified as some special particle type. Many of these originated from the clinker crusher or from the fly ash of steel plant processes. These particles usually contained a lot of Si, Fe, Ca, and Al.
4. Dust from the electric filter of the blast furnace was characterized by a low content of Ca compared to K and Mg.
5. Dust from the clinker of the blast furnace contained a lot of Ca, Si, Mg, and Al.
6. Dust from the clinker of the converter had a very high Ca content compared to Si and Fe.
7. Dust from the converter filter had a high Fe and Ca content and very little Si.
8. Gypsum from the desulfurization process was made up of Ca and S (plus oxygen).
9. Carbon particles originated from the coke dust or from incomplete burning processes.
10. Calcium-rich particles did not contain S or Fe, only O or C in addition to Ca.

3.3. Distribution of particles types

Particle types 1, 2, and 3 were most common on pine bark surfaces. However, big differences in the distribution of these and other particle types were observed around the study area. In Figure 5 the share of particle types is given on a 4 km long study line. Soil dust particles form the major part of all particles at both ends of the line, i.e., at the remotest places where the impact of steel plant emissions is lowest. On the other hand, iron-rich particles are very few in these plots. The situation is contrary near the plant: iron-rich particles are most abundant and soil dust makes up only a few percent of all particles. Unidentified emissions are relatively common in 'intermediate' plots, but their real quantitative amount is greatest near the plant, which can be seen from the distribution maps of Ca, Fe, Si and other elements. The relative and absolute amounts of some other particle types, such as dust from the converter clinker and the converter filter, are also large near the steel plant.

The results of some especially interesting sample plots are given in Figure 6. Sample plot 2 was situated near the clinker crusher. According to the moss-bag experiment (Kemppainen, 1998), this place collected the highest amounts of calcium but not very much of iron. Particle identification produced a composition which was very different from the other areas. Iron-rich particles were few, unlike in other plots near the plant. Dust particles from the blast furnace clinker (containing 0.4% Fe and 39% CaO) made up 22.5% of the total particle amount, whereas no other sample plot contained more than 5% of this particle type 9. Also, Ca-rich particles (type 12) and particles from the converter clinker dust (containing 62% CaO) were common in this sample plot, as well as dust from the converter filter (CaO 16%)

and unspecified emissions, which probably were partly composed of the crushed clinker dust. All of these results prove that dust emissions of the clinker crusher, using mainly Ca-rich and Fe-poor clinkers, make up the major part of particle deposition in this environment.

It was already stated that sample plot 5 contained many $CaSO_3$/$CaSO_4$ particles originating from the nearby desulfurization waste (gypsum) stockpile. The highest amount of converter clinker dust was also observed in this plot. It originated both from the clinker crusher and from the converter clinker pile that was near this.

Sample plot 13 was also an example of the dominating role of stockpile dust. 87.5% of all particles belonged to two types, namely iron-rich particles and converter filter particles (containing 50% Fe). Stockpiles of iron-rich wastes as well as those of the converter filter were close to this plot and the trunks of trees were colored red by iron-rich dust.

One more extraordinary sample plot (number 23) is represented in Figure 6. This place was situated on a seashore bank and the bark samples were taken from the seaside of the trunk. Taking into account the remote situation of this plot, the sample contained relatively numerous Fe- and Ca-rich particles. Also, the elemental concentrations of Fe and Ca were high. It was supposed that trees on the seashore bank are acting as a sink for particles coming there. The winds, usually blowing in a northeastern direction, transport steel plant emissions only in this direction. Additionally, particles deposited on the ice in winter are now and then blown to the shore and accumulate there.

4. Conclusions

The SEM/EDX methodology proved to be a useful tool in the identification and quantitative measurement of different particle types under complicated multi-pollutant conditions. Pine bark is a suitable natural material to study pollutant distribution since particulate emissions accumulate on it. The results revealed that particulate emissions from steel plant processes or from energy production were not the main dust source in the environment. Most emissions were produced by the clinker crusher. The clinkers contained a lot of calcium. Therefore Ca was found in very great amounts, having an alkalizing impact on the surrounding pine forests. Numerous stockpiles of industrial wastes and raw-materials give rise to particulate emissions as a result of wind erosion.

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