THE EFFECT OF TRACTION SANDING ON URBAN SUSPENDED PARTICLES IN FINLAND

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Abstract. Springtime urban road dust forms one of the most serious problems regarding air pollution in Finland. The composition and origin of springtime dust was studied in southern Finland with two different methods. Suspended particles (PM_{10} and TSP) were collected with high volume particle samplers and particle deposition was collected with moss bags. The composition of the $PM_{1.5-10}$ fraction was studied using individual particle analysis with SEM/EDX. The deposition in the moss bags was analysed with ICP-MS. The results showed that during the study period, approximately 10% of both $PM_{1.5-10}$ particles and the deposition originated from sanding. Other sources in the springtime $PM_{1.5-10}$ were e.g. asphalt aggregate or soil and combustion processes. It can be concluded that sanding produced a relatively small amount of particulate matter under the investigated circumstances.

Keywords: deposition, fugitive dust, mineral dust, moss bag method, paved roads, PM_{10}, SEM/EDX, studded tires, traction sanding

1. Introduction

In areas with cold winter climate a special air pollution problem exists. Particulate matter accumulates in the urban environment over the winter months. When the snow melts, the particles concentrated in the snow are suspended to the air. In the urban areas the PM_{10} levels rise drastically especially near roads with dense traffic when road surfaces dry out (Kukkonen *et al.*, 1999; Pakkanen, Loukkola *et al.*, 2001; Pakkanen, Kerminen *et al.*, 2001). Special meteorological conditions like ground based low-height inversion with low wind speeds are often related with the high PM concentrations (Pohjola *et al.*, 2000).

Mineral dust forms usually a major part of the PM of the springtime episodes (Ojanen *et al.*, 1998; Pakkanen, Loukkola *et al.*, 2001). The two main sources of mineral dust in the urban areas during winter are: asphalt aggregate worn by the winter tires and traction sand (Hosiokangas *et al.*, 1999; Kukkonen *et al.*, 1999). Winter tires are compulsory in motor vehicles during the winter months in Finland and approximately 90% of cars have winter tires that are equipped with metal studs. To further promote traffic safety sand is commonly applied on streets and roads after snowstorms. Also de-icing salt is used to prevent road slipperiness.



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To estimate the shares of the different mineral dust sources, like traction sand and asphalt aggregate is difficult, because the rock aggregates often have a similar chemical composition (Song *et al*., 1999). There are several factors that have an impact on the source contribution but very few studies exist. The shares of the different sources are affected by the amount of traffic, the speed limit, the amount of sand dispersed on the road and the share of studded tires in the traffic flow. The actual wearing process of the road surface is affected by the design of the tire and the studs. According to Lampinen (1993) the most important stud properties affecting the wear are: stud mass, protrusion of the stud, the studs' impact energy and the area of spike impact. Wearing occurs also because of the grinding effect of the sand material under the tire (Kanzaki & Fukuda, 1993, Lindgren, 1998). Some rocks resist the wearing processes better than others, so the morphologies and the mineralogy of the pavement and sanding aggregates have an effect on the source shares.

In Japan studded tires were banned in the beginning of 1990's largely because of the dust emissions. Fukuzaki *et al*. (1986) and Amemiya *et al*. (1984) concluded that during spring the levels of particulate matter in the ambient air were highest in areas where studded tires were used. Noguchi *et al*. (1995) reported that in Japan after the ban of studded tires, the overall dust concentrations and the road pavement derived alkaline dust in precipitation have declined. These studies did not report the use of traction sanding. Also in some states of the United States of America studs are prohibited. The benefits and setbacks of the use of studded tires and traction sanding have been discussed also in Scandinavia. In an American study, Kantamaneni *et al*. (1996) observed 40% higher PM_{10} emissions from a sanded road compared to an unsanded one. However, they did not report the use of studded tires.

Another interest towards the composition and sources of the springtime dust rises from the new EU limit values for airborne particles. If the EU limit values given in the Council Directive 1999/30/EC for thoracic particles (PM_{10}) are exceeded, member states must implement action plans in accordance with Council Directive 1996/62/EC for attaining the limit value within a specific time limit. However, the member states can designate the zones within which the limit values are exceeded due to the resuspension of particulates following the winter sanding of roads (Council Directive 1999/30/EC, article 5). A list of such areas must be provided with information of concentrations and sources of PM_{10}. It must be shown that the exceedances are due to road sanding and that reasonable measures have been taken to lower the concentrations. In Finland the PM_{10}-concentrations have exceeded the national guide values (Table I) largely because of the springtime mineral dust (Kukkonen *et al*., 1999) and there is a risk that also the new limit values will be exceeded especially when the stricter second phase will be implemented in 2010 (Table II).

The aim of this study was to estimate the share of the sanding component in the suspended particles in a situation where both studded tires and traction sand are in

TABLE I
The Finnish guide values for TSP and PM_{10}

The Finnish guide values	Daily ($\mu g\ m^{-3}$)	Statistical definition	Annual ($\mu g\ m^{-3}$)	Statistical definition
PM_{10}	70	second highest daily value of the month	–	–
TSP	120	98. percentile of the data	50	arithmetic mean of daily mean values in a year

TABLE II
The EU limit values for PM_{10} which are implemented in two stages

EU limit values (PM_{10})	Daily ($\mu g\ m^{-3}$)	Number of exceedances allowed	Annual ($\mu g\ m^{-3}$)
2005	50	35	40
2010	50	7	20

use. Estimations of other possible sources in the springtime dust were also made. The shares were estimated from ambient particles with individual particle analysis with SEM/EDX and from deposition samples with bulk analysis with ICP-MS

2. Materials and Methods

In the city of Hanko a special material was used for winter sanding of roads, namely clinker from a nearby iron factory. The elemental composition (XRF-analysis) was received from the laboratory of the Rautaruukki Group. The clinker has high contents of Ca and Mg but very little Fe (Table III). The Ca-, Fe- and Mg-content of 4 clinker samples was additionally analysed with AAS (Varian SpectrAA 300/400) using HNO_3-digestion similarly as in the analysis of the moss bags (see Section 2.2). The maximum concentration in the blank samples was less than 1% of the lowest clinker sample. During the winter 1999–2000, approximately 15–20 t km^{-1} of clinker sand was dispersed on the roads of Hanko and its influence was clearly detected in the chemistry of the local dust (See section 3.3).

TABLE III

The Ca, Fe and Mg concentrations (%) of the clinker analysed with AAS and XRF. SD calculated from 4 duplicates

	AAS		XRF
	%	SD	%
Ca	24.1	0.4	26.6
Fe	0.4	0.1	0.4
Mg	7.2	0.2	8.1

2.1. TSP AND PM_{10} COLLECTION WITH HIGH-VOLUME SAMPLERS IN HANKO AND ANALYSIS WITH SEM/EDX

During the spring 2000, ambient particles were collected with two high-volume particle samplers (TSP – Kimoto, and PM_{10} – gravimetric Wedding & Associates Sampler) in the city of Hanko. The samplers were placed by a paved road with an average traffic flow of 5,500 vehicles per day. The equipment was placed at a distance of 3 m from the road side at a height of 4 m. Particles were collected on glass fibre filters in 3-day periods, altogether nine filters from both devices. The particle concentration was measured gravimetrically.

The elemental composition of individual particles was studied from the PM_{10} samples with a scanning electron microscope (SEM – ZEISS DSM 962) coupled with an energy dispersive X-ray microanalyzer (EDX – LINK ISIS with ZAF-4 measurement program). This instrumentation has been used in several individual particle studies (e.g. Mamane *et al*., 1980; Kasparian *et al*., 1998; Ganor *et al*., 1998; Paoletti *et al*., 1999; Haapala & Kikuchi, 2000; Breed *et al*., 2002; Paoletti *et al*., 2002). The SEM/EDX samples were mounted on an aluminium stub by attaching the filter surface covered with particles, on a double sided tape (Scotch Ruban Adhesive) (Breed *et al*., 2002). A minimum of two samples from each filter was made. The samples were coated with carbon (Agar SEM Carbon Coater) to make the sample surface conductive (e.g. Katrinak *et al*., 1995; Jones *et al*., 2001; Paoletti *et al*., 2002). The accelerating voltage was 20 kV. The total X-ray count rate was calibrated with cobalt to approximately 1500 counts s^{-1}. From each particle an X-ray spectrum was collected with a preset time of 15 s.

The minimum geometric diameter for the analysed particles was 1.5 micrometers. This was thought to be the lowest limit to obtain reliable results from individual particles with this technique (Jambers *et al*., 1995). Such a limit did not cause problems for the study because the main focus was in the coarse mode mineral particles.

TABLE IV
The most abundant particle types and their main elements (oxygen excluded)

	Al	Ca	Fe	K	Mg	Na	S	Si	Ti
Quartz								X	
Plagioclase feldspar	X	X				X		X	
Potassium feldspar	X			X				X	
Biotite	X		X	X	X			X	X
Amfibols	X	X	X		X	X		X	
Clinker	X	X			X		X	X	
Fly ash/asphalt filler/tire dust[a]	X	X	X	X	X	X	X	X	X

[a] also carbon was observed.

From all 9 PM_{10}-filters altogether 971 individual particles were analysed with SEM/EDX. From each filter a minimum of 100 randomly picked particles were analysed with EDX and the elemental weight percentages for each particle were calculated with ZAF for Al, Ca, Cl, Fe, K, Mg, Na, O, S, Si, and Ti. If other elements were observed, they were also recorded. It is important to note that ammonium compounds are not included because of the poor efficiency of the EDX in detecting nitrogen and because ammonium nitrate particles are volatile and may be lost in the vacuum of the SEM (Katrinak *et al*., 1995; Kasparian *et al*., 1998; Paoletti *et al*., 2002). Similar approach has been used in airborne particulate studies by Paoletti *et al*. (1999 and 2002) and Breed *et al*. (2002). To obtain 'fingerprints' for comparison, separate samples of the filler material of asphalt, and the clinker sand were crushed to dust and a SEM/EDX-sample was prepared similarly as from the airborne samples.

Particles were classified according to their elemental combinations. Observed particle types and their main elements are shown in Table IV. The 'fingerprints', local mineralogy and earlier studies were used as a basis for the classification. Katrinak *et al*. (1995) have estimated the relative errors of the ZAF-procedure for the atomic fractions observed in the individual particles to be 10–20%. Because of the inaccuracies of the quantitative analysis method (ZAF) the weight percentages were used more in a semi-quantitative way: the elements with a weight percentage of over 1% were considered present (see Hoornaert *et al*., 1996).

2.2. Deposition collection with moss bags in Hanko and Tammisaari and analysis with ICP-MS

Particle deposition was collected with the moss-bag method (see Little & Martin, 1974; Vasconcelos & Tavares, 1998) in Hanko and Tammisaari. They are neighbouring towns having similar population (10,000–15,000), similar traffic and other

conditions relevant for this research. The difference was in the sanding material. The city of Hanko used clinker sand while in Tammisaari, a 'normal' ground stone material was used. The composition of the deposition in these two cities was analysed to see the effect of the clinker sand on the chemistry of the dust. The main focus was on Ca and Mg since they were present in high concentrations in the clinker.

The moss material (*Sphagnum girgensohnii*) was collected from a non-polluted bog in southern Finland and prepared according to the Finnish standard (Finnish Standards Association, 1994). The moss bags were placed at the height of 2.5 m, in Hanko by the same road as the samplers, and in Tammisaari in a similar location. There were 3 roadside sampling points in Hanko and 2 in Tammisaari. The bags were placed to the sites on March 17th 2000, and the samples were collected after 2, 4, and 6 weeks.

The sample bags were dried in 60 °C for 24 hr. For element analysis, approximately 0.3 g of dry homogenised moss matter was weighed to Teflon vessels and digested with 6 ml of nitric acid (BDH Aristar, 69% HNO_3) and hydrogen peroxide (Merck Perhydrol 30% H_2O_2) (5:1) and heated with a Milestone Ethos 1600-microwave. The concentrations of Al, B, Ba, Ca, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Pb, Sr, Zn were measured by a Perkin-Elmer Elan 6000 ICP-MS. This paper concentrates on the results on Ca, Fe and Mg, which were the indicator elements for the purpose of this study. The ash content of the samples was measured by burning dry homogenised moss in 500 °C for 4 hr. Each sample was prepared and measured in duplicate. The mean element concentrations (μg g^{-1}) of the blank moss bags with standard deviation were 239 (SD 23) Ca, 132 (SD 30) Fe and 32 (SD 2) Mg. The statistical calculations were performed with the Statistix 7 programme.

3. Results and Discussion

3.1. Vernal road dust episode

A particle episode was observed in Hanko in the spring 2000 (29th March, Figure 1). The daily national TSP guide value (120 μg m^{-3}) was exceeded (208 μg m^{-3}). The national PM_{10} guide value (70 μg m^{-3}) was not exceeded. The mean temperature in Hanko during the study was 0.3 °C (max 7.1 °C and min –6.4 °C) and the amount of precipitation during the study was approximately 50 mm. Only moderate winds were observed (mean wind velocity 5–6 m s^{-1}). In March, over 50% of the winds were from SW-NW. In April the easterly winds (NE-SE) dominated (Finnish Meteorological Institute, 2000a and 2000b).

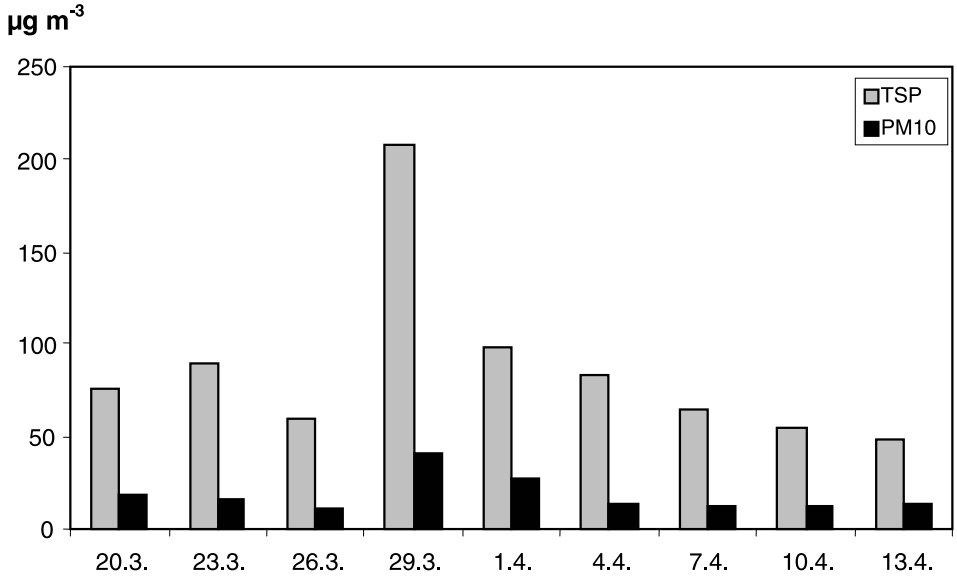

Figure 1. Mass concentrations for PM_{10} and TSP in Hanko. The mean values of three days, the last day marked on x-axis.

3.2. THE COMPOSITION AND SOURCES OF INDIVIDUAL $PM_{1.5-10}$ PARTICLES IN HANKO

The filters were divided into three classes: before (March 17–23, 2000), during (March 27 to April 1, 2000) and after (April 2–13, 2000) the episode. From each period over 300 individual particles were analysed. The observed particle types are shown in Table IV. The abundances (%) of the particle types were counted from the total number of the individual inorganic $PM_{1.5-10}$ particles analysed with the SEM/EDX (see section 2.1). The shares are given in Table V, with standard deviation calculated from the subsamples. Possible sources of the particle classes were estimated and thus source contributions were assessed, shown in Figure 2. It should be noted that particles with carbon as the main component were left out of the study because of analytical problems (possible influence of carbon coating and carbon tape) and because the main focus was on inorganic particles. Based on analyses of two PM_{10} filters, the possible share of carbon particles in $PM_{1.5-10}$ particles was estimated to be approximately 10% of the total number of particles.

The **sanding**-class consisted of the particles originating from the clinker sand that was used for winter sanding of the roads. The SEM/EDX-analyses showed that the clinker is relatively homogenous with a specific elemental composition shown in Table IV. The shape of the clinker particles was typically angular. The share of this particle class was approximately 10% of the total number of $PM_{1.5-10}$-particles, showing a decreasing trend during the study period.

TABLE V

The abundance (%) of the different inorganic particle types in the total number of the $PM_{1.5-10}$ with standard deviation before, during and after the particle episode

$PM_{1.5-10}$	March 17th – 26th		March 27th – April 1st		April 2nd – 13th	
	Mean %	SD	Mean %	SD	Mean %	SD
Clinker	12	1.2	11	1.7	6.3	5.1
Amfibols	3.0	1.2	1.7	0.6	3.5	0.8
Biotite	2.7	1.2	2	2.1	3.3	0.5
Potassium feldspar	14	1.5	13	1.0	11	1
Plagioclase feldspar	20	0	24	4.5	22	9.1
Quartz	10	3.1	13	0.6	9.3	3.3
Fly ash/asphalt filler tire dust	21	2.3	30	4.6	30	2.4
Other	17	1.0	11	0.6	17	0.8

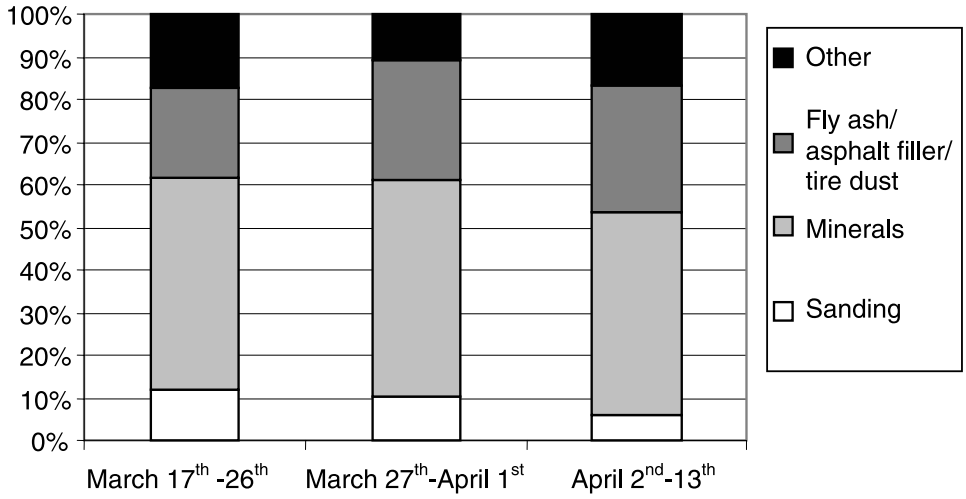

Figure 2. The abundance (%) of the different sources in the total number of the inorganic $PM_{1.5-10}$ particles before, during and after the particle episode.

The particles in the **minerals**-class made up to 50% of the inorganic $PM_{1.5-10}$-particles. They are minerals or mineral groups that are common in the Finnish bedrock like quartz, plagioclase feldspar, potassium feldspar, biotite and amfibols. They were angular in shape and were assumed to originate mainly from the stone material of the asphalt. Other soil sources may also have some contribution to this mineral particle class, namely particles from local unpaved roads (see Claiborn *et*

al., 1995), as well as from the unpaved shoulders of the studied road (Moosmüller *et al*., 1998).

Some of the particles were assumed to originate mainly from **combustion sources or asphalt filler** (with C and S from bitumen). Also **tire dust** has been reported to have similar composition (Rauterberg-Wulff *et al*., 1995; Camatini *et al*., 2001). This class made up to 20–30% of the inorganic $PM_{1.5-10}$-particles (Table V, Figure 2). It was not possible to identify separately these sources. Their chemical composition resembled also that of fly ash from different combustion sources (see Ganor *et al*., 1998; Valmari *et al*., 1999; Lighty *et al*., 2000), and on the other hand that of bitumen mixed with minerals possibly from filler or stone materials of the asphalt, based on analysis made for this study. The fly ash can originate from the residential area where the houses are heated mainly with oil and wood. The shape of the particles varied between angular and roundy forms. Also agglomerates of roundy particles were observed. However, coarse spherical particles usually found in fly ash (Kindratenko *et al*., 1994; Xie *et al*., 1994) were not present. On the other hand the temperature for the formation of compact spherical particles may not have been high enough (Valmari *et al*., 1999) in the residential boilers. Some of the particles might also have been agglomerates of mineral particles that had been 'contaminated' e.g. through coagulation with carbon or sulphur bearing particles (Maname *et al*., 1980) or by gaseous compounds through heterogenous nucleation.

Class **other particles** contained particles, which were observed in small quantities, and those whose origin could not be identified. This class included sea salt particles, as well as Na_2SO_4-particles, which might be originally sea salt, in which Cl has been substituted by SO_4 in the atmosphere (Kerminen *et al*., 1997; Song & Carmichael, 1998). Ca-S-O-particles were most likely $CaSO_4$, which can be of combustion origin or formed in the atmosphere from $CaCO_3$ (Hoornaert *et al*., 1996; Mori *et al*., 1998; Song and Carmichael, 1999). $CaCO_3$-particles could originate from concrete materials (De Miguel *et al*., 1997) or from the filler material of asphalt (Hoornaert *et al*., 1996). Particles with high iron content most probably originate from wearing and corrosion of the metal surfaces and brakes (Garg *et al*., 2000). It is important to note that ammonium particles could not be included due to analytical reasons (see Section 2.1). Particle class 'other' made up about 15% of the inorganic $PM_{1.5-10}$.

As a conclusion from the individual particle analysis with SEM/EDX, the share of particles from traction sanding was approximately 10% from the number of all inorganic $PM_{1.5-10}$ particles and 17% from the number of all $PM_{1.5-10}$ mineral particles.

3.3. THE CONTRIBUTION OF CLINKER SAND ON THE ROADSIDE DUST DEPOSITION

In order to get more information about the role of winter sanding material in vernal dust, we made comparative investigations of the dust in Hanko and Tam-

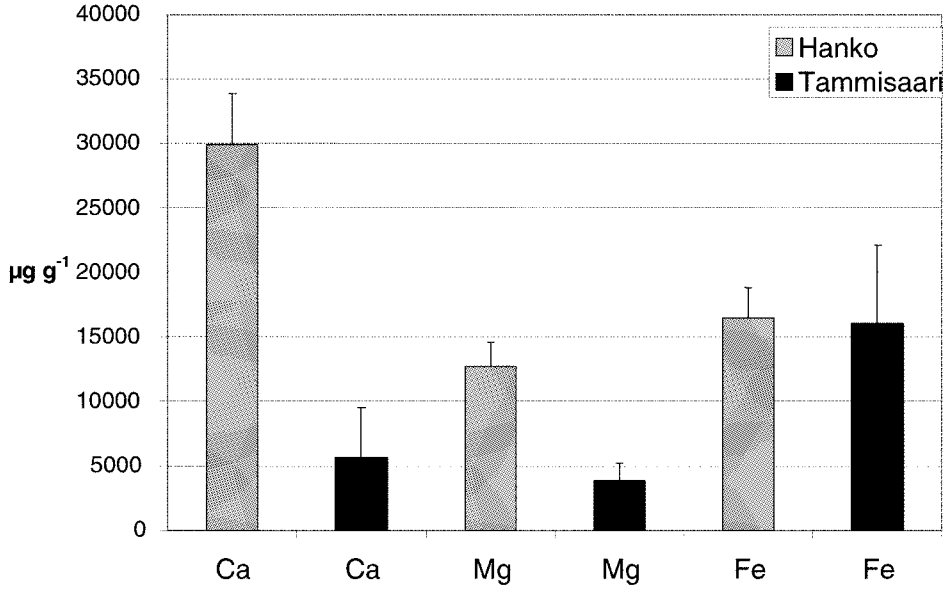

Figure 3. Accumulated Ca, Fe and Mg in the ash of moss-bags in Hanko and Tammisaari with standard deviations.

misaari. Road dust deposition was collected with moss bags, and the elemental and ash concentrations of the bags were analysed. The ash content represents the amount of dust accumulated in the moss bag. The measured elemental concentrations (μg g^{-1}) of the moss bags were divided with the ash content (g g^{-1}) to get the concentrations (μg g^{-1}) in the dust.

The concentrations of Ca, Mg and Fe in the dust (C_{tot}) are given in Figure 3. The results in Figure 3 are averages of 9 moss bags in Hanko and 6 in Tammisaari. The statistical analysis (Rank Sum Test) showed that the amounts of Ca and Mg in the dust were significantly higher in Hanko than in Tammisaari ($p = 0.0002$ for both metals). Fe did not show any significant difference ($p = 0.228$). The only big difference in the sources of these two cities was the traction sanding material. It was shown that the clinker sand that was used for traction sanding in Hanko, has high concentrations of Ca and Mg (see Section 2.). Thus the excess Ca and Mg have to originate from the clinker. The concentrations measured from the Tammisaari moss bags represent the composition of the dust from other sources than the clinker sand.

A simple mass balance approach based on the measured concentrations (Equations 1–3) was used for estimating the share (X, %) of the clinker sand in the dust deposition of Hanko. The concentrations of Ca and Mg in the dust (C_{tot}) (Figure 3) as well as the measured concentrations of these elements in Tammisaari (C_T) and in the clinker sand (C_S) were used for calculations.

$$C_{tot} = C_S * X\% + C_T * (100\text{-}X)\% \quad (1)$$

TABLE VI

The values used in the calculations (Equations 1–3) and the results for the share of dust from road sanding

	C_{tot} ($\mu g\ g^{-1}$)	C_S ($\mu g\ g^{-1}$)	C_T ($\mu g\ g^{-1}$)	X (% from sanding)
Ca	29 900	241 000	5 680	10
Mg	12 600	72 000	3 800	13

$$C_{tot} = C_S * X/100 + C_T * [(100\text{-}X)/100] \quad (2)$$

$$X = [(C_{tot} - C_T)/(C_S - C_T)] * 100 \quad (3)$$

C_{tot} = concentration of the element in the dust ($\mu g\ g^{-1}$)
C_S = concentration of the element in the dust caused by the clinker sand ($\mu g\ g^{-1}$)
C_T = concentration of the element in the dust caused by other sources ($\mu g\ g^{-1}$)
X = percentage from clinker sand

The values of C_{tot}, C_T and C_S used in the calculations and the share of the clinker, X are shown in Table VI. These results support the SEM/EDX studies, according to which about 10% of the total number of the inorganic $PM_{1.5-10}$ particles was from the sanding material. It is important to notice that the two quite different sampling and analysing methods gave similar results.

4. Conclusions

A special traction sanding material, clinker sand made it possible to estimate the contribution of the traction sand to road dust. SEM/EDX was used for classifying and quantifying the particles collected by a high-volume sampler, and to identify the origins of the particles. Moss bag method was used for collecting roadside particle deposition and ICP-MS for analysing its elemental composition. Both methods gave similar results, indicating that about 10% of the vernal road dust originated from the sanding material. A more significant share was from other sources.

The national guide value for TSP was exceeded during the studied springtime road dust episode in the coastal Finnish town Hanko. The results showed that only a minor part of the exceedance resulted directly from the sanding material. However, there are many factors that can have a significant influence in the composition of the dust (e.g. the pavement wearing effect of the sand rocks depending on the properties and amount of the sanding and pavement aggregates, traffic, meteorology and the influence of other sources). These factors have been studied very little and more information about their importance is needed to effectively mitigate the emissions.

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