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AN ATTEMPT TO IDENTIFY TRAFFIC RELATED ELEMENTS IN SNOW

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Abstract: The main objective of this article is an attempt to use snow as an indicator in the process of assessing and identifying vehicle-derived elements pollution. The aim of the present study is to characterize traffic-related elements in snow collected from three sites: a parking place, a highway and a relatively unpolluted airfield. Several recent studies suggest that road traffic is considered to be one of the major sources of environmental pollution in urban areas. In order to avoid the problem of low emission from household furnaces, samples were collected far away from residential buildings. Snow located near roads with heavy traffic seems to be a very useful tool and indicator of traffic-related elements released into the environment. Snow acts as a natural filter for various chemical elements and particles. Snow is an efficient scavenger of aerosol and air pollutants, usually remains on the ground for sampling after the event; moreover, snowmelt contaminates soil. In the present study filtered (0.45 µm) samples of melted snow were analyzed with ICP-MS. The results show significantly higher concentrations of elements in snow collected at the parking lot and at the highway when compared to samples taken from a relatively unpolluted airfield. Research on exploitation dust (break, tire, clutch) was performed with SEM-EDS.

Key words: traffic, parking lot, snow, vehicle-derived elements

INTRODUCTION

Snow collected from sites located in the vicinity of roads with heavy traffic seems to be a very useful tool and indicator of traffic-related elements released to the environment. Road traffic is associated with significant emissions of vehicle-derived elements contaminating soil and water, and at the moment is less controllable than other anthropogenic activities such as power plants, metallurgy and/or mining.

During springtime, melting snow and roadway runoffs significantly affect the receiving soil-water environment and river systems. Transport rate and travel distance of elements increase substantially when they reach river environments. In the terrestrial environment, metals and metalloil can penetrate deep below the surface and pollute groundwater. As a result, traffic-related pollution leads to a number of consequences for living organisms; for example some elements bioaccumulate in ecosystems (Basta et al. 2005). Studies of the chemical composition of snow as an indicator of anthropogenic environment pollution have increased in recent years due to higher awareness of health risks associated with its contamination with heavy metals. Similar research on snow contamination was carried out in China by Hu et al. (2012). The results obtained in the study have been presented in Table 1.

Table 1

Statistical parameters of selected heavy elements concentrations in snow – Beijing

Statistical parameters	Elements concentration [µg/L]						
	Al	Mn	Cu	Zn	As	Sb	Ba
Min	N.D.	1.16	N.D.	3.91	N.D.	0.548	2.80
Max	4.41	54.60	9.630	71.30	3.361	2.860	65.90
Average	1.75	15.80	1.340	36.60	0.878	1.230	17.10
Median	1.19	12.90	0.687	35.60	0.766	1.150	13.40

N.D. – not detected.

After analyzing PAHs in snow and moss, Viskari et al. (1997) confirmed that snow appears to be a good collector of pollutants from the atmosphere and can be used to monitor local airborne pollution from road traffic. Similarly, extensive research on PAH related to deposition of motor vehicle emission along roadside was carried out by Hautala et al. (1997). Bizzotto et al. (2009) ran studies on glacial and non-glacialfed streams to evaluate the loading of persistent organic pollutants (PCBs, HCB, HCHs and DDTs) accumulated in Alpine glaciers through seasonal snow/ice melt. A significant number of studies have been focused on natural areas attractive for tourists. Highly interesting results have been obtained in extensive research focusing on heavy metals in winter snow in the Dolomites; its statistical analysis of natural and anthropogenic contributions has been presented by Gabrielli et al. (2008). Magill & Sansalone (2010) analyzed particulate-bound metals for source area snow in the Lake Tahoe watershed. Sansalone & Buchberger (1996) analyzed distribution of traffic related metals and solids in urban highway snow and spring rainfall-runoff. Furthermore, Sansalone et al. (2003) ran detailed research on physical and chemical characteristics of deposition material generated from traffic activities after the melting as well as transport of the snow from the pavement shoulder. Reinosdotter & Viklander (2005) analyzed snow in Swedish municipalities, focusing on metals in melted water and particulate matter as well as granulometric analysis of urban snow residuals generated from traffic activities quality. Westerlund & Viklander (2006) reported concentrations of particles in different size fractions in snow and associated metals in road runoff during snowmelt and rainfall. Significant body of research has been focused on platinum-group elements (Rh, Pt, Pd) in snow. Platinum group elements (Pd, Rh), rare earth elements (Ce, Y), Ir, Os, Zr, As as well as Ce, Rb are associated primarily with the exploitation of catalytic converters. Platinum group elements and rare earth elements are extensively used as components of automobile catalytic converters. PGE has been reported to bioaccumulate in the environment and cause allergic reactions and various health problems. Palacios et al. (2000), Leśniewska et al. (2004) and Ward & Dudding (2004) in detail describe PGE distribution in different environmental samples (indicators) in many cities all over the world. A significant correlation of Pt, Pd, Rh indicates a common source for these metals. PGE correlate positively with Ce, Zr, Hf and Y – these positive inter-element correlations identify traffic as the main sources of PGE. Gregurek et al. (1999) analyzed Rh, Pt, Pd and Au in snow samples from the Kola Peninsula in Russia. Snow sampling has been used to assess the load of anthropogenic emissions and as an indicator to determine environmental loads of specific point sources, such as concentrating smelters and power plants (Ettala et al. 1986, Gregurek et al. 1998).

In order to understand the impact of road traffic emission on the environment, it is important to analyze and evaluate pollution originating through the wear of frictional elements in vehicles. Sampling in winter allows us to obtain representative samples of snow which is a good indicator of traffic derived elements, because it does not contain soil and other contaminants from the surrounding areas. Road dust contains potentially toxic pollutants originating from urban land uses and soil inputs from surrounding areas. Recent studies have shown that road dust primarily consists of soil derived minerals (60%) with quartz averaging 40–50% and the remainder being clay forming minerals of albite, microcline, chlorite and muscovite originating from surrounding soils. Organic matter primarily originating from plant matter constitutes about 2% of road dust, while potentially toxic pollutants represent about 30% of the build-up. These pollutants originate from brake and tire wear, combustion emissions and fly ash from asphalt. Heavy metals such as Zn, Cu, Pb, Ni, Cr and Cd primarily originate from vehicular traffic while Fe, Al and Mn mainly come from surrounding soils (Gunawardana et al. 2011).

Break materials and additives can be grouped according to their expected functions in the following categories: abrasives, friction modifiers, filters and reinforcements and binder materials. Today, friction materials in vehicle brake lining consist of a wide range of compounds, with fibers of steel, glass and plastic serving as reinforcements. Some compounds are used for their heat-conducting properties (brass chips) and good filling properties. Break dust consists mainly of particulate Al, Si, S, Ti, Fe as well as Cd, Cr, Ni, Pb and Zn (Adachi & Tainosho 2004, Hjortenkrans et al. 2007). Iron alloys also contain traces of S, Cu, Sb and Ba; some brake dust samples analyzed by researchers contained Zr, Cu to control heat transport, Sb to enhance stability and $BaSO_4$ to increase the density of the break pad (Blau & Meyer 2003, Hjortenkrans et al. 2006). Standard brake pads consist of: 48% barite, 14% vermiculite, 19% phenolic resin, 4.6% antminite, 7.5% rubber, 6.4% aramide, 0.3% sulphur. Österle et al. (2001) described chemical and microstructural changes induced by friction and wear of brakes, whereas composition, functions and testing of friction brake materials and their additives have been discussed in detail in Blau (2001). Typical composition of brake materials and additive functionality is presented in Table 2 (compiled based on Blau 2001).

Table 2

Brake materials and additive functionality

Material characteristics	Composition and consequences	Reference
Aluminum oxide	Hydrated form added as a polishing agent and for wear resistance, anhydrous form is more abrasive, fused is very hard and is most abrasive form	Nicholson 1995
Iron oxide	Hematite (Fe_2O_3) and magnetite (Fe_3O_4) can act as a mild abrasive	Nicholson 1995
Quartz	Crashed mineral particles	Erikssoon 2000
Silica	May be natural or synthetically produced	Hooton 1969
Zirconium silicate	$ZrSiO_4$	Jang 2000
Graphite	Used as a powder to control heat transport but can cause excessive cast iron wear	Spurr 1972, Nicholson 1995
Ceramic microspheres	Used as a powder to control heat transport but can cause excessive cast iron wear	PQ Corporation 1993
Copper	Used as a powder to control heat transport but can cause excessive cast iron wear	Nicholson 1995
Friction powder	May consist of Fe sponge, e.g. for semi-metallic brake pads, a number of particle sizes are available depending on requirements for surface area	Hoegenase 1990
Lead oxide	PbO has been used as friction modifier	Nicholson 1995
Metals-fluxing compounds	Pb, Sb, Bi, Mo as fluxing compounds serve as oxygen getters to stabilize friction-induced films.	Hooton 1969

Table 2 cont.

Metal oxides	Magnetite (Fe_3O_4) improves cold friction. ZnO lubricates but can cause drum polishing. Cr_2O_3 raises friction	Gudmand-Hoyer et al. 1999
Metal sulfides	PbS soft solids lubricant additive (2–8 mass %), MoS_2 (3–8 mass %), ZnS recommended for high load temperatures (5–10 mass %), metal sulfides mixtures are also used	BBU 1993
Antimony sulfides	Solid lubricant added to enhance frictional stability (>450°C)	Young 2000
Brass	62% Cu, 38% Zn, improves wet friction and recovery, common additive, sometimes used as chips	Nicholson 1995
Antioxidant	Graphite is commonly used in metal-ceramic composite brakes	Hooton 1969
Barium sulfate	Increases density and may aid in wear resistance, stable at high temperature	Nicholson 1995
Fibers – mixed oxide	A mixture of silca (45–50 mass %), alumina (5–15 mass %), calcia (34–42 mass %), magnesia (3–10 mass %) and other inorganics (0–7 mass %); function is to control fade and increase braking effectiveness	Sloss (no year)
Lime	$CaOH_2$ is used to avoid corrosion in Fe-additives	Nicholson 1995
Potassium titanate	Inert filler material; often used instead of asbestos	Young 2000
Zinc oxide	Imparts some wear resistance, but can polish drums	Nicholson 1995

A description of more than 100 formulations of patented friction materials was presented by Newman (1978). Fauser et al. (1999), Adachi & Tainoshob (2004), and Schauer et al. (2006) confirmed that tire dust contains significant amounts of Zn, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni and Pb. Snow is an efficient scavenger of air pollutants and it usually remains on the ground for sampling after the event. With rising temperatures pollution contained in snowmelt contaminates soil. Vasić et al. (2012) performed correlation analysis between concentration of metals and fine particles with the diameter lower than a few μm that stay suspended in snowmelt. The fine particles mass consisted of elements mainly originating from anthropogenic sources, and this conclusion was supported by the statistically significant positive correlation between Fe, Zn and Al and fine particles mass based on distribution parameters. In their earlier research the authors performed brake dust characteristics (Wieszała &Wyciślik 2008).

Granulometric analysis of dust formed through exploitation of the frictional parts of the car revealed that over 88% of the dust consist of particles in a fraction of up to 56 μm. Fractions under 20 μm, which have been found to present the highest health risk, form 17% of the exploitation dust.

STUDY AREAS AND SAMPLING

The aim of this study is to estimate the concentration of traffic-related metals in melted snow. Surface snow samples (about 20 cm depth) have been collected from three open fields: five samples were taken from 1 m^2 and then averaged. The first sampling point at the highway is located in Ruda Śląska-Halemba. There are three highway lanes in each direction. Highway test section is straight with the volume of traffic averaging at about 60 thousand vehicles per day, of which about 30% are trucks and coaches. The selected section of the highway is located at a considerable distance from both industrial plants and residential houses. Samples were collected at a distance of 3 meters away from the edge of the roadway. The next sampling point was located at the entrance to the parking lot in Katowice, Pukowca Street, at a distance of 1.5 m from the edge of the roadway. During the weekend this site serves more than 3 thousand vehicles, mainly individual cars. Speed at the parking lot is minimal and should be classified as starting speed. This particular location at the entrance to the parking lot has been selected also due to the fact that here cars form traffic jams while waiting to join the road. The third sampling point was located in close vicinity to an old airfield Katowice-Muchowiec surrounded by forest (huge meadow) at a distance of 300 m away from any road. Nearby there are no factories, houses or any other emission sources.

METHODS

Researchers focused on As, Ba, Be, Bi, Cd, Co, Cr, Cs, Cu, Fe, Mn, Mo, Ni, Pb, Rb, Sb, Te, Ti, Tl, V, W, Zn and Zr. In each sampling point field duplicates of snow samples were collected. Elements have been determined with ICP-MS application. In order to obtain unambiguous and unbiased ICP-MS results, the above-mentioned elements were additionally measured by ICP-OES, while Zn, Pb, Fe, Cu and Cd were measured by AAS. In order to estimate the accuracy and bias of the analytical method used in the study, reagents blanks and certificated reference material 1643d were used to assure that analytical results meet the required criteria. Table 3 presents detection limits for ICP-MS.

Table 3

Detection limits for ICP-MS

Element	Detection limits [mg/L]
As	0.01
Be	0.01
Ba	0.002
Bi	0.001
Cd	0.01
Co	0.0005

Table 3 cont.

Cr	0.04
Cu	0.01
Fe	0.01
Mo	0.01
Mn	0.0002
Ni	0.01
Pb	0.002
Pd	0.01
Sn	0.01
Tl	0.001
V	0.004
W	0.002
Zn	0.0002
Zr	0.01

RESULTS

Concentrations of elements obtained in the analysis of melted snow samples have been presented in Table 4.

Table 4

Concentrations of elements in melted snow samples

Element	Concentration [μg/L]		
	airfield	highway	parking lot
As	2.61	6.66	4.32
Ba	5.63	8.71	10.10
Be	N.D.	56.7	17.3
Bi	44.6	74.2	17.3
Cd	0.10	1.29	4.16
Co	10.80	9.50	1.57
Cr	3.62	3.15	17.04
Cs	2.04	1.89	12.90
Cu	3.16	7.14	6.33
Fe	15.5	22.7	392.0
Mn	4.19	1.29	6.38

Table 4 cont.

Element	Concentration [µg/L]		
	airfield	highway	parking lot
Mo	24.3	18.3	179.0
Ni	4.40	5.59	4.18
Pb	1.30	1.84	0.86
Rb	5.03	5.77	1.96
Sb	6.83	13.06	11.40
Te	2.65	2.55	3.57
Ti	4.71	6.82	5.91
Tl	6.47	7.19	10.00
V	2.19	4.27	18.00
W	1.70	2.59	1.93
Zn	2.58	9.37	25.70
Zr	9.12	26.65	9.90

N.D. – not detected.

Very high concentration of Zn (up to 25.7 µg/L) was found in samples collected in the parking lot. Such high concentration most likely was caused by tire wear. These samples also contained very high concentrations of Fe and Cr. These findings suggest that the main source of pollution was tire as well as brake-pad and clutch-plate wear. High concentrations of Zr, Ti and Sb are connected with friction modifiers, matrix and fillers. Predictably, the analysis of samples collected at the parking lot showed high concentrations of these elements.

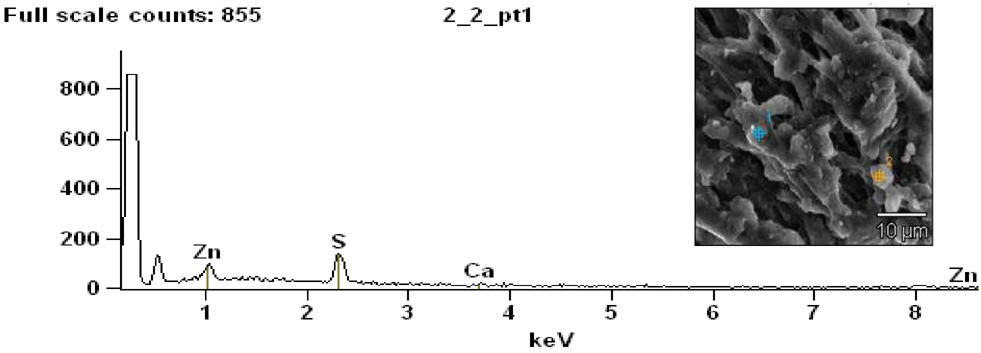

Fig. 1. Tire-derived – exploitation dust (fraction below 20 µm). Micro-photo of the surface (SEM) with a magnification 2000× spectrum characteristic of X radiation (EDS) (Wieszała 2006)

The characteristics of exploitation dust coming from a tire was shown in Figure 1. The research was conducted with the use of scanning microscope SEM: S4200 (HITACHI) in the following experimental conditions: the energy of primary electron beam 15 keV, cold cathode with field emission, the intensity of absorption current $1 \cdot 10^{-10}$ A. In order to take a micro-photo of the dust a signal of secondary electrons (SEM) was used. A magnification from 50× to 8000× was used. To the analysis of chemical composition of the dust a X-ray spectrometer with energy dispersion cooperating with a microscope was used (EDS): VOYAGER (NORAN, detector Si-Li, thin polymer window). During the tests a presence of such chemical elements was proved: Zn, S and Ca.

Micro-photo of exploitation dust coming from a break pads was shown in Figure 2. Dust coming from a car brake contains such elements as Cr, Fe, Cu, Zn, Mg, Al, Si , Zr , Sn and Cr.

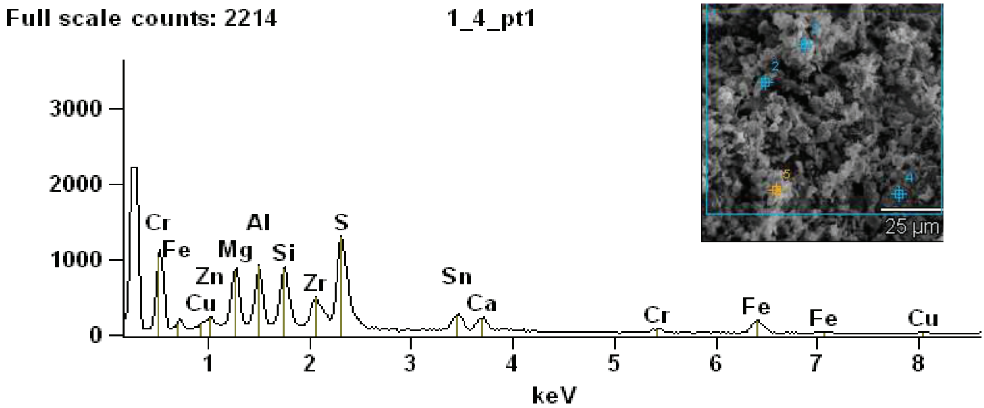

Fig. 2. Break-pads – exploitation dust (fraction below 20 μm). Micro-photo of the surface (SEM) with a magnification 1000× spectrum characteristic of X radiation (EDS) (Gajdzik et al. 2012)

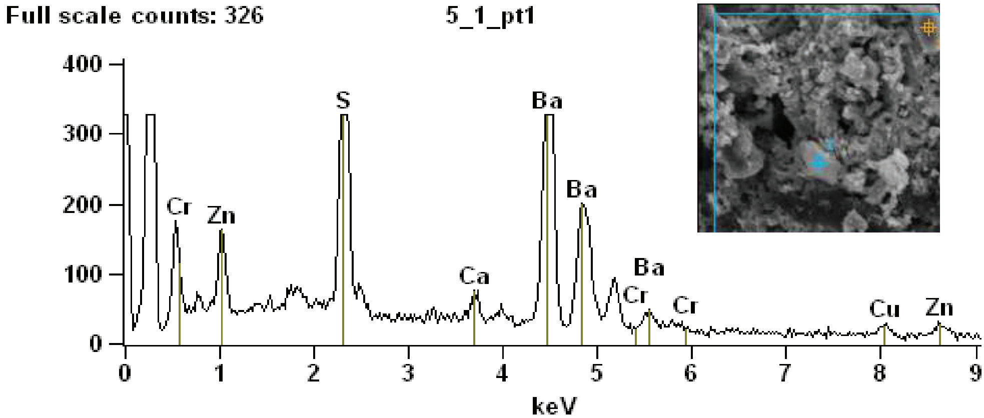

Fig. 3. Clutch plates – exploitation dust. Micro-photo of the surface (SEM) with a magnification 2000× spectrum characteristic of X radiation (EDS) (Gajdzik et al. 2012)

Relatively high concentrations of As, Ce, Cr and Zr seem to be typical for dust produced in the process of catalytic converter exploitation. The highest concentration of the above mentioned elements was found in dust samples collected on the highway. Clutch dust, in turn, contains significant amounts of Cr, Cd, Cu, Pb, Ba, Sb and Zn (see Fig. 3).

CONCLUSIONS

Snow is a good indicator of elements dissolved in snow which can later on easily penetrate into soil and water. Concentrations of elements analyzed in the study have been shown to be higher in samples collected on the highway and parking lot than in samples from a relatively unpolluted airfield areas in Katowice-Muchowiec. The fact of particular importance is that high concentrations of Cs, Sb, Zr, Zn, Cr and As have been found in snow samples from the parking lot in Katowice, Pukowca Street and the highway in Ruda Śląska-Halemba. Composition of elements found in contaminated samples may be a useful indicator of the source of pollution. The main sources of exploitation pollution formed in the process of vehicle operation are vehicle specific places: brake systems, tires, clutch plates and the erosion of the active layer of catalytic converters. Based on its composition, pollution found in samples collected on the highway seems to be predictably related to exploitation of catalytic converters, whereas elements found in contaminated samples from the parking lot are associated with tire, brake-pad and clutch-plate wear. Thus the composition of road pollution varies and seems to depend on the type of vehicle exploitation predominant in a given location.

Further research is indicated to analyze particulate matter in snow, focusing on geochemical composition and phase analysis in order to determine the proportion of elements in melted snow and particulate matter. In order to determine various forms of metals and estimate amount of metals that can be released from particulate matter to the soil-water environment – TCLP and BCR procedure should be performed. The Authors are fully conscious of the limitations associated with the process of sample collection. This study is exploratory in character more conclusive results will be obtained in model studies accounting for precise number of vehicles passing through the sampling area and "trap" sampling collection.

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