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The preliminary study of the selected environmental effects around the paper industry

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POTENTIAL INORGANIC POLLUTANTS IN THE SNOW SAMPLES FROM SE-LECTED LOCALITIES OF RUŽOMBEROK CITY

Introduction

This study focused on the evaluation of impacts of the cellulose and paper production in the company Mondi SCP on the deposition and distribution of inorganic pollutants (especially metals) in snowfall within the city and vicinity of Ružomberok given that the pulp and paper industry is responsible for releasing of different organic (solvents, alcohols, or chelating agents) or inorganic compounds ($NO_{2'} SO_{2'} CO_{2'}$ chlorites, or metals) into water sources or air.

Considering the fact that snow is an active media that transfers particulates and gases between the atmosphere and the landscape, snow can represent a suitable indicator of different particles and compounds deposition and accumulation over winter and spring. Snow chemistry therefore reflects changes in the chemistry of the atmosphere.

Material and Methods

Snow samples were obtained from two selected localities within the vicinity of the company Mondi SCP sampling locality Lisková and Hrboltová.

X-ray fluorescence spectrometry

For elemental analysis in snow water samples a hand-held X-ray fluorescence (XRF) spectrometer DELTA CLASSIC (USA) was used. The concentrations of the following elements were quantified: P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Rb, Sr, Zr, Mo, Ag, Cd, Sn, Sb, Ba, Hg, and Pb. Analytical modes utilize a Factory Grade Library consisting of a set of minimum and maximum values for each element in an alloy. If the content of an element was under the minimum or over the maximum limit of detection, it was cited as not detected element.

Data analysis and statistics

One-way analysis of variance (ANOVA) was performed to analyse significant differences between studied sampling sites and time deposited (i.e. length of time since the snow fell) in concentrations of Cl, K, Ca, or Ti in snow samples. The data analyses were carried out using the program STATGRAPHICS Centurion ver. 15 (Stat-Point Technologies, Inc., Warrenton, VA, USA).

Results

The determination of major and trace metals in surface snow can provide valuable information on the air mass transport and deposition processes, revealing sources and pathways of the elements at local and global scale. Snowwater analyses have been successfully used to estimate winter-time pollution loads on the terrestrial and maritime ecosystems (Benassai *et al.* 2005; Ming *et al.* 2007). The accumulation of atmospheric pollutants in the snow cover depends on weather conditions and the properties of underlying surface.

Our results (Table 1) of the measurement of selected elements in snow samples showed that from 25 analysed elements only eight elements (Cl, K, Ca, Ti, Rb, Mo, Sb, and Pb) were found in concentration above the limits of detection of used X-ray fluorescence spectrometry. Comparing concentrations of selected elements by one-way ANOVA for both sampling sites (Fig. 1), there was a statistically significant difference from the point of view of Ca and Ti presence in taken snow samples. These two elements were significantly more abundant in snow samples obtained from the sampling locality Lisková than in the case of snow samples originated from the locality Hrboltová. According to EPA (2010), make-up chemicals are a source of process-related CO₂ emissions at chemical pulp mills. Over time, small amounts of Na and Ca are lost from the recovery cycle at Kraft and soda facilities. Also, the pulp and paper industry uses titanium in process equipment exposed to corrosive media, such as sodium hypochlorite or wet chlorine gas (in the bleaching process).

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Sample No.	Cl	К	Ca	Ti	Rb	Мо	Sb	Pb
1	409 ±40	197 ±15	177 ±11	16 ±5	1.6 ±0.5	1.1 ±0.5	n.d.	n.d.
2	499 ±43	195 ±15	190 ±11	16 ±5	2.2 ±0.5	1.1 ±0.5	n.d.	n.d.
3	85 ±43	193 ±15	181 ±11	19 ±5	2.4 ±0.5	2.1 ±0.5	n.d.	2.6 ±1.
4	425 ±41	198 ±15	171±11	18 ±5	1.5 ±0.5	1.4 ± 0.5	n.d.	n.d.
5	427 ±41	196 ±15	185 ±11	18 ±5	2.0 ±0.5	n.d.	n.d.	3.0 ±1.
6	490 ±42	214 ±15	187 ±11	18 ±5	1.7 ±0.5	1.2 ±0.5	n.d.	2.2 ±1.
7	436 ±41	216 ±16	176 ±11	18 ±5	1.3 ±0.5	1.6 ±0.5	13 ±5	n.d.
8	454 ±41	213 ±16	202 ±11	18 ±5	1.4 ±0.5	1.3 ±0.5	n.d.	n.d.
9	426 ±41	191 ±15	185 ±11	15 ±5	1.2 ±0.5	n.d.	n.d.	n.d.
10	446 ±41	202 ±15	164 ±11	19 ±5	1.9 ±0.5	n.d.	n.d.	n.d.
11	452 ±41	208 ±15	179 ±11	18 ±5	1.6 ±0.5	n.d.	11 ±5	n.d.
12	477 ±42	201 ±15	176 ±11	11 ±5	n.d.	1.2 ±0.5	n.d.	n.d.
13	496 ±42	206 ±15	171 ±11	20 ±5	1.4 ±0.5	n.d.	13 ±5	n.d.
14	398 ±40	210 ±16	182 ±11	17 ±5	1.5 ±0.5	1.2 ±0.5	n.d.	n.d.
15	423 ±41	217 ±16	182 ±11	17 ±5	1.3 ±0.5	n.d.	n.d.	n.d.
16	464 ±42	193 ±15	181 ±11	19 ±5	1.9 ±0.5	1.1 ±0.5	n.d.	n.d.
17	406 ±40	203 ±15	188 ±11	18 ±5	1.9 ±0.5	n.d.	n.d.	n.d.
18	420 ±40	197 ±15	179 ±11	20 ±5	1.7 ±0.5	1.4 ±0.5	n.d.	2.8 ±1.
19	398 ±40	208 ±15	179 ±11			n.d.		n.d.
20	460 ±42	202 ±15	180 ±11			n.d.		
21	443 ±41	202 ±15	190 ±11					
22	478 ±42	202 =10 211 ±15	176 ±11		2.6 ±0.5	1.5 ±0.5		
23	483 ±42	193 ±15		20 ±5	1.4 ±0.5			
24	427 ±41	190 ±15	101 ±11		1.5 ±0.5	1.2 ±0.5		n.d.
25	398 ±40	100 ±15	189 ±11		1.7 ±0.5			
26	434 ±41	193 ±15	179 ±11		1.7 ±0.5		11 ±0	
20	448 ±42		173 ±11 188 ±11			1.5 ±0.5		
28	440 ±42	213 ±10 208 ±15	179 ±11			n.d.		
		208 ±15 197 ±15						
29 20	442 ±42						n.d.	n.d.
30	467 ±42	193 ±15	173 ±11					
31	500 ±43		171 ±11		1.6 ±0.5			
32	409 ±41					n.d.		
33	433 ±41		183 ±11		1.2 ±0.5			n.d.
34	526 ±43	208 ±15	171 ±11		1.9 ±0.5		n.d.	
35	429 ±41	209 ±16	184 ±11		1.9 ±0.5	1.5 ±0.5		
36	443 ±41	212 ±15	166 ±11		2.4 ±0.5			
37	458 ±42		181 ±11			n.d.		
38	421 ±41	223 ±16	173 ±11			n.d.		
39	446 ±41	197 ±15	176 ±11		1.8 ±0.5		n.d.	
40	482 ±42	191 ±15	177 ±11	17 ±5	2.1 ±0.5	n.d.	13 ±5	n.d.
41	454 ±41	214 ±15	177 ±11		1.3 ±0.5	1.1 ±0.5	13 ±5	n.d.
42	443 ±41	208 ±15	184 ±11	18 ±5	1.9 ±0.5	1.2 ±0.5	n.d.	n.d.
43	451 ±41	198 ±15	178 ±11	18 ±5	2.6 ±0.5	2.2 ±0.5	n.d.	n.d.
44	409 ±40	192 ±15	178 ±11	16 ±5	1.1 ±0.5	1.6 ± 0.5	n.d.	n.d.
45	431 ±41	198 ±15	178 ±11	17 ±5	1.6 ±0.5	1.4 ± 0.5	n.d.	2.2 ±1.
46	411 ±40	212 ±15	176 ±11	16 ±5	n.d.	n.d.	n.d.	n.d.
47	380 ±40	217 ±16	180 ±11	17 ±5	1.6 ±0.5	1.1 ±0.5	n.d.	n.d.
48	429 ±41	205 ±15	177 ±11	20 ±5	1.4 ±0.5	1.5 ±0.5	n.d.	n.d.

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	49	438 ±41	195 ±15	182 ±11	20 ±5	1.8 ±0.5	1.5 ±0.5	n.d.	n.d.
	50	427 ±41	201 ±15	169 ±11	16 ±5	1.8 ±0.5	1.1 ±0.5	n.d.	n.d.
	51	409 ±41	203 ±15	176 ±11	15 ±5	2.0 ±0.5	1.5 ±0.5	13 ±5	2.4 ±1.1
	52	440 ±41	215 ±16	171 ±11	13 ±5	1.7 ±0.5	1.2 ±0.5	n.d.	n.d.
	53	398 ±40	204 ±15	192 ±11	18 ±5	1.3 ±0.5	1.2 ±0.5	n.d.	n.d.
	54	421 ±41	203 ±15	190 ±11	17 ±5	1.8 ±0.5	1.8 ±0.5	15 ±5	n.d.
	55	453 ±41	197 ±15	179 ±11	15 ±5	1.6 ±0.5	1.6 ±0.5	n.d.	n.d.
-	56	417 ±40	187 ±15	171 ±11	12 ±5	1.7 ±0.5	n.d.	13 ±5	n.d.

Table 1. Elemental analysis of snow water samples obtained from Lisková (samples no. 1 - 30) and Hrboltová (samples no. 31 - 56) and carried out by X-ray fluorescence spectrometry (values in ppm ± standard deviation; S.D.; n.d. - not detected).

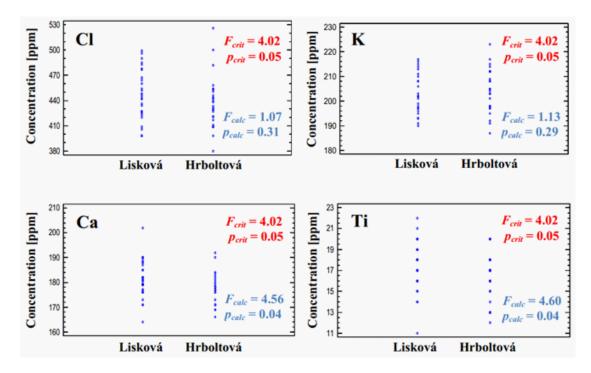


Fig. 1. One-way analysis of variance (ANOVA) of the data describing concentrations of Cl, K, Ca or Ti in snow samples obtained from sampling localities Lisková and Hrboltová.

Outside of the existence of point or nonpoint sources, the proportion of pollutants in snow/snow cover can be also affected by many factors, such as snowfalls yield, snow crystal morphology determined by temperature and water vapour supersaturation relative to ice, air currents, and age of snow. The last factor can be crucial not only in terms of level of pollutants deposition, but also from the point of view of vertical redistribution of pollutants within the snow cover. According to this fact, the effect of age of snow on the proportion of analysed elements in snow samples was evaluated. One-way ANOVA showed that with increasing age of snow the concentration of K in snow samples decreased (Fig. 2). For other studied elements, this relationship was not statistically confirmed.

Conclusions

In the further step of snow water samples evaluation, it will be advisable to use analytical method with lower limits of detection for metals, such

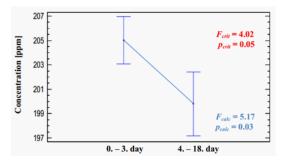


Fig. 2. Effect of age of snow on the concentration of K in snow samples obtained from Lisková and Hrboltová.

as atomic absorption spectrometry (AAS), inductively coupled plasma atomic emission spectrometry (ICP-AES) or inductively coupled plasma mass spectrometry (ICP-MS) as well as to obtain other parameters better describing physicochemical characteristics of snow water samples.

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POTENTIAL ORGANIC POLLUTANTS IN THE SNOW FROM EXPERIMENTAL STUDY AREA - RUŽOMBEROK

Introduction

Paper and pulp guidance note from Environment Agency of the UK government includes volatile organic compounds (VOCs) into main air pollutants (Environmental permitting regulations 2010). In the Kraft process of pulping, VOCs come from spent liquor evaporation, wood-chips digestion and bleaching (Ince *et al.* 2011). Other authors include persistent organic pollutants (POPs), notably, dibenzofurans (dioxins and furans) and polychlorinated dibenzodioxins into pollutants discharged to the effluents from pulp and paper mills (Ali and Sreekrishnan 2001).

Chemical compounds are transported from atmosphere to the surface by atmospheric deposition. Deposition can occur from sedimentation of particulate matters (dry deposition) or binding of chemical compounds on water drops or snowflakes (wet deposition) (Holoubek *et al.* 2001). Even light snowfall captures most compounds from the atmosphere. Snow is an effective absorbent and adsorbent of aerosols and gasses and it can be collected for samples collection even after snowfall (Vučinič Vasič *et al.* 2011). There are many studies about POPs and VOCs deposition, also in snow, for example Lei and Wania (2004), Kawamura and Kaplan (1986), Starokozhev *et al.* (2009), Franz and Eisenreich (1998) etc.

Material and Methods

Given the information about wind directions, two sampling sites were selected, one to the east of factory (Lisková) and one to the west (Hrboltová).

Gas chromatography with mass spectrometry analyses were carried out on an Agilent Technologies (Palo Alto, CA, USA). The analytes were separated on the capillary column from Agilent Technologies using helium as the carrier gas.

For preconcentration and isolation of organic compounds from water matrix, stir bar sorptive extraction (SBSE) was performed.

Results

The primary aim of screening was to identify the potential pollutants in snow from Hrboltová and Lisková. Results of samples are carried in Table 2.

Although 29 samples were collected from Hrboltová and 29 samples from Lisková, only 9 of them were analysed completely with usable results. Remaining samples were subject to mea-

	Hrboltová			Lisková					
	S1	S2	S3	S4	S1	S2	S3	S4	S5
Alkanes	Х	\checkmark	Х	Х	Х	Х	Х	Х	Х
Carboxylic acid	Х	\checkmark	Х	Х	Х		\checkmark	\checkmark	Х
Carboxylic acid esters - phthalates	Х	\checkmark	Х				\checkmark		\checkmark
Hydroxycarboxylic acid esters	\checkmark	\checkmark		Х			\checkmark	Х	Х
Terpenes	\checkmark	\checkmark					\checkmark		Х
Quinoline and derivatives	\checkmark	\checkmark	Х	Х			Х	Х	Х
Naphthalene and derivatives	\checkmark	\checkmark							
Fluorene and derivatives		\checkmark							
Phenathrene and derivatives		Х		Х	Х			Х	
Anthracene and derivatives									
Fluoranthene									
Pyrene									
Biphenyl and derivatives		Х	Х	Х	Х		Х	Х	

Table 2. Results from complete analyses of organic compounds in snow samples from Hrboltová and Lisková (S1-S4 – samples; $\sqrt{-}$ detected compounds; x - not detected compounds).

suring of LLE on n-hexane with subsequent GC-MS. We did not publish results from LLE measuring because of low detection rates. This could be caused by low effectiveness of LLE, but also by long time of storage of samples, even though samples were stored in optimal (cold and dark) conditions. We found only few compounds in each analysed sample. Moreover these compounds were insignificant. Nevertheless, published results from the complete analyses are of good M. Hudák. M. Randis. T. Bartaloš, M. Kmeťová, I. Kosová & M. Horník

quality and have informative value about organic pollution in snow from Lisková and Hrboltová. Though there are many studies on metal contaminants or suspended solids in snow, studies on organic pollutants are rarely reported. For example, the occurrence of perfluorinated compounds (Liu et al. 2009), and PAHs (Reinsdotter et al. 2006) in urban snow has been reported occasionally. Even more infrequent are screening studies of snow with several groups of organic pollutants investigation. The only one study with same subject we have found is from Gothenburg, Sweden (Björklund et al. 2011). This study aim was to investigate the anthropogenic organic compounds in the urban snow in Gothenburg and perform the non-target screening of semivolatile organic compounds and thereafter target analysis of specific compound groups. Despite problems with number of peaks in the total ion chromatograms, authors could clearly identify specific phthalates as diisobutyl phthalate, di-nbutyl phthalate, butyl benzyl phthalate and diethylhexyl phthalate, several phenols, and PAHs in the snow samples. The highest concentrations and also the most occurring PAHs were pyrene, phenathrene, fluorine and benzo[ghi]perylene. Brominated flame retardants and chlorinated paraffins were only sporadically detected. The most occurring group were PAHs and phthalates.

The most common compounds in our study PAHs are same as in Björklund et al. (2011). Similarities are also found in phthalates. In our work were identified diethyl phthalate, dibutyl phthalate, diisobutyl phthalate, butyl benzyl phthalate, and di-n-octyl phthalate. Phthalates were found in all of the samples from Lisková but only in two samples from Hrboltová. The number of relative intensity of peak is much higher in Lisková than in Hrboltová samples. This phenomenon could be affected by several factors which are beyond the scope of this study. Other groups of compounds investigated by Björklund et al. 2011 such as alkylphenols, brominated flame retardants, chlorinated paraffins and perfluorinated compounds were not detected in our study.

Conclusions

Despite of the relatively low number of analysed samples, we can conclude that most occurring group of pollutants were PAHs, which is similar to the only comparable published study we have found (Björklund et al. 2011). As in this study, phthalates were also detected. However, this study is an initial survey and should serve as the basis for future studies.

METAL CONTAMINATION OF APPLE FRUIT IN EXPERIMENTAL STUDY AREA RUŽOMBEROK

Introduction

The aim of this study was to detect the presence of heavy metals (Soják et al. 2002) and their accumulation in the fresh fruit selection on fruit trees (Mičieta and Murin 1996). We have chosen this crop because it is an important food crop and is of nutritional value (Boyer and Liu 2004). The results of this research can be applied to identify main sources of pollution and determine industry impacts on biota, and consequently the health (Wyzga and Folinsbee 1995) and quality of human life (Kampa and Castanas 2008).

The aim of the research is to study the specific microclimate in the Liptov basin in which the city of Ružomberok is located. The contributing emissions of industry can be seen in the mapping and the identification of the individual sources of pollution, and are identified by their occurrence in bodies of other organisms. Another benefit is to understand the process of pollution in ecosystems, and the transitions through trophic levels to humans.

Material and Methods

Samples from the apple trees (Malus domestica) were gathered in 2014 from the six different places: Lisková, Černová, Hrboltová, Štiavnička, Biely Potok and Vlkolínec that were located at different distances and directions from the largest industrial polluter Mondi SCP. All sites were located within the range of dispersed pollutans from Mondi SCP which may be enhanced by wind patterns in the basin.

Collected samples were cut into thin discs and they were dried at 65.3° C for 15 hours. Dried samples were crushed in the CryoMill. Homogenised samples were performed to X-ray analyses by hand-held X-ray fluorescence (XRF) spectrometer DELTA CLASSIC (USA).

The statistical analysis was realised by using the program CANOCO for Window. PCA analysis was performed to see relationship between detected elements in particular locations.

Results

Study focused on the elements S, Cl, K, Ca, Mn, Rb, Mo and Pb (Table 3) because of the value above

Locality	S	Cl	К	Ca	Mn	Rb	Мо	Pb
Lisková	178	378	9678	608	19.0	11.0	5.7	11.4
Černová	132	497	8667	706	12.3	14.2	5.0	8.7
Hrboltová	203	395	14752	1145	17.0	11.7	4.4	10.9
Štiavnička	124.5	414	10555	535	9.2	15.9	4.8	9.2
Biely Potok	162	415	10029	630	13.0	11.6	4.2	8.0
Vlkolínec	198	430	10879	1240	15.0	12.0	4.1	7.7

Table 3. The final measured concentrations of detected elements, ppm.

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Environmental effects around the paper industry the detection limit. Other elements such as P, Ti, Cr, Fe, Co, Ni, Cu, Zn, As, Se, Sr, Zr, Ag, Cd, Sn, Ba, and Hg were not detected.

Figure 3 shows statistical PCA analysis when sum of all eigenvalues was 0.028 at an significant level p =0.05. The first principal component (Axis 1) describes an inverse relationship between S and K, Rb, Cl, Pb, Mo. It is noticible that the increase of K, Pb, Cl Rb, Mo related to Ca mainly occurred in the village Štiavnička which is located very close to Mondi SCP. The second principal componet (Axis 2) denotes an inverse relationship between Ca and Mn, Cl, Rb, Pb, Mo accumulation in the apples. Apples from the villages Vlkolínec and Hrboltová contained relatively more Ca and less Mn, Rb, Cl, Pb, Mo. In the villages Biely Potok, Černová and mainly in Lisková, the situation was opposite.

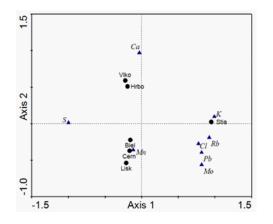


Fig. 3. PCA analysis: representation of the detected elements to locations.

Conclusions

We consider that increased amount of elements in pulp of apples, mostly sulphur and chlorine (Vlkolínec, Biely Potok) may come from Mondi SCP, which produce air pollutants and further contamination of apples by lead in Lisková mostly comes from traffic pollution, cause (Pb) was independent from TRS. Our work can serve as an initial study on the sources of pollution identification of the air in Ružomberok and results may serve to other similarly targeted research as a source of valuable information about industrial pollution in this area.

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