Chemical pollutants in Finnish and Karelian snow

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Abstract

Snow samples were collected in March 2001 in 10 sites in the vicinity of the Kostomuksha factory in Karelia and in Finland longitudinally from the South to the North. GC-MS and ICP-MS allowed to identify and quantify 72 chemical elements and more than 200 individual organic compounds. A list of toxicants emitting with the factory exhausts, a list of priority pollutants for Kostomuksha and a list of pollutants of concern for Finland were proposed. High level of phthalates in the sample collected at the most northern site may be treated as an example of the "cold finger effect".

1 Introduction

There are several approaches to estimate atmospheric pollution. One of them deals with the long-term air pollution by collecting snow samples on the site. Snow is a perfect matrix to study depositions of the pollutants. It is static in
contrast to the air or water. It does not require any complicated sample preparation and due to the sub-zero temperatures even rather unstable organic compounds remain unchanged for a long time. In Finland and Russia this type of analysis is particularly valuable, since 5-6 months fall-outs may be studied. Quite interesting results were obtained applying this method earlier [1-7].

Besides the immediate influence of ecotoxicants on the nature while in the atmosphere, these chemicals may also be harmful after the snow melts. To estimate this danger the concentrations of the toxicants were compared with the standard values of their Maximal Allowable Concentrations (MAC). In Russia there are two sets of these values. The first one involves the danger of chemicals towards aquatic ecosystems (MAC_f - for fishery water basins), the second one (MAC_s - for sanitary and hygiene) – the danger towards mammals (humans). If the level of a chemical in water is below its MAC value this compound cannot harm aquatic system or human being during the whole life water consumption.

The factory in Kostomuksha was built in 1982. According to Russian and Finnish reports it seriously aggravated the anthropogenic burden on the forests and water basins of the region. The main product of the factory is beneficiated iron ore. The principal atmospheric exhausts involve sulfur dioxide (60,000 tons/year) and technogenic dust (5,000 tons/year).

The main tasks of the present study were as follows:

1. To find out pollutants coming to the environment with the exhausts of the Kostomuksha factory.
2. To compose a list of priority pollutants for the Kostomuksha region.
3. To confirm or to reject transboundary transfer of pollutants from the Kostomuksha factory to Finland via atmosphere.
4. To screen the Finnish territory from the North to South to select the most important organic and inorganic air pollutants.
5. To confirm or to reject the "cold finger" effect.

2 Methods and materials

For the present study snow samples were collected at the end of March 2001 at 10 sites in Finland and Karelia. Sample 1 was collected on the Finnish territory few kilometers to the west from the state border; sample 2 - in the National Resource Park in Karelia near the state border; sample 3 - in the city park (20 km west to the factory); sample 4 – 4 km to north-east from the factory (direction of predominant winds); sample 5 – 4 km to south-west from the factory to the city; sample 6 - close to the border with Finland (near the railroad and autostrada). Sample 7 was collected near Savonlinna (south of Finland), sample 8 – near Kuopio (center of Finland), sample 9 – near Kemijarvi (on the Northern Polar Circle) and sample 10 – near Inary at the far north of Finland.

ICP MS was used to measure the integral concentrations of 72 elements in the samples, while GC-MS was applied to identify and quantify the semi volatile organic pollutants (including priority pollutants from the US EPA list). Snow samples were kept in polyethene bags and transported on ice to the laboratory. For the ICP-MS analysis nitric acid was added to 10 ml of melted snow to adjust its pH to 2 and kept for 10 hours prior to the injection into the
Air Pollution X

instrument. Standard solutions were used for the quantitative analysis. PlasmaQuad (VG, UK) ICP-MS instrument was used with the following parameters: mass range – 6 - 242D; number of channels – 2048; number of scans – 100; time of measurement for one channel – 320 mksec; type of registration – counting of impulses.

For the GC-MS analysis the melted snow (1 l) was saturated at room temperature with solid NaCl. To extract the base/neutral pollutants, 60 ml dichloromethane was added to the samples adjusted to pH 11 with NaOH (1N), shaken to rinse also the inner surface and transferred to the separatory funnel (for 2 liters). The funnel was shaken for 15 min. with periodic venting to release excess pressure. After the layers were separated, the dichloromethane extract was collected into a 250 ml flask. The procedure was repeated three times, i.e. finally 180 ml of the dichloromethane extract were obtained.

For the extraction of acidic pollutants pH of the aqueous phase was adjusted to 2 with 1N solution of sulfuric acid. Extraction procedure was the same as for the neutral and basic compounds. Both base/neutral and acid fractions were separately dried over anhydrous sodium sulfate and concentrated in Kuderna-Danish concentrator to reach a final volume of 1 ml. Just prior to the GC-MS analysis, the base/neutral and acid extracts were combined.

Three internal standards were used (D8-naphtalene, D10-phenananthrene and D12-perylen). These internal standards covered the range of the masses and retention times of the compounds of interest, while response factors calculated on their bases satisfy the requirements of the US EPA method 8270 for the semivolatile priority pollutants.

Hewlett-Packard 5989 “Engine” GC-MS system was used. The following parameters were applied during the analyses: mass range 25-550 D; scanning rate - 0.5 sec; ionization energy - 70 eV (electron impact); chromatographic column HP – 5 (30 m x 0.53 mm x 1.5 μm); temperature program: initial temperature 50°C (4 min.), gradient 8/min., final temperature 300°C (20 min.).

The software provided by Hewlett-Packard enabled to reconstruct Extracted Ion Current Profiles (EICP) and to integrate the abundances in any EICP between specified time or scan limits. The Wiley mass spectra library for 275,000 compounds was used for the qualitative analysis.

3 Results and discussion

3.1 Kostomuksha factory.

3.1.1 Metals

Rather low MAC values for lithium (MACf = 80mg/l, MACp = 30mg/l) make treat this metal as a regional priority pollutant. Its concentration in the vicinity of the factory was two orders of magnitude higher than at the background sites (for the most polluted sample MACp value was exceeded more than 8 times). High level of Li in sample 6 may be accounted for the fact that it was collected near the road. Thus transport could be considered as a possible source.
Although boron levels do not exceed MAC values, the influence of the factory on the environment was obvious. Boron concentration in the contaminated sites 3-6 was 2 orders of magnitude higher than in the background sites.

It is rather difficult to draw any precise conclusion on the regional contamination with aluminum. All 6 samples contained aluminum in quantity notably exceeding MAC$_f$ (40 mkg/l) and in some cases MAC$_{a-h}$ (500 mkg/l). However the samples in the vicinity of the factory were cleaner than the others. Therefore aluminum should penetrate the environment from other sources.

Another metal of concern is manganese. Although its MAC$_{a-h}$ (500 mkg/l) value was not exceeded, much tougher MAC$_f$ value (10 mkg/l) makes this metal one of the priority pollutants for the region. The most polluted site was 2. This fact is rather strange and requires further study.

It is hardly possible to avoid contamination with iron working with iron ore. Concentration of the metal in the samples collected in the vicinity of the factory exceeded MAC$_f$ value (100 mkg/l). Nevertheless it is worth mentioning that a priori we expected higher concentrations of this metal in the samples.

Although concentration of cobalt was far below its MAC levels (10 mkg/l) in all the samples, this metal comes into environment from the factory, being a constituent of the iron ore. That is why its concentration in samples 5 and 4 was much higher than in the background samples. On the other hand the amount of Co in Finnish sample 1 was 8,1 mkg/l – notably higher than in the vicinity of the factory. Definitely there is another source of this metal nearby.

Levels of nickel exceeded MAC$_f$ value (10 mkg/l) for 4 of 6 samples reaching maximum (7.4MPC) for the most contaminated site 4 km to the north-east from the factory. Again the influence of the factory was obvious while nickel should be treated as a priority pollutant.

The major discrepancy between MAC$_f$ and MAC$_{a-h}$ values (1.0 and 1000 mkg/l correspondingly) for copper demonstrates the danger of this metal for aquatic organisms. Taking into account that MAC$_f$ value was exceeded in all the samples including the most clean Finnish one it is possible to suppose that the increased copper level in the air is a regional characteristic. However the highest concentration (86.2 mkg/l) was detected for the most polluted site on the north-east from the factory proving that the factory is a source of copper in the air.

Similar conclusion may be derived concerning zinc. Its levels were higher than MAC$_f$ value (10 mkg/l) at all the sites reaching maximum for the most contaminated sample 4. Even MAC$_{a-h}$ value (500 mkg/l) was exceeded for this site. Definitely this metal exhausts with the dust during the processing of the ore. MAC$_f$ value for molybdenum is 1 mkg/l. The factory may be considered as a source of contamination of the environment with this metal. The most polluted sample 4 contained 9 MPC. Mo is apparently a priority pollutant for the region.

Kostomuksha factory emits tin with the dust. The level of tin in the most contaminated site 4 was 10 times higher than in other samples. Nevertheless this metal may not be included into the regional list of priority pollutants so far as its MAC$_{f}$ value (112 mkg/l) is significantly higher than the detected levels.

Mercury is a priority pollutant all over the world. Amounts of this metal in the Karelian samples were quite significant especially taking into account that MAC$_{a-h}$ value is 0.5 mkg/l, and MAC$_f$ is 0.1 mkg/l. However it is hardly
possible to connect the presence of this metal in the samples with the activity of the factory, since the profile of the mercury depositions over the territory differs greatly with that of Mo, Zn or Cu. There should be other sources of this metal. The last metal worth discussion is lead. It could appear due to the activity of the factory since its concentration is highest for sample 4. However similar value was obtained for sample 3, which was collected in the city. The most probable source of this element is transport rather than the factory. MAC value for lead is 30 mkg/1, while MAC is 6 mkg/1. Thus it was not exceeded for any sample. It should be noted that lead concentration in Finnish sample 1 was 4.7 mkg/1, i.e. higher than in any Karelian sample. Therefore one can propose that lead does not come to Finland with the factory exhaust. There should be a source of this metal in the vicinity of the sampling site.

3.1.2 Organic pollutants

More than 200 individual organic compounds have been detected and identified in the snow samples by means of GC-MS. These chemicals include rather toxic anthropogenic pollutants as well as natural compounds. The presence of the persistent organochlorines (including PCB, BCH, DDT with metabolites, etc.) and other hazardous compounds from the US EPA lists of priority pollutants was specially checked. Fortunately only few chemicals from these lists were detected. MAC value for the sum of hydrocarbons is 50 mkg/1. Table 1 shows that it was exceeded in all the samples. Samples 5 and 4 collected in the vicinity of the factory, were twice more contaminated than the others. The majority of these hydrocarbons are readily biodegradable. They will be destroyed by sunlight and bacteria in spring. Thus there is no direct danger for the environment so far. However petroleum hydrocarbons compose a portion of the factory exhausts and should be controlled on the regular basis in the environment.

Similar conclusions may be derived for organic acids and alcohols. MAC values were not elaborated for these chemicals. This fact hinders the evaluation of their environmental danger. However it is also known that these compounds are not highly toxic and readily degradable.

An important group of organic toxicants includes antioxidant 2,6-ditertbutyl-4-methylphenol (ionol), its by-products and degradation products. Actually these compounds can be found worldwide being added to plastic materials and other products. They are quite stable and may be accumulated in organisms under constant exposure. Fortunately they are not very toxic. The levels of the compounds of this group were not high and they cannot be connected only with the activity of the factory since the lowest level was found in sample 4. Although the major concentrations of phthalates were detected for samples 5 and 4, it is not correct to treat the factory as the only source of these toxicants. Phthalates are used as plasticizers all over the world. Being constituents of plastics they appear in environment due to the most various sources. They are reasonably volatile and rather stable. Actually it is difficult to collect an environmental sample free of these chemicals. MAC values exist for dimethylphthalate, dibutylphthalate and bis(2-ethylhexyl)phthalate, being 500, 1 and 10 mkg/1 correspondingly. The observed levels of phthalates make them regional priority pollutants.
No reliable conclusion could be drown for polycyclic aromatic hydrocarbons (PAH). These compounds are included into the lists of priority pollutants all over the world. The detected levels of these chemicals demonstrate that they are spread randomly over the studied territory. The overall amount of PAH was higher for samples 4-6, however this was mainly due to naphthalene. Concentrations of other PAH were almost equal for all the samples. Transport could be one of the principal sources of these compounds. The factory may be blamed for the environmental contamination with naphthalene, although its level was significantly lower than MPC value.

On the contrary phenols appeared in the environment due to the activity of the factory. Although only two representatives of this group were detected their levels exceeded MAC value, which is 1 mkg/l. The highest levels were measured for samples 5 and 4, which were collected in the vicinity of the factory. In the other samples the presence of these toxicants was not so pronounced while they were not detected at all in sample 1. Although being very toxic phenols are not stable enough in the environment. Therefore they hardly may do any serious harm to the nature in such amounts after the snow melts. Nevertheless similarly to petroleum hydrocarbons they should be controlled on the regular basis.

Summarizing the results on the influence of the factory on the environment it is possible to conclude that the exhausts of Kostomuksha factory contaminate the surrounding territory with Li, B, Mn, Fe, Co, Ni, Cu, Zn, Mo, Sn, Pb, petroleum hydrocarbons, organic acids, phthalates and phenols. These contaminants may be treated as priority pollutants for the region. On the other hand these ecotoxicants may be divided into two groups. The first one contains Li, Mn, Fe, Ni, Cu, Zn, Mo, phenols and phthalates; the second - B, Co, Sn, Pb, hydrocarbons and organic acids. Concentration of the toxicants in the first group exceeds the established MAC values. It is necessary to carry on an extensive search to decrease the amount of these ingredients in the factory exhausts. The second group contains toxicants which levels are not high enough to trigger some negative changes. Nevertheless these chemicals should be controlled on the regular basis as priority pollutants. The list of priority pollutants for the region including their MAC values and their levels in the samples is presented below as Table 1.

Mercury should be definitely included into the list of priority pollutants for the region, as its concentration in all the Karelian samples significantly exceeded the MAC value. However Kostomuksha factory may hardly be treated as the only source of mercury in the environment. There should be other sources. Similar situation is with aluminum. The MAC values for this metal are exceeded, but the presence of this metal in the snow samples cannot be connected only with the functioning of the factory. There should be other sources of this metal.

The most contaminated site lies toward the north-east from the factory. The sample collected at the same distance (4 km) from the factory to the opposite direction was less contaminated. The Finnish sample collected near the Russian border did not contain any of the priority pollutants emitted by the factory at significant level. Actually these chemicals were detected at the background level. On the basis of the results obtained it is possible to conclude that the impact of the exhausts of Kostomuksha factory on the environment in Finland is relatively
small. Even the increased concentrations of Zn and Cu exceeding MAC values cannot be connected only with the activity of the factory in Kostomuksha, as in other samples (7-10) collected in Finland at the same time the levels of these metals were much higher (Table 2).

3.2 Finland

In contrast to the Karelian samples the Finnish ones covered a vast territory more than 1000 km from the North to the South. There are many different sources of air pollution emitting a great variety of chemicals. Nevertheless it is possible to make some estimations and to select chemicals of special concern.

3.2.1 Metals

Contamination of the Finnish territory with metals differed significantly in comparison with that of the Karelian Kostomuksha region. For example Li or B were detected in all the Finnish samples at background level and definitely do not represent any danger to the environment. On the other hand Cr should be referred as metal of concern (Table 2).

The most contaminated samples were 7 and 10, representing the extreme northern and southern sampling sites. The metals of concern are Al, Cr, Mn, Fe, Ni, Cu, Zn, Mo, Pb (Table 2).

Concentration of aluminum exceeds both MAC$_f$ and MAC$_{\text{3h}}$ values in all the samples. It is impossible to connect the presence of this metal in the environment with any definite source. It should be also taken into account that the data obtained for this metal could represent an artifact, as the sampling device was made of aluminum.

Chromium concentration in sample 7 comprises 113 ppb (1.6 MAC$_f$), while it is less than 4 ppb for samples 1,8,9. Some industrial exhausts should be responsible for the high level of this metal in the southern region.

Concentration of manganese exceeded MAC$_f$ value in samples 7,9,10, being close to that value for others. Site 9 is rather clean in general, thus the presence of the highest detected level of this metal in that sample (Table 2) is quite difficult to rationalize without additional studies.

MAC$_f$ for iron was exceeded in samples 7,8,10, reaching 7MAC for sample 7. This level is even higher than that detected for the most polluted site in Kostomuksha region (Table 1). The same sample (No 7) contained 17 MAC$_f$ of nickel. Even the MAC$_{\text{3h}}$ value for this metal was exceeded.

All the Finnish samples contained notable amounts of copper, which exceeded its MAC$_f$ value. In this case it is possible to conclude that all the territory is contaminated with copper. On the other hand this increased concentration may represent a natural level. To check this hypothesis another set of samples should be collected. It would be worth to collect water and soil samples as well.

Only sample 9 contained zinc in concentration below MAC$_f$ (Table 2), while the record belongs to sample 10 (5 MAC$_f$).

MAC$_f$ for molybdenum was exceeded only for sample 7 (3.8 MAC$_f$). Thus situation with this metal was much better than in Kostomuksha region. On the other hand the levels of lead appeared to be higher than in Karelian samples. For
the most polluted sites (7 and 10) the MAC\textsubscript{f} value for this very dangerous element was exceeded.

In contrast to Karelian samples silver should be mentioned. Its concentration for 4 samples notably exceeded the background level, which is <0.1 ppb (sample 9). For sample 7 the detected level was 17.4 ppb. Taking into account that MAC\textsubscript{h} value for this metal was 50 ppb, the presence of Ag should be treated with more attention. Similarly it is worth to pay attention to the increased values of yttrium, lanthanium, cerium, uranium and some other rare metals in sample 7 (Table 2). They may appear due to the vicinity of Sankt-Petersburg, or it may come with the exhausts of some Finnish enterprises. To answer these questions another set of samples should be collected on the smaller territory from Sankt-Petersburg to Lahti and Savonlinna in Finland.

Similar conclusion could be derived for sample 10. The influence of the Russian ore refinement factories on Cola peninsula on the increased metal concentrations at this site should be proved by collecting another set of samples in the vicinity of the factories and along the way from the factories toward Inari lake.

### 3.2.2 Organic pollutants

The levels of organic pollutants in the Finnish samples were rather low. Similarly to metals the most polluted samples were 7 and 10. The MAC\textsubscript{f} value for hydrocarbons is slightly exceeded only for sample 7 collected in the South-Eastern corner of Finland. Taking into account low stability of the majority of these compounds there is no serious danger for the environment. Similar conclusions may be derived for organic acids, alcohols and antioxidants. These chemicals are not toxic. Besides they were detected in smaller amounts than in the case of the Karelian samples. More dangerous phenols were not detected in the Finnish samples at all.

The levels of phthalates were lower in the Finnish than in the Karelian samples. They spread rather randomly over the territory of sampling with usual maximums at sites 7 and 10. Very low MAC\textsubscript{f} value for the most widespread dibutylphthalate allows to consider this compound as a pollutant of concern, as its measured levels were higher for all the samples. Without mentioning the sources of these chemicals in the environment it is necessary to treat them as regional priority pollutants.

### 3.2.3 "Cold finger" effect

Trying to detect "cold finger" effect we expected to detect increased levels of toxicants in sample 10, collected in the most northern site. Actually the levels of some metals as well as of alcohols and phthalates were higher for this site. However in all these cases the difference with other samples was not clearly pronounced. Nothing definite could be proposed for metals, since the vicinity of the industrial region at Cola peninsula could be a cause of their increased levels. Similarly alcohols are not considered to be environmental pollutants and the sources of their appearance in the samples are not obvious. The case of phthalates may be treated as a possible example of the "cold finger" effect. They are quite volatile and persistent enough to be transported with air to the northern territories. It was already mentioned above that these pollutants are widely
Air Pollution X spread all over the world. To prove the "cold finger" effect another sites for sample collection should be selected. These sites should be far away from the industrial or agricultural enterprises, cities, villages or roads. On the contrary site 10 was selected first of all to estimate the influence of the ore refinement factories on the Cola peninsula. Hence it contains a notable contribution of chemicals coming from the east rather than from the south.

4 Conclusions

1. The exhausts of the Karelian Kostomuksha factory contaminate the environment with Li, B, Mn, Fe, Co, Ni, Cu, Zn, Mo, Sn, Pb, petroleum hydrocarbons, organic acids, alcohols, phthalates and phenols.
2. The mentioned ecotoxicants together with Hg and Al coming from unknown sources could be treated as priority pollutants for the region.
3. Li, Mn, Fe, Ni, Cu, Zn, Mo, Al, Hg, phenols and phthalates are the pollutants of major concern since their concentration in the snow samples exceeds the safe levels (MAC values).
4. Due to the predominant south-west winds the most contaminated site lies toward the north-east from the factory. The levels of contaminants in the opposite direction are lower and decrease towards the Finnish border. As a result the impact of the exhausts of the Kostomuksha factory on the environment in Finland is relatively small.
5. The long distances between sampling sites in Finland do not allow to draw definite conclusions about the sources of contaminants in the samples. Smaller regions should be selected for this purpose. Al, Cr, Mn, Fe, Ni, Cu, Zn, Mo, Pb, and phthalates may be treated as contaminants of concern.
6. The case of phthalates may be treated as a possible example of the "cold finger" effect. However, to prove the effect reliably another sites for sample collection should be selected. These sites should be far away from the industrial or agricultural enterprises, cities, villages as well as roads.

5 References and notes

Table 1. The list of priority pollutants (levels in ppb) for Kostomuksha region

<table>
<thead>
<tr>
<th>N</th>
<th>Element</th>
<th>MAC&lt;sub&gt;f&lt;/sub&gt;</th>
<th>MAC&lt;sub&gt;ph&lt;/sub&gt;</th>
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<th>2</th>
<th>3</th>
<th>4</th>
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**DBP** - dibutyl phthalate  
**BEHP** - Bis(2-ethylhexyl) phthalate

Table 2. The list of pollutants of concern (levels in ppb) for Finland.

<table>
<thead>
<tr>
<th>N</th>
<th>Element</th>
<th>MAC&lt;sub&gt;f&lt;/sub&gt;</th>
<th>MAC&lt;sub&gt;ph&lt;/sub&gt;</th>
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**DBP** - dibutyl phthalate  
**BEHP** - Bis(2-ethylhexyl) phthalate