

CONTAMINATION BY PERSISTENT ORGANIC POLLUTANTS AND HEAVY METALS



IN THE SURROUNDINGS OF
THE WASTE TREATMENT
FACILITY HŮRKA

Václav Mach



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Mgr. et Mgr. Václav Mach, Ph.D.





Summary

We carried out a single monitoring of presence of persistent organic pollutants and heavy metals in the surroundings of the waste treatment facility Hůrka. The purpose of the monitoring was to obtain data on pollution in the surroundings of the premises of the facility of interest, and on possible origin of the pollution. The waste disposal facility Hůrka is operated by the company Quail spol. s r. o., carrying out biodegradation and stabilisation of waste there. The resulting output are certified products serving as a filling layer under a biological layer, or for direct reclamation of sludge-drying beds, mines, and waste landfills. The monitoring focused on polychlorinated biphenyls, dioxins, polycyclic aromatic hydrocarbons, and heavy metals, in samples of sediments taken in the surroundings of the facility of interest. Further, assessment of wastes accepted by the facility was carried out from the point of view of presence of persistent organic pollutants, on the basis of the waste records, and the overall balance of inputs and outputs concerning dioxin contents was calculated.

From a comparison of the measured concentrations of contaminants with reference sites and/or with long-term average concentrations measured in a number of various sites, it followed that many-times higher concentrations of all the monitored contaminants were present in the site of interest. In the case of dioxins, the concentrations were by one to two orders of magnitude higher, in comparison with reference values of an unburdened background site in Košetice. The concentrations of polychlorinated biphenyls and polycyclic aromatic hydrocarbons, found in the taken samples, were comparable with the values from highly burdened sites - with the Elbe river sediments in Ústí nad Labem (polychlorinated biphenyls) and the Černý Potok stream sediments in Ostrava (polycyclic aromatic hydrocarbons). By comparing the measured contaminant concentrations with legislative criteria, it was found that concentrations of substances ranked among polycyclic aromatic hydrocarbons and dioxins, and, further, of arsenic, lead, and antimony, exceeded indicators of soil pollution for other areas, in at least one of the places where samples were taken.

The waste treatment facility Hůrka treats waste containing the monitored contaminants. Releases of the stored materials may take place, and took place occasionally, from the facility in question. The occurrence of the contaminants in the individual places where samples were taken suggests that material was transported in the direction away from the facility of interest. In the surroundings of the site, no other potential source of the monitored contaminants is known. From these reasons, a conclusion may be drawn that the source of the contaminants found in the taken sediment samples was, with the highest likelihood, the waste treatment facility Hůrka. This conclusion is in accordance with the previous results of the Arnika Association from 2009, 2010, 2012, and 2014, presented in the study entitled „Pollution by POPs in the Surroundings of the Quail spol. s.r.o. Facility, Hůrka near Temelín“.

According to the records, fly ash from flue gas treatment from municipal and hazardous waste incinerators was accepted into the waste treatment facility Hůrka in 2014 and 2015. The PCDD/F concentrations in the fly ash from hazardous waste incinerators were in the range of 15,000 – 100,000 ng I-TEQ/kg. From this, it follows that the waste treatment facility accepted wastes exceeding the „low POPs content“ for PCDD/Fs, namely its current value set according to the Article 6 of the Stockholm Convention.

For the period of 2014 and 2015, the estimated amounts of PCDD/F inputs into the facility were in total 32.67 – 33.5 g I-TEQ. The estimated amount of PCDD/Fs leaving the facility of interest in its certified product was 3.62 – 4.02 g I-TEQ, as a sum for the both years. Thus, the total estimated input of PCDD/Fs was eight-times higher than the estimated output in the certified product, in the same period. The fate of the remaining 28.65 – 29.88 g I-TEQ of PCDD/Fs is not clear, in 2014 and 2015.

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1. Introduction

Persistent organic pollutants (POPs) and heavy metals are contaminants to which considerable attention is paid. The reason is, in addition to their toxic properties, in particular their long-term persistence, and, in some cases, also their accumulation in the environment. Persistent organic pollutants are organic compounds that are, under common conditions, very resistant to decomposition. The half life, depending on the specific compound, may be in the order of several years to decades. For these reasons, the most problematic persistent organic pollutants are regulated at the international level. Heavy metals rank among the most common environmental contaminants originating from anthropogenic activities. If released into the soil, they may persist hundreds of years there.

In addition to high resistance to decomposition, persistent organic pollutants are characterised by certain other common properties, such as the ability of bioaccumulation and biomagnification. These properties were observed also in the case of certain heavy metals. Bioaccumulation occurs when a living organism absorbs a potential contaminant into its body more quickly than it is removed by decomposition and excretion. The individual persistent organic pollutants and heavy metals show different abilities to accumulate in living tissues, depending on their chemical properties. [1] [2] [3] Biomagnification occurs when the concentration of a potential contaminant grows with the growing trophic level of the organism in question. It is, essentially, growth of a pollutant's concentration in the food chain. The highest concentrations of a number of contaminants, such as dioxins (PCDD/Fs), mercury, and polychlorinated biphenyls (PCBs), were found in bodies of organisms at the top of the food chain, where human beings are placed, too. [4] [5] [6]

Due to the ability of long-range transport, PCBs and PCDD/Fs, produced almost solely by human activities, are practically ubiquitous in very low concentrations all over the world nowadays, and are present even in the areas where industrial processes, during which they are produced, were never operated, and where these substances were not used either. [7] [8] [9] In contrast with that, polycyclic aromatic hydrocarbons (PAHs) and heavy metals form part of the environment, too, but human activities contribute significantly to increasing their concentrations in various environmental components. [10] [11]

There was found that PCBs, PCDD/Fs, PAHs, and heavy metals, pose a number of health risks for humans, such as teratogenicity, neurotoxicity, carcinogenicity, disruption of hormonal activities, and damage to internal organs. [12] [13] [14] For the above-mentioned reasons, PCBs, PCDD/Fs, and certain other persistent organic pollutants, were included, in 2001, into a list of substances manufacturing (in the case of PCBs and certain other substances) and unintentional production of which is regulated by the Stockholm Convention, representing an international legally binding agreement for prevention of spreading persistent organic pollutants. The occurrence of PAHs and heavy metals is monitored and regulated by a number of national legislative regulations, setting also their limit values in various environmental components and consumer goods. In spite of the fact that PAHs are not regulated by the Stockholm Convention, they belong among persistent organic pollutants subject to the international Convention on Long-range Transboundary Air Pollution (CLRTAP), namely its Protocol on POPs. [15] From the heavy metals, the highest attention is paid to heavy metals that are also regulated by the Convention on Long-range Transboundary Air Pollution (CLRTAP), namely its Protocol on Heavy Metals. The internationally legally binding

Minamata Convention focuses on one heavy metal only, mercury, and regulates its emissions from human activities.

Waste containing persistent organic pollutants and heavy metals is produced by a long number of human activities. Such kinds of waste may be products of waste incineration and flue gas treatment (ash, slag, fly ash, dust captured in separators and filters), waste water treatment sludge, and a number of other kinds of waste that may subsequently represent a risk for the environment. In order to reduce hazardous properties of these kinds of waste, various specialised facilities are operated, working at the basis of reducing hazardous properties of the waste to the level that is acceptable according to the legislation.

Waste treatment facility Hůrka is one of the facilities utilising waste containing persistent organic pollutants and heavy metals. In the facility, the waste is further reprocessed, and certified products are manufactured of it, serving subsequently for reclamation of sludge-drying beds, mines, and waste landfills. Products manufactured in the facility are used, in particular, for reclamation of sludge-drying beds in Mydlovary near České Budějovice, where the state company for uranium ore treatment, MAPE, was in operation in 1962 – 1991. At present, lagoons in Mydlovary are being filled by various materials, including mixtures produced in the waste treatment facility Hůrka. According to the estimates of the Arnika Association of 2004, up to a quarter of all fly ash from the Czech incinerators ended in the Mydlovary lagoons. Treatment of fly ash and other kinds of waste containing toxic substances, and their subsequent incorporation into construction materials and reclamation mixtures, represent a potential contamination source in the surroundings of the treatment facilities, as well as of the constructions where such reprocessed waste is used, and it should be controlled consistently. [16] [17]

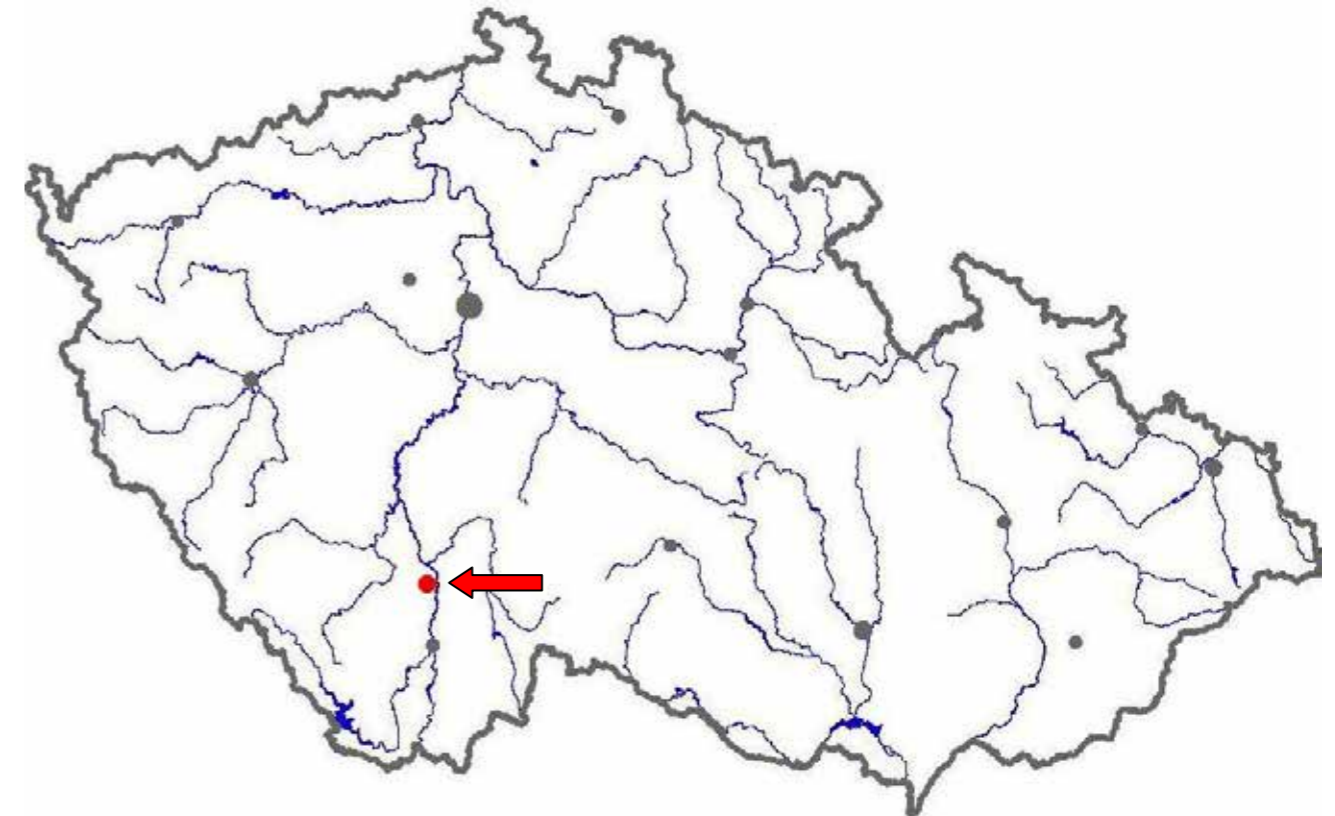
This study summarised results of a single monitoring of persistent organic pollutants and heavy metals in the sediments sampled in the surroundings of the waste treatment facility Hůrka in 2016. Its purpose was to examine pollution in the surroundings of the premises of the waste treatment facility Hůrka, and to verify possibilities of releases of persistent organic pollutants and heavy metals from the facility. A further purpose of the study was to verify validity of certain conclusions of former investigations carried out in 2009, 2010, 2012, and 2014, results of which were summarised in the study „Pollution by POPs in the Surroundings of the Quail spol. s.r.o. Facility, Hůrka near Temelín“. Further, the study aimed to make and assess a balance of inputs into, and outputs from, the waste treatment facility Hůrka concerning PCDD/Fs and other persistent organic pollutants, namely PCBs, hexachlorocyclohexane (HCH), tetrachlorobenzene (TCB), pentachlorobenzene (PeCB), hexachlorobenzene (HCB), and hexachlorobutadiene (HCBD). This study forms part of long-term activities of the Arnika Association in the field of protection of the environment against toxic substances, and implementation of the Stockholm Convention.

2. Site

The site of interest, the waste treatment facility Hůrka, is found in the South Bohemian Region, approximately 40 km to the north from České Budějovice and 5 km to the south-west from Týn nad Vltavou. Localisation of the site on the map of the Czech Republic is given in Figure 1. The premises of the waste treatment facility Hůrka are located in isolation in the cadastral territories Březí and Knín in the municipality Temelín. The premises are found in a slightly rolling landscape in the altitude of 465 m above sea level. The terrain in the surroundings of the premises has a slight slope

to the south. The municipality closest to the site of interest is Litoradice, located 1.5 km to the north-east from the facility of interest. The nuclear power plant Temelín is located approximately 1.5 km to the north-west, and the former municipality Knín, abandoned nowadays, was located about 1 km to the west. The fortress Býšov, a historic building and a tourist destination, is located about 1 km to the south from the facility.

Figure 1: Localisation of the waste treatment facility Hůrka on the map of the Czech Republic. The site is marked red.



The close surroundings of the premises of the facility of interest are formed predominantly by agricultural land with small forested areas and several fish-farming ponds. The ponds Starý, Barbora, and Pohrobný, are located close to each other in the distance of about 500 m to the south from the premises of the facility, in the direction of natural flow of surface water. In the direction from the north to the south, a small stream of the size of a drain, having no name, flows through the ponds, and its two branches flow past the premises of the facility. The branch flowing past the west side of the premises of interest communicates with the sedimentation tank of the facility of interest. The ponds Starý, Barbora, and Pohrobný, are then further drained by small streams (Strouha, Rachačka) into the river Vltava.

The waste disposal facility Hůrka is operated by the company Quail spol. s r. o., carrying out biodegradation and stabilisation of waste there. The premises of interest are surrounded with a fence and have an area of approximately 3.5 hectares. The majority of the facility premises is formed by buildings and stabilised areas. On the stabilised areas, there are found a decontamination area with lagoons (1.9 hectares), a line for intensifying waste biodegradation, a facility for collection and purchase of waste, a washing platform, a filling station, above-ground tanks of technological water, a mobile emergency station, a digital weighbridge, a sewerage and a sewage water tank, an operation building, propane storage tanks, and internal service roads. The

rest of the facility premises is formed by lawns, a fire water reservoir, and a technological drainless tank. A sedimentation tank is located outside the area surrounded with a fence, to the south of the edge of the premises of interest. The premises of the facility in question are depicted in Figure 2.

Figure 2: Aerial photograph of the waste treatment facility – Hůrka. Source: mapy.cz



The facility serves for utilisation of hazardous and other waste through reprocessing to products, using the biodegradation method (waste contaminated, in particular, by hydrocarbons), and the stabilisation method (waste contaminated, in particular, by heavy metals). Further, physical treatment of waste through crushing and sizing on a mobile crusher is carried out in the facility occasionally. The resulting products are certified products designated Q.I.-1 and Q.I.-2 (loose filling materials) serving as a filling layer under a biological layer, or for direct reclamation of sludge-drying beds, mines, waste landfills, etc. A detailed description of the technological processes is given in the Decision on an Application for Granting an Integrated Permit. [18]

Biodegradation of waste containing hydrocarbons, polycyclic aromatic hydrocarbons, and phenols, is carried out through spraying by a solution of microbial inoculum in the decontamination area. In particular, contaminated soils and surface-contaminated concrete, crushed in advance in the technological area, are treated in this way. Waste stabilisation is carried out by a number of physical and chemical processes, with the aim to prevent or slow down transfer of hazardous contaminants into the environment when the treated waste is used as a raw material. The waste is crushed and mixed, in order to amend the water content, with binders and fillers (fly ash, lime) and water. Stabilisation of fly ash is carried out in an isolated mixing station, where fly ash is filtered, and sprayed by technological water, by means of a moistening screw. The resulting product is

moistened fly ash, stored in storage areas for subsequent stabilisation of other kinds of waste. Stabilisation of liquid waste is carried out using moistened fly ash in an accessory lagoon.

3. Methods

All the sediment samples were taken during an only visit to the site of interest, on September 12, 2016. During the visit, three sediment samples were taken in the site, in three sampling places, by a worker of the company ALS Czech Republic s.r.o., a holder of a certificate on accreditation for sampling of earths, sediments, and soils. A list of the taken samples, including basic data on the sampling and geographic coordinates, is given in Table 1. Each of the samples was composed of 3 to 6 partial samples. The partial samples were taken from the bottom using a scoop, a sampling shovel, a beaker, and a telescopic bar. The individual partial samples were taken randomly in irregular distances. The partial samples from each of the sampling places were placed into a homogenisation vessel and they were homogenised there. More detailed data on taking the individual samples are given in the Protocols on Sampling of Bottom Sediments. After sampling, the samples were stored in a mobile insulated box with cooling inserts, in which the samples were transported into the laboratory. A part of the samples was transported by a passenger car into the laboratory ALS Czech Republic s.r.o., and another part of the samples was transported into the laboratory Axys Varilab s.r.o.

Table 1: List of taken samples and basic data on them

| Working title of the sample | Sample designation | Sampling protocol number | GPS | Description |
|-----------------------------|-----------------------|--------------------------|----------------------------|---|
| Above the Tank | sediment HÚ 2016 I. | 029/MAR/2016 | N 49.1654928, E 14.4018969 | under the hill of the facility Hůrka, sediment at the foot of the hill from the facility premises, wet area above the sedimentation tank |
| Western Stream | sediment HÚ 2016 II. | 030/MAR/2016 | N 49.1650683, E 14.4019936 | watercourse downstream of the sedimentation tank discharge (flowing past the west side of the premises, communicating with the tank), sediment from the stream bottom |
| Eastern Stream | sediment HÚ 2016 III. | 031/MAR/2016 | N 49.1652544, E 14.4021008 | second branch of the watercourse, not in contact with the sedimentation tank (flowing past the east side of the premises), sediment from the bottom of the clogged stream channel |

Concentrations of seven indicator PCB congeners (I-PCBs), concentrations of twelve dioxin-like PCB congeners (DL-PCBs), concentrations of seventeen toxicologically important dioxin congeners (PCDD/Fs), concentrations of sixteen homologues of polycyclic aromatic hydrocarbons (PAHs), and concentrations of, in total, twenty heavy metals, were determined in the taken samples. Chemical analyses for determination of concentrations of I-PCBs, DL-PCBs, PCDD/Fs, and PAHs, in the taken samples were carried out in the accredited laboratory Axys – Varilab spol. s.r.o. in Vrané nad Vltavou. The analyses in the laboratory Axys – Varilab spol. s.r.o. were carried out by the gas chromatography / high resolution mass spectrometry method, using the mass spectrometer

Autospec Ultima. All the analyses carried out in the laboratory Axys – Varilab spol. s.r.o. were made by accredited tests. Chemical analyses for determination of concentrations of twenty heavy metals were carried out in the laboratory ALS Czech Republic s.r.o. in Prague. Analyses for determination of metals in the laboratory ALS Czech Republic s.r.o. were carried out by the inductively coupled plasma emission spectrometry method, made after sample homogenisation and mineralisation by aqua regia. Hexavalent chromium was determined by ion chromatography with spectrophotometric detection. All the analyses carried out in the laboratory ALS Czech Republic s.r.o. were made by accredited tests, too. More detailed data on the used analytical methods are given in the Test Protocols.

Because different DL-PCBs and PCDD/Fs show different toxicity, they were converted using toxic equivalency factors (TEFs). For this purpose, international toxic equivalency factors (I-TEFs) were used, preferred for abiotic matrices. [19] The toxicity equivalency factors express the level of toxicity of the specific PCB congener, or PCDD/F congener, in relation to the most toxic PCDD/F congener, namely 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD). The resulting values are expressed in international toxic equivalents (I-TEQs), enabling comparison of toxic effects of samples having different composition of the individual PCB and PCDD/F congeners. [20]

In order to calculate the overall toxicological balance of the waste treatment facility Hůrka, the estimates of total inputs (E_n) and outputs (E_x) into, and out of, the facility were calculated for PCDD/Fs and other persistent organic pollutants within a specific time period. Weights of waste obtained from the individual waste suppliers were used as an input information for calculating the estimate of inputs in the monitored period, according to the Report on Waste Production and Management the operator has to draw up according to Decree No. 383/2001 Coll. Data on concentrations of PCDD/Fs and other persistent organic pollutants in the waste inputs from various suppliers were obtained from Protocols on Chemical Analyses, provided by the Regional Authority of the South Bohemian Region, and the Czech Environmental Inspectorate. The estimates of inputs into the facility, concerning PCDD/Fs and other persistent organic pollutants in the waste, were calculated using the following formula:

$$E_n = \sum m_i * c_i$$

wherein

E_n is the estimated weight of a specific contaminant that entered the waste treatment facility Hůrka together with the waste, in the monitored period. In the case of PCDD/Fs, it is expressed in grams of I-TEQ, in the case of the other persistent organic pollutants (HCH, TCB, PeCB, HCB, PCBs, and HCBD) in grams.

m_i is the weight of waste from the supplier i , used for the production of the certified product in the monitored period. It is expressed in tons of waste.

c_i is the concentration of the specific contaminant in the waste from the supplier i , used for the production of the certified product. In the case of PCDD/Fs, it is expressed in mg I-TEQ/kg of dry matter, and in the case of the other persistent organic pollutants (HCH, TCB, PeCB, HCB, PCBs, and HCBD) in mg/kg of dry matter.

The estimated weight of the product, according to Sampling Protocols, and concentrations of PCDD/Fs and other persistent organic pollutants, according to Protocols on Chemical Analyses,

were used as input information for calculating the estimate of outputs in the monitored period. The Sampling Protocols, and the Protocols on Chemical Analyses, were provided by the Regional Authority of the South Bohemian Region. According to the Protocols on Chemical Analyses, concentrations of HCH, TCB, PeCB, HCB, PCBs, and HCBD, were below the detection limits. Thus, the estimated output was calculated for PCDD/Fs only. The estimate of output of PCDD/Fs out of the facility in waste was calculated using the following formula:

$$E_x = \sum m_i * c_i$$

wherein

E_x is the estimated weight of a specific contaminant that left the waste treatment facility Hůrka in the certified product, in the monitored period. It is expressed in grams of PCDD/F I-TEQ.

m_i is the weight of the certified product batch i , production of which was finished in the monitored period. It is expressed in tons of the product.

c_i is the concentration of the specific contaminant in the certified product batch i , that left the waste treatment facility Hůrka in the monitored period. It is expressed in mg PCDD/F I-TEQ/kg of dry matter.

Due to the nature of the processed input data, the estimates of the inputs of contaminants in waste could be calculated for the individual years 2014 and 2015 separately, but the estimate of the outputs in the product was calculated in total for the two-year period of 2014 and 2015. The estimates of PCDD/F input and output into/out of the facility were calculated in variants, as the minimum estimated input/output and the maximum estimated input/output, on the basis of the lower and upper values of the results of chemical analyses. If data on contaminant concentrations were not available for the year in question, available data for another period were used, that caused a higher error level of the resulting estimate.

4. Results

Concentrations of the seven I-PCB congeners in the taken sediment samples were in the range from 110 to 155.6 µg/kg of dry matter. The I-PCB concentrations found in the individual sediment samples are given in Table 2. The pattern of the individual I-PCB congeners in the samples is shown in Graph 1. The concentrations of the DL-PCB congeners in the individual sediment samples are given in Table 3. The pattern of the individual DL-PCB congeners in the samples is shown in Graph 2. The concentrations of the PCDD/F congeners in the individual sediment samples are given in Table 4. The pattern of the individual PCDD/F congeners in the taken samples is shown in Graph 3. The sum of I-TEQ for DL-PCBs and PCDD/Fs in the taken sediment samples was in the range from 409.8 to 566.8 ng/kg of dry matter. Toxic effects, I-TEQs, concerning DL-PCBs and PCDD/Fs, and their total sum in the taken sediment samples, are summarised in Table 5. The concentrations of the sixteen PAH homologues in the taken sediment samples were in the range from 107,950 to 6,298 µg/kg of dry matter. The concentrations of the PAH homologues in the individual sediment samples are given in Table 6. The pattern of the individual PAH homologues in the samples is shown in Graph 4. The concentrations of the heavy metals in the individual sediment samples are given in Table 7.

Table 2: I-PCB concentrations in the sediment samples in µg/kg of dry matter.

| | Above the Tank [µg/kg of dry matter] | Western Stream [µg/kg of dry matter] | Eastern Stream [µg/kg of dry matter] |
|------------------------|---|---|---|
| PCB 28 | 24 | 53 | 9 |
| PCB 52 | 3.1 | 20 | 3.9 |
| PCB 101 | 1.6 | 4.7 | 1 |
| PCB 118 | 7.3 | 28 | 7.9 |
| PCB 153 | 20 | 8.1 | 48 |
| PCB 138 | 32 | 19 | 85 |
| PCB 180 | 22 | 3.3 | 0.77 |
| ∑ 6 PCBs ¹⁾ | 102.7 | 108.1 | 147.67 |
| ∑ 7 PCBs ²⁾ | 110 | 136.1 | 155.57 |

¹⁾ The designation ∑ 6 PCBs means the sum of six congeners, PCB 28, PCB 52, PCB 101, PCB 138, PCB153, and PCB 180.

²⁾ The designation ∑ 7 PCBs means the sum of seven congeners, PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB153, and PCB 180.

Graph 1: Pattern of I-PCB congeners in the taken sediment samples in %

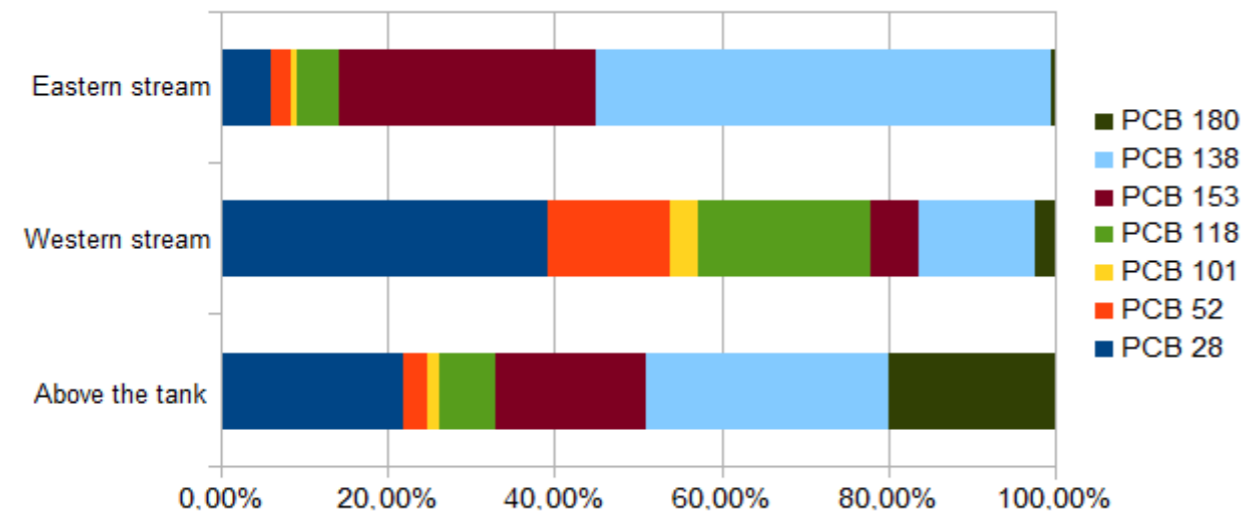


Table 3: DL-PCB concentrations in the sediment samples in µg/kg of dry matter.

| | Above the Tank [µg/kg of dry matter] | Western Stream [µg/kg of dry matter] | Eastern Stream [µg/kg of dry matter] |
|---------|---|---|---|
| PCB 81 | 0.046 | 0.16 | 0.04 |
| PCB 77 | 2.4 | 3 | 0.88 |
| PCB 126 | 0.09 | 0.11 | 0.027 |
| PCB 169 | 0.096 | 0.0068 | < 0.002 |
| PCB 123 | 0.65 | 0.23 | 0.16 |
| PCB 118 | 7.3 | 28 | 7.9 |
| PCB 114 | 0.081 | 1.6 | 0.42 |
| PCB 105 | 2.7 | 15 | 4.4 |
| PCB 167 | 1 | 0.9 | 3.4 |
| PCB 156 | 3 | 3.3 | 19 |
| PCB 157 | < 0.02 | 0.83 | 5.2 |
| PCB 189 | 0.19 | < 0.03 | < 0.02 |

Graph 2: Pattern of DL-PCB congeners in the taken sediment samples in %.

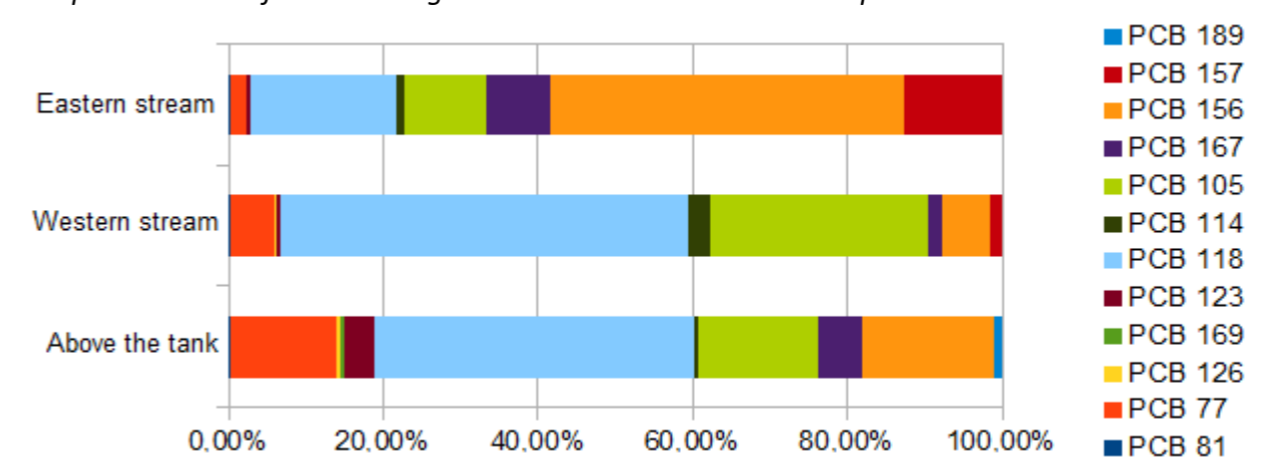


Table 4: PCDD/F concentrations in the sediment samples in ng/kg of dry matter.

| | Above the Tank [ng/kg of dry matter] | Western Stream [ng/kg of dry matter] | Eastern Stream [ng/kg of dry matter] |
|--------------------------|---|---|---|
| 2378-TeCDD | 14 | 1.7 | 0.76 |
| 12378-PeCDD | 32 | 4.1 | 2.2 |
| 123478-HxCDD | 57 | 5.7 | 1.2 |
| 123678-HxCDD | 100 | 8 | 2.1 |
| 123789-HxCDD | 85 | 7 | 2 |
| 1234678-HpCDD | 130 | 68 | 11 |
| OCDD | 4300 | 310 | 44 |
| 2378-TeCDF | 190 | 27 | 9 |
| 12378-PeCDF | 190 | 43 | 8.4 |
| 23478-PeCDF | 560 | 98 | 16 |
| 123478-HxCDF | 53 | 20 | 12 |
| 123678-HxCDF | 32 | 17 | 8.7 |
| 234678-HxCDF | 69 | 13 | 8.3 |
| 123789-HxCDF | 7.1 | 6.8 | 1.6 |
| 1234678-HpCDF | 410 | 56 | 13 |
| 1234789-HpCDF | 53 | 6.2 | 0.9 |
| OCDF | 8200 | 39 | 8.8 |
| ∑ 3 HxCDDs ¹⁾ | 242 | 20.7 | 5.3 |

¹⁾ The designation ∑ 3 HxCDDs means the sum of the three dibenzo-p-dioxin congeners having six chlorine atoms (hexachloro-dibenzodioxins) that were determined, specifically, 123478-HxCDD, 123678-HxCDD, and 123789-HxCDD.

Graph 3: Pattern of PCDD/F congeners in the taken sediment samples in %.

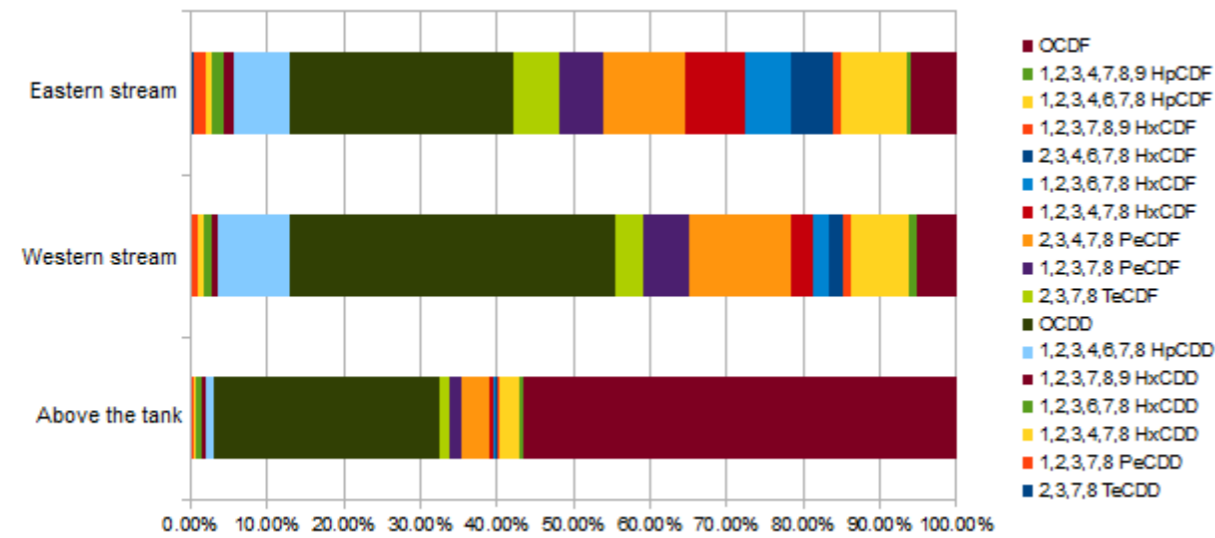


Table 5: Toxic effects, I-TEQs, of PCDD/Fs and DL-PCBs in the sediment samples in ng/kg of dry matter.

| | Above the Tank [ng/kg of dry matter] | Western Stream [ng/kg of dry matter] | Eastern Stream [ng/kg of dry matter] |
|-------------------------------|---|---|---|
| DL-PCB I-TEQ | 149.9 | 499.8 | 394.7 |
| PCDD/F I-TEQ | 397.2 | 67 | 15.1 |
| Σ I-TEQs ¹⁾ | 547.1 | 566.8 | 409.8 |

¹⁾ The designation Σ I-TEQs means the sum of DL-PCB I-TEQ and PCDD/F I-TEQ.

Table 6: PAH concentrations in the sediment samples in $\mu\text{g}/\text{kg}$ of dry matter.

| | Above the Tank [$\mu\text{g}/\text{kg}$ of dry matter] | Western Stream [$\mu\text{g}/\text{kg}$ of dry matter] | Eastern Stream [$\mu\text{g}/\text{kg}$ of dry matter] |
|--------------------------------|--|--|--|
| Naphthalene | 3 100 | 2 300 | 190 |
| Acenaphthylene | 570 | 100 | 55 |
| Acenaphthene | 4 500 | 450 | 110 |
| Fluorene | 3 400 | 480 | 120 |
| Phenanthrene | 7 800 | 1 100 | 320 |
| Anthracene | 3 400 | 300 | 110 |
| Fluoranthene | 17 000 | 2 000 | 880 |
| Pyrene | 56 000 | 1 300 | 2 900 |
| Benzo[a]anthracene | 3 500 | 440 | 360 |
| Chrysene | 2 300 | 390 | 240 |
| Benzo[b]fluoranthene | 920 | 170 | 150 |
| Benzo[k]fluoranthene | 590 | 120 | 110 |
| Benzo[a]pyrene | 2 300 | 360 | 340 |
| Indeno[1,2,3-c,d]pyrene | 1 000 | 190 | 160 |
| Benzo[g,h,i]perylene | 1 300 | 290 | 200 |
| Dibenzo[a,h]anthracene | 270 | 35 | 53 |
| Σ 16 PAHs ¹⁾ | 107950 | 10025 | 6298 |
| Σ 12 PAHs ²⁾ | 99210 | 8960 | 5960 |

¹⁾ The designation Σ 16 PAHs means the sum of all the PAH homologues that were determined and listed in the table..

²⁾ The designation Σ 12 PAHs means the sum of anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(ghi)perylene, benzo(k)fluoranthene, fluoranthene, phenanthrene, chrysene, indeno(1,2,3-cd)pyrene, naphthalene, and pyrene.

Graph 4: Pattern of PAH homologues in the taken sediment samples in %.

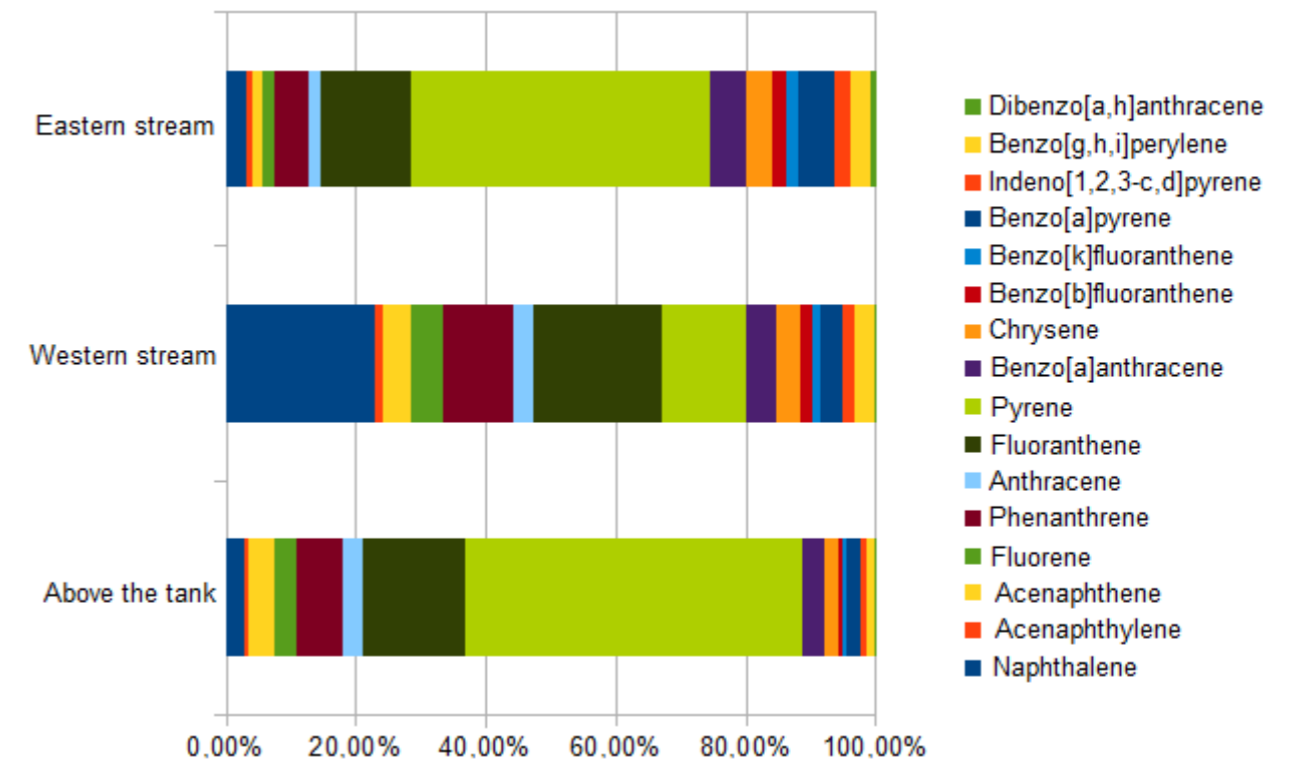


Table 7: Concentrations of heavy metals in the sediment samples in mg/kg of dry matter.

| | Above the Tank [mg/kg of dry matter] | Western Stream [mg/kg of dry matter] | Eastern Stream [mg/kg of dry matter] |
|--|---|---|---|
| Silver (Ag) | 2.62 | < 0.5 | < 0.5 |
| Arsenic (As) | 53.1 | 10.8 | 3.29 |
| Boron (B) | 69 | 6.2 | 2.3 |
| Barium (Ba) | 410 | 224 | 163 |
| Beryllium (Be) | 4.93 | 1.16 | 0.542 |
| Cadmium (Cd) | 20.8 | 2.39 | 0.55 |
| Cobalt (Co) | 15.6 | 12.5 | 8.97 |
| Hexavalent chromium (Cr ⁶) | 0.232 | 0.106 | 0.104 |
| Copper (Cu) | 248 | 52 | 29.7 |
| Iron (Fe) | 32700 | 23800 | 20200 |
| Mercury (Hg) | 2.69 | < 0.2 | < 0.2 |
| Manganese (Mn) | 1070 | 631 | 453 |
| Molybdenum (Mo) | 4.84 | 1.42 | < 0.4 |
| Nickel (Ni) | 75.7 | 38 | 22.4 |
| Lead (Pb) | 497 | 84.3 | 27.6 |
| Antimony (Sb) | 71.5 | 9.67 | 1.58 |
| Selenium (Se) | 2.6 | < 0.2 | < 0.2 |
| Tin (Sn) | 121 | 14.4 | 3.6 |
| Vanadium (V) | 109 | 54 | 39.8 |
| Zinc (Zn) | 3100 | 502 | 160 |

5. Discussion

Interpretation and assessment of importance of the measured concentrations of the monitored substances in the sediments may be carried out on the basis of comparison with reference values. As the reference values, there may serve: 1) concentrations of the monitored contaminants in reference sites or 2) legislative criteria concerning the monitored contaminants. The concentrations of the monitored contaminants in the reference sites may help to interpret whether the concentrations of the monitored contaminants presented in our study are at the usual levels, or whether the site is burdened by significant presence of the monitored contaminants.

5.1 Comparison of Concentrations of the Monitored Contaminants in the Reference Sites

As the reference values, there may serve concentrations of the monitored contaminants: 1) in unburdened sites without significant pollution sources, 2) in sites in areas burdened by industry, but without significant acute contamination by the monitored substances, 3) calculated as an average from a higher number of sites, and 4) in contaminated or significantly polluted sites. All these comparisons may specify the level of pollution of the monitored site more exactly, and determine importance of the potential pollution.

During ten years of measurements (1998 to 2008) in the site Košetice, regarded as an unburdened site, the average found concentration of seven I-PCBs in the watercourse sediment was 2.2 µg/kg of dry matter. [21] If this concentration were used as the reference value, it could be said that all the three taken sediment samples showed at least 50-times higher concentrations of the seven I-PCB congeners than the one found outside areas with industrial facilities and potential pollution sources. From this, it may be concluded that the site of interest is polluted with PCBs. The concentrations of the seven I-PCB congeners in all the three sampling places may be regarded as higher than average also in comparison with the long-term average in sediments of watercourses in the Czech Republic. For the purposes of comparison, there may be used data determined by the Brno branch of the Central Institute for Supervising and Testing in Agriculture that found the average concentration of the sum of the seven I-PCB congeners in sediments of watercourses of 14.3 µg/kg of dry matter between 1995 - 2014. [22] The concentrations of the seven I-PCB congeners in the monitored samples exceeded these average values several-times, too. Thus, the site of interest may be regarded as significantly polluted with PCBs. The I-PCB concentrations in the monitored site were roughly comparable to the values found in the rivers Bílina and Elbe in the surroundings of Ústí nad Labem in the last year (35.6 – 360.7 µg of the seven I-PCBs/kg of dry matter). [23] The above-mentioned rivers rank among the Czech watercourses showing the highest pollution with PCBs. Thus, the PCB concentrations found in the site of interest may be considered significant.

In Košetice, regarded as an unburdened background site for the Central Europe, the PCDD/F concentration of 1.4 ng I-TEQ/kg of dry matter was found in the watercourse sediment. [24] The samples taken in the site of interest showed ten- to hundred-times higher concentrations than this reference value. We would come to a very similar conclusion, if we used, as the reference values, average PCDD/F concentrations in industrial conurbations of Zlín (1.64 ng I-TEQ/kg of dry matter) and Beroun (1.83 ng I-TEQ/kg of dry matter). [24] At best, the lowest PCDD/F I-TEQ values measured in the site of interest were still more than eight-times higher than the above-mentioned

reference background values. From this, it may be concluded that the site of interest is significantly polluted with the presence of PCDD/Fs. In the National Implementation Plan of the Stockholm Convention on Persistent Organic Pollutants [25], the average PCDD/F concentration in soils was regarded to be the result of the monitoring carried out in 38 sites in the Czech Republic in 2001, during which the average value of 3.1 ng I-TEQ/kg of dry matter was found. The present samples exceeded the average values of the monitoring several-times, and the lowest presently found value was at the level of the maximum found by the monitoring (14.3 ng I-TEQ/kg). If we used the reference value for soils, the PCDD/F concentrations in the present sampling places would be ten- to hundred-times higher, too.

During ten years of measurements (1998 to 2008) in the site Košetice, the average found concentration of sixteen PAH homologues in the watercourse sediment was 210 µg/kg of dry matter. [21] If this concentration were used as the reference value, it could be said that it was exceeded many-times in all the three monitored sampling places. In the sampling place showing the lowest concentration of the sixteen PAH homologues (Eastern Stream), it was exceeded almost thirty-times, however, in the sampling place showing the highest concentration of the sixteen PAH homologues (Above the Tank), it was exceeded more than five hundred-times. All the three sampling places showed significantly higher PAH concentrations than the ones found in unburdened sites. However, in comparison with the average concentrations in sediments of watercourses, PAH contamination in the monitored site was not so great. For comparison with the average PAH concentrations in sediments of watercourses, there may serve data found by the Water Research Institute that found the average concentration of the sum of the twelve PAH homologues of 26,300 µg/kg of dry matter. [22] In this comparison, the sampling places designated "Eastern Stream" and "Western Stream" did not exceed the average. The sampling place designated "Above the Tank" contained concentrations almost four-times higher than this value. The concentration of the sixteen PAH homologues in the sampling place designated "Above the Tank" exceeded even the value found in the stream Černý potok in Ostrava (90,200 µg ∑ 16 PAHs/kg of dry matter) burdened by long-term operation of a former coking plant. [23]

Comparison with the average concentrations of heavy metals in sediments of watercourses may be carried out on the basis of data found by the Water Research Institute. Results of the Institute's monitoring, carried out for several years, that concerned nine of the twenty heavy metals analysed in the site of interest are found in Table 11.

Table 11: Average concentrations of heavy metals in sediments of watercourses (1995 – 2014), given in mg/kg of dry matter after extraction by aqua regia. [22]

| As | Cd | Co | Cu | Hg | Ni | Pb | V | Zn |
|------|------|------|------|------|------|------|------|-----|
| 11.4 | 0.63 | 11.1 | 33.3 | 0.15 | 31.3 | 44.6 | 44.9 | 157 |

From comparison of the concentrations of the heavy metals, it is obvious that concentrations of cadmium and zinc in two of the sampling places ("Above the Tank" and "Western Stream") were several-times higher than the average values found in sediments of watercourses. In the case of six further heavy metals (arsenic, copper, mercury, nickel, lead, vanadium), several-times higher concentrations were found in the sampling place designated "Above the Tank". In this context, it should be noted that arsenic concentrations may differ in the individual places greatly due to its different contents in the bedrock. Only in the case of cobalt, the average concentration in sediments of watercourses is roughly comparable to the values found in the present sampling places.

5.2 Comparison with the Legislative Criteria Concerning the Monitored Contaminants

The Czech legislation does not comprise explicit limit values for presence of the contaminants monitored by us in sediments of watercourses. In spite of that, comparison with certain criteria used by the state administration may be made. These criteria are as follows: 1) pollution indicators given in the Ministry of Environment Guidelines [26], and 2) requirements on pollutant contents in waste used on the surface of the ground, according to Table No. 10.1 in Annex No. 10 to the Decree No. 294/2005 Coll., on the conditions of depositing waste in landfills and its use on the surface of the ground and amendments to Decree no. 383/2001 Coll., on details of waste management. [27] The overview of the above-mentioned criteria is given in Table 12.

In view of absence of limit values for acceptable pollution of sediments of watercourses, comparison with pollution indicators according to the Ministry of Environment Guidelines may be the best way for assessing the pollution level in the site of interest. In spite of the fact that the pollution indicators according to the above-mentioned Guidelines are used for assessing presence of contaminants in soils, they may be, according to the written statement of the director of the Water Protection Department of the Czech Environmental Inspectorate, Regional Inspectorate Ostrava, used also for assessing sediments of watercourses.

The soil pollution indicators are set for: 1) industrially used areas (including production areas and technical infrastructure areas), and 2) other areas outside industrially used areas (for example, residential areas, public utility areas, mixed areas, etc.). According to the Land Register, the sampling places designated "Western Stream" and "Eastern Stream" are found in plots of land used as permanent grassland, and protected as agricultural land resources. These two sampling places may be undoubtedly classified as other areas. According to the Land Register, the sampling place designated "Above the Tank" is found in plots of land without specified use and protection, however, the actual sedimentation tank of the facility in question, in close neighbourhood to the above-mentioned plots of land, is found on plots of land recorded as permanent grassland and protected as agricultural land resources. From the available information, it cannot be decided clearly whether the sampling place designated "Above the Tank" should be regarded as other area or an industrially used area. In spite of the fact that it would be proper to classify the above-mentioned place rather as other area, it was, due to the ambiguity, compared with the pollution indicators in the both above-mentioned categories.

The requirements on pollutant contents in waste used on the surface of the ground, according to the Decree No. 294/2005 Coll., do not directly legally apply to the sediments in the sampling places, however, for the reasons specified below, they are interesting for the comparison purposes.

Table 12: Overview of criteria used by the state administration concerning presence of the contaminants monitored by us.

| | Soil – other areas ⁶⁾ | Soil – industrially used areas ⁶⁾ | Surface of the ground in landfills ⁷⁾ |
|----------------------------------|----------------------------------|--|--|
| Individual I-PCB congener | 110 µg/kg d.m. | 380 µg/kg d.m. | - |
| Σ 6 PCBs¹⁾ | 220 µg/kg d.m. | 740 µg/kg d.m. | - |
| Σ 7 PCBs²⁾ | - | - | 200 µg/kg d.m. |
| 2,3,7,8-TCDD³⁾ | 4.5 ng/kg d.m. | 18 ng/kg d.m. | - |

| | Soil – other areas ⁶⁾ | Soil – industrially used areas ⁶⁾ | Surface of the ground in landfills ⁷⁾ |
|--|----------------------------------|--|--|
| Σ HxCDDs⁴⁾ | 94 ng/kg d.m. | 390 ng/kg d.m. | - |
| Naphthalene | 3,600 µg/kg d.m. | 18,000 µg/kg d.m. | - |
| Acenaphthene | 3,400,000 µg/kg d.m. | 33,000,000 µg/kg d.m. | - |
| Fluorene | 2,300,000 µg/kg d.m. | 22,000,000 µg/kg d.m. | - |
| Anthracene | 17,000,000 µg/kg d.m. | 170,000,000 µg/kg d.m. | - |
| Fluoranthene | 2,300,000 µg/kg d.m. | 22,000,000 µg/kg d.m. | - |
| Pyrene | 1,700,000 µg/kg d.m. | 17,000,000 µg/kg d.m. | - |
| Benzo[a]anthracene | 150 µg/kg d.m. | 2,100 µg/kg d.m. | - |
| Chrysene | 15,000 µg/kg d.m. | 210,000 µg/kg d.m. | - |
| Benzo[b]fluoranthene | 150 µg/kg d.m. | 2,100 µg/kg d.m. | - |
| Benzo[k]fluoranthene | 1,500 µg/kg d.m. | 21,000 µg/kg d.m. | - |
| Benzo[a]pyrene | 15 µg/kg d.m. | 210 µg/kg d.m. | - |
| Indeno[1,2,3,-c,d]pyrene | 150 µg/kg d.m. | 2,100 µg/kg d.m. | - |
| Dibenzo[a,h]anthracene | 15 µg/kg d.m. | 210 µg/kg d.m. | - |
| Σ 12 PAHs⁵⁾ | - | - | 6,000 µg/kg d.m. |
| Silver (Ag) | 390 mg/kg d.m. | 5,100 mg/kg d.m. | - |
| Arsenic (As) | 0.61 mg/kg d.m. | 2.4 mg/kg d.m. | 10 mg/kg d.m. |
| Boron (B) | 16,000 mg/kg d.m. | 200,000 mg/kg d.m. | - |
| Barium (Ba) | 15,000 mg/kg d.m. | 190,000 mg/kg d.m. | - |
| Beryllium (Be) | 160 mg/kg d.m. | 2,000 mg/kg d.m. | - |
| Cadmium (Cd) | 70 mg/kg d.m. | 800 mg/kg d.m. | 1 mg/kg d.m. |
| Cobalt (Co) | 23 mg/kg d.m. | 300 mg/kg d.m. | - |
| Hexavalent chromium (Cr⁶⁾) | 0.29 mg/kg d.m. | 5.6 mg/kg d.m. | - |
| Copper (Cu) | 3,100 mg/kg d.m. | 41,000 mg/kg d.m. | - |
| Iron (Fe) | 55,000 mg/kg d.m. | 720,000 mg/kg d.m. | - |
| Mercury (Hg) | 10 mg/kg d.m. | 43 mg/kg d.m. | 0.8 mg/kg d.m. |
| Manganese (Mn) | 1,800 mg/kg d.m. | 23,000 mg/kg d.m. | - |
| Molybdenum (Mo) | 390 mg/kg d.m. | 5,100 mg/kg d.m. | - |
| Nickel (Ni) | 1,500 mg/kg d.m. | 20,000 mg/kg d.m. | 80 mg/kg d.m. |
| Lead (Pb) | 400 mg/kg d.m. | 800 mg/kg d.m. | 100 mg/kg d.m. |
| Antimony (Sb) | 31 mg/kg d.m. | 410 mg/kg d.m. | - |
| Selenium (Se) | 390 mg/kg d.m. | 5,100 mg/kg d.m. | - |
| Tin (Sn) | 47,000 mg/kg d.m. | 610,000 mg/kg d.m. | - |
| Vanadium (V) | 390 mg/kg d.m. | 5,100 mg/kg d.m. | 180 mg/kg d.m. |
| Zinc (Zn) | 23,000 mg/kg d.m. | 310,000 mg/kg d.m. | - |

¹⁾ The designation Σ 6 PCBs means the sum of six congeners: PCB 28, PCB 52, PCB 101, PCB 138, PCB 153 a PCB 180.

²⁾ The designation Σ 7 PCBs means the sum of seven congeners: PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153 a PCB 180.

³⁾ The designation 2,3,7,8-TCDD is an abbreviation for 2,3,7,8-tetrachloro-dibenzodioxin.

⁴⁾ The designation Σ HxCDDs means the sum of the dibenzo-p-dioxin congeners having six chlorine atoms (hexachloro-dibenzodioxins).

⁵⁾ The designation Σ 12 PAHs means the sum of anthracene, benzo(a)anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(ghi)perylene, benzo(k)fluoranthene, fluoranthene, phenanthrene, chrysene, indeno(1,2,3-cd)pyrene, naphthalene, and pyrene.

⁶⁾ Ministry of Environment Guidelines: Pollution Indicators 2013, Annex No. 1: Overview of Indicator Values for Pollution of Soils, Soil Air, and Underground Water. [26]

⁷⁾ Table No. 10.1 in Annex No. 10 to the Decree No. 294/2005 Coll., on the conditions of depositing waste in landfills and its use on the surface of the ground and amendments to Decree no. 383/2001 Coll., on details of waste management. [27]

Neither the concentrations of the individual PCB congeners nor their total sum exceeded the pollution indicators in any of the two above-mentioned categories. However, concerning the concentration of 2,3,7,8-tetrachloro-dibenzodioxin, the sampling place designated "Above the Tank" exceeded the soil pollution indicator several-times in the category of other areas, and reached almost 80 % of the pollution indicator in the category of industrially used areas. The sampling places designated "Eastern Stream" and "Western Stream" were below the level of the soil pollution indicator concerning the concentration of 2,3,7,8-tetrachloro-dibenzodioxin. In the case of the sum of hexachloro-dibenzodioxin congeners, the results of the comparison with the soil pollution indicators were similar, with the only difference that the concentration of this indicator in the sampling place "Above the Tank" reached 60 % of the relevant indicator for industrially used areas. From the above-mentioned facts, it follows that the sampling place "Above the Tank" did not meet the soil pollution indicators concerning PCDD/F contents in the category of other areas.

Concentrations of, in total, four PAH homologues (benzo[b]fluoranthene, benzo[a]pyrene, indeno[1,2,3,-c,d]pyrene, and dibenzo[a,h]anthracene) exceeded soil pollution indicators in all the three sampling places, in the both above-mentioned categories. The benzo[a]anthracene concentration in the sampling place "Above the Tank" exceeded the soil pollution indicator in the category of industrially used areas, and in the two remaining sampling places (Western Stream and Eastern Stream) it exceeded the soil pollution indicator in the category of other areas. The remaining seven PAH homologues did not exceed the soil pollution indicators in any of the three sampling places.

Arsenic concentrations exceeded the soil pollution indicators in all the three sampling places, in the both above-mentioned categories. In the case of arsenic, concentrations higher than the above-mentioned pollution indicators commonly occur in the Czech Republic, because of the geochemical conditions in the geological environment. In such cases, pollution is indicated only by arsenic concentrations exceeding the natural background values in the locally specific conditions of the site to be assessed. In spite of the fact that increased arsenic content in agricultural soils was not found in the cadastral territory of the monitored site, according to the data of the Central Institute for Supervising and Testing in Agriculture in Brno [28], the possibility of natural occurrence of this element cannot be assessed objectively without further input data. Lead and antimony concentrations in the sampling place designated "Above the Tank" exceeded the soil pollution indicators in the category of other areas. The concentrations of the remaining heavy metals were lower than the indicator values of soil pollution in all the three sampling places.

The purpose of the pollution indicators is identification of places where chemical substances are present in amounts requiring further investigation and assessment whether the pollutant occurrence does not represent a risk for human health. If a pollution indicator value is exceeded, it does not automatically mean the necessity of carrying out remedial measures. It is only an indication that the found pollution level may potentially have adverse impacts on human health and/or ecosystems, and it is necessary to further investigate and assess the importance of the risk. From this, it may be concluded that at least in the case of five different PAH homologues, but, probably, also in the case of PCDD/Fs, lead, and antimony, it is necessary to carry out a more

detailed monitoring of the site of interest, on the basis of which remedial measures could be determined.

In spite of the fact that criteria of Table No. 10.1 in Annex No. 10 to the Decree No. 294/2005 Coll., on the conditions of depositing waste in landfills and its use on the surface of the ground and amendments to Decree no. 383/2001 Coll., on details of waste management, do not apply directly to sediments of watercourses, the taken sediment samples were compared, for working reasons, also with requirements on pollutant contents in waste used on the surface of the ground according to the above-mentioned Decree. The assessment may be interesting, in particular, in comparison with the products produced by the waste treatment facility Hůrka, because the above-mentioned Decree sets legislative requirements for some kinds of their utilisation.

Concentrations of the seven I-PCBs in sediments in all the three sampling places would meet the requirements on pollutant contents in waste used on the surface of the ground according to the Decree No. 294/2005 Coll. Concentrations of the twelve PAH homologues in sediments in the sampling places designated "Above the Tank" and "Western Stream" would not meet the requirements on pollutant contents in waste used on the surface of the ground. From the point of view of arsenic and cadmium contents, sediments in the sampling places designated "Above the Tank" and "Western Stream" would not meet the requirements on pollutant contents in waste used on the surface of the ground. Sediments in the sampling place designated "Above the Tank" would not meet the requirements on pollutant contents in waste used on the surface of the ground from the point of view of mercury and lead contents. From the metal element contents, only vanadium and nickel contents would meet the requirements on pollutant contents in waste used on the surface of the ground in sediments in all the sampling places.

5.3 Source of the Sediment Contamination

With the highest likelihood, the source of various contaminants, found in the taken sediment samples, is operation of the waste treatment facility Hůrka. This conclusion may be drawn on the basis of several facts discussed below.

The facility in question uses materials and waste demonstrably containing some of the monitored contaminants. In spite of the fact that data on all the contaminants of interest in all the accepted kinds of waste used in the facility are not at our disposal, presence of PCDD/Fs, PCBs, and other persistent organic pollutants has been proved in some kinds of the waste. Specifically, this concerns solid waste from flue gas treatment (List of Wastes number 19 01 07), and solid wastes from gas treatment containing dangerous substances (10 02 07). Data on the contents for PCDD/Fs and other persistent organic pollutants in waste accepted into the facility of interest are given in Table 14, and they are further discussed below (Chapter 5.5). Presence of significant concentrations of heavy metals can be expected in the above-mentioned kinds of waste, and, simultaneously, their significant presence was proved in the Protocols on Chemical Analyses. Data that could prove significant presence of PAHs in waste used in the facility of interest were not verified, however, in spite of that, potential presence of PAHs can be also expected in a broad spectrum of the kinds of waste used in the facility of interest.

Releases of the stored materials may take place, and took place occasionally, from the waste treatment facility Hůrka. The premises of the facility are not sufficiently secured in order to prevent transport of dust particles away from the premises by wind. In view of the current technical

measures and amounts of the stored materials, stored in the open air, transport of dust particles away from the premises cannot be prevented efficiently.¹ In the past, just release of dust particles from the stored materials was noted in the place above the sedimentation tank, as illustrated by Figure 3.

Figure 3: Release of dust particles from the waste treatment facility Hůrka (photograph: Arnika 2010).



The occurrence of the contaminants in the individual sampling places suggests that material was transported just in the direction away from the waste treatment facility. The highest concentrations of PCDD/Fs, PAHs, and heavy metals, were found in the sampling place designated "Above the Tank", located in a close vicinity (in the order of meters) from the fence surrounding the facility. This sampling place was located at the edge of the sedimentation tank, and, in the case of heavy rains, material from this place may be transported even into the tank itself. From the comparison of concentrations of PCDD/Fs, PAHs, and heavy metals, found in the sampling places designated "Eastern Stream" and "Western Stream", it is obvious that higher concentrations of the above-mentioned contaminants, with the exception of one of the PAH homologues (pyrene), were always found in the sample taken in the western stream. Just this branch of the watercourse,

¹ The operator of the facility of interest prepared, on the basis of a complaint of the Arnika Association concerning an amendment of the integrated permit, a proposal of measures for reducing dust releases. The measures became part of the amended integrated permit, issued in February 2017 by the Regional Authority of the South Bohemian Region. Because of that, transport of dust particles away from the premises should be reduced in the future.

flowing past the west side of the premises of interest, is interconnected with the sedimentation tank. From this, it may be deduced that potentially higher amounts of material released from the facility in the direction away from the sedimentation tank may be transported to the sampling place "Western Stream" than to the sampling place "Eastern Stream". PCBs are the only exception in the trend in concentrations of the monitored contaminants when comparing the individual sampling places. PCB concentrations were highest in the sampling place "Eastern Stream", and they decreased in the direction to the sampling place "Above the Tank". The pattern of the seven I-PCB congeners showed higher similarity between the sampling places "Above the Tank" and "Western Stream" that differ considerably from the sampling place "Eastern Stream".

In the surroundings of the site, no other potential source of the monitored contaminants is known. In the drainage area of the stream flowing past the facility of interest, there are only the nuclear power station Temelín, Hůrecký pond, small forested areas, and fields, that are not known as significant sources of any of the contaminants monitored by us. The only potential source is just the waste treatment facility Hůrka that, moreover, treats materials containing probably all the monitored contaminants.

From the above-mentioned information, it follows clearly that the contamination found in the sampling places originated in the waste treatment facility Hůrka with the highest likelihood, or almost surely.

If we accept the conclusion that the contamination by the monitored substances originated in the waste treatment facility Hůrka, it is also possible to comment the comparison of the found results with the requirements on pollutant contents in waste used on the surface of the ground according to Table No. 10.1 in Annex No. 10 to the Decree No. 294/2005 Coll. Concentrations of twelve PAH homologues exceeded the requirements set by the above-mentioned Decree in the sampling places "Above the Tank" and "Western Stream". The situation concerning arsenic and cadmium concentrations was similar. Mercury and lead concentrations exceeded the above-mentioned requirements according to the Decree in the sampling place "Above the Tank". The sampling place "Eastern Stream" would meet the requirements set by the Decree concerning all the monitored parameters, and all the sampling places would meet them in the following parameters: contents for nickel, vanadium, and seven I-PCB congeners. The above-mentioned requirements set by the Decree do not concern pollutant contents in sediments of watercourses, but pollutant contents in materials placed on the surface of the ground. Certified products of the waste treatment facility Hůrka are supplied for reclamation of sludge-drying beds in Mydlovary where uranium ore was treated by the state company MAPE in the past. Within the framework of the reclamation, carried out by the state company DIAMO at present, the certified products Q.I.-1 and Q.I.-2, obtained from the facility of interest, are used as a reclamation material. In Annex No. 9 to the EIA documents concerning the plan for termination of uranium ore treatment in Mydlovary, there is stated that the company Quail offered, in the past, the above-mentioned certified products from the facility in Hůrka as the so-called covering material that should meet the criteria according to Table No. 10.1 in Annex No. 10 to the Decree No. 294/2005 Coll. It means that contaminant concentrations in sediments of watercourses in the sampling places designated "Above the Tank" and "Eastern Stream" would not meet the requirements that have to be met in the case of the potential use of the products produced in the facility of interest. In other words, if the polluted sediment of the watercourse were a product, it could not be used as a covering material within the framework of reclamation of the sludge-drying beds in Mydlovary.

5.4 Comparison with Results of the Previous Investigations

In the surroundings of the facility of interest, previous investigations of presence of persistent organic pollutants in watercourses and ponds were carried out by the Arnika Association in 2009, 2010, 2012, and 2014. The results of the investigations were presented in the study entitled „Pollution by POPs in the Surroundings of the Quail spol. s.r.o. Facility, Hůrka near Temelín“ in 2016. The author of the study stated in her conclusions that pollution of the sediments with persistent organic pollutants (specifically, PCDD/Fs and PCBs) was caused by the operation of the facility of interest. The persistent organic pollutant concentrations found during the above-mentioned investigations are summarised in Table 13. The concentrations of PCDD/Fs and DL-PCBs found in 2014 were comparable to, or lower than, the concentrations found by the present study, what could show continuing input of the contaminants into the watercourses in the surroundings of the facility of interest. In particular, increase of concentrations could be noted in the sampling places designated "Eastern Branch of the Watercourse Having No Name" and "Western Branch of the Watercourse Having No Name" that approximately corresponded to the sampling places designated "Eastern Stream" and "Western Stream" according to the present study. In spite of all the shortcomings and improper formulations in the study „Pollution by POPs in the Surroundings of the Quail spol. s.r.o. Facility, Hůrka near Temelín“, it may be agreed to the study conclusion that the source of contamination by PCDD/Fs was the facility of interest.

Table 13: Overview of concentrations of persistent organic pollutants in sediment samples in the surroundings of the waste treatment facility Hůrka, found in 2009 to 2014.

| Site and date of sampling | ∑7I-PCBs [µg/kg d.m.] ¹⁾ | DL-PCB I-TEQ [ng/kg d.m.] | PCDD/F I-TEQ [ng/kg d.m.] | DL-PCB + PCDD/F I-TEQ [ng/kg d.m.] | DL-PCB + PCDD/F DR CALUX TEQ [ng/kg d.m.] | ∑3HCHs [µg/kg d.m.] ²⁾ | HCB [µg/kg d.m.] | PeCB [µg/kg d.m.] |
|--|-------------------------------------|---------------------------|---------------------------|------------------------------------|---|-----------------------------------|------------------|-------------------|
| Retention tank (December 2009) | NA | NA | NA | NA | 14 | NA | NA | NA |
| Watercourse having no name (December 2009) | NA | NA | NA | NA | 4.5 | NA | NA | NA |
| Retention tank (June 2010) | 3.9 | NA | NA | NA | 16.5 | < 4.5 | < 1.5 | < 1.5 |
| Watercourse having no name (June 2010) | < 11 | NA | NA | NA | 1.1 | < 4.5 | < 1.5 | < 1.5 |
| Hůrecký pond (June 2010) | < 11 | NA | NA | NA | NA | < 4.5 | < 1.5 | < 1.5 |
| Tributary to the Pohrobný pond (September 2012) | NA | NA | NA | NA | 4.9 | NA | NA | NA |
| Effluent from the Pohrobný pond (September 2012) | NA | NA | NA | NA | 5.4 | NA | NA | NA |

| Site and date of sampling | ∑7I-PCBs [µg/kg d.m.] ¹⁾ | DL-PCB I-TEQ [ng/kg d.m.] | PCDD/F I-TEQ [ng/kg d.m.] | DL-PCB + PCDD/F I-TEQ [ng/kg d.m.] | DL-PCB + PCDD/F DR CALUX TEQ [ng/kg d.m.] | ∑3HCHs [µg/kg d.m.] ²⁾ | HCB [µg/kg d.m.] | PeCB [µg/kg d.m.] |
|---|-------------------------------------|---------------------------|---------------------------|------------------------------------|---|-----------------------------------|------------------|-------------------|
| Wetland near the facility (September 2012) | NA | NA | NA | NA | 16 | NA | NA | NA |
| Wetland near the watercourse having no name (September 2012) | NA | NA | NA | NA | 7.7 | NA | NA | NA |
| Wetland near the facility (September 2014) | 81.7 | 111.29 | 292.88 | 404.17 | NA | NA | NA | NA |
| Watercourse having no name (September 2014) | NA | 228.79 | 303.51 | 532.3 | NA | NA | NA | NA |
| Eastern branch of the watercourse having no name (September 2014) | NA | 1.85 | 4.82 | 6.67 | NA | NA | NA | NA |
| Western branch of the watercourse having no name (September 2014) | NA | 17.37 | 22.52 | 39.89 | NA | NA | NA | NA |

¹⁾ The designation ∑ 7 PCBs means the sum of seven congeners: PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, and PCB 180.

²⁾ The designation ∑ 3 HCHs means the sum of three hexachlorobenzene isomers: α, β, γ.

5.5 Contents of Persistent Organic Pollutants in the Accepted Waste

According to the records, in total 69 kinds of waste having the total weight of 101,600 tons were accepted by the waste treatment facility Hůrka in 2014, and 65 kinds of waste having the total weight of 120,604 tons a year later. From the point of view of persistent organic pollutant contents, especially two kinds of waste were interesting.

The first one of these kinds of waste was solid waste from flue gas treatment (List of Wastes number 19 01 07), supplied to the facility from four incinerators in total. Two of them were big commercial municipal waste incinerators, and two of them were hazardous waste incinerators. The first municipal waste incinerator was ZEVO Malešice, operated by the company Pražské služby a. s. Its total capacity is 310,000 tons of waste per year. The second municipal waste incinerator, SAKO

Brno, is operated by a joint stock company having the same name, and its capacity is 248,000 tons of waste per year. The both hazardous waste incinerators are operated by the company RUMPOLD s.r.o. These incinerators are Spalovna Strakonice (capacity 1,500 tons of waste per year), and Spalovna Jihlava (capacity 1,900 tons of waste per year). The both hazardous waste incinerators incinerate, in particular, medical waste, but also waste oils and industrial waste.

The second accepted kind of waste containing persistent organic pollutants was waste from gas treatment containing dangerous substances (10 02 07), produced by dedusting in ironworks in Hrádek (Železářny Hrádek). Table 14 states Waste List numbers, weights of the accepted waste, and persistent organic pollutant concentrations, concerning the individual producers of waste accepted by the facility of interest in 2014, 2015, and 2016.

Table 14: Data on producers, Waste List numbers, weights, and persistent organic pollutant contents, concerning waste accepted by the waste treatment facility Hůrka in 2014 to 2016. The abbreviation MD means "missing data". The source of the data on persistent organic pollutant contents were Protocols on Chemical Analyses provided by the Regional Authority of the South Bohemian Region and the Czech Environmental Inspectorate.

| Year | Producer | Waste List number | Waste weight [t] | PCDD/Fs [ng I-TEQ/kg d.m.] | | ∑3TeCBs [mg/kg d.m.] | PeCB [mg/kg d.m.] | HCB [mg/kg d.m.] | ∑4HCHs [mg/kg d.m.] | ∑6PCBs [mg/kg d.m.] | HCBd [mg/kg d.m.] |
|------------------|---------------------|-------------------|------------------|----------------------------|---------|----------------------|-------------------|------------------|---------------------|---------------------|-------------------|
| | | | | Min | Max | | | | | | |
| 2014 | Spalovna Jihlava | 19 01 07 | 26.61 | 32,000 | 32,000 | 1.53 | 0.624 | 0.117 | < 0.04 | < 0.12 | < 0,01 |
| | Spalovna Strakonice | 19 01 07 | 53.7 | 20,000 | 20,000 | 1.29 | 0.49 | 0.0704 | < 0.04 | < 0.12 | < 0,01 |
| | SAKO Brno | 19 01 07 | 7,139.87 | 408 | 415 | MD | MD | MD | MD | 0.000936 | MD |
| | ZEVO Malešice | 19 01 07 | 3,871 | 2,200 | 2,200 | MD | MD | MD | MD | 0.018 | MD |
| | Železářny Hrádek | 10 02 07 | 1,170.58 | MD | MD | MD | MD | MD | MD | MD | MD |
| 2015 | Spalovna Jihlava | 19 01 07 | 23.77 | 36,000 | 36,000 | MD | MD | MD | MD | MD | MD |
| | Spalovna Strakonice | 19 01 07 | 46.46 | 100,000 | 100,000 | MD | MD | MD | MD | MD | MD |
| | SAKO Brno | 19 01 07 | 6,884.58 | 493 | 504 | MD | MD | MD | MD | 0.00226 | MD |
| | | | | 517 | 517 | < 0.03 | < 0.01 | < 0.005 | < 0.04 | < 0.12 | < 0,01 |
| | ZEVO Malešice | 19 01 07 | 2,985.08 | 324 | 324 | MD | MD | MD | MD | MD | MD |
| | | | | 528 | 528 | MD | MD | MD | MD | MD | MD |
| Železářny Hrádek | 10 02 07 | 1,342.48 | MD | MD | MD | MD | MD | MD | MD | MD | |
| 2016 | Spalovna Jihlava | 19 01 07 | 14.64 | 15,000 | 15,000 | MD | MD | MD | MD | MD | MD |
| | Spalovna Strakonice | 19 01 07 | 12.88 | 99,000 | 99,000 | MD | MD | MD | MD | MD | MD |

| | | | | | | | | | | | |
|------------------|----------|----|-------|-------|----|----|----|----|----|-------|----|
| SAKO Brno | 19 01 07 | MD | MD | MD | MD | MD | MD | MD | MD | MD | MD |
| ZEVO Malešice | 19 01 07 | MD | MD | MD | MD | MD | MD | MD | MD | MD | MD |
| Železářny Hrádek | 10 02 07 | MD | 3,763 | 3,763 | MD | MD | MD | MD | MD | 0.318 | MD |

The fact that just these kinds of waste are problematic is confirmed also by a German project that tried to carry out a complex emission inventory of PCDD/Fs from industrial and non-industrial sources in 1990 to 1995 in the territory of Norway, Switzerland, and 15 states of the European Union at that time. It concluded that the biggest source of PCDD/Fs at the all-European level was just municipal waste incineration, with iron smelting narrowly in the second place. [29]

In spite of the fact that reduction of PCDD/F contents in solid residues of waste incineration took place in the past three decades [30], the presence of PCDD/Fs in this waste has continued to be discussed internationally. Table 15 states PCDD/F concentrations in solid residues from flue gas treatment in municipal and hazardous waste incinerators in various countries of the world, where the above-mentioned decrease of PCDD/F contents is obvious, too. The PCDD/F concentrations in fly ash from flue gas treatment in municipal waste incinerators were approximately in the range from 100 to 11,000 ng I-TEQ/kg in the past fifteen years. PCDD/F concentrations in fly ash from municipal waste incinerators, supplied to the facility of interest, did not deviate from this range, and are closer to its lower limit. According to the Protocols provided by the Regional Authority of the South Bohemian Region and the Czech Environmental Inspectorate, PCDD/F concentrations found in the fly ash from flue gas treatment in the municipal waste incinerators (ZEVO Malešice and SAKO Brno) were in the range from 324 to 2,200 ng I-TEQ/kg in the period of 2014 to 2016.

Table 15: PCDD/F concentrations in solid waste from flue gas treatment in municipal and hazardous waste incinerators in various countries of the world.

| Country | Publication year | Type and number of incinerators where samples were taken | PCDD/F range [ng I-TEQ/kg] unless otherwise stated | PCDD/F average [ng I-TEQ/kg] unless otherwise stated | Source |
|-------------|------------------|--|--|--|--------|
| Germany | 1991 | 11 municipal waste incinerators | 1,000 – 28,000 | 4,000 | [31] |
| Denmark | 1997 | 19 samples from municipal waste incinerators | 100 – 3,800 | 1,600 | [32] |
| South Korea | 1999 | 11 municipal waste incinerators | 130 – 21,000 | - | [33] |
| Spain | 2001 | 6 municipal waste incinerators | - | 100 - 2,400 ² | [34] |
| Japan | 2001 | 3 municipal waste incinerators | 1,500 – 6,700 | - | [35] |
| Sweden | 2001 | Municipal waste | 1,300 – 3,800 | - | [36] |

² Range of averages for six incinerators from in total 50 measurements..

| Country | Publication year | Type and number of incinerators where samples were taken | PCDD/F range [ng I-TEQ/kg] unless otherwise stated | PCDD/F average [ng I-TEQ/kg] unless otherwise stated | Source |
|-----------------------|------------------|--|--|--|--------------|
| | | incinerator Renova | | | |
| United Kingdom | 2002 | 11 municipal waste incinerators | 330 – 5,800 | - | [37] |
| Italy | 2002 | Municipal waste incinerator | - | 193.8 ³ | [38] |
| Europe, Asia | 2001 - 2004 | 47 municipal waste incinerators | 100 - 9,400 | | [30] |
| China | 2004 | Municipal waste incinerator in Shanghai | 970 – 1,500 | - | [39] |
| South Korea | 2005 | 3 municipal waste incinerators | 244 – 24,786 | - | [40] |
| China | 2007 | Municipal waste incinerator Harbin | - | 798.2 | [41] |
| China | 2008 | 5 municipal waste incinerators | 137.8 – 2,680 | - | [42] |
| Czech Republic | 2008 - 2010 | Municipal waste incinerator Liberec | 1,000 – 11,000 ⁴ | - | [43] [44] |
| Taiwan | 2011 | 16 municipal waste incinerators | 781 – 2,866 | 1,870 | [45] |
| China | 2013 | 15 municipal waste incinerators | 34 – 2,500 [ng WHO-TEQ/kg, for PCDD/Fs and DL-PCBs] | - | [46] |
| Denmark and Greenland | 2016 | 3 municipal waste incinerators | 180 – 2,010 | - | [47] |
| China | 2007 | 3 medical waste incinerators | 9,547 – 15,619 | 12,179 | [48] |
| China | 2008 | Medical waste incinerators | 20,397 | - | [42] |
| Colombia | 2009 | Hazardous waste incinerator in Medellin | 181,535.8 [ng/ WHO-TEQ/kg] | - | [49] |

Hazardous waste incinerators produce waste from flue gas treatment showing significantly higher PCDD/F concentrations. PCDD/F concentrations in the range from 15,000 to 100,000 ng I-TEQ/kg were found in fly ash from flue gas treatment in the hazardous waste incinerators (Spalovna

³ Concentration in fly ash from the fabric filter. However, the incinerator produces also sludge from wet flue gas treatment, showing the concentration of 604 ng I-TEQ/kg.

⁴ Šyc et al. [43] state the values up to 1,000 ng I-TEQ/g in the case of fly ash from the electrostatic filter, the amount of which is significantly higher, and 1,000 – 10,000 ng I-TEQ/g in the case of fly ash from the catalytic filter. A previous study of the same authors [44] states up to 11,000 ng I-TEQ/g in the case of fly ash from the catalytic filter.

Strakonice and Spalovna Jihlava) in the period of 2014 to 2016. These concentrations are several-times higher than the ones in the case of municipal waste incinerators. Also these values generally correspond to the ranges of PCDD/F contents in fly ash from flue gas treatment in hazardous waste incinerators in other countries of the world (see Table 15).

Because the Czech Republic ranks among countries that ratified the Stockholm Convention, the waste treatment facility in Hůrka should not treat waste showing higher PCDD/F contents than the so-called „low persistent organic pollutants content“ according to Article 6 of the Stockholm Convention. Such limit value is also laid down in Annex IV to the Regulation of the European Parliament and of the Council No. 850/2004 of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC. In the case of PCDD/Fs, this concerns waste showing concentrations higher than 15,000 ng WHO-TEQ/kg. The international conventions laid down this concentration temporarily, with the possibility of further tightening in the future. Waste exceeding the „low POPs content“ of PCDD/Fs laid down by the Stockholm Convention may be used only „in such a way as to ensure that the persistent organic pollutant content is destroyed or irreversibly transformed“. The facility of interest accepted waste exceeding the „low POPs content“ of PCDD/Fs from hazardous waste incinerators in Jihlava and Strakonice till May 2016.

The company referred to the fact that waste was treated, in the facility, by one of the methods of the category of physico-chemical treatment, use of which is enabled in the case of such kinds of waste according to Part 1 of Annex No. V to the Regulation of the European Parliament and of the Council No. 850/2004 of 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC. Actually, the process of stabilisation was carried out in the facility of interest. The process utilises compounds that chemically or physically bind hazardous substances present in waste, and reduce solubility of the contaminants in this way. This process is unsuitable from the point of view of increase of the total waste volume. [50] Because only reduction of leachability of heavy metals⁵ and reduction of contaminant concentrations by mixing with other materials takes place during the stabilisation, this process cannot be regarded as an irreversible transformation of the persistent organic pollutant content. Moreover, the process is not on the list of technologies for treatment of waste with dioxin contents according to the Technical Guidelines of the Basel Convention [51].

The product itself – the certified product – is subsequently used as a filling layer during decontamination and reclamation of sludge-drying beds from uranium ore leaching in the area of Mydlovary. From the drawn up report of October 2004, it was obvious that the amounts of the offered materials for reclamation exceeded the required amounts, in the both variants of reclamation duration - 20 or 15 years. The price of the material (without transport) was in the range from 10 to 70 CZK per ton. Obviously, the crucial factors for the choice of utilisation of the certified product from the facility of interest for reclamation of sludge-drying beds in Mydlovary were, in particular, its low purchase price and low transport costs, caused mainly by low distance of the facility of interest from the sludge-drying beds in Mydlovary where reclamation should be carried out. [52] This contrasts especially with the fact that waste containing PCDD/Fs and other persistent organic pollutants is transported into the facility of interest from five regions of the Czech Republic.

⁵ Reduction of leachability of persistent organic pollutants was not investigated in the material resulting from the process.

5.6 Balance of Inputs and Outputs of Persistent Organic Pollutants

A balance of inputs and outputs of contaminants was calculated for 2014 and 2015, on the basis of data on amounts of waste forming inputs into the facility, the amounts of products forming outputs of the facility, and concentrations of persistent organic pollutants contained in them. Graph 5 shows estimates of amounts of PCDD/Fs that entered the facility of interest together with the waste from the individual waste producers, in the individual years. Similar estimates, expressed in percents, for the individual years and as an average for the both years, are given in Table 16. In interannual comparison, the total estimated amount of PCDD/Fs entering the facility of interest increased slightly, however, the share of the individual waste producers, in percents, changed significantly in the case of the incinerators ZEVO Malešice and Spalovna Strakonice only. In the case of ZEVO Malešice, the share expressed in percents decreased approximately seven-times, and in the case of Spalovna Strakonice it increased about six-times. The interannual change in the estimated amounts of inputs of PCDD/Fs from Spalovna Strakonice was caused by different PCDD/F contents in the waste, because the weight of the accepted waste did not change much. On the other hand, the interannual decrease in the estimated amounts of PCDD/F inputs from ZEVO Malešice was caused partially by decrease of the weight of the accepted waste, too. Further, it follows from the data that approximately a third of the total estimated PCDD/F amount entered the facility in question from the ironworks Železářny Hrádek in average for the both years, approximately a quarter from the hazardous waste incinerators (Strakonice and Jihlava), and approximately 45 % from the municipal waste incinerators (ZEVO Malešice and SAKO Brno).

Graph 5: Estimated amounts of PCDD/Fs entering the waste treatment facility Hůrka in waste from various waste producers in 2014 and 2015.

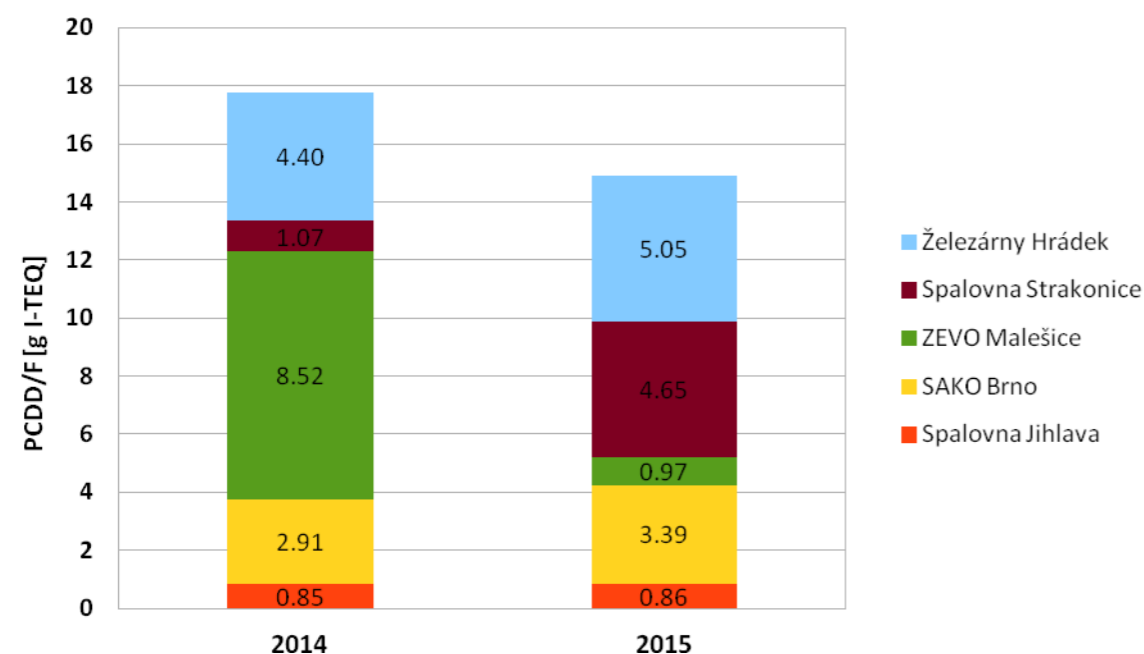


Table 16: Estimate of share of the individual waste producers concerning the total sum of PCDD/F I-TEQ entering the waste treatment facility Hůrka in waste in 2014 and 2015.

| | 2014 | 2015 | Average |
|--|------|------|---------|
|--|------|------|---------|

| | | | |
|-------------------------|------|------|-------|
| Spalovna Jihlava [%] | 4.8 | 5.7 | 5.25 |
| SAKO Brno [%] | 16.4 | 22.8 | 19.6 |
| ZEVO Malešice [%] | 48.0 | 6.5 | 27.25 |
| Spalovna Strakonice [%] | 6.0 | 31.2 | 18.6 |
| Železářny Hrádek [%] | 24.8 | 33.9 | 29.35 |

Estimates of annual amounts of persistent organic pollutants (TeCB, PeCB, HCB, HCH, PCBs, and HCBd) entering the waste treatment facility are given in Table 17, for the individual years and their sum. Because data are missing for a number of accepted kinds of waste, the estimates presented below may be regarded rather as the lower limit of their actual amounts entering the facility in the waste.

Table 17: Estimates of the annual amounts of persistent organic pollutants that entered the waste treatment facility in Hůrka in waste in 2014 and 2015.

| | Σ 3 TeCBs ¹⁾ | PeCB | HCB | Σ 4 HCHs ²⁾ | Σ 6 I-PCBs ³⁾ | HCBd |
|-----------|--------------------------------|-------|-------|-------------------------------|---------------------------------|------|
| 2014 [g] | 109.99 | 42.92 | 34.38 | 0.00 | 457.39 | 0.00 |
| 2015 [g] | 96.30 | 37.60 | 27.25 | 0.00 | 504.46 | 0.00 |
| Total [g] | 245.30 | 95.96 | 64.24 | 0.00 | 963.40 | 0.00 |

¹⁾ The designation Σ 3 TeCBs means the sum of three tetrachlorobenzene isomers (1,2,3,4-TeCB, 1,2,3,5-TeCB, 1,2,4,5-TeCB)

²⁾ The designation Σ 4 HCHs means the sum of four hexachlorocyclohexane isomers (α , β , γ , δ)

³⁾ The designation Σ 6 PCBs means the sum of six PCB congeners (28, 52, 101, 138, 153, 180)

The total estimated amounts of PCDD/Fs entering the facility of interest in waste were 17.76 - 17.81 g I-TEQ in 2014 and 14.91 - 15.69 g I-TEQ in 2015. This finding corresponds to estimates carried out by the Arnika Association in 2004 that between 1.5 and 27 g PCDD/F I-TEQ entered the facility of interest in 2003 [53]. The present calculation makes the estimate much more precise. The estimated amount of PCDD/F input into the facility was 32.67 - 33.5 g I-TEQ in the two-year period. This is at least eight-times more than the amount of output from the facility in the certified product in the same period. The estimated amount of PCDD/Fs leaving the facility in its certified product was 3.62 - 4.02 g I-TEQ, as a sum for the both years. The fate of the remaining 28.65 - 29.88 g of PCDD/F I-TEQ is not clear, because the technology operated in the facility is not based on destruction of persistent organic pollutants, and PCDD/Fs show half life in the order of decades.

If the total estimated amount of PCDD/Fs entering the facility of interest were preserved and it left the facility after dilution with other materials perfectly mixed in the product, the expected PCDD/F concentration in the certified product would be 178.43 ng I-TEQ/kg, as the average for the both years. However, the PCDD/F concentrations in the certified product, found by measurements, were in average 18.94 - 21.08 ng I-TEQ/kg. The balance of PCDD/F inputs and outputs into/out of the facility of interest in 2014 and 2015 is shown in Figure 4. Further data on inputs and outputs into/out of the facility of interest are presented in Table 18. A part of the difference between the PCDD/F inputs and outputs into/out of the facility of interest may be attributed to inaccuracies of the estimates, PCDD/F releases into the surrounding environment, finishing production in the subsequent year, and alternative utilisation of a part of the contaminated waste. However, because of such a huge difference, it will be necessary to pay higher attention to processes and actions taking place inside the facilities treating waste in this way. For example, the calculations may be made much more precise by means of regular and more frequent measurements of concentrations of PCDD/Fs and other contaminants in the accepted waste and the products leaving the facility.

Figure 4: Balance of PCDD/F inputs in waste and outputs in product into/out of the waste treatment facility Hůrka in the two-year period 2014 to 2015.

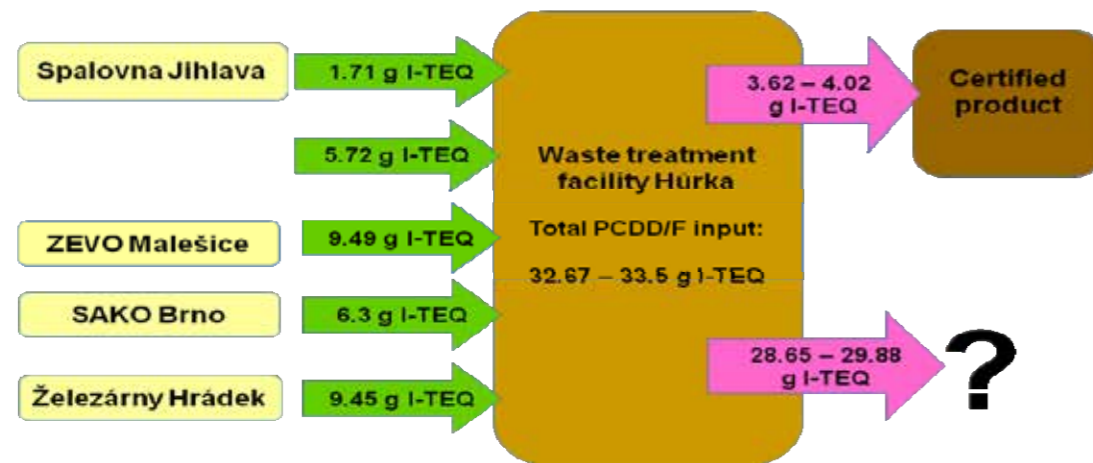


Table 18: Estimated balance of material (waste/product) flow and PCDD/F flow into/out of the waste treatment facility Hůrka in 2014 and 2015.

| | 2014 | 2015 | Total |
|---|-----------|----------|-----------|
| Inputs – waste and technological water, total [t] | 101,600 | 120,604 | 222,204 |
| Inputs – fly ash from incinerators 19 01 07 [t] | 11,091.18 | 9,939.89 | 21,031.07 |
| Inputs – dust from ironworks 10 02 07 [t] | 1,170.58 | 1,342.48 | 2,513.06 |
| Fly ash from incinerators (19 01 07) weight share of the total waste input [%] | 10.92 | 8.24 | 9.58* |
| Dust from ironworks (10 02 07) weight share of the total waste input [%] | 1.15 | 1.11 | 1.13* |
| Minimum estimated amount of PCDD/Fs entering the facility [g I-TEQ] | 17.76 | 14.91 | 32.67 |
| Maximum estimated amount of PCDD/Fs entering the facility [g I-TEQ] | 17.81 | 15.69 | 33.5 |
| Expected PCDD/F concentration in the output [ng I-TEQ/kg of dry matter] | 200.86 | 155.99 | 178.42* |
| Minimum estimated amount of PCDD/Fs entering the facility without the incinerators Jihlava a Strakonice [g I-TEQ] | 15.83 | 9.41 | 25.24 |
| Expected PCDD/F concentration in the output without the incinerators Jihlava a Strakonice [ng I-TEQ/kg of dry matter] | 179.03 | 98.45 | 138.74* |
| Outputs – certified product [t] | 88,421 | 95,584 | 184,005 |
| Minimum estimated amount of PCDD/Fs leaving the facility in the certified product [g I-TEQ] | - | - | 3.62 |
| Maximum estimated amount of PCDD/Fs leaving the facility in the certified product [g I-TEQ] | - | - | 4.02 |
| Average minimum PCDD/F concentration in the certified product [ng I-TEQ/kg of dry matter] | 16.38 | 21.5 | 18.94* |
| Average maximum PCDD/F concentration in the certified product [ng I-TEQ/kg of dry matter] | 18.75 | 23.4 | 21.08* |

* in the case of the marked values, average, not sum, was calculated for the both mentioned years.

The Regional Authority of the South Bohemian Region issued, on the basis of review of the Integrated Permit on Integrated Pollution Prevention and Control (IPPC) a Decision on the Integrated Permit Amendment, according to which waste produced by hazardous waste incineration was excluded from the list of the kinds of waste accepted into the waste treatment facility of interest. This concerned solid waste from flue gas treatment (List of Wastes number 19 01 07), fly ash containing dangerous substances (19 01 13), and boiler dust containing dangerous substances (19 01 15) originating from hazardous waste incinerators. [54] However, the adopted measure applies neither to fly ash from flue gas treatment produced by municipal waste incinerators, nor to solid waste produced by dedusting in ironworks, that contain PCDD/Fs and other persistent organic pollutants, too. The same measure was adopted in the facility of interest by the operator itself in May 2016 already.

The measure adopted within the framework of the integrated pollution prevention and control did not solve the overall situation concerning the PCDD/F inputs into the facility of interest. According to the data presented in Table 16, the hazardous waste incinerators (Jihlava and Strakonice) contributed to the total estimated amount of PCDD/Fs entering the facility by less than 25 % only. In the absolute values, this means that the inputs into the facility of interest would be 25.24 g PCDD/F I-TEQ in 2014 and 2015, instead of the actual 32.67 g PCDD/F I-TEQ.

6. Conclusion

Three sediment samples, taken in the surroundings of the waste treatment facility Hůrka, were analysed in order to check the contamination, and, further, to verify possibilities of contamination releases from the facility in question. The spectrum of the monitored chemical substances included PCBs, PCDD/Fs, PAHs, and heavy metals. From the results, it follows that sediments in the surroundings of the facility of interest were polluted with almost all the monitored substances, some of them exceeding limits set by the legislation, and that the source of the contamination was, with the highest likeliness, the waste treatment facility Hůrka.

From comparison of the measured contaminant concentrations with reference sites and average concentrations in a number of various sites, it followed that many-times higher concentrations of PCBs, PCDD/Fs, PAHs, and heavy metals, were found in the site of interest. In the case of PCDD/Fs, the concentrations were ten-times up to hundred-times higher than the typical values. The measured PCB and PAH concentrations in the taken samples were comparable with the values in highly burdened sites, namely, Elbe and Bílina sediments in Ústí nad Labem in the case of PCBs, and Černý Potok stream sediments in Ostrava in the case of PAHs. By comparing the measured contaminant concentrations with legislative criteria, it was found that concentrations of substances ranked among PAHs and PCDD/Fs, arsenic, lead, and antimony, exceeded indicators of soil pollution for other areas, in at least one of the places where samples were taken. Only the measured PCB concentrations met the soil pollution indicators in all the places. From the point of view of the Decree No. 294/2005 Coll., on the conditions of depositing waste in landfills and its use on the surface of the ground, contaminant concentrations, in certain sediment samples, did not meet the requirements valid for the potential utilisation of the products produced by the facility of interest.

The waste treatment facility Hůrka treats waste containing the monitored contaminants. Releases of the stored materials may take place, and took place, at least occasionally, from the facility of interest. The occurrence of the contaminants in the individual sampling places suggests that

material was transported in the direction away from the facility of interest. In the surroundings of the site, no other potential source of the monitored contaminants is known. From these reasons, a conclusion may be drawn that the source of the contaminants found in the taken sediment samples was, with the highest likeliness, the waste treatment facility Hůrka. This conclusion is in accordance with the previous results of the Arnika Association from 2009, 2010, 2012, and 2014, presented in the study entitled „Pollution by POPs in the Surroundings of the Quail spol. s.r.o. Facility, Hůrka near Temelín“.

According to the records, two kinds of waste containing persistent organic pollutants were accepted into the waste treatment facility Hůrka in 2014 and 2015: 1) solid waste from flue gas treatment, supplied to the facility by two municipal waste incinerators and two hazardous waste incinerators, and 2) waste from gas treatment containing dangerous substances, originating from dedusting in ironworks. The PCDD/F concentrations in the range from 324 to 2,200 ng I-TEQ/kg were found in fly ash from flue gas treatment from municipal waste incinerators. In the case of fly ash from hazardous waste incinerators, the PCDD/F concentrations were in the range from 15,000 to 100,000 ng I-TEQ/kg. Thus, the facility of interest had accepted waste exceeding the „low POPs content“ of PCDD/Fs according to the Article 6 of the Stockholm Convention, up to May 2016 when the company operating the facility of interest stopped this practice.

The estimated amount of PCDD/F input into the facility was in total 32.67 - 33.5 g I-TEQ in the period of 2014 and 2015. The estimated output of PCDD/F from the facility of interest in the certified product was 3.62 - 4.02 g I-TEQ, as the sum for the both monitored years. The total estimated input of PCDD/Fs was eight-times higher than the estimated output in the certified product, in the same period. The fate of the remaining 28.65 – 29.88 g I-TEQ of PCDD/Fs is not clear, in the two years. In February 2017, the Regional Authority of the South Bohemian Region issued, on the basis of the Act No. 76/2002 Coll. on integrated pollution prevention and control, on the Integrated Pollution Register, and on amendment to certain acts, a Decision on the Integrated Permit Amendment, according to which waste produced by hazardous waste incineration was excluded from the list of the kinds of waste accepted into the waste treatment facility of interest. However, hazardous waste incinerators contributed to the total estimated PCDD/F amount entering the facility by less than 25 % only, according to the calculations.

From the conclusions, it follows that the waste treatment facility Hůrka should adopt necessary measures to prevent further contamination of its surroundings. Firstly, suitable measures preventing transport of dust particles away from the premises should be implemented. These measures form part of the amended Integrated Permit of the facility of interest already. Further, treatment of waste containing higher concentrations of persistent organic pollutants should be stopped in the facility, above the requirements set by the valid legislation and the valid integrated permit. Waste with higher concentrations of persistent organic pollutants should be rather treated by non-combustion technologies for their complete destruction. [51] [55] [56] [57]

7. Literature

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english.arnika.org

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Contact: Arnika – Toxics and Waste Programme
Dělnická 13
170 00 Prague 7
Czech Republic
e-mail: toxic@arnika.org