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HIGH-CONTAMINATED SOIL WITH MERCURY IN BAY OF VLORA (ALBANIA) AND ITS POSSIBLE REMEDIATION

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Abstract: High-contaminated soil with mercury in bay of Vlora (Albania) and its possible remediation. Most of mercury pollution was caused by Chlorine-alkali unit where mercury cathode was used for NaCl brine electrolyses and by PVC unit where $HgCl_2$ was used as a catalyst for the C_2H_3Cl monomer synthesis. We evaluated the mercury pollution within a former PVC (polyvinyl chloride) plant area situated 3 km west of Vlora centre and gave the solution for its rehabilitation. In this aim, 40 soil samples, 14 sludge samples, 4 leachate samples and 10 groundwater samples were analyzed with respect to content of mercury, in the framework of the project: "Environmental Clean Up of the PVC Plant in Vlora". The highest (>20.0 mg/kg) mercury concentrations are found in surface soils around the buildings of former electrolysis and de-mercurisation plants and they show a roughly downward decrease. For a definitely remediation of the PVC territory, it is proposed to be constructed a Confined Disposal Facility (CDF) where all the contaminated soils containing over 10 mg/kg Hg will be landfilled.

Key Words: Mercury, contamination, sludge, PVC plant, landfill, CDF.

1. INTRODUCTION

The former PVC plant, that is situated 3 km west of Vlora city (Albania), represents one of the numerous hot spots in Albania. It was part of the larger industrial complex that produced chlorine, alkali, vinyl chloride monomer, PVC, hydrochloric acid, soda and a variety of other related chemicals. This complex was closed in 1992, and its buildings have been completely destroyed since that time. The PVC plant area (about 50 ha) was known for high pollution of soil with mercury, which have an important impact on water, air and biota contamination (Lazo et. al., 2006). The contamination of soil and water with mercury was due to technological looses and non controlled waste discharges. The quantity of mercury discharged in the environment only during period 1977-1983 was estimated about 65 tons. Most of mercury pollution was caused by Chlorine-alkali unit where mercury cathode was used for NaCl brine

electrolyses and by PVC unit where $HgCl_2$ was used as a catalyst for the C_2H_3Cl monomer synthesis (Kotorri, 2006). The contamination of Vlora bay with mercury attracted many researchers, who studied it.

Most of studies were focused on pollution of marine environment, sediments, seawater and biota (Babi, 1996; Çullaj et.al. 2000; Lazo & Çullaj, 2002; Lazo et.al. 2003). However, the most completed study concerning the PVC area was carried out in 2002 by GEOtestBRNO Czech Company. Its geochemical investigation was focused on the determination of mercury concentrations in ambient air, soil, groundwater, sea water and marine deposits. On the basis of the accomplished geochemical investigation, they found that the mercury contamination in soils is principally bound to the unsaturated (1.2 to 2.0 m depth) zone (Průša, 2006). In addition, they distinguished two heavily (> 75mg/kg Hg) contaminated areas with mercury, which correspond to electrolysis and de-mercurisation buildings, respectively.

From these studies was concluded that this area presents an unacceptable threat to the environment, not only for 180 families living now on and around the former plant but also for development of tourism, because this territory is situated right by the beach. On July 2002, a mission of UNEP/MAP (GEF Project GF/ME/6030-00-08) had identified this area as a "hot spot" and recommended a rehabilitation study to be carried out.

In the framework of the project Environmental Clean-up of the PVC plant in Vlora (December 2007 - January 2008), a geochemical investigation was carried out in order to study the current concentrations of the mercury in soil, water, sludge, leaching, air and to estimate the volumes of contaminated soils to be placed within a CDF. The analyzed soils show contents of Hg which do not coincide entirely with the respective Geotest Brno mercury levels, probably due to the heterogeneous spatial distribution of the mercury in soil. On the other hand, our analytical results confirm the same downward depletion trend of the Hg concentration, may be due to both soil structure and free groundwater fluctuations.

Based on the geological drillings, four lithogical layers with slightly different physical-mechanical features were distinguished. From the surface towards the depth a slightly increase of the silt and clay fraction was observed. Thus, the content of silt fraction ranges from 7.4% through 17.3% to 45% in the second, third and forth layer, respectively.

All the contaminated soils containing more than 10 mg/kg Hg will be landfilled within a Confined Disposal Facility (CDF) for the remediation of the PVC plant territory. The most contaminated (>75mg/kg) soils shall be washed for separating the metallic mercury, whereas fine sludge shall be stabilized with a sufficient amount of concrete or with elementary sulfur in stoichiometric amounts. The deposition of all the contaminated soils within the CDF will impede the mercury source for further contamination of water, air and biota.

2. AREA DESCRIPTION AND SAMPLING

2.1. Area description

The Vlora area, that is part of the Pre-Adriatic structural-facial zone, represents

a depression which, from the top to the bottom, consists of sands, silts and silty clays.

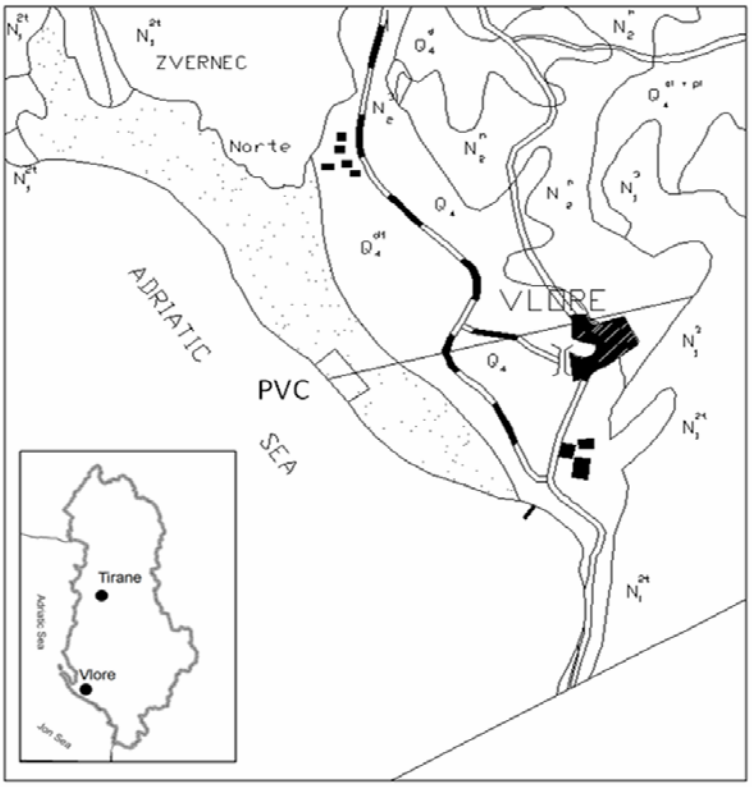

Figure. 1. Geological map of Vlora Bay (PVC location is pointed). *Q_4^{dt} – Quaternary marine deposits; N_2^P – Pliocenic clays; $N_1^3 – N_1^{2t}$ – Miocenic molasses; Dotted field – sandy beach.*

The above soft horizontal formations of Quaternary overly the folded molasses of Upper Miocene (N_1^3) and Pliocene (N_2^{pl}), mostly consisting of clay. The coast line of Vlora bay presents two main morphological types: the shallow, sandy, accumulative coast (Frasheri at al., 2002), where the PVC is placed and the high, rocky coast that is located in the southwestern sectors of the Bay (Fig.1).

The Quaternary deposits of Vlora bay belong to marshy, marine and marine lagoon type which consists of silty clay, sand - silty sand and silty clay – clay, respectively. On the other hand, the sands are aeolian or marine. The yellow aeolian sands contain small shell pieces, whereas their bottom is limited by the groundwater table that, in turn, coincides with the sea level. The marine sand that range from yellow (upper part) to grey-blue (lower part), are around 15.0 m thick. Based on the content of decomposed organic matter, two marine sand horizons can be distinguished: i) the upper horizon consisting of pure sand without clay material, where the fraction 0.01-0.25 mm represents around 75% and ii) the lower silty sand horizon containing up to 15% seaweeds, where the fraction 0.1-0.05 mm dominates (>75%).

The above downward increase of the fine fraction (clay-silt) was well observed during the drilling of five geotechnical wells (depth 10 m) within the territory of the

PVC. Four different layers have been distinguished (Fig. 2) based on their geotechnical features, where the particle size distribution and compactness extent seems to be the most important geotechnical parameters, from the geochemical point of view:

Layer 1 (1.0 – 1.5 m thick) represents a mixture of limestone clasts, building rubbish and loam with soft sand.

Layer 2 (1.3 – 2.4 m thick) consists of poorly graded sands with silt. From the top to the bottom the sand ranges from soft (weekly dense) to moderately dense. The content of dust and clay is 7.4% and 2.3%, respectively.

Layer 3 (5.0 – 6.2 m thick) consists of poorly graded sands with silt which are moderately dense, whereas the content of dust and clay arrives up to 17.3% and 2.7%, respectively.

Layer 4 (7,0-8,0 m thick) consists of silty sands, moderately dense, containing 44.8% dust and 2.7% clay.

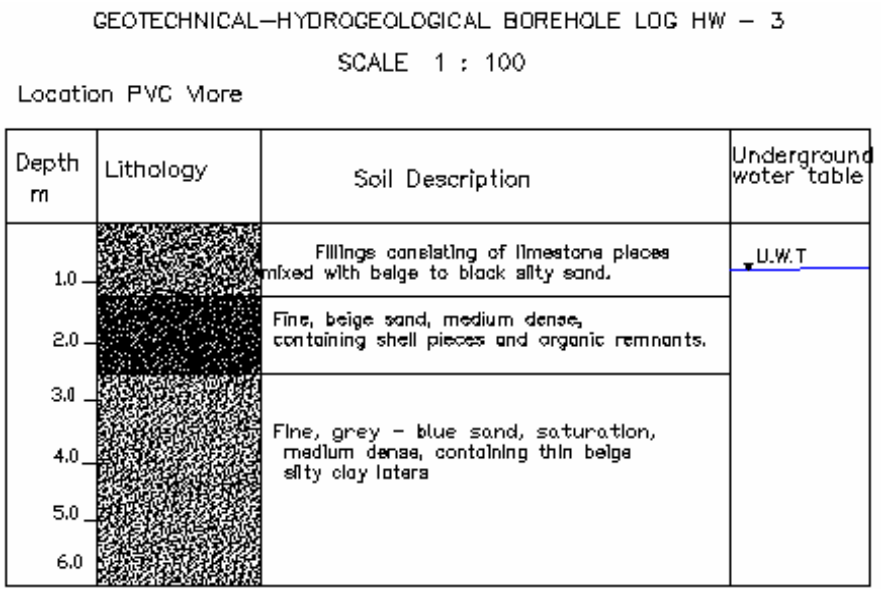

Figure.2. Geotechnical-Hydrogeological log (layer composition and underground water table (UWT) are shown)

Land cover of the study area consists of sandy soils developed in marine deposits. The soils vary from undeveloped sandy soils (beach sands) to sandy soils of old dunes. The land within the PVC territory is with sparse vegetation, while, the land around the industrial site is under planted and natural vegetation. According to the land suitability classification, the soils of the PVC territory could be classified as soils with limited suitability (Gjoka & Cara, 2003).

From the hydrogeological point of view, the PVC site consists mainly of water - bearing sandy section. This aquifer is characterized by low permeability features which are conditioned by its particle size composition. Thus, its hydraulic conductivity, that is less than 3-5 m/day ($6*10^{-3}$ m/s), coincides very well with the silty sand section of the site. Groundwater flows mainly towards the sea which represents the principal drainage area. The low rates of the flow are due to the sea water pressure, also.

As it can be seen in the Table 2 the groundwater level measured by us lies at depths below the ground that range from 0.6 to 1.3 m which are shallower than those measured by GEOtestBRNO group (up to 2.0 m below). This upgrade of groundwater table was probably due to the heavy precipitations. In fact, the precipitations represent the main recharge source of the unconfined sandy aquifer. The lack of an impermeable clay top soil and the flat surface of the site favored high infiltration rates of the water towards the aquifer that caused a fast upgrade of the groundwater table. The mean value of the seasonal groundwater fluctuation is found to be around 0.7 m.

2.2. Sampling

In order to assess the volumes of the contaminated soils and to evaluate the possible spatial migration of mercury since 2002, an extensive sampling of soil (48 samples), groundwater (10 samples), sludge material (14 samples), leaching (4 samples) was carried out (Fig.3)

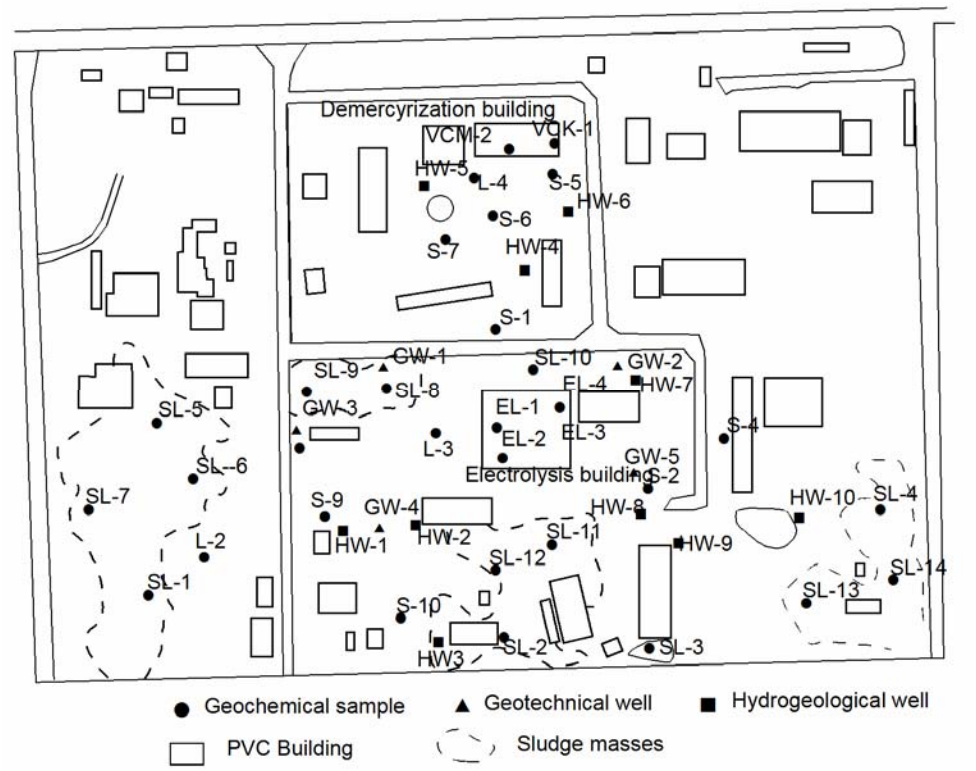

Figure 3. Schematic map of the former PVC plant with location of sample points and wells

Soil and groundwater samples were taken in the periphery of the two most contaminated areas of the PVC site that are de-mercurisation and electrolysis former plants. Soils were collected only from the unsaturated zone, from four depth levels of 0.0 m, 0.5 m, 1.0 m and 1.5 m, respectively. Groundwater was sampled about two weeks after the drilling of the hydrogeological wells in order to avoid any influence of

the suspended material in Hg content in water. Water samples are sampled on acid precleaned glass bottles and transported within day in laboratory.

The leaching samples were taken in the electrolysis (two), VCM (vinyl chloride monomer) (one) and chemical storage (one) buildings. A leachate sample represents a composite sample taken from 10 different points in the surface or 20 – 30 cm depth, with a distance of 5.0 m from each other. The same conservation procedure as for water samples is followed for leaching samples.

A sludge sample represents a mixture of 10 subsamples taken from 10 adjacent sludge piles, at depth 20-30 cm below the surface.

Six measurements of the mercury concentration in the PVC ambient air were carried out. Time duration of air sampling was 6 hours.

3. ANALYTICAL PROCEDURE

3.1. Determination of total Hg in soil & sludge samples:

An analytical procedure based on UNEP/IAEA Reference Method for Pollution Studies* is used. About 3 g sub sample is weight out accurately in glass beaker and is treated with 30 ml mixture of HNO_3+HCl (9:1); after 24 hours at laboratory temperature, it is heated in a hot plate at about 80 ^{0}C for 3 hours. After cooling, 2 ml of 5% $K_2Cr_2O_7$ is added and water up to 100 ml. Depending on mercury content in the sample, an aliquot of clear solution is used for mercury determination by Cold Vapor Atomic Absorption Spectrometry (CV-AAS) using a Varian SpectrAA 10 Plus apparatus and a home-made cold vapor accessory, having sensitivity about 0.2 ppb Hg. After the measurement of mercury absorbance in sample solution, an appropriate volume of a mercury standard solution is added in the sample matrix and the absorbance of standard is measured. The method of standard additions is used to calculate the concentration of mercury in sample solution. All the mercury results are reported in basis of dry matter. For a quality control, a Certified Reference Material (CRM) Sediment IAEA 356 purchased from IAEA Monaco is introduced for mercury analysis for every set of unknown samples.

3.2. Determination of total Hg in water samples

The water was filtered through 45 μm glass membrane filter and was conserved with HNO_3 to pH < 2 and stored at 4°C. Analysis was performed by Cold Vapor Atomic Absorption Spectrometry (CV-AAS) on 20-100 ml water samples, depending on level of mercury. After the measurement of mercury absorbance in sample solution, an appropriate volume of a mercury standard solution is added in the sample matrix and the absorbance of standard is measured. The method of standard additions is used to calculate the concentration of mercury in sample solution. All reagents used are "low in Hg" quality (from Merck).

3.3. Toxicity Characteristic Leaching Procedure (TCLP) for mercury

The test is implemented according to European Standard EN 12457-3 (2002). The sample material, having a particle size below 4 mm, is brought into contact with

water. This test is a serial batch leaching test consisting of two steps: first step is implemented using a solid to liquid ratio of 2 l/kg dry matter and agitation for 6 h ± 0.5 h and, subsequently, the second step is implemented using a solid to liquid ratio of 8 l/kg and agitation for 18 h ± 0.5 h. After filtration over a 45 µm membrane filter using a vacuum filtration device, mercury concentration along with temperature, pH, conductivity, redox potential was determined for each eluate.

3.4. Determination of metallic and non-metallic mercury in sludge masses

10 g subsample was placed in one 400 ml beaker in an oven for 48 hours at 150 ^{0}C to liberate elemental mercury from samples. The Hg content in the heated samples was determined and this concentration was compared with total content of Hg in the original sample (with no heating): the difference is elemental mercury.

4. EXPERIMENTAL RESULTS AND DISCUSSION

4.1. Hg content in soil.

The GeotestBrno geochemical investigations found the presence of two heavily contaminated areas with mercury within the territory of the PVC plant. They correspond to electrolysis and demercurization buildings, respectively. According to this study the concentration of Hg in the soil surface ranges from < 10 mg/kg to > 75 mg/kg (Průša, 2006). Our samples, which were taken around the periphery of the above two most contaminated areas, show contents of Hg which do not coincident entirely with respective results of GeotestBrno sample points. Our soil results are presented in table 1.

Table 1. Hg content (mg/kg DW) in soil, PVC in Vlora

No. sample	Sampling depth			
	0.0 m	- 0.5 m	-1.0 m	-1.5 m
S1	53.3	1.6	0.2	1.2
S2	76.1	26.4	9.3	n.d.
S3	20.6	15.1	13.6	6.7
S4	9.8	0.75	0.33	0.84
S5	30.0	15.7	3.0	0.39
S6	34.8	0.17	<0.1	0.9
S7	8.7	0.8	<0.1	0.3
S8	84.5	6.9	1.2	0.34
S9	3.4	1.8	0.6	0.21
S10	1.65	0.52	0.58	1.6

The lack of a perfectly correspondence between our analytical results with those of the Geotest Brno may reflect the heterogeneous spatial distribution of the mercury in soil. This can be explained by its random loose (particularly because the great part of mercury is in liquid metallic form), non regular surface water flow and the heterogeneous soil structure, but also by the displacements of surface soils during this period due to the new construction activities.

Our analytical results confirm the same downward depletion trend of the Hg concentration reported by Průša, 2006. In fact, the Hg content in the surface samples is clearly very high and it shows a roughly downward depletion (sample S8: 84.5mg/kg, 6.9 mg/kg, 1.2 mg/kg and 0.34 mg/kg in soil depth 0.0 m, 0.5 m, 1.0 m. and 1.5 m, respectively). However, the intensity of the downward Hg content decrease is not the same for different sampling points. As shown in the table 1, in contrast with the general tendency, a gradual downward decrease of Hg content in soil was observed in the sampling points S3 and S9 (Fig. 4), which occur in the northwestern part of the electrolysis building. The very low concentration of mercury toward the depth along with its limited vertical migration may be due to both silt content of the mostly sandy section and the fluctuation of the free groundwater table. Çullaj et al., (2004) found that the fine size fraction accumulates the major part of Hg, thus acting as barrier against the downward migration of mercury. On the other hand, the migration of mercury from surface to depth 2.0 m coincides perfectly with high seasonal fluctuations of groundwater level that range from 0.0 m to 0.6 m during the heavy precipitations up to 2.0 m during the dry season.

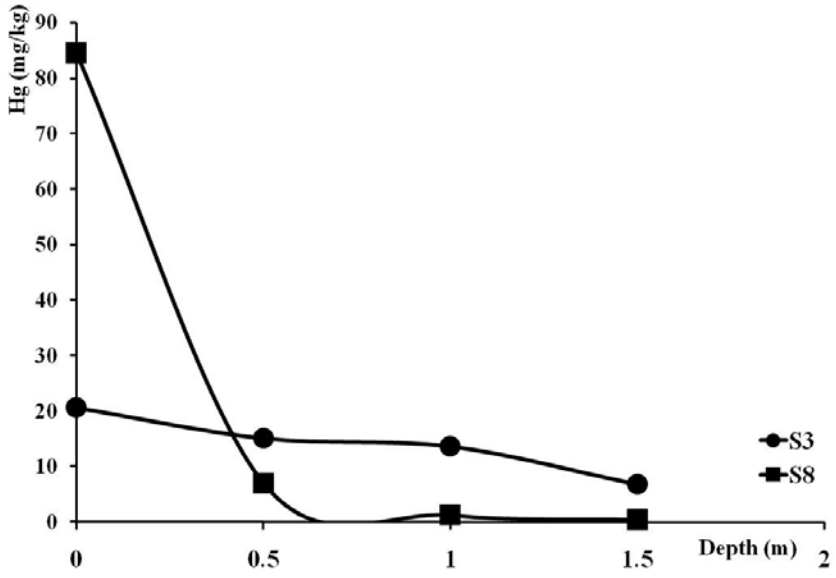

Figure 4. Downward depletion of Hg concentration in soil

4.2. Hg content in groundwater

The content of the mercury in the tested groundwater samples ranges from < 0.2 μg/L to 12.2 μg/L (Table 2). These contents are higher than those (<1.0 – 3.5 μg/L) reported by Geotest Brno company (Novotná et al., 2006). The evident Hg enrichment in groundwater can be addressed to two factors:

The first factor is the position of the well with respect to most contaminated areas. Apart from other factors, it could be expected that the concentration of mercury

in groundwater to be positively correlated with its content in soil. Higher is the Hg content in the soil, higher will be its content in the corresponding groundwater. This is particularly valid for the medium with low permeability as it is our case. Our wells are drilled just in the periphery of the two most contaminated areas.

Table 2. Hg content in groundwater

No. well	GWL below the ground (m)	No. sample	Hg content (μg/L)
HW-1	0.1	GW1	< 0.2
HW-2	0.7	GW2	11.7
HW-3	0.6	GW3	0.38
HW-4	1.1	GW4	1.9
HW-5	1.3	GW5	0.25
HW-6	1.0	GW6	< 0.2
HW-7	1.3	GW7	< 0.2
HW-8	0.4	GW8	12.2
HW-9	0.9	GW9	1.42
HW-10	1.1	GW10	< 0.2

The highest Hg contents (11.7μg/L and 12.2 μg/L) in groundwater belong to wells HW-2 and HW-8, respectively, which are located in the proximity of the western side of the electrolysis building. On the other hand, the lowest Hg contents are found in the wells HW-1, HW-6, HW-7 and HW-10 because of their position out of contaminated groundwater flow.

The second factor is the season when the water sampling was carried out. The groundwater sampling by part of Geotest Brno was performed during the dry summer time. In this season the head of groundwater is probably below the most contaminated (up to 2.0 m deep) topsoil. Thus, the quantity of the soluble Hg in water will be decreased. In addition, the missing of precipitations does not favor the washing and solution of the mercury in groundwater. On the contrary, we sampled the groundwater from the boreholes in January 2008, after a period of heavy raining. This later had a significant impact to the upgrade of the groundwater head. Both the downward infiltration of the surface water and the upward moving of the free groundwater table have favored the solution of the mercury.

4.3. Hg content in leachate

The content of the mercury in leachate ranges from 0.74. 10^{-3} mg/kg to 5.53. 10^{-3} mg/kg. The highest values correspond to samples from the vicinity of electrolysis and VCM buildings. The content of mercury in leachate is strongly lower with respect to the corresponding content in soil. It means that the soil of the PVC territory represents very low leachibility features probably due to both its silt content in the sandy section and the very low content of organic matter that favors the transfer of the metallic mercury in solution.

4.4. Hg content in sludges

The sludge represents technological waste of the PVC plant. They consist of carbide dust, precipitate of the generators of the acetylene gas production and precipitate of the brine. This later is responsible for the contamination of the sludge with mercury. The content of mercury ranges from 0.33 to 156.0 mg/kg (Tab. 3). Such an extremely heterogeneous distribution of mercury in sludge masses is related with different content of Hg in the brine precipitate. In addition, there is no vertical distribution trend in mercury concentration because each layer corresponds to a cycle of the deposition of sludge that contained a different amount of mercury (Průša, 2006).

Table 3. Hg content in sludge samples

No.	No. Sample	Hg content (mg/kg)
1	SL-1	18.3
2	SL-2	0.65
3	SL-3	18.0
4	SL-4	0.33
5	SL-5	156.0
6	SL-6	108.6
7	SL-7	17.9
8	SL-8	4.9
9	SL-9	3.2
10	SL-10	60.6
11	SL-11	0.8
12	SL-12	1.3
13	SL-13	67.6
14	SL-14	94.5

4.5. Content of Hg compounds

Three soil samples, two from VCM building and one from chemical storage, were tested for the presence of Hg compounds. The concentration of total mercury in these samples was: 2748-2840 mg/kg (VCM-1), 627.5-650 mg/kg (VCM-2) and 1.52-2.0 mg/kg (CS-1). The above samples do not contain measurable concentrations of the metallic mercury, in distinction of soil samples where the great part of mercury is in forms of tiny liquid drops. They contain only chemical compounds of mercury which are stable in relatively high temperature. This is because the mercury in the VCM samples occurs in the form of mercury chloride which was used for the production of the PVC.

4.6. Hg content in air

Five measurements of the Hg content in air within the territory of the PVC plant and one about 1.2 km away from it were carried out. The content of mercury in the air of the PVC territory ranges from 1.42 to 1.83 $\mu g/m^3$. These values correspond to the mean Hg concentration (0.01 – 30 $\mu g/m^3$) of Hg in air reported by Geotest Brno

(Průša, 2006). The weather, when the measurements were performed (February 2008), was typically characterized by a high atmospheric pressure that has probably favored such a concentration of the mercury vapor in the air of the area of concern.

5. CLEAN-UP OF THE PVC PROCESS AREA

In order to immobilize mercury in the contamination source from the surrounding environment (air, water, soil, biota), the clean-up of the contaminated PVC area was proposed as the best way for its environmental remediation. Selection of the remediation technology depends on the remediation target, which means the establishment of the maximum acceptable residual contamination content in soil and on the remediation deadline (Jaroslav, 2006; Canstein & Timmis, 1999). A combination of conservation – decontamination technologies is proposed which consists in contamination encapsulation, gravity separation, excavation of contaminated soil and their landfill deposition. The CDF construction represents the best solution for the PVC area remediation because of the variability of contaminated materials to be cleaned up. They consist of contaminated soil, sludge conglomerates, debris from factory buildings, underground cables and pipes, chemicals, etc. An evaluation of respective volumes of the contaminated masses is given in the table 4. On the other hand, the mostly elemental form of mercury excludes the possible use of biochemical remediation technologies (Wagner-Döbler, 2003).

All the contaminated soils containing more than 10 mg/kg Hg will be landfilled within a Confined Disposal Facility (CDF). The most contaminated (>75mg/kg) soils shall be washed before being landfilled in order to separate the metallic mercury. If during the washing, sludge is formed, which has a particle size too small for a successful washing, it shall be stabilized before depositing in the CDF.

Table 4. Volumes of contaminated masses

1	Contaminated soils over10 mg/kg	24000 m^3
2	Washed soils <75 mg/kg	4000 m^3
3	Soils >75 mg/kg with Hg compounds	1000 m^3
4	Stabilized wash sludge	500 m^3
5	Electrolysis house foundation	500 m^3
6	Contaminated external masses	20 000 m^3
7	Total	50 000 m^3

The stabilization shall be done with a sufficient amount of concrete to chemically bind the metallic mercury and compounds in the structure. It is recommended to add elementary sulfur in a small excess of stoichiometric amounts for further immobilization of mercury. The isolation of the CDF will be done according to EU directives for hazardous landfills. The main component for both top and bottom layers is the natural clay, with permeability less than 4×10^{-10} m/s (Fig. 5). The Pliocene clay, that crop out on the eastern hills of PVC area, is suitable to be used for the CDF isolation (Beqiraj et al., 2007). The deposition of all the contaminated masses

within the perfectly isolated CDF will avoid the possible source for further contamination of water, air and biota with mercury.

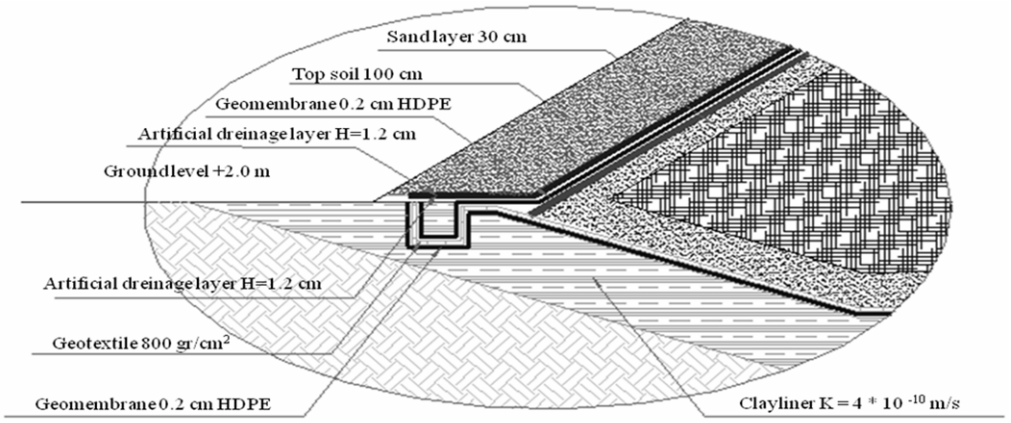

Figure 5. Transversal section of CDF structure at its edge

6. CONCLUSIONS

The territory of the former PVC plant represents a high-contaminated area with mercury, caused by technological looses and non controlled waste discharges. It was placed on a sandy basement which shows a downward increase of silt (7.4% - 45%) content. The electrolysis and demercurization buildings represent the most contaminated (> 75 mg/kg Hg) areas of the PVC territory. In the framework of the heterogeneous spatial distribution in soil, the Hg content shows a roughly downward depletion trend due to the increase of silt content and fluctuations of the free groundwater table. The highest Hg contents (11.7µg/L and 12.2µg/L) in groundwater are found in the proximity of the heavily contaminated electrolysis building. The soil of the PVC territory represents very low ($0.74.\ 10^{-3}$ - $5.53.\ 10^{-3}$ mg/kg) leachibility features probably due to both high silt and very low organic matter content. The content of mercury in sludge shows an extremely heterogeneous (0.33 to 156.0 mg/kg) distribution because of different content of Hg in the brine precipitate. In the VCM building, the mercury occurs entirely of mercury chloride which was used for the production of the PVC. The content of mercury in the air of the PVC territory ranges from 1.42 to 1.83 µg/m^3. The clean-up of the contaminated PVC area, through a combination of conservation – decontamination technologies, was proposed as the best way for its environmental remediation. The deposition of all the contaminated masses within a perfectly isolated CDF will avoid the source for further contamination of water, air and biota with mercury

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