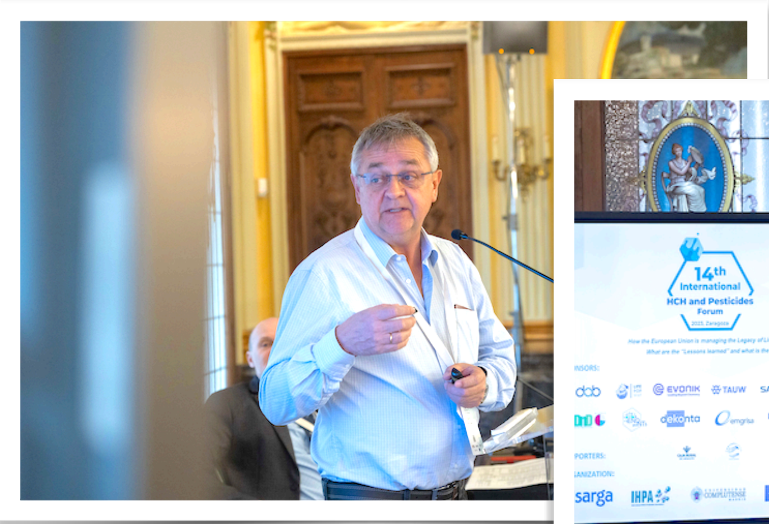
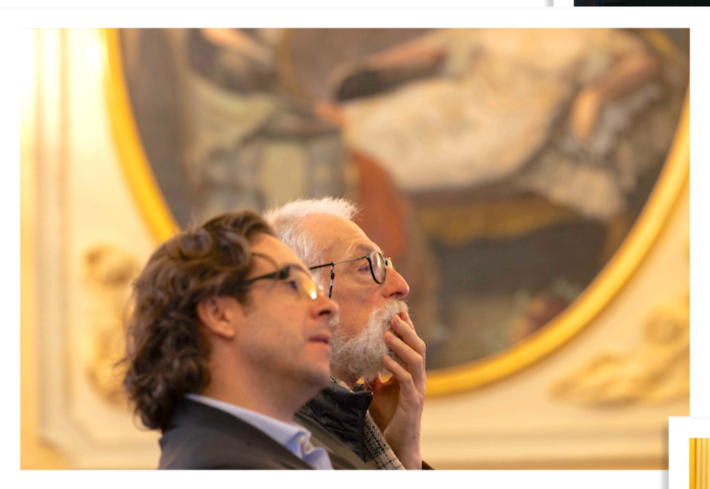
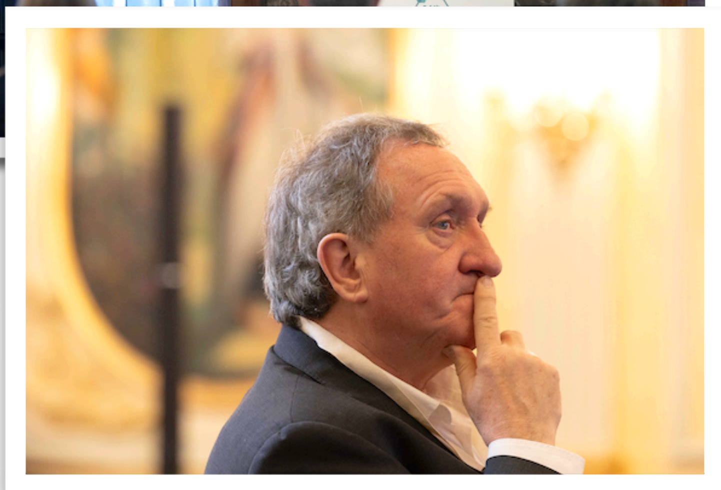
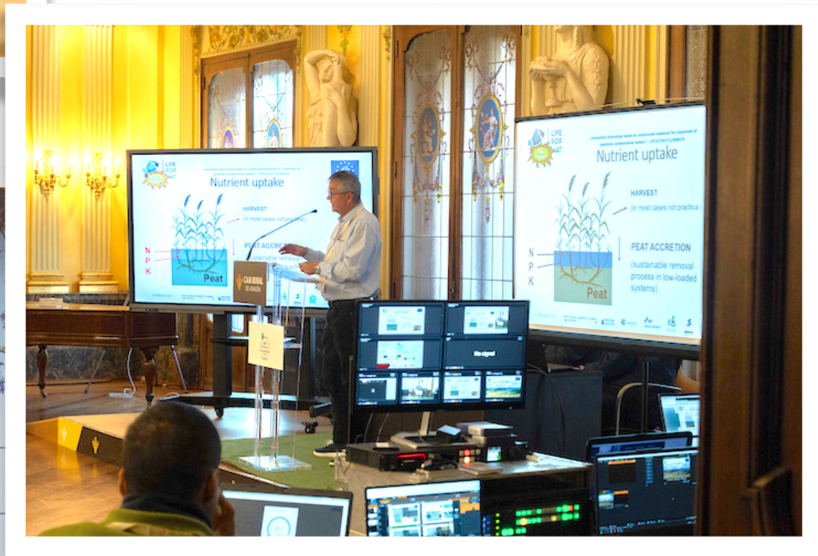
Block 7

LIFEPOPWAT









WETLAND+® TECHNOLOGY: TREATMENT OF HCH CONTAMINATED WATER BY A PASSIVE BIOLOGICALLY BASED REMEDIATION SYSTEM

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Summary

LIFEPOPWAT is a European project focusing on innovative technology based on constructed wetlands for the treatment of pesticide contaminated waters, funded by the European Union LIFE Programme under grant agreement number LIFE18 ENV/CZ/000374. The Wetland+® system is robust, low maintenance, and sustainable treatment can be deployed in remote locations where access to infrastructure may be limited. The Wetland+® technology is based on the use of oxidation-reduction and biosorption methods. The first prototype was installed at Hajek (the Czech republic). The drainage water, after the removal of dissolved Fe in *the sedimentation tank*, enters the first reactive stage - *a permeable reactive barrier* filled with Fe chips, where it is deoxygenated and converted back to the reduced state. Subsequently, HCHs are partially dechlorinated, and chlorobenzenes (ClB) are formed. The second reactive step is the *biosorption unit*, where HCH compounds are sorbed and subsequently degraded by present microorganisms. The last step is the *aerobic wetland*, where the plant root system purifies the water, and the concentration of HCHs and their daughter products decrease below specified limits. The technology no needs additional chemicals and energy, and the whole process is naturally based. The system was finished in September 2021 with a capacity of 3 L⁄s. The initial efficiency of our systems was 97.3% for ClB and 81.5 % for HCHs. During the next months, due to a tunning of the system, the efficiency was increased to almost 100% for ClB and 97 % for HCHs. The system is ready for replications in other sites with similar HCH contamination [1], [2].

Keywords

Wetland+, HCH remediation, oxidation-reduction processes, bioremediation, passive remediation system

The site

The project's primary pilot site is the former uranium mine and its repository in Hajek (CZ). In the 1960s (and until 1971), uranium mining was carried out on the site. In parallel with uranium mining, kaolin was mined, and basalt and later bentonite were mined in the foreground. Between 1966 and 1968, the state authorities decided to dispose here of the ballast HCH isomers and waste chlorobenzenes from the chemical production of lindane (γ-HCH) in Spolana Neratovice (CZ). These substances were placed in various parts of the Hajek dumpsite in metal drums or even in paper packaging or in bulk. The estimated quantity of these chemicals deposited in this way is 3,000-5,000 tonnes. The drainage system was installed to collect drainage water from the site.

In 1977, a landslide occurred on the spoil heap (at an area of about 10-12 ha), and a part of stored chemical waste was exposed. The landslide was remedied by the construction of a weighting bench of crushed aggregate into which a drainage system consisting of pipe drains was incorporated. Since January 1989, the concentrations of hexachlorocyclohexane (HCH) isomers and chlorinated benzenes (ClB) have been monitored and documented at the outlet of the drainage system and it the downstream water bodies (creeks and lakes). A sampling of boreholes drilled in 1994 showed that groundwater contamination with organic substances had already exceeded the contour of the spoil heap and was spreading mainly toward the flooded Hajek quarry.

Since 1991, hydrological, climatological, and hydrochemical monitoring has been carried out in the area of interest. Between 1999 and 2002, DIAMO carried out Phase I remediation work. This stage consisted of the construction of a sealing and covering element over the landslide area (laying of 0.3 m thick bentonite with a 0.45 m thick cover of heap material).

Technical and Economic Study

In 2015, Aquatest a.s. prepared a Technical and Economic Study (TES) for DIAMO on the reclamation of the Hajek Quarry drainage cleanup. This TES addressed five remedial measures. Specifically, these were:

1. Drainage effluent treatment: the most appropriate method of passive treatment of the drainage water from the spoil body was designed to remove contaminants such as isomers of hexachlorocyclohexane (HCH) and chlorobenzene (ClB) from the water before it enters a natural receiving water body.
2. Ensuring the stability of the spoil body: the deformations measured so far have been verified, and a design for the final stability of the dump body has been developed.

3. Avoiding the influence of infiltration and atmospheric precipitation: The influence of infiltration of atmospheric precipitation into the dump body on its stability and on the quality and quantity of drainage water was verified, and technical measures to prevent the infiltration of atmospheric precipitation were proposed.
4. Elimination of the impact of groundwater inflow: The inflow of groundwater into the landfill body has been verified, and the technical measures to eliminate them have been proposed.
5. Verification of possible HCH deposition and proposal of measures: It was not possible to verify the locations of possible deposition of ballast isomers of HCH in the body.

Geochemical parameters of drainage water

Basic geochemical characteristics of the drainage systems at the Hájek dump are the following: The water flow is approximately 3 l/s with relatively high annual fluctuation. The average content of HCH and ClB is about 100 µg/l and 600 µg/l, which are formed by the transformation of HCH, respectively. These pollutants, with a mass flux of approximately 25 g HCH and 150 g ClB per day, flowed from the site via the Ostrovský brook to the Hájek preserve and the adjacent breeding ponds Horni Stit and Dolni Stit. The drainage water had the character of mine water with high mineralization of about 1.5 g/l and approximately neutral pH. The predominant anions were SO_4^{2-} (600 mg/l) and HCO_3^- (300 mg/l), and the predominant cation was Ca^{2+} (180 mg/l). A major complication for the treatment of the drainage water was the higher Fe^{2+} content (20 mg/l as Fe_{tot}). Precipitates of Fe oxyhydroxides and siderite covered all surfaces in contact with water in the aerated parts of the drains and in the downstream Ostrovsky Creek. The groundwater in the body of the dump was classified in detail in [3], and the history of the laboratory [4] and pilot testing of the various stages of the cascade at the site, and the design details of the first Wetland+® prototype were described in [5].

System Wetland+®

Based on the results of the pilot project, a system for the full operation was designed. This system was built within the LIFEPOPWAT project in 2021. The system (Fig. 1) has 4 stages:

A) sedimentary tank

The aeration and sedimentation unit consists of aeration steps and a tank for the deposition of ferrous sludge with a total area of 313 m² and a depth of 0.5 m. The size of this unit is small, causing relatively short water residence time and low efficiency of a sludge separation. The water flow time through the unit is about 23 hours for the flow 3 l/s.

B) permeable reactive compartment

The permeable reactive barrier (PRB) is formed by three parallel branches, each of them constructed by two basins. The basins were filled by Fe chips (Fig. 2). Here the treated water loses dissolved oxygen and gains a low redox potential (ORP) for HCH dichlorination. This step is mainly added to decompose the most persistent beta, gamma, and epsilon HCH isomers under the sulfate-reducing condition. For a better flow of water in the Fe chips and a longer residence time, the basins are divided by wooded boards to force water to flow up and down. The total area of the PRB is 540 m² and the total volume 360 m³. The water flow time through the unit is about 10 hours for the flow 3 l/s.

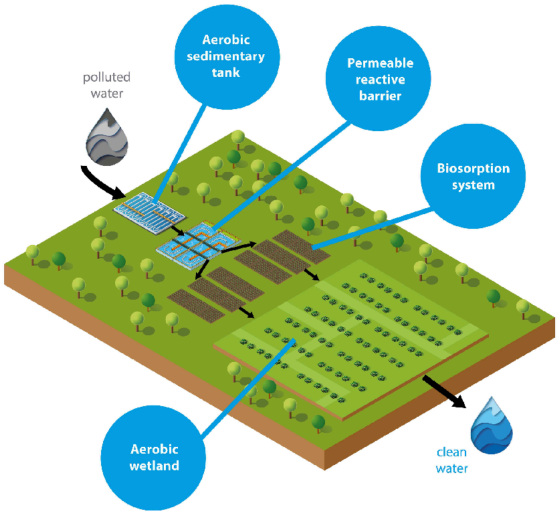

FIGURE 1. PROTOTYPE OF THE WETLAND+®

FIGURE 2 FILLING CAST IRON SPONGES INTO MODULE B (+ DETAIL)

C) Bioreduction and sorption unit

The treated water continues to the next compartment – the biosorption modules. There are two parallel anaerobic sorption modules C1 and C2 filled with a mixture of peat (40%), crashed stones (30%), loamy soil (20%) and wooden chips (10%). The unit is covered with reed canary grass and common reed. The total area of the tanks is 650 m², the total volume is 480 m³. Massive precipitation of Fe oxyhydroxides occurs on the substrate surface in modules C, resulting from the change in redox conditions between stages B and C (Figure 3). The water flow time through the unit is about 10 hours for the flow 3 l/s.

FIGURE 3. ROOT FILTER MADE OF WOODY PLANTS, ANTLERS IN THE UNDERGROWTH, MODULE D

D) Aerobic wetland system for final treatment

The wetland is characterized by a high biodiversity of plants reintroduced from the surrounding wetland reserves. In addition to the emergent vegetation of classic and rare wetland species planted in the summer of 2021, transverse sills (root filters) of wetland alder and willow species, designed as emergent with a short rotation period, were added in the spring of 2022. Totally, three to four plants per m^2 were planted. Plant samples from the HCH and ClB phytoaccumulation monitoring are still in the process of laboratory analysis, as are bacterial DNA samples from the rhizosphere of wetland plants from Stages C and D. The aerobic wetland system is comprised of one compartment with a total area of 2,669 m^2, and the total aerobic wetland volume of 1,600 m^3. The bottom of the wetland was filled with loamy soil (50%), crashed stones (30%), compost (10%), and wooden chips (10%). This stage serves for the final removal of organic substances, suspended substances, and decomposition products of HCH, especially chlorobenzenes. The aerobic wetland was filled with good quality soil as a growth medium for the wetland plants (approximately 40 cm high), and the water level above is limited to a maximum of 20 cm. The water flow time through the unit is about 65 hours for the flow 3 l/s.

Conclusions

The Wetland+® technology based on oxidation-reduction and biosorption methods was installed at Hajek (the Czech republic). The system consists A) *a sedimentation tank,* for the removal of dissolved Fe, B) *a permeable reactive barrier* filled with Fe chips, for HCHs partial dechlorination, C) *a biosorption unit*, where HCH compounds are sorbed and subsequently degraded by present microorganisms, and D) *an aerobic wetland*, where the plant root system purifies the water, and the concentration of HCHs and their daughter products decrease below specified limits. The initial efficiency of our systems was 97.3% for ClB and 81.5% for HCHs. During the next months, due to a tunning of the system, the efficiency was increased to almost 100% for ClB and 97% for HCHs.

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ADAPTATION OF METHODOLOGICAL ASSUMPTIONS FOR DESIGN OF PILOT SCALE WETLAND+ INSTALLATION FOR WATER TREATMENT FROM HCH TAKING INTO ACCOUNT PRACTICAL LESSONS FROM DIFFICULT FIELD CONSTRUCTION PROCESS IN JAWORZNO, POLAND (LIFEPOPWAT PROJECT)

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The construction of the pilot scale Wetland+ installation for water treatment from HCH in Jaworzno, Poland is one of the key elements of international project LifePOPWAT implemented by 7 partners from 4 countries (Czech Republic, Poland, France and Denmark) under coordination of Technical University of Liberec (CZ). The Municipality of Jaworzno (PL) is responsible for design and installation of the pilot scale treatment installation while PhotonWater (CZ) and Central Mining Institute (PL) provide methodological assumptions for the design. The original design developed in late 2020 have been the subject of construction works performed from August 2021 till June 2022 by contracted company. However, the contracted company finally failed to deliver the proper installation according to the design from 2020. Therefore, there was an urgent need to adapt at first the methodological assumptions and then to develop the updated design. The updated methodological assumptions had to take into account the lessons from the previous failed construction of the pilot system. The current conference paper describes the lessons learnt while transferring the methodology developed in Czech Republic into a real field pilot scale installation in Poland. The finally adapted methodological assumptions developed for the construction design are also presented. The design changes concerned technical aspects such as adapting the installation design to the materials available on the Polish market, improving the connection between the tanks, implementing solutions to protect against vandalism and damage. Technological aspects were also refined to increase the functionality of the Wetland+ system: a change of the installation control system, a change of the installation monitoring system, an adjustment of the filling of the chambers with reactive material to Polish conditions, a change of the aeration and sedimentation system, a verification of plant species for planting.

Keywords

Design, wetland, permeable reactive barrier, PRB, implementation, scaling up, construction

1. Introduction

An uncontrolled migration of pesticides in groundwater poses a serious threat to drinking water resources. One of the most toxic pesticides produced in Europe since the 1940s until the 1980s was lindane, or γ-HCH. The γ-HCH isomer has insecticidal properties. The other HCH isomers do not have such properties. The production of lindane proved to be the biggest problem, as for every 1 tonne of γ-HCH, 8 tonnes of the other HCH isomers were produced, which also have toxic properties (Dominguez et al., 2016; Dominguez et al., 2018; Wacławek et al., 2019; Kończak et al., 2022). Both lindane and its post-production wastes were often disposed of inappropriately, resulting in uncontrolled migration of these compounds into the soil and water environment. In Poland, it has been estimated that 35,000 tonnes of HCH waste is still present, of which the largest amount is stored in Jaworzno. Therefore, there is a need to find suitable technologies to reduce the concentration and eco-toxicity of these pesticides. Due to the complexity of the contaminants stored in Jaworzno and the presence of other persistent organic pollutants such as chlorobenzens (CBs), chlorophenols (CF), dichlorodiphenyldichloroethylene (DDE), dichlorodiphenyldichloroethane (DDD), dichlorodiphenyltrichloroethane (DDT), chloroethenes and chloromethane, the technology used should be based on both chemical oxidation of contaminants, physical treatment methods through sorption processes and biological treatment methods using plant biomass and coexisting microorganisms. An example of such technology is the Wetland+ technology being developed within the LIFEPOPWAT project. The project LifePOPWAT is implemented by 7 partners from 4 countries (Czech Republic, Poland, France and Denmark) under coordination of Technical University of Liberec (CZ). The Municipality of Jaworzno (PL) is responsible for the design and installation of the pilot scale treatment installation (named P2 wetland system) while PhotonWater

(CZ) and Central Mining Institute (PL) provide methodological assumptions for the design.

2. Pilot site description

The site is located in Jaworzno in southern Poland (Figure 1) where the Chemical Plant "Organika-Azot" produced lindane from 1965 till 1982 (AMIIGA 2017, Kończak et al., 2022).

FIGURE 1. THE LOCALIZATION OF STUDY AREA (AFTER KOŃCZAK ET AL., 2022)

The P2 wetland system is located on the south of the Wąwolnica stream where the concentration of the sum HCH is even above 300 µg/L, including α-HCH, γ-HCH and δ-HCH over β-HCH. Such isomer composition with a low β-HCH share is typical for relatively "fresh" contamination (Gzyl et al. 2014).

The pilot system is located between trenches R2 and R3 which are the part of the network of dewatering trenches constructed on the bottom of former sand pit “Rudna Góra” which is currently partially filled in by HCH waste (Figure 2). This drainage system is managed by the Chemical Plant "Organika-Azot". The advantages of this localization is that it is possible to direct the water of the different level of contamination to P2 wetland system. Thetrench R3 is collecting heavily contaminated water where the concentration of sum of HCH in 2020 was ca. 79 µg/L and the trench R2 is collected less contaminated water with the concentration of HCH was ca. 1 µg/L (Figure 3).

3. The original design of P2 wetland system with different level of HCH contamination

The original design of P2 wetland system was developed in late 2020. It included:

- two pumping **wells** located in the bottom of trenches R2 (well SW1) and R3 (well SW2)
- the mixing tank in which the water of desired level of contamination would be obtained and then flow gravitationally along the further elements of the system
- **oxidation and sedimentation module** for the precipitation of excessive iron content from the water
- zero-valent iron (ZVI) compartment for chemical reduction – **ZVI module**
- anaerobic wetland for further reduction and biosorption (**Biosorption module**)
- aerobic wetland for final bioremediation (Z1)
- infiltration wetland for reverting the treated water into groundwater and for increasing biodiversity (Z2)

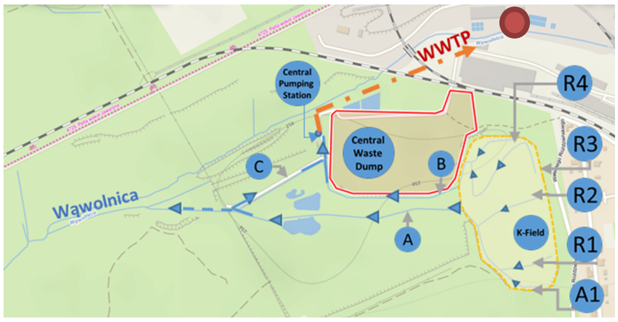

FIGURE 2 THE LOCALIZATION OF P2 WETLAND SYSTEM

Legend: Red dot – P2 wetland system localization; R1-R4 and A1, A, B, C – the trenches collected contaminated water to wastewater treatment plant (WWTP) and partially to Wąwolnica stream

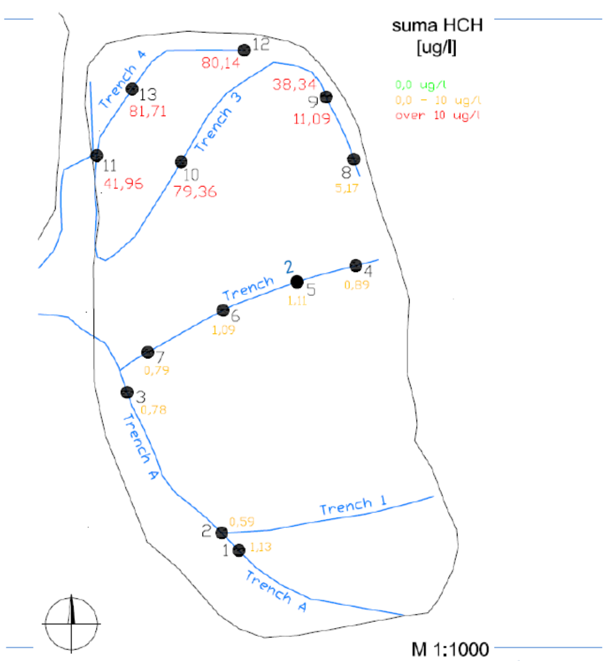

FIGURE 3 THE CONCENTRATION OF HCH SUM IN TRENCHES

The preparation for the construction implementation has started in June 2020. Several legal permits were needed prior to the start of field works. It was needed to get the notification of 1) the construction works from the Mayor of Jaworzno; 2) power connection condition from Tauron Dytrybucja SA.; 3) the permission for felling trees and shrubs from the Marshal of the Silesian Voivodship; 4) the water legal permit from the State Water Management Company Polish Water.

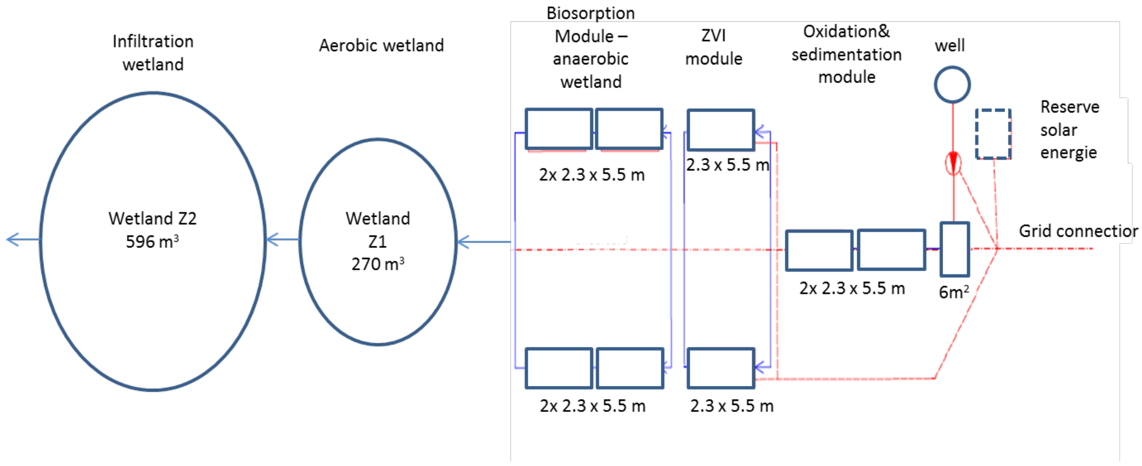

FIGURE 3 THE ORIGINAL DESIGN OF P2 WETLAND SYSTEM

The construction itself have been postoned several times which resulted in significant delays in its completion and commissioning. The main problems were at first re-tendering of the construction contractor due to obtaining in the 1st tender very high prices of the offers that exceeded the budget limits.

Nevertheless, the contractor failed to deliver the installation and was finally removed for the site in June 2022. This resulted in the need to launch a new tender procedure for the completion of the construction.

In July 2022 the works commenced to assess the current status and prepare the design documents for the tender to complete the construction. Finally, in October 2022, the tender was issued and in November the new contraction company was chosen. Finally, in October 2022, the tender was issued and in November the new construction company was chosen. It is expected that the complete P2 installation will be ready in the very end of December 2022.

4. The changes in a design of P2 wetland system

The new situation after removing the previous contractor and the need for preparation of new construction design for the new tender enforced many changes compared to original design. A checklist was prepared with a specific task plan for the completion of the construction. With the help of this checklist many changes have been implemented including construction materials, practical aspects like endurance and vandal-proof solutions as well as technological aspects. They are described in details in the following chapters.

4.1. Adaptation of construction materials to existing conditions

During the COVID pandemic, problems with steel production and supply in the intra-EU market began, exacerbated by the war in Ukraine and the rising costs of energy, gas and raw materials for steel production. Steel producers are not taking measures to increase capacity utilization and thus steel availability, due to the significant increase in production costs. Steel mills and distributors are primarily focused on fulfilling long-term contracts. As a result, this has reduced steel availability for the current steel purchases.

FIGURE 4. THE UNCOMPLETED P2 WETLAND SYSTEM IN JUNE 2022

It was decided to change the construction materials of the tanks. Currently, the tanks are made of concrete. Some of the tanks were purchased as ready-made tanks available on the Polish and European markets. Other tanks with non-standard dimensions were tailor-made to the needs of the project.

4.2. Improvement the design of the installation in practical aspects

The field inventory showed many technical deficiencies necessary to improve both at the stage of the installation design, as well as its reconstruction and construction (Figure 6). First of all, the connections between the tanks were faulty - made of materials not adapted to work outside (Figure 5). It was also shown that there is a need to change the construction and protection of the system of connections between the tanks (valves and connections of distribution pipes will be placed in closed and secured boxes), which is aimed at both improving the functionality of the system and protecting against vandalism. The method of covering the tanks has also been changed. In the original design, some of the tanks were uncovered. The construction company delivered on the site the concrete tanks with concrete covers. The new project solution is to cover the tanks with a tarpaulin, which on the one hand is to protect against vandalism, against contamination with plant material (e.g. leaves), and on the other hand is meant to facilitate access to the tanks.

4.3. Adaptation of the technological aspects to the local conditions

4.3.1. Changing the aeration system

According to previous design, the first containerized module was the oxidation and sedimentation unit. The previous design assumed that there will be a need for active oxidation in order to eliminate the excessive iron content from contaminated water prior to treatment in further modules. Now, using the experiences from already functioning Wetland + system in Hajek (CZ) and taking into account lower values of iron in water in Jaworzno (PL) compared to Hajek (CZ), this idea has been changed.

FIGURE 5. THE UNACCEPTABLE CONNECTION BETWEEN TANKS PERFORMED BY PREVIOUS CONTRACTOR

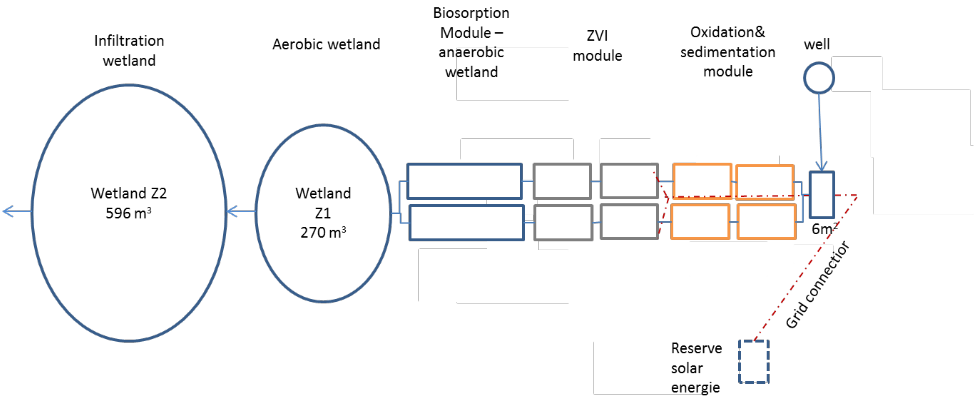

FIGURE 6. NEW SCHEME OF P2 SYSTEM

The results from Hajek shown that it is impossible to enforce full precipitation and sedimentation inside the module. The process is starting in the oxidation and sedimentation module but it continues also throughout all downstream modules. In this way the primary goal of oxidation and sedimentation module, which is to avoid precipitation downstream, is not fulfilled. On contrary, the active oxidation is causing in fact more problem than without this model being in use. Moreover, the next module is for chemical reduction using zero valent iron. Therefore, it is much better idea to prevent the contact with oxygen and to prevent iron precipitation than to oxidize the water actively. Therefore in the new adapted version of design in Jaworzno the first module is dedicated only for sedimentation of suspended matter, without oxidizing the water.

4.3.2. Adaptation of filling materials to existing conditions

During the construction works, there was also a problem with the availability of iron chips (as a source of reactive iron) for filling the permeable reactive barrier (PRB) tank. The contractor provided iron chips from wet metalworking for testing. The results from preliminary test showed the contamination of the iron chips with an oil fraction. This rules out the possibility of using the system for pesticide treatment, as it poses the risk of additional water pollution. In addition, the status of the iron chips was the waste and its use for the installation requires a legal permit with difficult and long procedure. Therefore, in the new design project, it was decided that iron chips should be replaced with commercially available product. In Poland, Pol-Aura is the only supplier of micro-iron chips. However, the unit price of iron exceeds 80 euro per kg. On the European market exist micro-iron producers such as Hoganas (Sweden) and

Nanoiron (Czech Republic). The micro iron could be also obtained from the Hepure company (US). It should be noted that despite the availability of micro-iron on the European market, there is a difficulty in supplying them to the Polish market, mainly due to the fact that these companies don't have representative offices in Poland, which will make it difficult to purchase, transport and receive this product.

4.3.3. Optimization of the monitoring system

The design of the P2 system after its update assumes two separate technological lines- each of them equipped with a system of samplers and flow meters, which will allow for more flexible operation of the system and the possibility of adapting its operation to current research needs.

4.3.4. Verification of plant species for planting

The construction company prepared tanks Z1 (aerobic wetland) and Z2 (infiltration & biodiversity wetland) according to the original design. These tanks were prepared in the form a wetland system. Such species as: *Mentha aquatic, Lemma mino, Acorus calamus, Phragmites australis, Typha latifolia L., Juncus sp.Iris pseudacorus L., Nuphar lutea, Nymphaea alba L., Filipendula ulmaria* were planted in tank Z1.

The greenery inventory carried out in July 2022 showed that most of the plants had developed properly, and additional plant species had appeared, including strictly protected species such as *Uticularia sp.* The replacement of plantings of *Mentha aquatic* and *Filipendula ulmaria* were found to be necessary, as well as the removal of invasive plants such as *Bidens frondosus*.

Because the P2 installation has not been completed and wetland Z2 has not been continuously supplied with water - some plant species have died (Figure 8). There are areas in the wetland Z2 that are devoid of vegetation and require replanting according to the Table 1. Some species are not available on the Polish market at the moment, so replacement plantings are proposed as *follow Juncus effuses, Ranunculus lingua, Variegata' (Glyceria maxima).*

TABLE1. THE RESULTS OF GREENERY INVENTORY OF

Plant	Abundance on site	Proposed action
Carex cespitosa	lot	No
Juncus articulatus L.	sparse	Replacement planting with Juncus effusus
Ranunculus repens L.	none	Replacement planting with Ranunculus lingua and Variegata (Glyceria maxima)

5. Conclusion

The experiences from the construction of the Wetland+ pilot plant in Poland showed that, despite the extensive methodological assumptions validated by the technology suppliers (PhotonWater from the Czech Republic), it is necessary to revise the design in order to adapt it to the local conditions. In particular, this concerns the materials available on the local market and the optimization of the control and monitoring system of the installation to increase its flexibility and efficiency. The experiences presented in this paper can provide guidance for future technology designers, especially in scaling up and transferring the technology to other locations.

FIGURE 7. WETLAND – Z1

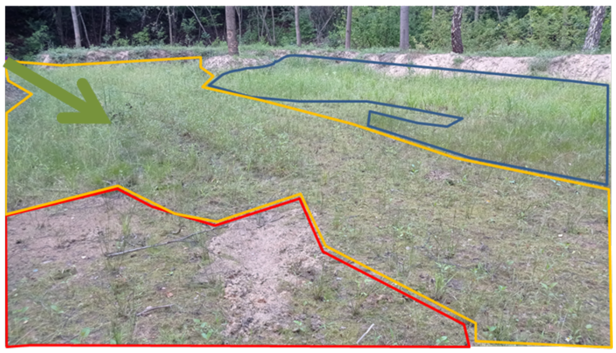

FIGURE 8. WETLAND – Z2

Legend: red area – dry places without plants, blue area – lot of Carex Sp., yellow area – mixed plants; green arrow – some species of Juncus articulatus L.

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EXPERIENCE FROM OPERATION AND TUNNING OF WETLAND+® TECHNOLOGY FOR TREATMENT OF HCH-CONTAMINATED WATER

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Summary

At the Hájek site (Czech Republic) approximately 5,000 tons of HCH residue were disposed of in a dump of overburden of a caoline pit mine in the 1970´s. Dump leachate impacted mainly by hexachlorocyclohexane (HCH) isomers and chlorobenzenes (ClB) discharged into the Ostrovsky Creek and contaminated its ecosystem. Within the LIFEPOPWAT project Wetland+® demonstration prototype for treatment of the dump leachate has been installed. The Wetland+® cascade contains 4 sequential stages: (A) aeration and sedimentation module, (B) permeable reactive modules with zerovalent iron fill, (C) biosorption module, and (D) aerobic wetland module. During the testing period of 14 months (the test is ongoing) the total concentrations of HCH isomers at the inlet to the Wetland+® varied from 52 to 265 µg/l, total concentrations of chlorobenzenes (ClB) varied from 103 to 1330 µg/l. During the first 5 months of operation the removal efficiency of HCH showed a descending trend from 97% to 54%.

The analysis of chemical data and geochemical modelling revealed that the descending efficiency is mainly result of prevailing aerobic conditions in the B modules that led to clogging of the reactive fill by ferric hydroxide and oxyhydroxide precipitates. In addition, original design of the B modules led to an improper flow pathway of contaminated water through these modules. Bypassing the A module and modification of B modules suppressed the unwanted geochemical processes and resulted in increase in the overall HCH removal efficiency of Wetland+® to 95%. Removal efficiency was not uniform for individual HCH isomers but exhibited the trend: $\alpha = \gamma = \delta > \beta = \varepsilon$. As a consequence, while δ-HCH isomer dominates in the inflow, ε-HCH prevails in the outflow from Wetland+®.

The operation of the Wetland+® led to decrease in HCH mass discharge to the Ostrovský Creek from 23 to 25 g/day prior to the Wetland+® down to 0.8 – 0.9 g/day (approximately 97% decrease).

Keywords

hexachlorocyclohexane, lindane, treatment, wetland, ZVI

Introduction

Prior to the start of the Wetland+® test operation (September 2021), the drainage systems of the Hájek dump discharged approximately 1 - 3 l/s of leachate with an average content of 100 µg/l of HCH and 600 µg/l of chlorobenzenes (ClB), which are intermediates of HCH transformation. These pollutants discharged into the Ostrovský Creek that feeds the Upper and Lower Štít fish breeding ponds. The dump leachate has high mineralisation of about 1.5 g/l and approximately neutral pH. The predominant anions are sulphate (600 mg/l) and bicarbonate (300 mg/l), the predominant cation is calcium (180 mg/l). The dump leachate has high content of dissolved iron (20-30 mg/l) and manganese (3-4 mg/l).

Wetland+® demonstration prototype

Within the LIFEPOPWAT project Wetland+® demonstration prototype for treatment of the dump leachate has been installed. The Wetland+® cascade contains 4 sequential stages: (A) aeration and sedimentation module, (B) permeable reactive modules with zerovalent iron fill, (C) biosorption module, and (D) aerobic wetland module. The scheme of the Wetland+® prototype is depicted in Figure 1, the function and description of individual modules of the prototype are given in Table 1.

TABLE 1: BASIC PARAMETERS OF WETLAND+® MODULES

Module	Fill	Function
A - aeration and sedimentation	none	Removal of iron
B - permeable reactive module	ZVI (iron chips) in four modules connected in two parallel branches	Abiotic degradation of HCH
C - biosorption	20% loamy soil 40% peat 30% crashed stones 10% wooden chips In two modules connected in parallel	Biosorption and degradation of residual HCH and intermediates of HCH degradation
D – aerobic wetland	50% loamy soil 10% compost 30% crashed stones 10% wooden chips	Mainly removal of intermediates of HCH degradation

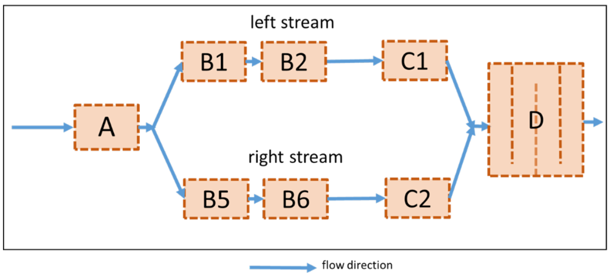

FIGURE 1. THE SCHEME OF THE WETLAND+® PROTOTYPE OPERATED PRIOR TO THE MODIFICATION

Operation and tunning of the Wetland+® demonstration prototype

The test operation of the Wetland+® prototype started in September 2021. During the first 5 months of operation the removal efficiency of HCH showed a descending trend from 97% to 54% mainly due to low and decreasing efficiency of B modules. The analysis of chemical data and geochemical modelling revealed that the lack of efficiency of B modules is mainly result of prevailing aerobic conditions in the B modules that led to clogging of the reactive fill by ferric hydroxide and oxyhydroxide precipitates. These precipitates were also present in the form of colloids in water feeding the B modules as the sedimentation of iron precipitates in the A module was not satisfactory.

In the following period starting in June 2022, corrective measures have been comparatively tested with the aim to maintain the B modules in the reducing state and to minimize production of iron precipitates. Within these measures module B5 was directly fed by the dump leachate (aeration module A was bypassed) in order to keep water in the anoxic state. Furthermore, modules B5 and B6 were equipped with a floating foil and partitions to limit diffusion of atmospheric oxygen into water and to increase contact of contaminated water with the reactive fill (iron chips), respectively.

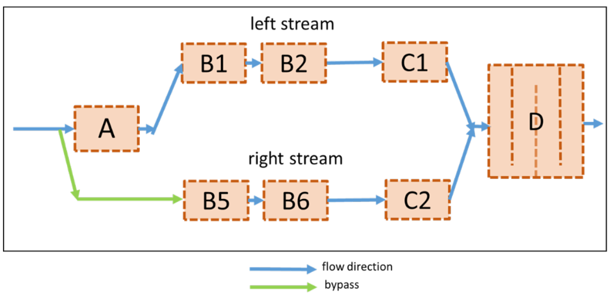

FIGURE 2. THE SCHEME OF THE WETLAND+® PROTOTYPE OPERATED AFTER THE MODIFICATION

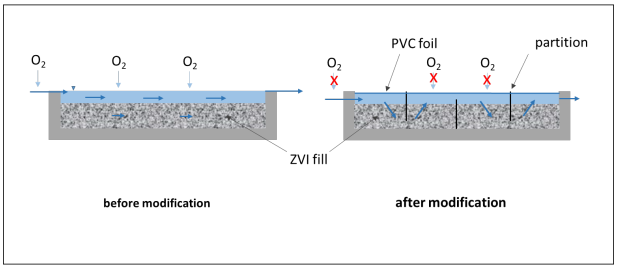

FIGURE 3. SCHEMATIC CROSS SECTION OF B MODULE BEFORE (LEFT) AND AFTER (RIGHT) THE MODIFICATION

The above-described modification led to much more reducing conditions within modules B5 and B6. Redox potential (ORP) of water leaving the modified B6 module was lower by 246 mV than ORP at the outlet of the unmodified module B2. As depicted in Figure 4, concentrations of colloid iron in both modified modules B5 and B6 were approximately 50% of colloid concentrations found in unmodified modules B1 and B2, despite of much higher concentrations of dissolved (i.e., divalent) iron.

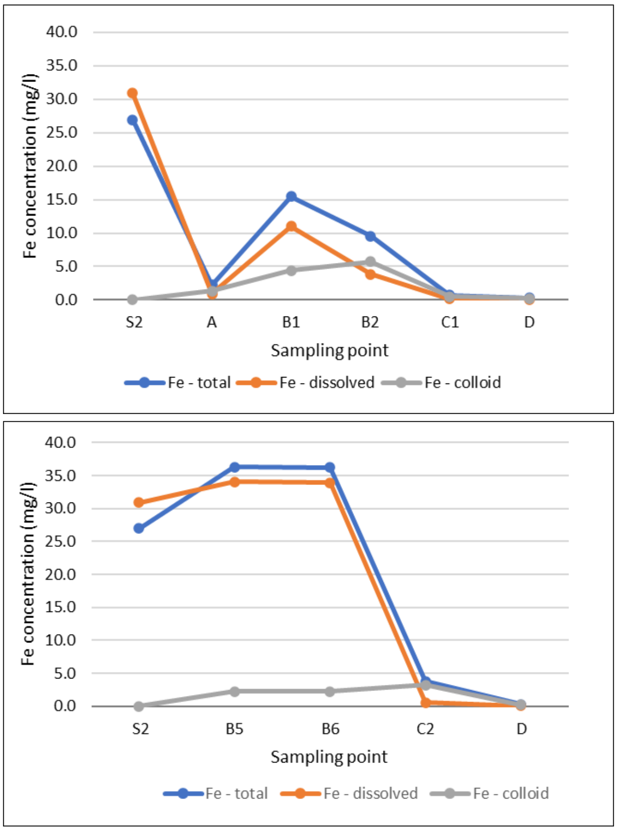

FIGURE 4. CONCENTRATION OF TOTAL, DISSOLVED AND COLLOID IRON IN UNMODIFIED (LEFT) AND MODIFIED (RIGHT) FLOW STREAMS OF THE WETLAND+® PROTOTYPE

Better conditions for chemical reduction of HCH by iron chips in modified modules B5 and B6 resulted in increased HCH removal efficiency (76% in comparison to 40% efficiency in unmodified modules B1 and B2) and contributed to the increase of the overall efficiency of the Wetland+® prototype up to 95%.

The overall removal efficiency is not the same for individual HCH isomers. As shown in Figure 5, isomers α-HCH, γ-HCH and δ-HCH have very high degradation efficiencies (97% - 99%). On the other hand, isomers β-HCH and ε-HCH have very low efficiency (43% and 59%, respectively), of which only ε-HCH has more significant concentration in the original mixture of HCH isomers. Thus, whereas δ-HCH isomer dominates at the inlet to the Wetland+® prototype, ε-HCH dominates at its outlet.

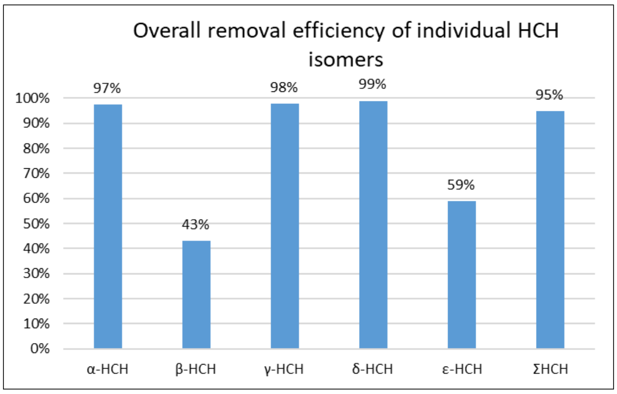

FIGURE 5. OVERALL REMOVAL EFFICIENCIES OF INDIVIDUAL HCH ISOMERS

The recipient of the dump leachate – the Ostrovský Creek is a sensitive watercourse as it feeds the Horní and Dolní Štít ponds used for carp fish breeding. The operation of the Wetland+® led to decrease in HCH mass discharge to the Ostrovský Creek from 23 to 25 g/day prior to the Wetland+® down to 0.8-0.9 g/day (approximately 97% decrease).

Summary

During the first 5 months of the operation of Wetland+® prototype the removal efficiency of HCH showed a descending trend from 97% to 54%.

The analysis of chemical data and geochemical modelling revealed that the descending efficiency is mainly result of prevailing aerobic conditions in the B modules that led to clogging of the reactive fill by ferric hydroxide and oxyhydroxide precipitates. In addition, original design of the B modules led to improper flow pathway of contaminated water through these modules.

Bypassing the A module and modification of B modules suppressed the unwanted geochemical processes and led to an increase in the overall HCH removal efficiency of Wetland+® prototype up to 95%.

Removal efficiency was not uniform for individual HCH isomers but exhibited the trend: α = γ = δ > β = ε. As a consequence, while δ-HCH isomer dominates in the inflow, ε-HCH prevails in the outflow from Wetland+®.

The operation of the Wetland+® prototype also led to a decrease in HCH mass discharge to the Ostrovský Creek from 23 to 25 g/day prior to the Wetland+® down to 0.8 – 0.9 g/day (approximately 97% decrease).

BENTHIC DIATOMS AS INDICATOR OF ENVIRONMENTAL IMPACT OF WETLAND+® TECHNOLOGY FOR TREATMENT OF HCH-CONTAMINATED WATER

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Summary

The Wetland+® demonstration prototype for the treatment of HCH-contaminated dump leachate has been installed at the Hájek site (Czech Republic). Among other indicators, benthic diatoms are used as indicators of the environmental impact of Wetland +® on the water environment. Phytobenthos samples were surveyed along the Ostrovský Creek that is the recipient of HCH-contaminated leachate as well as in the Wetland+® prototype. Already before Wetland+® was put into operation (August 2021), the results showed an increasing trend of the number of diatom species identified in profiles along the Ostrovský Creek (in direction of surface water flow: 0, 3, 30, and 35 species in profiles 1, 2, 3, and 4, respectively). It fits well with the descending trend of HCH concentration in surface water due to dilution and attenuation processes. The monitoring campaign carried out after the year of operation of the Wetland+® exhibited a markedly higher number of diatom species in Ostrovský Creek profiles 1 and 2 (3 and 14 species in profiles 1 and 2, respectively). The numbers of diatom species at sites 3 and 4 were similar before and after the start of Wetland+® (30 and 35 species before and 29 and 30 species after in profiles 3 and 4, respectively). Monitoring of the Ostrovský Creek showed an increasing trend of diatom species before and after the Wetland+® was started in operation in correlation with the decreasing trend of probably iron precipitates at the bottom of the Ostrovský Creek rather than the concentration of HCH, which did not change significantly in the Ostrovský Creek. In summary, phytobenthos acts as a suitable indicator of water quality and of the impact of the operation of Wetland+® on the water ecosystem.

Keywords

hexachlorocyclohexane, treatment, wetland, phytobenthos, diatoms, diversity

Introduction

The Wetland+® demonstration prototype for the treatment of water contaminated with dangerous HCHs was put in operation in September 2021 at the Hájek quarry dump near the village of Hroznětín in the Karlovy Vary region. The Ostrovský Creek is the recipient of HCH-contaminated leachate. The small tributary of the Ostrovský Creek was chosen as the reference locality.

The use of Wetland+ technology improves water quality and environmental conditions for organisms in the Ostrovský Creek. In addition to other monitoring, long-term monitoring of changes in the composition of phytobenthos communities is used to assess water quality after the implementation of the Wetland+ system.

Phytobenthos are microscopic and macroscopic algae that live attached to submerge substrates or plants. The main group of phytobenthos is benthic diatoms, microscopic unicellular algae (*Bacillariophyceae*), which are common in almost all types of water. They are sensitive to various environmental factors such as light, temperature, current speed, oxygen content, pH, salinity, nutrient and organic matter concentration, pollution with toxic substances, etc. Therefore, diatoms are very good bioindicators of changes in the local environment. While evaluation of water quality based on physicochemical examination detects water quality only at the time of measurement, phytobenthos analysis assesses water quality in the longer term. In addition, phytobenthos communities are affected by their competitors and predators and also by various disturbances such as larger flows, turbidity, drying, mud cover, etc. This must be taken into account when evaluating the results.

Several authors have studied how pesticides affect diatoms (Goldsborough et al. 1986, Peres et al. 1996, Berard et al. 2004, Schmitt-Jansen and Altenburger 2005, Debenest et al. 2008, Rimet and Bouchez 2010). The effect of pesticides on diatoms can be different; for example, maleic hydrazide caused an increase in deformed diatom frustules (Debenest et al. 2008) and diuron, azoxystrobin, and tebuconazole affected diatom life forms (Rimet and Bouchez 2010). Generally, pesticide exposure alters the species diversity of diatom communities, the abundance of diatoms and it can cause abnormalities in the shape of their frustules.

There is only few information in the literature about the effect of HCH on diatoms, but it is known that diatoms are able to accumulate HCH (Trautmann and Streit 1979, Łukowski and Ligowski 1987, Bystrzejewska et. al, 1993, Łukowski et al. 1997).

Diatoms have been used for many years to assess organic and nutrient pollution and diatom indices have been developed for a practical evaluation of water quality. The European Water Framework Directive (European Commission, 2000) requires evaluation of water quality in rivers in EU countries and the ecological state of rivers in the EU is also evaluated by using diatom indices alongside other bioindicators, such as macrophytes, macro-invertebrates, and fish. Diatom indices are mainly based on the sensitivity of diatoms to the trophic state (the concentrations of phosphorus and nitrogen) and the saprobity (the concentration of organic substances). In addition to these traditional indices, evaluation based on the sensitivity of diatoms to pesticide pollution is beginning to be used (Morin et al., 2009). It has been suggested that diatoms can be used as good indicators of pesticides in water (Rimett et al., 2011); thus indices using the sensitivity of diatoms to pesticides are still being developed.

Methods

Phytobenthos samples were surveyed along the Ostrovský Creek, which is the recipient of HCH-contaminated leachate in August 2021 and 2022 as well as in the Wetland+® prototype in August 2022 (Figures 1, 2). The tributary of the Ostrovský Creek was chosen as the Reference Creek. This nameless creek is not contaminated and flows out of the Ztracený pond (Figure. 2).

The sampling sites for the diatoms surveyed were the same as those for surface water sampling for chemical analysis (Figures 1, 2). Samples were taken from submerged stones and from various surfaces (submerged plants, leaves, and branches of trees and shrubs) and from the mud surface layer with fine detritus when no stones were present.

The determination of the diatom species was processed by light microscopy using standard European methods. The relative abundance of diatoms (the proportional representation of different diatom taxa within the community) was evaluated by enumeration of frustules.

For evaluation of diversity of the diatom community, the Shannon diversity index was used. This index expresses the diversity of species where it takes into account the number of species (richness) and their relative abundance (evenness). The Shannon diversity index values were counted using the OMNIDIA 6.1.2 software (France). The Shannon diversity index increases with the number of species and the evenness of their abundance. The minimum value of the Shannon diversity index is 0 when there is no diversity (only one species is presented in the community). The maximum value becomes when all species have the same number of individuals.

Results and Discussion

Phytobenthos samples were surveyed from the Ostrovský Creek in 4 profiles in August 2021 and 2022 and in 9 profiles in the Wetland+® prototype in August 2022 (Figures 1 and 2). The Reference Creek (nameless tributary of the Ostrovký Creek) was sampled only in August 2021, it was dry in 2022. Each benthic sample contained 0 to 35 species of diatoms and values of the Shannon diversity index ranged from 0 to 4.72 (Figures 1 and 2). The fewest diatom species (none) were observed in profile 1 in the Ostrovský Creek in 2021 and at the beginning of section C1 and C2 in 2022. The most species of diatoms and the highest diversity (35 and 4.72, respectively) were observed in profile 4 in Ostrovský Creek in 2021.

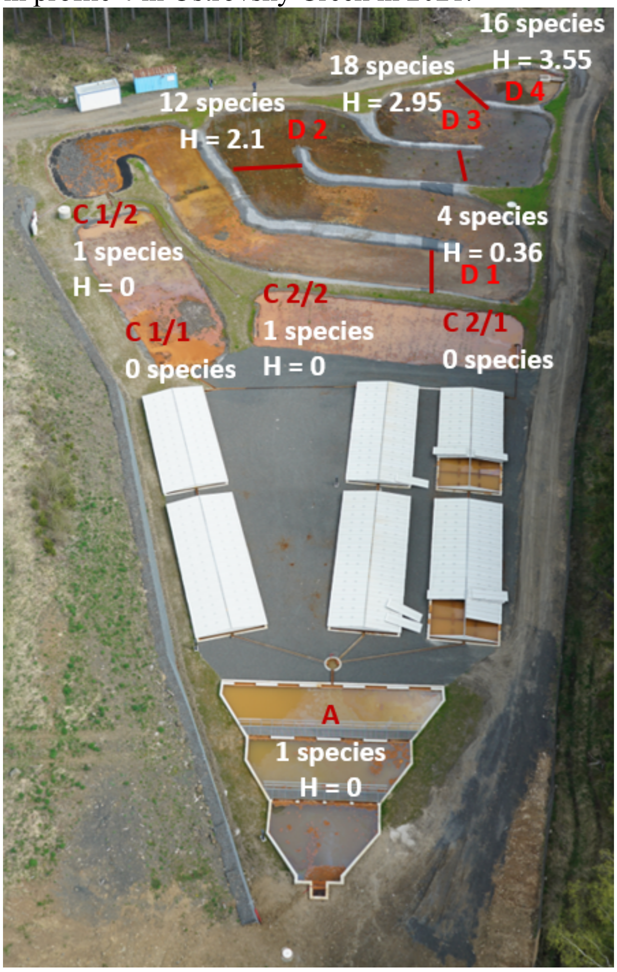

FIGURE 1. DIATOM SAMPLING SITES IN THE WETLAND+® PROTOTYPE WITH THE NUMBER OF SPECIES AND THE VALUE OF THE SHANNON DIVERSITY INDEX (H) IN SEPTEMBER 2022

The diatom monitoring performed after the year of Wetland+® operation exhibited a higher number of diatom species and their diversity in Ostrovský Creek profiles 1 and 2 (Figure 2). The numbers of diatom species and their diversity in Ostrovský Creek profiles 3 and 4 were similar before and after the start of Wetland+® (Figure 2).

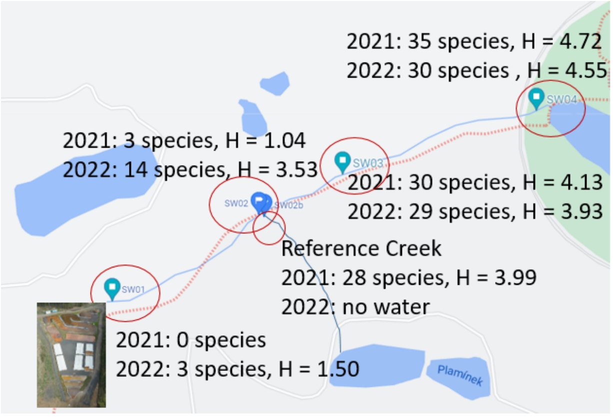

FIGURE 2. THE MAP OF MONITORING SITES IN THE OSTROVSKÝ CREEK AND THE REFERENCE CREEK WITH THE NUMBER OF DIATOM SPECIES AND THE VALUE OF THE SHANNON DIVERSITY INDEX (H) IN SEPTEMBER 2021 AND 2022

Monitoring of the Ostrovský Creek showed an increasing trend of diatom species before and after the Wetland+® was put in operation, in correlation with the decreasing trend of probably iron precipitates at the bottom of the Ostrovský Creek rather than the concentration of HCH, which did not change significantly in the Ostrovský Creek.

59 species of diatoms were observed in the four sampled profiles in the Ostrovský Creek in August 2021 and 28 species of diatoms were observed in the Reference Creek in August 2021. There were a total of 3 species in profiles 1 and 2, and 58 species of diatoms were detected in profiles 3 and 4 in September 2021.

80 species of diatoms were observed in nine sampled profiles in the Wetland+® prototype and in four sampled profiles on the Ostrovský Creek in August 2022 (Figures 3, 4, 5). 32 species of diatoms were found in the Wetland+® prototype and 64 species were observed in the Ostrovský Creek. There were a total of 15 species on profiles 1 and 2, and 53 species of diatoms were detected on profiles 3 and 4 in September 2022.

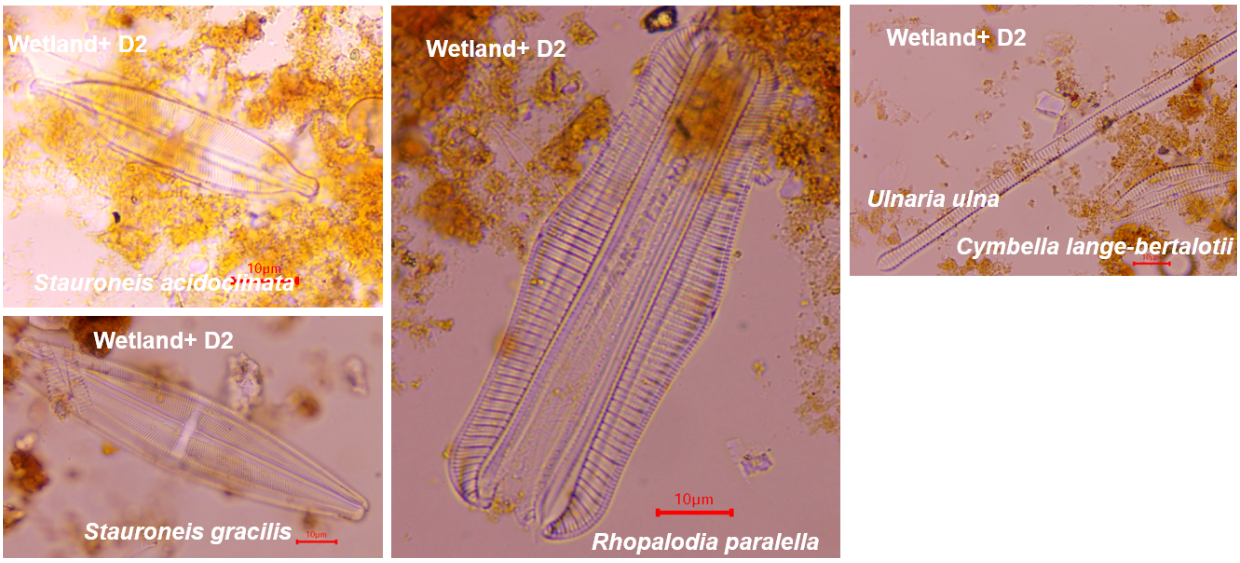

FIGURE 3. VARIOUS DIATOM SPECIES FROM THE WETLAND+ D2

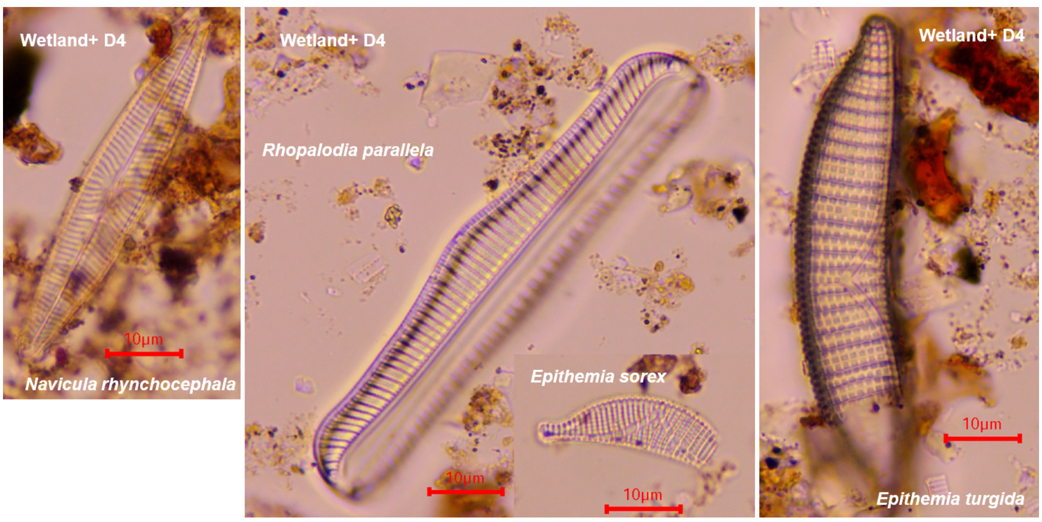

FIGURE 4. VARIOUS DIATOM SPECIES FROM THE WETLAND+ D4

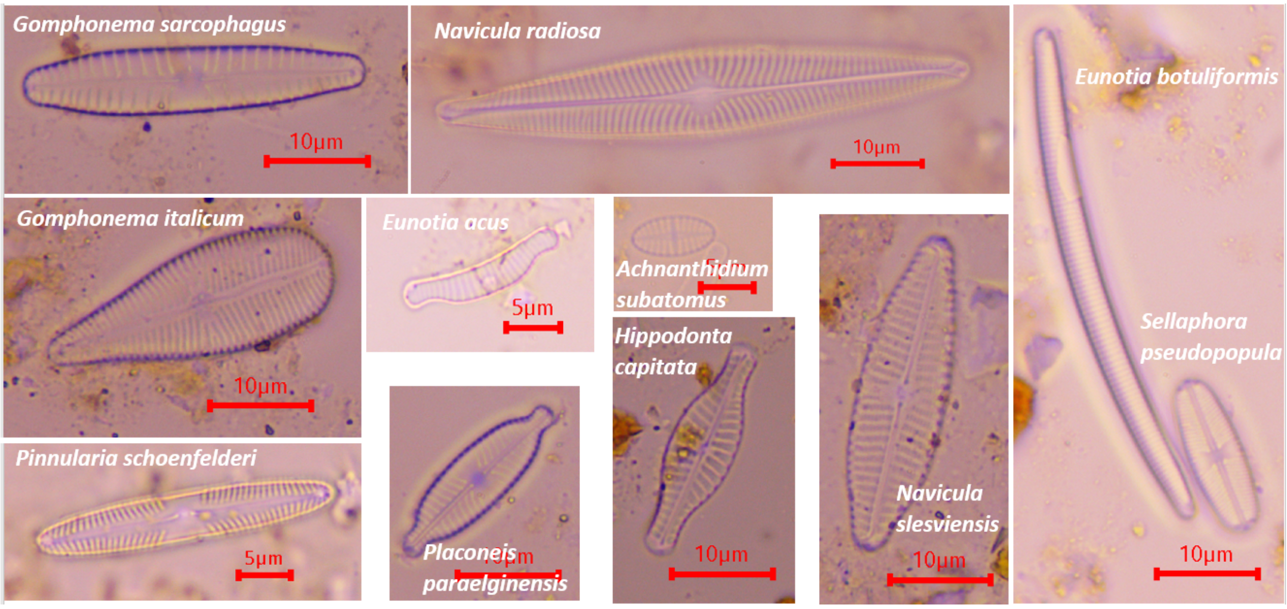

FIGURE 5. VARIOUS DIATOM SPECIES FROM THE OSTROVSKÝ CREEK

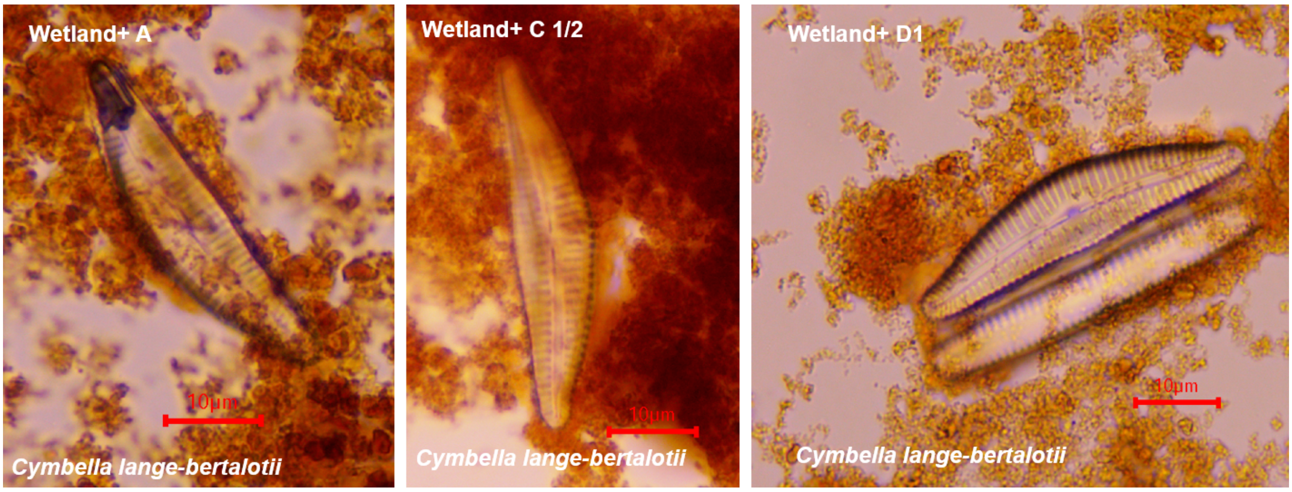

FIGURE 6. CYMBELLA LANGE-BERTALOTII FROM THE WETLAND+

Although Debenest et al. (2008) and Schmitt-Jansen and Altenburger (2005) have indicated that pesticides cause abnormalities in the diatom frustule, in our study no abnormalities in frustule shape were observed in the Wetland+ profiles.
We monitored changes in the composition of the diatom community and in diatom cell density before and after the start of Wetland+®, thus long-term monitoring in the changes of composition of phytobenthos communities seems to be a suitable tool to assess water quality after starting the Wetland+® system.

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BENEFITS OF THE PRESENCE OF PLANTS IN WETLAND+ SYSTEM, TREATING HCH POLLUTED SITES

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Summary

Hexachlorocyclohexane (HCH) is an insecticide banned around the world, but it is still a worldwide problem since there are many legacy sites that are leaching polluted waters to the environment. The most common solution to limit the leaching has been confining the site to retain the pollutant, but the solution does not treat the source and no actual treatment results from the procedure. Pilot plant studies have shown the capacity of treatment wetlands to remove HCH, and as result a LIFE EU project, LIFEPOPWAT has financed the establishment of two sites, one in the Czech Republic and one in Poland as well as the performance of parallel studies to assess the capacity of planted system to treat the pollutant. Treatment wetlands take advantage of the combination of process through the intervention of water, media, biofilm and plants to remove pollutants.

The LIFEPOPWAT project has developed a wetland treatment system (Wetland$^+$), where the combination of planted anaerobic and anaerobic environments speed up the removal of HCHs. The systems are planted with selected species of plants to improve the performance. The presence of plants in wetlands is known to contribute to treatment since they perform processes that speedup the removal process including: the transport of oxygen to the root zones, improving hydraulics in the reactors, while the roots can release a cocktail of low molecular carbon compounds (exudates) that benefit the removal of the pollutants. Additionally, the presence of plants increases the density of the biofilm community attached to roots and stems, which results in an increase biological activity and the presence of degrader microorganisms and therefore, increases the rate at which the pollutants are removed. Furthermore, plants can uptake, translocate the pollutants to the plant organs, metabolize, and store the compounds, and in case of further treatment, the management becomes easier that dealing with diffuse pollution. Another advantage of the plants in the system is the increase of biodiversity, since plants attract pollinizers and increase the presence of fauna. The presence of plants in the system helps to integrate the sites to the landscape, resulting in acceptance from the local populations and can be used as a site for research, education and leisure.

Keywords

Treatment wetlands, plant selection, pollutant translocation, metabolites, rhizosphere.

Regarding the use of treatment wetlands and the roll of plants for the removal of pesticides, few experiences are available in the literature although wetlands have been used for the purpose. When performing a search in publications on "pesticides and organochlorine and constructed wetlands", the results show total, 643 documents found, of which 232 were published by China, 103 by the USA and 76 in India. In the case of Europe, France and Germany lead the publication.

In the USA, 90% of the articles published are reviews on the effects, presence, and toxicity. Zhao et al. (2014) show results of bioremediation of water with the presence of Endosulfan in a vertical flow TW using *Phragmites australis* as vegetation at laboratory scale. The results show removals of up to 98% of the pesticide in a period of 20 days. They attributed the results to the addition of adapted bacteria that served as catalysts and biostimulation using sucrose. Further research has been performed in countries with pesticides legacy sites such as India and Brazil using wetlands but the rol of the plants tehemselves is hardly approach.

Using treatment wetland systems have lately been used more frequently to mitigate contamination by POPs, being these an ecological, sustainable and low-cost strategy (Matamoros et al., 2020, Lui et al., 2018). The removal of the pesticides using treatment wetlands has shown to be a sustainable solution due to the very nature of the system; where the combination of mechanisms take place. The mechanisms occur by the interaction of the components of the systems resulting in an effective elimination of the compounds (Jing et al., 2021).. The mechanisms may include adsorption to the filter material, absorption translocation and metabolization by the plants in different plant tissues, hydrolysis, and the intervention of bacteria consortiums attached to the plant tissues, photodegradation.

The presence of plants also stimulated a high level of biodiversity, CO_2 sequestration, and nutrient cycle (Lorenzo et al., 2019). The development of robust root system by the plants produce changes in the rhizosphere areas modifying the sediments and soil microorganisms in the wetland resulting in beneficial impact enhancing the biogeochemical cycle of pollutants. (Girones et al., 2021; Hu et al., 2020).

Research using different plant spices for the removal of pesticides have shown that the selection of the plant species does have an impact on the performance. Lyu et al 2018 tested five different plant species *Typha latifolia, Phragmites australis, Iris pseudacorus, Juncus effusus* and *Berula erecta*, showing phytoaccumulation concentration in the aboveground tissue of the different plants ranging from 0.7 to 0.8 mg kg^{-1} DW equivalent to around. 3.6% to 12.1% of the pesticide removal at the end of the experiment.

FIGURE 1. TESTING HCH PLANT REMOVAL CAPACITY

Within the LIFEPOPWAT project the plant selection has been elected done after evaluating the potential of the removal by testing different plant species in growth chambers and planting different species and ran under controlled growing conditions that included temperature, light exposure, and humidity. The plants were acclimated before being exposed to different concentrations of HCH. The plants were tested for the accumulation of HCH and metabolites in the areal organs and for physiological parameters using Infrared Gas Analysis techniques to evaluate not only the capacity for the removal of HCH but also the plants health (Figure 1). The plants tested included *Alnus glutinosa, Phragmites australis, Juncus lacustris and Thypa latifolia*, plants commonly used in treatment wetlands.

The plant selected for the actual site included the ame species and where planted during the construction of the Hajek site. The planted species included the ones tested as well as other local plants to integrate the site to the landscape. Besides removal capacity and performance by the plants, beautification and integration are of utmost importance and must be considered when establishing remediation wetland systems. Figure 2 presents the site once the plants where established.

After establishment and a growing season the plants will propagate and cover the entire wetland, resulting in improving the removal as higher plant density is achieved.

Conclusions

Plants in treatment are a main component of the system and enhances the removal performance of HCH compounds through their functions that include: Growth and biomass production, Photosynthesis, Nutrient uptake, Water uptake, Oxygen transport, Metabolism, CO2 sequestration, food chain support and integration to the landscape. As the LIFEPOPWAT project develops, the plants are being tested to determine the effect of the plants in the removal of the HCH pollutants.

FIGURE 2. SYSTEM AT HAJEK ONCE PLANTED

GROUNDWATER HCH INDICATION VIA PHYTOSCREENING OF TREES

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Summary

The topic of this paper is the detection of HCH contamination in groundwater using tree biomass. The term phytoscreening can be used for pharmaceutical search for phytochemicals with potential medical use. However, neither medical phytoscreening nor phytoremediation will be covered. Briefly, our contribution focuses on an indirect and non-invasive alternative to reach the groundwater level, which is the use of trees - HCH phytoscreening (tree coring). Because of evapotranspiration from the leaf area, trees massively transport groundwater and deep soil water up into their above-ground biomass, which can, therefore, be used as a pollutant groundwater presence indicator.

In this sense, the term "phytoscreening" was established by the work of Sorek in 2008 [1] to indicate VOC contamination. Since then, more than 30 scientific papers have appeared describing the sampling methodology, data interpretation, and specific outcomes from tree studies at contaminated sites. Looking closely, we see that the authors have studied the patterns of VOC uptake by trees, the distribution of VOCs in wood biomass and its age/height/seasonal dependence, the applicability of phytoscreening to delineate the contamination plume, and the involvement of green analytical techniques (e.g., SPME).

In the case of HCH, the scientific literature and groundwater contamination investigations using phytoscreening are very limited. Some historical manuals even listed HCH as a pollutant unsuitable for phytoscreening. Despite that, we claim phytoscreening of trees for the purpose of indicating HCH in groundwater is a very viable approach. Data from the growth chamber and from 2 HCH contaminated sites in the Czech Republic and Poland confirm that (i) trees do uptake and phytoaccumulate HCH in their above-ground biomass, (ii) HCH phytoaccumulation is isomer-specific, (iii) HCH phytoaccumulation is species-, age-, seasonal-, direction-, health status-, habitat-, and height-dependent, (iv) different genotypes of the same species provide different phytoaccumulation rates, (v) SPME is a useful green technique for HCH analysis in woody biomass and (vi) birch sap monitoring has only limited applicability. All of these aspects of HCH phytoscreening will be demonstrated using data from HCH contaminated sites. Nondestructive analysis of tree biomass can be used to indicate groundwater pollution and help delineate HCH contamination plumes.

Acknowledgement

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HCH phytoaccumulation by trees

A very limited number of studies deal with this topic. So far, tree investigation studies have been published from Bitterfeld-Wolfen site [2] and from Sacco river valley [3, 4]. Samples of *Prunus* sp., *Robinia* sp., and *Crataegus* sp, were analyzed at the German site, whereas *Juglans regia* and *Poplar* sp. were studied in Italy. However, none of the studies targeted HCH phytoscreening.

TUL started investigating tree samples from the Hájek site (Czech republic) in 2016 and from the Jaworzno site (Poland) in 2018. Basic tree models were phreatophytic *Alnus glutinosa* (alder) and a specie with a more ubiquitous habitat - *Betula pendula* (birch). But HCH was found in all sampled species (*Quercus* sp., *Pinus* sp., *Poplar* sp., *Salix* sp…).

Alder biomass samples from Hájek mine heap with buried HCH waste were analyzed with freshly implemented analytical protocol on classical solvent extraction bases and matrix-matched calibration approach. Results proved HCH absence in control (reference) alder individuals growing at a 2 km distance from the dump, whereas the first units of $\mu g.g^{-1}$dw ΣHCH (α+β+γ+δ+ε) were present in biomass from the heap. Maximum concentrations exceeded 10 $\mu g.g^{-1}$dw ΣHCH in trees growing spontaneously from the drain water spring swamp. As for HCH transformation products, only traces of di- and tri-chlorobenzenes were found rarely, indicating a different uptake mechanism or a quick metabolism/excretion of chlorobenzenes. Figure 5 shows options for tree sampling.

Findings gained at Hájek were later extended by sampling campaigns in Jaworzno, which is, due to the Wawolnica river floodplain and overall aquifer geometry, an excellent phytoscreening model site. Maximum ΣHCH concentration exceeded 300 $\mu g.g^{-1}$dw in Jaworzno. It was confirmed by bark analysis (< LOD, 0.01 $\mu g.g^{-1}$dw) that root transport takes place.

Isomer-specific phytoaccumulation of HCH

At both pilots, δ-HCH dominates the groundwater and drain water. However, δ-HCH takes only a small fraction (less than 5%) of ΣHCH in tree biomass. There were some exclusions in the source zone of Jaworzno: a few birches contained δ-HCH up to 20% of the HCH sum. The majority of bioaccumulated HCH had the β-isomer. Together with ε isomer, they usually reach about 90% of ΣHCH in tree biomass.

Species-, age-, season-, direction-, health status-, habitat-, and height-dependence

Spontaneously vegetating trees, as we usually find them on HCH dumps, are of various species. Of course, there are interspecies differences in HCH uptake, with root system shape and root membrane selective permeability being responsible for the uptake differences. Almost one order of magnitude lower HCH concentration was found in *Pinus silvatica* compared to *Betula pendula* trees growing together in Jaworzno. Pines are well known for their massive taproot, so rather higher HCH uptake by birch root membrane makes the difference in this case. Anyway, interspecies differences are very important and any areal study of HCH site should therefore deal with one ubiquitously growing specie, preferably of a broadleaf tree.

Age difference of HCH uptake is very hard to study because of limitations in available tree individuals for experimental work and more or less similar age of trees in forests at dump sites. The older the tree, the higher the evotranspirative suction of HCH-contaminated groundwater in vegetation season. At higher age, tree heartwood loses conductivity and decays by the activity of parasitic fungi (see Figure 1). Roten heartwood samples should be avoided in phytoscreening campaigns. Azimuthal phytoscreening (direction of tree coring) was claimed important by Limmer et al [5]. At studied sites, however, the tree direction difference of ΣHCH did not match any known groundwater gradient. Seasonality effects were studied in a group of 20 years old alders at Hájek site. In accordance with the Guideline of project Timbre [6], seasonality was found to influence ΣHCH concentrations, with spring maximums at quadruple values of the vegetative winter rest (1.2, resp. 4.7 $\mu g.g^{-1}dw$. Figure 2).

Intuitively, ΣHCH concentration in tree biomass decreases with tree height (Figure 3). This was repeatedly proven by the whole tree harvest and its trunk profiling. It is therefore important to keep the established tree coring height of 130 cm above the ground.

Tree habitat parameters, such as tree position on the forest edge or in the forest inside, directly influence evapotranspiration intensity and tree groundwater suction. Actually, even at wet weather trees prefer water uptake from shallow soil layers so that groundwater suction differs from dry weather.

All the above-mentioned parameters disturb the match between groundwater- and tree biomass HCH concentrations.

Genotype-dependent HCH uptake of alder seedlings

In our current study[7], we compared HCH uptake by germinating seeds and young seedlings of 6 *Alnus glutinosa* genotypes, that were bred for the resistance against the pathogen *Phytophthora alni* (a parasitic fungus). Interestingly, the results show that the seedling presence enhanced the soil δ-HCH loss by 21 – 36%. Seedlings probably stimulated microbial activity by rhizospheric exudates. Individual genotypes differed seriously in stress adaptation (phytohormone levels) and HCH uptake. Of the 6 genotypes, 2 were selected as most suitable for HCH uptake and tolerance.

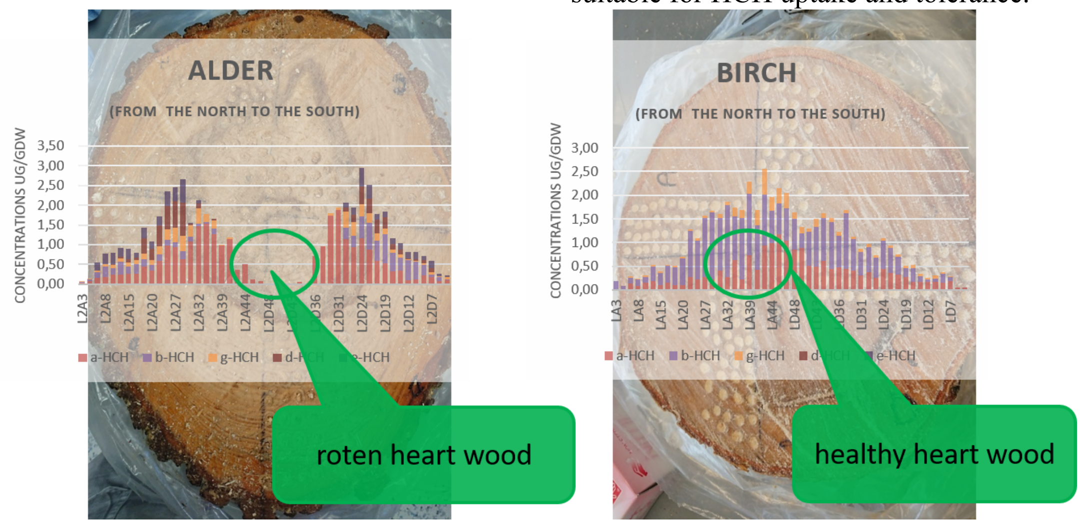

FIGURE 1. HCH CONCENTRATION PROFILES THROUGH TREE TRUNKS WITH THE DIFFERENT HEALTH STATUSES OF THE HEARTWOOD

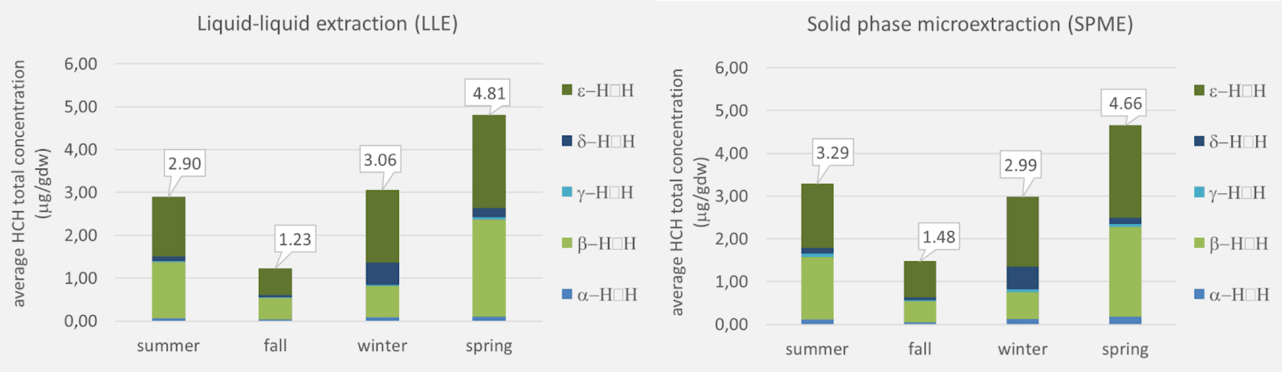

FIGURE 2. HCH SEASONALITY – MEAN ΣHCH CONCENTRATION (LEFT – SOLVENT EXTRACTION, RIGHT – SOLID PHASE MICROEXTRACTION)

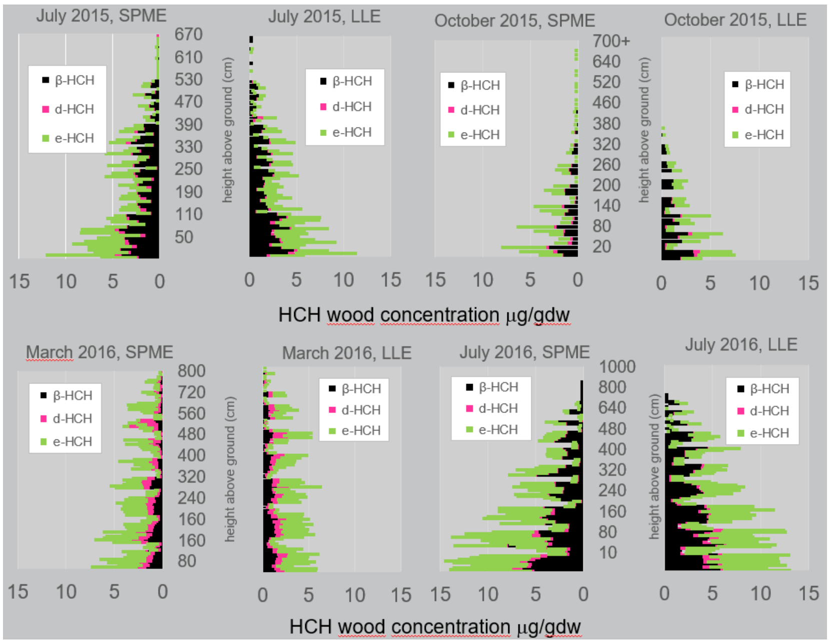

FIGURE 3. ΣHCH CONCENTRATION – HEIGHT DEPENDENCE, ALDER, HÁJEK, SEASONALITY, AND PARALLEL EXTRACTION

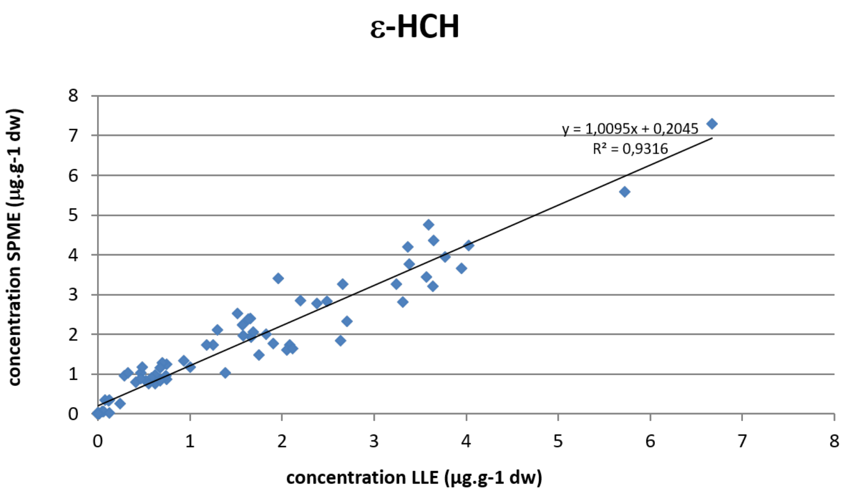

FIGURE 4. HCH CONCENTRATIONS IN ALDER DETERMINED BY SPME AND SOLVENT EXTRACTION (LLE) IN PARALLEL

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SPME for HCH phytoscreening

At early stages of HCH phytoscreening research, TUL developed a green, solvent-free analytical protocol of plant biomass analysis. In many parallel experiments, SPME was confirmed a reliable technique, analytically comparable to classical solvent extraction. As an example of results obtained in parallel by both techniques, data from Hájek alder are shown in Figures 3 and 4.

Birch sap monitoring

In the early spring, birches are oozing sap from their buds for a short period of few weeks. If the birch trunk is drilled inside within this period, birch sap can be collected and used for pollutant analysis [8]. The sap is an alternative matrix to tree biomass core. Birch sap sampling was performed at Hájek site in 2021 from 16 trees growing on the landfill, along the local stream, and some background trees. The only HCH presence in sap samples was found in 2 birches growing close to the contaminated drain water spring. ΣHCH in sap was very small, even unquantifiable (between LOD and LOQ value).

ΣHCH in solid biomass of these trees reached 0.5 $\mu g.g^{-1}$dw. This means HCH are strongly adsorbed from the sap to the wood. From the sensitivity point of view, solid birch biomass is much more suitable matrix than birch sap in case of HCH phytoscreening.

Conclusions

HCH tree phytoscreening can be, under suitable circumstances, a useful tool to indicate groundwater HCH contamination. There are many reasons, why it is only indicative, and its suggestions should be further confirmed by robust hydrogeological survey data. Lab-scale experiments tell convincingly that HCH are readily uptaken be trees. However, little is known about the fate of HCH bioaccumulated in tree biomass after the groundwater HCH plume disappears and HCH uptake stops. This lingering trace can theoretically provide false positive indication of groundwater HCH contamination (historical instead of current contamination).

Biomass sampling with an increment borer

Biomass sampling with a cordless drill

Birch sap sampling

Germinating alder seeds exposed to δ-HCH

Young alder seedlings exposed to δ-HCH

2-years old alder seedlings exposed to HCH

FIGURE 5. FIELD SAMPLING AND LABORATORY EXPERIMENTS RELATED TO HCH PHYTOSCREENING

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SURVEY ON SOCIO-ECONOMIC IMPACT FOR WETLAND+®

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Summary

Sustainable remediation is the practice of demonstrating, that the benefit of undertaking remediation is greater than its impact and that the optimum remediation solution is selected through the use of a balanced decision-making process. Assessing sustainable remediation is site and project specific, and is strongly multifactorial across a wide range of categories, which may or may not be readily quantifiable. The applied socio-economic survey framework is based on the 2020 SuRF-UK guidance [1-3]. The 15 broad categories of indicators, which revolves around three main elements of sustainability (environment, society, and economy) were established. The approach develops the guidelines set out in the 1987 Brundtland Report (UN 1987) and subsequent UN Sustainable Development Goals or SDGs (UN 2015). The chosen framework considered both input and output effects within the sustainability assessment. According to the methodology used, the socio-economic analysis was divided into 15 broad categories. In the initial assessment, out of a total of 73 criteria, 15 from the Environment category, 19 from Economic and 11 from Social were identified as relevant.

Keywords

Wetland+, HCH remediation, socio-economic, survey, decision-making process, SURF-UK

Introduction

LIFEPOPWAT, the EU LIFE project (agreement number LIFE18 ENV/CZ/000374), is focusing on the treatment of HCH-contaminated water by innovative technology based on constructed wetlands. The HCH-contaminated sites in most cases need additional drainage to collect HCH leakages and subsequent treatment. In the LIFEPOPWAT project, the drainage water is treated by an integrated pretreatment and nature-based treatment system (Wetland+®), which was designed, tested, and installed on a full scale at Hajek (the Czech Republic). Besides technical and technology assessment of this technology with classical treatment by a conventional wastewater treatment plant (WWTP), in this study, the socio-economic impact of the Wetland+® is compared to WWTP and the no-intervention scenario (as a baseline).

The sustainability assessment is based on the applied socio-economic survey framework according to UK guidance [2,3] published by the *UK Sustainable Remediation* Forum (SURF-UK). The approach develops the guidelines set out in the 1987 Brundtland Report (UN 1987) and subsequent UN Sustainable Development Goals or SDGs (UN 2015), and meets the international standard on Sustainable Remediation according to [4]. The basis of the SuRF-UK is an assessment of indicators across 15 broad categories around three basic elements of sustainability (environment, society, and economy).

Site and the water treatment Methods

The Hajek site

The site is located in Western Bohemia (the Czech Republic) near the Karlovy Vary spa. In the 1960s (and until 1971), uranium was exploited in the neighborhood Hajek quarry and for the slag, the slag heap Hajek slag heap was established. In parallel with uranium, kaolin, basalt, and later bentonite were also mined here. Between 1966 and 1968, the state authorities decided to dispose of the ballast HCH isomers and chlorobenzene (CB) from the chemical production of lindane in Spolana Neratovice (CZ) into here. These wastes were placed on various heap parts in metal drums, paper packaging, or bulk in the total estimated quantity of 3,000-5,000 tonnes.

Since January 1989, the concentrations of HCH isomers and CB have been monitored and documented at the outlet of the drainage system. Since about beginning of the century, the average content of HCH and ClB is about 100 µg/l and 600 µg/l, respectively, and the mass flux of approximately 25 g HCH and 150 g ClB per day. The contaminated drainage water flowed via the Ostrovský brook to the Hájek preserve and the adjacent breeding ponds Horni Stit and Dolni Stit. A sampling of boreholes drilled in 1994 showed that groundwater contamination with organic substances had already exceeded the contour of the spoil heap and was spreading mainly toward the flooded Hajek quarry.

Description of the Wetland+® system

The Wetland+® technology for the treatment of water containing HCH substances, developed by the Technical University of Liberec and AQUATEST a.s., is based on the use of oxidation-reduction and biosorption processes. The technology is composed of *a sedimentation tank, a permeable reactive barrier* filled with Fe chips*, a bioreduction and biosorption unit*, and *an aerobic wetland.* After one year of the pilot operation (the system is tuned for better efficiency) the HCH and CB removal efficiency exceeded 97% and 99%, respectively. The project also assumes regular maintenance of the system, including the eventual replacement of the iron chips in the permeable reactive barrier; however, the whole technology is without further input and no waste is generated during normal operation.

An alternative method in waste removal

As can be seen from the above brief overview of the situation at the Hájek site, it is not possible to locate the contaminated areas within the Hájek quarry tailings. Detailed exploratory work would be required to locate the contaminated area precisely and even then the chances for the removal of all sources of contamination are very small. The complete disposal of contaminants in the tailings body would involve the processing of at least several tens of thousands of tonnes of soil. Even then the sources of HCH and CB would not be eliminated, because during the last decades, the contaminated water was spread in the bigger volume of the repository. Moreover, once the detected contaminated sediments are excavated, they must either be redeposited in another secure landfill or cleaned up. Thermal desorption, catalytic oxidation or soil washing can be considered as possible methods for cleaning HCH-contaminated sediments. At the cost of current soil decontamination technologies, which are in the order of several hundred € per tonne of soil, the cost of soil decontamination can be estimated at several tens of millions of €. The estimated costs relate only to the removal of contamination in the soils and do not include a full remediation of the site, which would include the precise delineation of the contaminated areas, their exposure and the subsequent reshaping of the tailings. This would increase the cost of a full remediation by an order of magnitude. Therefore, the removal of the contaminants was not considered as an alternative for site remediation and was not considered in the socio-economic survey.

An alternative method in WWTP

Another option is to build a wastewater treatment plant directly on the Hájek site for long-term treatment of drainage water. As these are persistent organic contaminants, methods for their disposal are limited. The method considered here is based on pH adjustment separation and sorption of dissolved contaminants on activated carbon. The disadvantage of this method is the high consumption of this sorbent, which has to be subsequently deposited as a contaminant. The capacity of such a wastewater treatment plant would be at the level of the long-term average outflow of this water, which is about 3 l/s, i.e. 260 m^3/day. With an expected residence time of about 1 day, this is a relatively large technology. The initial estimate for the construction of such a plant is about € 500 thousand. However, this cost is not significant in terms of overall costs, as its long-term operation would be considerably more financially demanding. Only the chemical, electricity and waste transport can be estimated at about 1.7 €/m^3 of treated water, which makes €160 thousand/a. Additional costs are the operational costs and maintenance at about €100 thousand/a.

Alternative no-intervention scenario

The third scenario considered for the socio-economic survey is the no-intervention scenario. This scenario is not acceptable to the owner of the site (DIAMO s.p.) as well as to the supervising state authorities. Its inclusion in the scenarios is due to the assessment of the two above-mentioned scenarios (Wetland+® and WWTP) against the status quo (no-intervention scenario). It can be expected that in an assessment process where the different assessment parameters considered have equal weight, this scenario may perform better than the active treatments in some of the assessment criteria (e.g. waste production and disposal). However, this has no bearing on the unacceptability of this solution for a real application on the site.

The methodology for assessment of socio-economic impacts

SuRF-UK defines sustainable remediation as the practice of demonstrating, in terms of environmental, economic and social indicators, that the benefit of undertaking remediation is greater than its impact and that the optimum remediation solution is selected through the use of a balanced decision-making process, [2]. Detailed procedural guidance for the assessment process is based on 2020 SuRF-UK guidance [2,3].

The SuRF-UK guidance provides a process for determining the scope of sustainability for the purposes of land contamination management, and a checklist of possible sustainable remediation indicators/criteria, shown in Table 1. This guidance follows the structure and logic of sustainable development as set out in the 1987 Brundtland Report (UN 1987) and subsequent UN Sustainable Development Goals or SDGs (UN 2015).

Assessing sustainable remediation is site and project specific, and is strongly multifactorial across a wide range of these categories, which may or may not be readily quantifiable. Some outcomes are technique related but also strongly related to the site or specific factors, for example carbon sequestration vs carbon emissions (e.g. as a part of ENV1:Emissions to air). Hence single metrics such as a carbon footprint are inevitably highly reductive and do not represent "sustainability" in any kind of overall way (e.g. [5]).

TABLE 1. SOCIO-ECONOMIC CATEGORIES INCLUDED IN THE EVALUATION PROCESS

Environmental	Social	Economic
ENV1: Emissions to air	SOC1: Human health and safety	ECON1: Direct economic costs and benefits
ENV2: Soil and ground conditions	SOC2: Ethics and equity	ECON2: Indirect economic costs and benefits
ENV3: Groundwater and surface water	SOC3: Neighbourhood s and locality	ECON3: Employment and employment capital
ENV4: Ecology	SOC4: Communities and community involvement	ECON4: Induced economic costs and benefits
ENV5: Natural resources and waste	SOC5: Uncertainty and evidence	ECON5: Project lifespan and flexibility

It is for this reason that SuRF-UK recommends taking a tiered approach to sustainability assessment for remediation, beginning with a qualitative stage that enables the most wide-ranging scope of sustainability to be considered with a more readily manageable degree of effort. In this tiered approach, detailed assessments are only needed where a simpler approach has not been able to yield clear decision support.

General process of assessment

The approach taken is incremental with three steps. The first step is an initial sustainability assessment being developed by a small core tea (for Hajek site the project principal investigator, an SEI expert, and an expert on water treatment technology). In the second one the assessment is done by the project of all beneficiaries. The final assessment is by a broad range of external stakeholders (planned later). The methodology is based on the comparison of the selected scenarios on individual criteria. The scenario that best meets a given criterion gets a score of 1-best and the others 2, 3,... depending on the number of scenarios. In the case the scenarios are equal on the criterion, both scenarios receive the same grade and the other two grades lower (e.g. 1,1,3). The results of each criterion are averaged for each socio-economic category and the averaged rankings is shown in a radar plot (see Figure 1). The smaller the area in the radar plot the higher (i.e. better) the overall ranking. One radar plot is generated for each of the overall categories - for environmental, social and economic elements of sustainability.

Results

According to the methodology used, a process for determining the socio-economic impact is based on a survey, where the questions cover 15 broad socio-economic categories (5 for each part of the survey). In **the initial assessment** provided by the core of the LIFEPOPWAT team, out of a total of 73 criteria, 45 criteria were identified as relevant (15 from the Environment, 19 from the Economic, and 11 from the Social category). The scenarios were compared comprehensively - in terms of the construction phase, the operation phase, and the dismantling phase.

In the second step, the previous results were consulted by the whole LIFEPOPWAT project team. All seven stakeholders had at least one representative in the discussion. The collective consisted of experts in the broader field of water treatment and remediation, as well as economists. The results were not significantly changed after the second step.

The ranked answers were summarized in the categories and plotted as radar graphs in Fig. 1. In this initial qualitative assessment, Wetland+® outranked the use of conventional WWTP for most of the 15 general categories defined in Table 1, and where it did not outrank, the ranking was very similar. The results for the categories are described below.

Environmental criteria

If we look in detail at the overall Environmental category, then in all criteria Wetland+® is the highest-rated scenario. No-intervention is the worst with the exception of natural resources and waste where WWTP would generate waste that would have to be disposed of.

Emissions to air: The construction of Wetland+® generates greenhouse gases, but over the lifetime is sequestered so the overall benefit is positive. WWTP will use fossil carbon in energy and resources. Fugitive emissions of volatile organic compounds (HCH isomers) will be greatest in the no intervention scenario.

Soil and ground conditions: The Wetland+® improves topsoil biology.

Groundwater & Surface Water: Both the Wetland+® and WWTP improve water quality in the stream and downstream water bodies. Additionally, Wetland+® is an ecological process and its outflow water will contain a range of aquatic microfauna and plankton that the WWTP would not.

Ecology: Wetland+® creates significant habitat and biodiversity benefit over other options. No change perpetuates ecosystem degradation in the surrounding environment and ongoing accumulation of pesticides along food chains, potentially affecting for example, birdlife. Additionally, the operation of cWWTP crates local disruption to ecology (for example, light and noise).

Natural resources and waste: Both treatments will help reduce orange (iron) staining in the stream bed. WWTP consumes some water (e.g. for washing filters) and also generates waste. Wetland+® includes the use of locally generated recyclates/renewables (such as woodchip).

Economic criteria

Direct economic costs and benefits: Wetland+® is cheaper over the long term than WWTP, although "no change" obviously has no direct costs. WWTP will have a higher degree of control, daily operations, and monitoring than Wetland+®. The benefit is comparable for both installations, significantly higher than no-intervention.

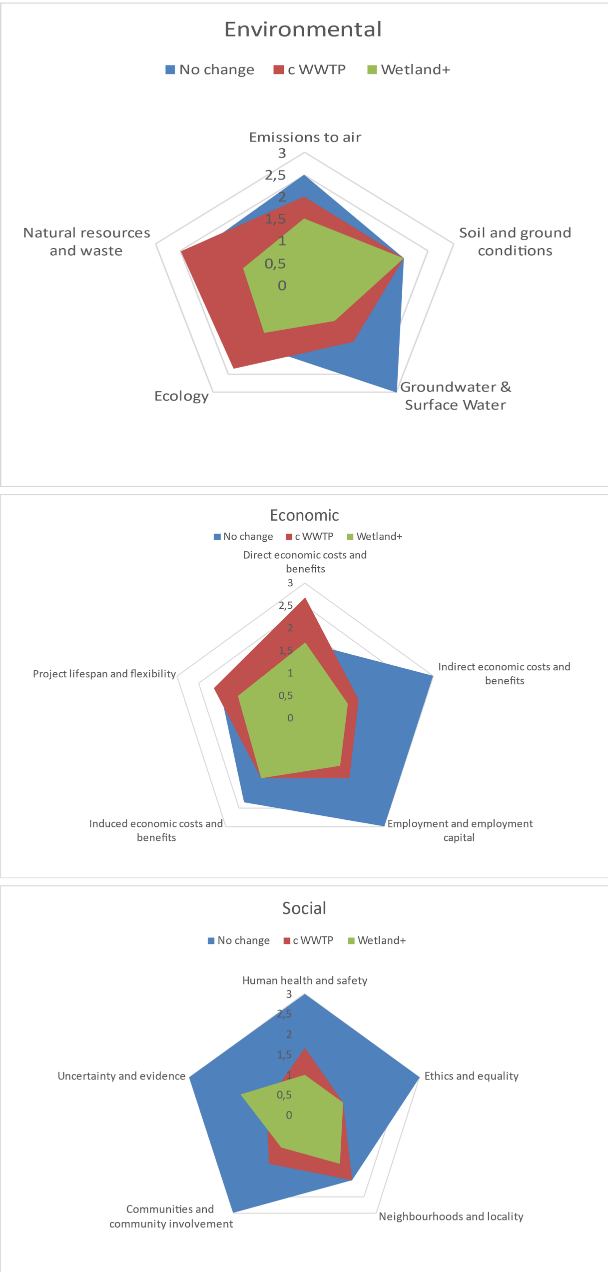

FIGURE 1. SUSTAINABILITY ASSESSMENT OUTCOMES FROM HAJEK (ENVIRONMENTAL, ECONOMIC, SOCIAL)

Indirect economic costs and benefits: Wetland+® has intrinsic value in making the area both safer and look safer compared with WWTP. Wetland+® may be closer to the sustainability aspirations of Diamo to show that the organizations involved share a goal of low input and sustainable solutions. Whereas no change is damaging to its reputation. Treatment also protects fisheries and hunting.

Employment and employment capital: Conventional WWTP likely has the greatest local job creation potential (e.g. for a technician). Wetland+® creates some maintenance needs. Wetland+® offers the widest and most attractive range of opportunities for school visits, and pathways for education and offers a platform for student projects.

Induced economic costs and benefits: Improvement of the stream, either by Wetland+® or WWTP, improves the attractiveness of the area downstream of treatment for a wider range of land use opportunities. Wetland+® offers the provider the chance to improve innovation and skills and provides a platform for replication elsewhere.

Project lifespan and flexibility: WWTP has the greatest process control, and likely is best understood by regulators for permitting purposes. However, where a problem does occur, Wetland+® has the greatest internal resilience and is likely to be most resilient to any constraints on the operator to pay. Wetland+® also is likely to have the smallest intuitional burden (e.g., maintenance of process control records). No action postpones the solution to the environmental problem to the future likely increasing both the cost of the solution and the economic impact of the damage being caused.

Societal criteria

Human health and safety: Both treatments meet the necessary surface water treatment criteria reducing risks. Risks to site workers are likely to be greatest for the WWTP as it is an operating process plant. Both treatments improve the amenity value of the local area, making it more attractive for physical recreation. Wetland+® acts its own right as a destination to encourage people outside.

Ethics and quality: No change means that the polluter does not pay and postpones the problem to future generations.

Neighbourhoods and locality: Given the remoteness of the location there are no likely neighbourhood concerns over effects from dust, light, noise, odour, vibrations and traffic. A remote WWTP is potentially an attractive destination for thieves and vandals in a way that Wetland+® is not. Both treatments provide local improvement for hunting, fishing, wildlife based recreation.

Communities and community involvement: Both treatments improve the amenity of the river downstream benefiting local communities. Treatment of the problem is in line with local planning and environmental policies, with

Wetland+® offering the greatest linkage to sustainable development policy goals.

Uncertainty and evidence: The robustness and rigour of the information for option design provided is greatest for WWTP because it is so well established, and it has more straight forward process control at present, and its outcomes more readily validated. No change is obviously a completely uncontrolled process.

The general category rankings are averages of individual rankings made for each of the criteria shown in Table 1. Each individual ranking is supported by a specific rationale. The next stage of assessment will bring in wider stakeholders within the LifePopWat project to scrutinize and review these individual rankings and their rationales.

Conclusions

This paper provides the assessment framework for the sustainability evaluation of Wetland+® implementations in Hajek. The proposed framework builds upon the 15 broad categories of indicators addressed by the SuRF-UK Indicator checklist, which revolves around three main elements of sustainable development: environment, society and economy. The approach taken in this project develops the guidelines set out in the 1987 Brundtland Report (UN 1987) and subsequent UN Sustainable Development Goals or SDGs (UN 2015). The chosen framework considered both input and output effects within the sustainability assessment. The initial assessments compared Wetland+® with the conventional WWTP and no-intervention scenario. The assessment was provided by a small group of the project team. Wetland+® outranked the use of WWTP, and both performed significantly better than the no-intervention scenario. This assessment was in the second step evaluated by the remaining team members from all stakeholders. In the third step, the external stakeholders will be involved (later this year).

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PROTOCOLS OFFER TO THE CLIENTS FOR WETLAND+® REPLICATION

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Abstract

For partners and clients interested in Wetland+® technology, iteratives service packages are proposed in order to go step-by-step from laboratory test up to scale 1. Also, GO/NO GO are present along this iterative reflexion to check the relevance of Wetland+ techniques vs other techniques (like coagulation/precipitation, activated carbon) in order to propose the best solutions in terms of costs but also in terms of sustainability for the treatment of an effluent (resurgence, GW, surface water) impacted in HCH or pesticides.
The cost for CAPEX roughly depends on the compartments needed (anaerobic with permeable reactive barrier with ZVI and/or biosorption wetland/aerobic wetland) and the total surface necessary (directly dependent on flow rate and pollutant concentrations). OPEX is relative to operational monitoring that should be low as Wetland+ is a robust and rustic passive treatment.