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Research article

HCH phytoremediation potential of native plant species from a contaminated urban site in Turda, Romania



Balázs E. Helga^{a,b}, Christoph A.O. Schmid^a, Ioana Feher^c, Dorina Podar^d, Paul-Marian Szatmari^{b,e}, Olivian Marincaş^c, Zoltan R. Balázs^d, Peter Schröder^{a,*}

^a Helmholtz Zentrum München, Comparative Microbiome Analysis, Ingolstädter Landstr. 1, 85764, München, Germany

^b Botanical Garden "Alexandru Borza", 42 Republicii St., 400015, Cluj-Napoca, Romania

^c National Institute for Research and Development of Isotopic and Molecular Technologies, 67-103 Donat St. 400293, Cluj-Napoca, Romania

^d Babeş-Bolyai University, Department of Molecular Biology and Biotechnology, 1 Kogălniceanu St., 400084, Cluj-Napoca, Romania

^e Biological Research Center, Botanical Garden "Vasile Fati", 16 Wesselényi Miklós St., 455200, Jibou, Romania

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ABSTRACT

Current physical or chemical methods used for remediation of soils contaminated with hexachlocyclohexane (HCH), leave behind significant levels of pollutants. Given the compound's volatility and persistence in the environment, sites contaminated with HCH remain a concern for the population living in nearby areas. By making use of both the recovery capacity and the pollutant uptake ability of spontaneously growing vegetation, our study aimed to identify native plant species able to cover and moreover take up the HCH left at a former lindane production unit in Turda, Romania. The results showed that dominant species across the study site like *Lotus tenuis, Artemisia vulgaris* or *Tanacetum vulgare*, were capable of taking up HCH in their tissues, according to different patterns that combined at the scale of the plant cover was characteristic for vegetation succession on disturbed soils of the Central European region. Finally, we conclude that plant species which grow spontaneously at the HCH contaminated site in Turda and are capable of taking up the pollutant, represent a self-sustainable and low maintenance phytomanagement approach that would allow for the reintegration of the site in the urban or industrial circuit and nevertheless would reduce the toxicity risk to the neighboring human inhabitants.

1. Introduction

Hexachlorocyclohexane (HCH) was one of the most popular pesticides produced for worldwide agricultural use between the 1950s and 1980s. From the mixture of 8 isomers that HCH comprises of, only the y-isomer (lindane) has insecticidal properties. Separating each ton of 99% purity lindane from the total HCH, results in approximately 8 tons of waste isomers (Vijgen et al., 2005). At lindane production sites around the world, 7-8 million tons of HCH isomer waste were generated, containing between 60 and 70% $\alpha\text{-HCH},$ 10–12% $\beta\text{-HCH},$ 12–16% γ-HCH, 6-10% δ-HCH, and 3-4% ε-HCH (Vijgen et al., 2005, 2011; Willett et al., 1998). Inadequate waste-disposal techniques and the generalized use of this compound created a global environmental contamination issue (Vijgen et al., 2005). For now, organochlorine pesticides remain the most widely encountered pollutants, with high resistance to physical, chemical, and biological degradation (Weber et al., 2013). Although severely restricted or banned, there are still countries that continue to produce and/or use HCH (Vijgen et al.,

* Corresponding author. E-mail address: peter.schroeder@helmholtz-muenchen.de (P. Schröder).

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2011). Romania last reported lindane synthesis in 2006 (Romania, 2008) and it was noted that the production of this compound has officially stopped (UNEP, 2006). Nevertheless, illegal trades with old waste HCH stocks were still reported after that time (Mogos, 2016).

Suspected to have carcinogenic and endocrine disrupting properties (Ogbeide et al., 2016; UNEP, 2005, 2007a; 2007b), HCH has recently been flagged for regulatory intervention and elimination (European Parliament, 2016a; Vijgen et al., 2011). The remediation of HCH contaminated soils is needed to limit the dispersion of the pollutant via air or water transport into other compartments of the environment and avoid its accumulation onto and into human food resources (Dubey et al., 2014). The industries that created the pollution problems have gone out of business, and there are no funds available for a rigorous clean-up and restoration of the former industrial sites. A series of physical (vapor stripping, soil washing, incineration) and chemical processes (super fluid extraction, oxidation, photochemical degradation) aim at the removal of organic contaminants from soils, but they have the strong disadvantage of high costs (Hamby, 1996) and are

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limited to small areas. Moreover, they cause the soil to lose most of its inherent properties, killing the soil life (Semple et al., 2001). Diffuse levels of pollutants that are left in the soil after the physical or chemical reclamation of the hotspots are too expensive to be further handled by these methods (Weber et al., 2011). Thus, recently bioremediation (Salam et al., 2013) and phytoremediation (Nurzhanova et al., 2013), were proposed as a sustainable, environmental-friendly and cost-efficient alternative (Singh and Singh, 2017), being especially suitable to tackle diffuse levels of soil contamination that are left behind by physical and chemical soil remediation. The Turda Chemical Plant (Romania) is a former lindane production site, a brownfield situated in an urban area, which represents an ongoing threat for human and environmental health. Therefore, an environmental-friendly management of this site is preferred to contain and reduce the pollutant levels, and to create the possibility of its re-integration into the urban or industrial circuit. The disadvantage of present phytoremediation methods is their focus on reducing the contamination levels in soil by using monocultures of fast growing invasive/alien plant species, while ignoring the improvement of soil quality and functions which are vital for any ecofriendly soil restoration process. As a matter of fact, phytoremediation using native spontaneous vegetation might prove more efficient, since these species are already adapted to the local conditions (Nouri et al., 2011). A high diversity plant cover is likely to be self-sustainable due to the interactions that develop between the different plant species or between plants and microorganisms in terms of nutrient use and cycling (Bender et al., 2016). On the contrary, as experienced in agriculture, monocultures exhaust an already disturbed and contaminated soil, requiring therefore higher maintenance cost (Figuerola et al., 2015; McDaniel et al., 2014).

In recent years, an increasing number of examples of HCH isomers uptake and accumulation by plants has become available from local spontaneous floras which adapted to the high toxicity of these sites (Abhilash et al., 2008; Becerra-Castro et al., 2013; Pereira et al., 2006; Salam et al., 2017). Additionally, previous studies have used the vegetation survey method on heavy metal (Heckenroth et al., 2016) and HCH contaminated soils (Tripathi et al., 2014), to infer the suitability for phytoremediation of the most abundant plant species spontaneously growing at these sites.

Our first hypothesis is that the vegetation cover that develops spontaneously at the HCH contaminated site in Turda provides a pool of novel plant species to be used in phytoremediation. For this purpose, we used the vegetation survey method combined with HCH accumulation measurements, to assess if the highly abundant species that grow within the contaminated site are able to tolerate and take up the pollutant. The second hypothesis underlines the possibility of the local vegetation to undergo spontaneous succession in pollution conditions. Such a development of the vegetation might indicate that phytoremediation using a high diversity plant cover, represented by species from the local flora growing on the HCH contaminated soil, is a low maintenance and self-sustainable method for pollutant removal. Considering the presence of numerous old waste HCH deposits all over the globe onto which a native vegetation cover likely developed, this self-sustainable approach on phytoremediation could be applied worldwide.

2. Materials and methods

2.1. Site description

The former Turda Chemical Plant $(N = 46.557192^{\circ})$, $E = 23.781689^{\circ})$, located within the precincts of Turda city, is a brownfield that has not been reclaimed since the activity ceased in 1998 (Fig. 1a). The factory was estimated to have produced about 60,000 tons of HCH waste isomers, which were inadequately disposed at several dump sites within and around the city. The proximity of the waste dumps and the easy access for private persons on the grounds of

the former chemical plant, make them a continuous pollution source and health concern for the neighboring residential area.

The study area includes two presumed HCH hotspots. One is a waste dump on the right bank of the Arieş river with a surface of 4000 m^2 , estimated to contain 14,800 tons of industrial waste with a maximum HCH concentration of 472,914 mg kg⁻¹ of substrate (Prodan et al., 2011; Proorocu, 2005). The other presumed hotspot is located at the former lindane production department (Filip D., Personal communication, 2015). The current aspect of the site is a mosaic of soil, industrial substrates (e.g. limestone waste from calcium carbide production) and rests of demolition. Nevertheless, the area was colonized by spontaneous vegetation, and therefore was selected for phytosociological and HCH pollution level assessment.

2.2. Sampling design and phytosociological surveys

Samples were collected based on a random stratified model (Taylor and Ramsey, 2005). The optimal number and the spacing of the sampling locations in order to find the precise location of the hotspots were calculated by estimating the size and shape of the presumed hotspot (Department of the Environment, 1994; Ferguson and Abbachi, 1993). This resulted in a regular grid with 17 square plots with side lengths of 60 m (Fig. 1b). A HCH-free control site in the nearby hills and canyons, which is similar enough to our study site in environmental parameters or land use, was not available. The difference in bedrock, microclimate, available seed pool or land use (e.g. pasture, natural reserve or urban area) would have hampered the comparison of the vegetation between these sites, due to a multitude of confounding variables. Furthermore, phytosociological surveys in the area, dating prior to the construction of the Turda Chemical Plant were inexistent. Therefore, the sampling design could not be replicated. To assess the existence of a vegetation succession on the HCH contaminated site and to be able to adapt a possible future phytoremediation management to the changes occurring in time in the area, solely the phytosociological survey was repeated in 2017, without being followed by HCH measurements in soil and plants.

For the present study, only the already existing vegetation at the former chemical plant was considered. To identify the most abundant plant species present throughout the plots, two circular shaped phytosociological surveys of 10 m diameter each were conducted at randomly selected locations within each plot. For the list of all the identified plant species at the Turda site in 2015, please refer to supplementary material 3. Following the phytosociological surveys, species chosen for further study were: Pastinaca sativa for plot G01, Artemisia vulgaris for G02, G03, G07, Lotus tenuis for G04, G06, G15, G18, Atriplex nitens for G05 and G17, Tanacetum vulgare for G08, G09, G12, G13, G14, and Erigeron annuus for plot G20. The sampling points for soil and plant material were selected within the limits of each plot, at the place where the targeted species were found in highest abundance. The identity of the plant species was confirmed using the flora by Sârbu et al. (2013), while the covering percentage of each plant species in the survey was assessed using the Braun-Blanquet coefficients for the cover-abundance scale in vegetation analysis (Braun-Blanquet, 1964) as follows: 0 = 0% coverage; r = single individuals; + = very few individuals; 1 = < 10%cover; 2 = 10-25% cover; 3 = 25-50% cover; 4 = 50-75% cover, 5 = > 75% cover. The coefficients were transformed into numeric data for further analysis using the bb2num function of the R package Simba V. 0.3-5 (Jurasinski and Retzer, 2012). The total vegetation cover (i.e. the visual estimation of the surface covered by vegetation in each plot, expressed in percentage), the total rock (concrete)/waste cover, as well as the GPS location of each plot center were recorded during the vegetation surveys. For a comprehensive assessment of the soil characteristics across the studied area, we made use of Ellenberg's Indicator Values (EIVs) (Ellenberg, 1992), with adaptation to Romanian flora (Sârbu et al., 2013). Each plant species shows a constant preference for a distinct range of edaphic parameters. EIVs (i.e. light, temperature, humidity, reaction and nitrogen) describe the edaphic preferences of

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