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Published online: 29 November 2023

https://doi.org/10.5802/crgeos.237

Part of Special Issue: Human Environment Observatory
Guest editors: Robert Chenorkian (Professeur Émérite Aix-Marseille Université, Conseiller Scientifique CNRS-INEE pour les OHM),
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Les Comptes Rendus. Géoscience — Sciences de la Planète *sont membres du Centre Mersenne pour l'édition scientifique ouverte*
www.centre-mersenne.org
e-ISSN : 1778-7025

Comptes Rendus
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Human Environment Observatory

OHMi Estarreja: an ecosystem highly anthropized where contamination is the unifying element towards interdisciplinarity

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Abstract. The Estarreja area (North of Portugal) has been subject to intense industrial pollution and host the second largest chemical industry complex in the country, which produced mainly ammonium sulphate, nitric acid and ammonium nitrate, but also synthetic resins. The CNRS' Human–Environment International Observatory of Estarreja (OHMi Estarreja) aims to study the multiple effects of this industrial activity on the man-environment relationship, considering the temporal

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dimension, marked by changes in industrial practices. Another OHMi Estarreja originality is that human health projects are being carried out at the local level, under a double geochemical and socio-behavioral approach. Lastly, the Estarreja region is also part of one of the most important wetland ecosystems from Portugal, the Baixo Vouga Lagunar. This area is highly vulnerable and currently faces a set of natural and anthropogenic pressures. The overall interdisciplinary understanding of the risk induced by the chemical complex was possible by several complementary approaches: (a) spatial-temporal evolution of the contamination level, (b) health studies of the Estarreja population, and (c) societal changes in Estarreja area. The results showed a complex relationship between industrial pollution and health risks. During these first 10 years of OHMi existence, the bibliographic research as well as the study of the titles of the funded projects clearly show the interdisciplinarity in place to address this complex issue of human–environment interactions.

Keywords. Bibliography, Environment, Health, Pollutions, Projects, Risk assessment, Society.

Published online: 29 November 2023

1. Estarreja Chemical Complex (ECC) industries (history, production)

Estarreja is a municipality of about 27,000 inhabitants with an area of 108.40 km^2, located nearby the largest brackish water lagoon in Portugal (Ria de Aveiro—Central Region of Portugal) (Figure 1). The Ria de Aveiro is an ecologically rich environment, a natural habitat for many aquatic and terrestrial species, and is defined as a Special Protection Area under the European Directive for the protection of birds [ELA-European Lung Association, 2013]. Despite the significant urbanization of the region, a rural character, marked by agricultural activity and diversified fishing activity in the lagoon and in the sea, persists.

Since the early 1950s, the Estarreja Chemical Complex (ECC), has an intense industrial activity with negative impacts on air, soil, sediment, surface water and groundwater. Notwithstanding, population historically relies on groundwater as a source of water supply for human, cattle, and agricultural uses.

During five decades, the liquid effluents from the Estarreja Chemical Complex (ECC) were discharged to the Ria de Aveiro lagoon or deposited in evaporation ponds, through open channels and pipelines [Costa and Jesus-Rydin, 2001]. These effluents were rich in benzene, monochlorobenzene, mononitrobenzene, aniline, methanol, hydrocarbons, mercury, arsenic, zinc, lead, chlorides, sodium, sulphates, nitrates, nitrites, nitrophenols, formaldehyde and aniline [Costa and Jesus-Rydin, 2001, Ordens, 2007]. Solid wastes were also deposited for decades, without waterproofing, in three different parks: (1) around 5 ha containing 300,000 t of calcium hydroxide sludges with small amounts of carbide and acetylene impurities resulting from the vinyl chloride monomer manufacturing; (2) around 60,000 t of sludges rich in mercury, sodium chloride, calcium sulphate, calcium carbonate, magnesium hydroxide, calcium hydroxide and iron resulting from chlorine and caustic soda production; and (3) around 150,000 t of sludge rich in arsenic, mercury, zinc, copper, vanadium, and iron resulting from the treatment of pyrites [Costa and Jesus-Rydin, 2001, Ordens, 2007]. In addition to the identified contaminants, many others were present in raw materials (as principal product or as co-contaminants) or could be produced as by-products or may even result from degradation of major contaminants. Some of these were identified (e.g., polychlorobiphenyls (PCBs), hexachlorobenzene and volatile organic compounds (VOCs)) [Costa and Jesus-Rydin, 2001, Ordens, 2007, Cachada et al., 2012, Grilo et al., 2013, Nunes et al., 2013], but many others could be unknown.

Since the implementation of the project ERASE (concluded in 2005), which included the construction of a landfill for the pyrite residues and contaminated sludges, together with the improvements of waste management and of the industrial processes, and concomitantly to the existence of legislation and a more restrict control of it, it would be expected that releases of contaminants from the ECC have been decreased. However, recent studies still report signs of contamination in the area, which are likely to be related with the legacy left by past management measures [Cachada et al., 2012, Marques et al., 2018, Cabral Pinto et al., 2020a,b].

Despite the several studies performed in the last decades to evaluate the organic and inorganic contamination of different compartments on the

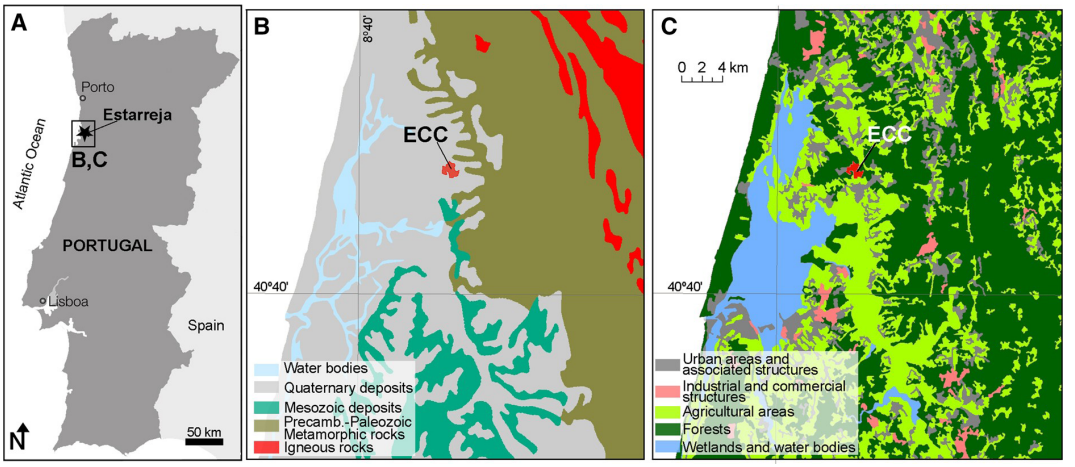

Figure 1. Location (A), geological (B) and land-use (C) maps of the OHMi area from Cabral Pinto et al. [2018].

terrestrial environment nearby the ECC, such as: atmosphere [Borrego and Pio, 1991, Nunes et al., 2013], groundwater [Barradas et al., 1992, Reis et al., 2009, Marques et al., 2018, Cabral Pinto et al., 2020b], soil [Barradas et al., 1992, Costa and Jesus-Rydin, 2001, Cachada et al., 2012, Anjum et al., 2013, Coelho et al., 2018, Cabral Pinto et al., 2020a], street dusts [Patinha et al., 2015a,b, Marinho Reis et al., 2018], and biota [Cabral Pinto et al., 2020b, Reis et al., 2009, Anjum et al., 2013, Coelho et al., 2018], a clear picture of the current situation is still missing. Indeed, a holistic risk assessment analysis for this area has never been done. Even though studies are increasingly comprehensive and interdisciplinary, most of them are still focused on a specific question, sometimes limited to only one matrix and to a small area, resulting in scattered data. Indeed, this is a very complex problem since several issues must be considered, i.e., the environmental compartments affected, the types of contaminants present (with different behavior, sources, and pathways), the land uses and receptors affected and the legacy *vs* current emissions.

2. Genesis of OHMi Estarreja

We approach the very tough complexities of extensively anthropized ecosystems whose understanding demands interaction between the full range of environmental sciences, from geology, life up to social sciences. Chernokian [2021] described it well and underlined that interdisciplinarity is a structural constraint in such observatories. Thus, a comprehensive interdisciplinarity facility within the French CNRS was implemented since 2007 to address with this situation: the Human–Environment Observatories (Observatoires hommes-milieux, OHMs). Thus, the OHMs are dedicated to the study of heavily anthropized sites hit by a "disrupting event", which has disturbed the ecological, economic and social balances that have developed over decades in the local socio-ecological conditions.

In this context, the OHMi Estarreja researches done in this area previous to 2010, were focused on the impact of the industrial activity on the geochemical aspects of contamination [Hall et al., 1985, 1987, Pio and Anacleto, 1988, Pio et al., 1989, Barradas et al., 1992, Inácio, 1993, Tavares, 1995, Pereira et al., 1998, Cachada et al., 2009]. Studies regarding the relationship between industrial activities, populations, their health status, and biological ecosystems remained in an early state. In fact, a comprehensive vision of the multiple impacts of the industrial complex was lacking. Thus, the human factor can no longer be considered either as a variable that distorts a natural ecosystem, or as a component of a socio-ecosystem in which an "anthroposystem" interacts with a natural biophysical "ecosystem". It must be seen as one of the components of a single ecosystem, in which the anthropic interacts directly with the abiotic and the biotic to constitute what we will call a Single EcoSystem (SES) [Chernokian, 2021].

Moreover, each of the studies was limited in time, and not allowed the understanding of the qualitative evolution of human–environment relations over several decades. The "public health" dimension seemed particularly neglected.

At this stage the binary relationship between anthropogenic contamination and health appeared too simplistic to account for the complexity of the balances between the industrial complex, humans, and the fragile surrounding environment. The need of interdisciplinary inputs and contributions from the social and health sciences was obvious to enlarge the scope of the research carried out in Estarreja. The French Observatoire Hommes-Milieux of Estarreja International (OHMi Estarreja) was created by the CNRS in March 2010, and co-constructed with Portugal (U. Aveiro researchers) and France (University of Paris cité, Babel researchers) in March 2010. Initially structured around the two teams that initiated the program: the UPR 2147 of the CNRS and the Department of Geosciences of the University of Aveiro, the OHMi very quickly involved other departments of this university (Departments of Biology, Chemistry and Land Management) and the Research Institutes GEOBIOTEC and CESAM, as well as several French research units, notably linked to the OHM Bassin Minier de Provence (OHM BMP).

The OHMi Estarreja initially focused on questions concerning the overall interdisciplinary understanding of the risk induced by the ECC, which was possible by several complementary approaches: (a) spatial-temporal evolution of the contamination level, (b) health studies of the Estarreja population, and (c) societal changes in Estarreja area (Figure 2). The results already achieved showed a complex relationship between industrial pollution and health risks. This does not depend exclusively on the environmental concentration of contaminants, but also on multiple factors that includes domestic (water use) and agricultural (crops cultivation and consumption), and practices that are influenced by the risk perception (variable within the population), based on studies at the local level, and as close as possible to the population. In order to understand the relevance of the factors that control the effects of environmental contaminants on human health (such as, concentration and bioavailability of contaminants, diet and lifestyle, age and gender, individual biological susceptibility, etc.), the OHMi Estarreja approach has been multidisciplinary from the beginning, with: (a) the collection of different types of geochemical data concerning the concentration, distribution and variation over time of potential toxic elements to humans; (b) eco-biological data concerning the accumulation and persistence of pollutants in the animal and plant biological chains; (c) medical data (biological analyses, prevalence rates of different diseases, morbidity and mortality rates); and (d) sociological data (lifestyles, dietary habits, family structure and risk perception). Another specificity of this approach was considering the long-term (more than two decades), which is rare in impact studies of industrial contamination. The access to retrospective data, both geochemical (previous work of colleagues from the University of Aveiro), and medical (national statistics and local databases) allowed to constitute a solid base of information on which our own work could be based.

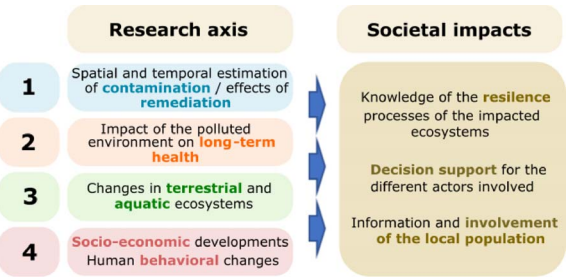

Figure 2. Present OHMi Estarreja structure axis around several major research areas.

Thus, knowledge of contamination as well as the impact of them on, notably, health for the population and the remediations done were in fact some backgrounds of the birth of OHMi Estarreja.

Moreover, from the beginning, entities of the civil society were involved in the studies of the OHMi Estarreja, such as, the Health Center of Estarreja depending on the Ministry of Health, the Environmental Service of the Estarreja Municipality and the PACOPAR (the Community Advisory Panel of Estarreja's Responsible Care® Programme—a group of entities that act in commitment with the local community to improve Estarreja's life quality, applying the commitments of Responsible Care®).

3. Development and evolution of the OHMi Estarreja

From the first studies, the ecological, economic, and social balances that have developed over decades at the local socio-ecological level progressively led to the structuring of the OHMi around several major research axes, each axis corresponding to a level of integration of the disciplines involved (Figure 2).

The methodology of the research areas have been listed and schematized in Figure 2 in order to clarify and help the management of the OHMi without forgetting think in a Global Ecology (an understanding from the interactions of all environmental sciences).

- The first research axis is the spatial and temporal estimation of environmental contamination. This axis is fundamental and constitutes the scientific base on which the other studies carried out within the OHMi are based. It essentially calls upon the skills of geochemists as well as some remediation techniques and constitutes the first level of study in the OHMi;
- The second axis concerns the health of populations in the Estarreja region. It links data on anthropogenic contamination with health biological and/or medical data;
- The third axis is related with the observed modifications of the terrestrial and lagoon ecosystems, notably the Baixo Vouga lagunar, in connection with the different human activities. It calls upon the work of biologists, but also researchers in human sciences (work on land use), and integrates the geochemical data of the first axis;
- Finally, a fourth research axis concerns the societal changes in the Estarreja region. It includes the economic and demographic evolutions on the long-term, as well as the changes in lifestyles during the last decades, in connection with the variations of the industrial activity.

Different projects have allowed the OHMi to put forward this interdisciplinarity and this "interactive/interdisciplinary" vision of contamination in the SES Estarreja.

The progressions, both in terms of structuring of the research in the OHMi and of the selected projects, gave rise to participative interactions with the population (actions with schoolchildren, information meetings), and offered to the local private or institutional actors (municipalities, industries, etc.) a scientific evidence base that decision-makers can consider. The influence of the OHMi's work is not always visible, with imperatives other than scientific (social, economic) being put forward by decision-makers. But the regular dissemination of scientific results to decision-makers allows and modifies their relationship to socio-environmental issues.

The initial OHMi core has expanded to other Portuguese institutions (Universities of Coimbra, Lisbon, Porto, among others) and French teams (University of Marseille-Aix, University of Montpellier, etc.) as the OHM's research area has expanded. Since 2019, the Observatory is jointly led by the Department of Geosciences of the University of Aveiro and the UMR 5023 LEHNA (University of Lyon).

4. Estarreja's projects: a way to illustrate the interdisciplinary Observatory development between SES studies

From the beginning until 2020, the OHMi Estarreja has financed a total of 62 research projects. Considering its main research axis, we are going to present the publications, as well as a focus on the words of all project titles, supported by OHMi Estarreja. These approaches can illustrate the OHMi evolution, but also the continuous focus on the contamination and health risk in a such anthropo-ecosystem.

4.1. *A zoom on the @10 years of OHMi: data compilation for a holistic environmental and human health risk assessment*

This project objective was to identify relevant scientific papers published in the last decade and review what is known about contaminants, areas of concern and potential risks to the environment and human health. At the end it was expected to identify knowledge gaps (e.g., matrixes that need to be further evaluated, compounds that need to be studied and exposures that need to be characterized).

4.1.1. *Methods*

The bibliographic search was first conducted in Scopus with the keywords "Estarreja AND (contamin* OR pollut*)". In addition, the OHMi Estarreja

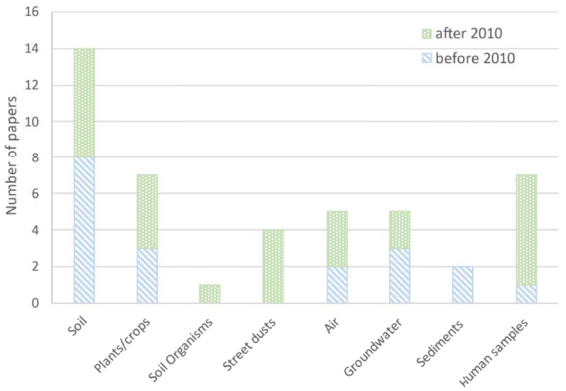

Figure 3. Number of papers published since 1989 for each matrix selected.

collection of HAL-Archive ouverte was also consulted. Two-time frames were considered: before and after 2010. For the selection of the papers the following criteria was used: present new data about contaminants for at least one of the terrestrial matrixes considered (air, soil, dusts, groundwater, stream sediments, and human biological samples); present a clear localization of the study area; perform an evaluation of levels found and/or identification of contamination sources.

4.1.2. *Main results obtained*

Despite the high number of studies related with the ECC contamination problems performed in the last decades, most of them were focused on the nearby lagoon, which was the final sink of the contaminated effluents. Indeed, a simple Scopus search with the keywords "Ria de Aveiro AND (contamin* OR pollut*)" came out with 185 results published since 1989, whereas using the keywords "Estarreja AND (contamin* OR pollut*)" resulted in 50 publications (with six of them being related with the lagoon). Thus, it is evident that the terrestrial environment is much less studied than the aquatic.

Thirty papers with relevant data about contaminant's levels in Estarreja terrestrial environment, published since 1989, were identified (after bibliographic search and cross-referencing). Most of them related with soil contamination, followed by levels of contamination in plants (including crops) and human biological samples (Figure 3).

For the other matrixes less than six papers have been published, with data about contaminant's levels in soil organisms being presented only in one paper. Looking to the data by period (before and after 2010), one can conclude that in the first 20 years research was focused on soil contamination and groundwater, whereas in the last decades several studies have been more dedicated to human health effects or matrixes that can directly affect it.

In Table 1 it is presented a summary of the selected studies published in the last 10 years.

Results show that, with exception of the atmospheric compartment, studies were mainly focused on inorganic contaminants, with only one study presenting data for two groups of organic contaminants (PAHs and PCBs) in soils. Other limitations are related with the fact that in several cases the sampling date is not known and most of the studies are focused on a restricted area, with a limited number of samples.

Soil studies conducted in the last ten years were mostly focused in a restricted area or in specific locations, previously identified as contaminated. Indeed, from the six studies published after 2010 (Table 1), only three gave a broader picture of the spatial distribution of contaminants, even that the study of Cachada et al. [2012] was more focused on the urban area and the ones from Inácio et al. [2014] and Cabral Pinto et al. [2020a,b], which are related to the same sampling campaign, had a higher intensity of sampling points nearby the ECC. Despite this, the median values observed for the potentially toxic elements (PTEs) studied in both areas are similar.

From PTEs evaluated, it was possible to identify the following contaminants of potential concern for this area: As, Cd, Cu, Hg, Pb and Zn. Cachada et al. [2012] also refer the polychlorobiphenyls (PCBs) as a group of concern. These studies indicate that median concentrations of PTEs are relatively low in the study areas and cannot be considered a concern, comparing for example with the soil quality guidelines for protection of human health from Canada [Cabral Pinto et al., 2020a], or with the Netherland Target Values [Cachada et al., 2012], or even with the recently proposed reference values for Portugal [APA-Agência Portuguesa do Ambiente, 2019].

However, these studies refer a high variability of results and the presence of some hotspots of contam-

Table 1. Relevant scientific papers containing data of contaminant's levels in different environmental compartments of Estarreja area, published since 2010 and ordered by date

Area	Sampling date	Land use	Matrix	No samples	Contaminants	References
ECC vicinity; steiro	n/a	Industrial; Riverbank	Soil; Plants	4	Zn	(1)
Urban area; ECC vicinity	n/a	Agricultural; Urban	Soil	26	As, Cu, Pb, Zn, Ni, Cd, Mn, Cr, PAHs, PCBs	(2)
Vala de S. Filipe	n/a	Agricultural	Soil; Plants	12	As, Cu, Hg, Pb, Zn	(3)
ECC vicinity (100 km^2)	2012	Industrial; Forest; Urban; Traffic; Rural	Air	128	NO_2, 17 VOCs	(4)
ECC vicinity (20 km^2)	2011	Agricultural	Soil; Crops	26	As, Cd, Cr, Cu, Hg, Ni, Pb, Zn	(5, 6)
Urban area; ECC vicinity	n/a	Urban	Dust	21	Al, Ba, Ca, Co, Cu, Fe, Mn, Ni, P, Pb, Sb, Ti, Zn	(7, 8)
Urban area; ECC vicinity	n/a	Urban	Dust; Toenails	18; 30	Cu, Mn, Pb, Zn	(9, 10)
ECC vicinity	n/a	n/a	Urine	103; 31	Al, As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Se, Zn	(11)
ECC vicinity	n/a	Agricultural; Industrial; Forest; Prairie	Soil; Earthworms	4	Cd, Fe, Cu, Ni, Pb, Zn	(12)
ECC vicinity	1989–2016	n/a	Groundwater	n/a	Fe, Cl, Mn, SO_4, Zn	(13)
ECC vicinity	n/a	n/a	Fingernail	58	Al, As, Ba, Cd, Co, Cr, Cu, Fe, Hg, Li, Mn, Ni, Pb, Sb, Se, Sn, Sr, Ti, V, Zn	(14)
ECC vicinity	n/a	n/a	Hair	79	Al, Mn, Fe, Cu, Zn, Hg, Pb	(15)
ECC vicinity and background	2013	n/a	Groundwater; Hair; Urine	19 + 75 + 103	Al, As, Cd, Hg, Ni, Pb, Zn	(16)

(1) Moreira et al. [2011]; (2) Cachada et al. [2012]; (3) Anjum et al. [2013]; (4) Nunes et al. [2013]; (5) Cabral Pinto et al. [2020a]; (6) Inácio et al. [2014]; (7) Patinha et al. [2015a]; (8) Patinha et al. [2015b]; (9) Marinho Reis et al. [2018]; (10) Reis et al. [2015]; (11) Cabral Pinto et al. [2018]; (12) Coelho et al. [2018]; (13) Marques et al. [2018]; (14) Cabral Pinto et al. [2019a]; (15) Cabral Pinto et al. [2019b]; (16) Cabral Pinto et al. [2020b].

ination, in which levels clearly exceed the reference values. These hotspots are especially located nearby Vala de São Filipe and values as high as 6746 $mg{\cdot}kg^{-1}$ for As, 1592 $mg{\cdot}kg^{-1}$ for Cu, 170 $mg{\cdot}kg^{-1}$ for Hg, 1295 $mg{\cdot}kg^{-1}$ for Pb, and 1485 $mg{\cdot}kg^{-1}$ for Zn, have been reported [Anjum et al., 2013]. Since soils from Estarreja region are sandy soils, acidic to neutral, with low contents of organic matter [Cachada et al., 2012, Inácio et al., 2014], there is a risk of elements transfer in the soil–water-plant system, which is of special concern in these areas with high levels of contaminants. Nevertheless, less than 1 km away from Vala de S. Filipe levels decrease between 20 and 370 times [Anjum et al., 2013]. This is in accordance

with what is known about the waste disposal management practices in the past, that leave a legacy regarding the contamination of the channels used to transport effluents until Ria de Aveiro.

Two studies assessed the soil-plant transference and concluded that, for some plant species, levels of PTEs (namely, Zn, As, Cu, Pb, and Hg) in aerial parts were above those considered as normal and that turns them unsuitable for animal consumption [Moreira et al., 2011, Anjum et al., 2013]. Nevertheless, these two studies were conducted in very restricted areas where the hotspots are located, and they may not be representative.

Another study investigated the concentrations of PTEs in vegetables (cabbage, tomato, and potato) collected in kitchen gardens/small farms from the vicinity of the ECC [Inácio et al., 2014, Cabral Pinto et al., 2020a], covering a wider area. The authors concluded that, despite levels of As, Cd, Cr, Cu, Ni and Zn above the proposed as normal for terrestrial plants, only in 4% of cabbage leaves samples the levels of As, Cd and Hg were above (or within) the limits for human consumption.

Only one study was focused on the potential adverse effects to soil biota [Coelho et al., 2018] and concluded that there was no bioaccumulation of the studied PTEs in earthworms. However, despite being conducted nearby the ECC, the four sites chosen presented low levels of PTEs and some of the contaminants of concern, such as As and Hg, were not considered.

Data about groundwater contamination in the last decade is very scarce, with only two studies published. The study of Cabral Pinto et al. [2020b] evaluates the levels of PTEs and reports median values higher than the aquifer natural background for As and Zn, but, as observed for soil samples, the data is characterized by a high variability, with some extreme concentrations that largely exceed these background values. The authors also compared the obtained values with several guidelines for human consumption and concluded that despite median values do not exceed these, there are several hotspots of contamination, especially for As, Hg and Zn. Similarly, Marques et al. [2018], which performed a temporal evaluation between 1989 and 2016 for some contaminants, observed that the there is a clear degradation of water quality nearby the channels (Breja and Vala de S: Filipe).

Regarding air quality, only one study was done, and almost 10 years ago, but it covered a representative area and an important group of contaminants, the VOCs, which may have negative human health effects. In this study, Nunes et al. [2013], concluded that the air quality of Estarreja region during the spring of 2012 was significantly influenced by ECC, with the highest concentrations of many VOCs like BTEX and NO_2 observed close to its vicinity. Nevertheless, the authors highlight that other sources contributed as well for the levels of VOCs and NO_2 in the area. However, it remains to be understood the implication of the levels found and the extent to which they may or not pose any risk to the environment and human health.

The four studies about levels of PTEs in street dusts are related with two different sampling campaigns (Table 1). The studies of Patinha et al. [2015a,b] did not find a clear link between studied elements and the ECC, but the urban dust samples showed a clear contribution of traffic-related particles in their composition, particularly related with brake dusts. Reis et al. [2015] and Marinho Reis et al. [2018] could not find a specific anthropogenic source of PTEs for outdoor dusts.

Regarding human health studies (human biological samples), Cabral Pinto et al. [2018] reported urinary levels of several PTEs (including As and Hg) and concluded that on average, they exceed those reported in the literature for healthy people. However, looking to the median values it is of note that, with exception of Al, Cd, Mn, and Zn, they fall within the expected range of values. In another study [Cabral Pinto et al., 2020b] the same authors showed the results by localities and it was observed that levels of PTEs found in urine were quite variable, and different elements had different trends showing that the origin and exposure pathway could be different for each element.

Levels of Cu, Mn, Pb and Zn in toenails were assessed in two studies [Reis et al., 2015, Marinho Reis et al., 2018] and concluded that the exposed group (27 individuals living nearby the ECC) are similar to the ones determined in toenails of the non-exposed participants (3 individuals). However, levels of Pb and Zn were slightly higher than the ranges found for healthy people available from the literature. On the other hand, Cabral Pinto et al. [2019a] concluded that PTEs contents (including Hg but not As)

in fingernails of Estarreja group fall within the range of values reported in the literature for non-exposed people. However, for Hg, levels in the study group were above the mean content found for non-exposed population. Similar results were observed by the same authors [Cabral Pinto et al., 2019b, 2020b] for hair samples. Moreover, some extremely high levels of As in inhabitants hair living closer to ECC and in areas where the hotspots of groundwater contamination were observed [Cabral Pinto et al., 2020b], warning that this issue should be further investigated.

4.1.3. *Conclusion in the context of OHMi*

The existence of sites with concentrations above the recommended value for soils, crops, and groundwater, especially for pollutants such as As, Hg and Pb, can represent a potential risk to the ecosystem and to human health. Indeed, some studies tried to predict the potential risks for human health, based on existing risk models and concluded that they may exist. Nevertheless, these models have to be used with care, and the following issues should be considered in the future: the use of specific input parameters for the local populations; the source–receptor pathway for each land use/matrix; the physical–chemical properties of contaminants, especially speciation studies of As and Hg, since highly influence their toxicity. Indeed, data related with human biological samples does not clearly point to the existence of a problem for human health, despite the levels of Hg and As found suggest that their source–pathway–receptor route should be investigated. The apparent contradiction obtained between risk models and data from human biological samples or even plants/crops may be related to the fact that highly contaminated areas are restricted to a very specific and small area and therefore do not have a major impact on human health. Moreover, the groundwater is not likely to be used for human consumption, even that it could be used to irrigation purposes. Nevertheless, all these issues should be taken into consideration in future studies.

Despite the paradigm shift since the implementation of OHMi, work has still to be made. In order to have a comprehensive overview of the contamination problematic it would be important to obtain updated data about the distribution of contaminants of concern, since the most recent data for soils and groundwater have almost ten years. It would be of great importance to have spatial distribution maps covering a wide area, in order to identify clearly the areas of concern (based for example on guideline values) and manage them accordingly. It is also crucial to identify the organic contaminants of concern (if there are). After identifying the most problematic areas, a specific risk assessment should be performed, considering the receptors at risk and the specific routes of exposure.

4.2. *OHMi-funded projects analyzed over 10 years: an illustrative approach through the study of project titles highlighting the founding element of contamination related to human–environment interactions*

An observatory is a privileged tool for monitoring a territory both spatially and temporally. Research through the titles of the research programs financed by the observatory allows us to highlight the axes put forward by the scientific committee of the observatory during the annual calls for projects, but also the "heart" of the socio-ecosystem and its "founding event".

The objective of this study was to identify relevant words in the projects' title in the last decade and listed what word was used about contaminants, areas of concern and potential risks to the environment and human health. At the end it was expected to identify if some of them was more or less present and/or if some new word was detected (that is to say new thematic and/or area of investigation).

4.2.1. *Methods*

This analysis of the words in the project's titles was also gathered over two periods, a first from 2011 to 2015 (Figure 4a) and a second from 2016 to 2020 (Figure 4b). Approximately 321 words were gathered for the 2011–2015 period (=25 projects), whereas for the 2016–2020 period 333 words were found (=37 projects). From these words we have performed "word clouds" using the software Iramutec©.

To make a "Word cloud" with the corpus of project titles between 2011 and 2020; the variable used *year; lemmatization performed. From each word the active forms were excluded (e.g., with, of, at, from). Some project titles were in French or Portuguese (as Baixo, complexo… or agriculture, sol, etc.), appeared also in the figure 4 and were pooled with

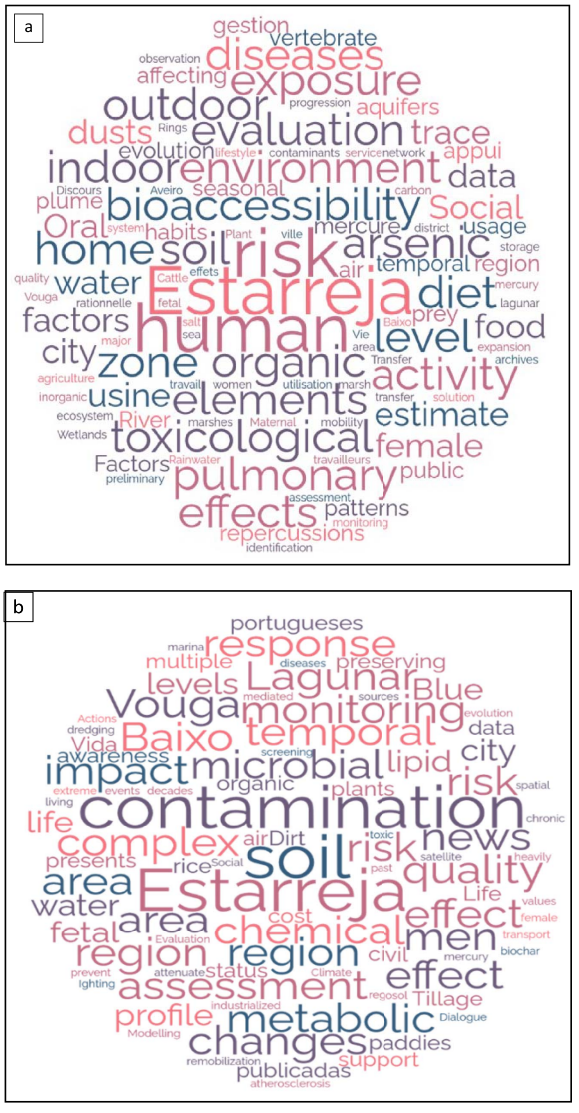

Figure 4. (a) Graphic representation of the cloud of words contained in the titles of funded projects and their frequency (the bigger the most cited) for the period (a) 2011 to 2015. (Iramuteq©; *n* total word = 321); (b) for the period 2016 to 2020. (Iramuteq©; *n* total word = 333).

similar English words (Table 2). Each word was listed and grouped together according to the general thematic or meaning (Table 2). Thus, with words by "themes" or "affinities/meaning" (Table 2) such as the environment or the link with humans, we gathered 61.6 and 62.8% of all the words forming the titles of funded projects respectively for the first and second periods.

4.2.2. *Main results obtained*

The word cloud, from all the projects over the 10 years, showed the word "Estarreja", but also "health" and "human" as well as to a less extent "contamination" or "contaminant", and also "soil(s)", "Aveiro", "risk" and "assessment" (Figure 4).

Other words associated with "local/spatial specificity" such as "*lagunar*, *baixo* or *complexo* or type of pollutants", such as arsenic and organic as well as "approaches/types/objects of study" such as pulmonar, monitoring, temporal or evolution were also commonly found.

The obtained word cloud illustrates well the intersection of questions and research topics addressed between environmental sciences, in particular via historical chemical contamination, and the specific lagoon ecosystem (Ria de Aveiro) as well as the study of societies and health.

The observatory is indeed a territory, particularly when the words "Estarreja" and/or "Aveiro" are found in the project titles. These two words (and their equivalents/synonyms, see Table 2) stand out in the two sequences analyzed (Figure 4; Table 2). Thus, they are used 15 and 17 times for Estarreja and 9 and 10 times for Aveiro, respectively for 2011/2015 and 2016/2020, i.e. between 7.5 and 8% of all words illustrating the strong link of the SES specific localization.

In the first studied period (Figure 4a), the titles of the projects express an important part to the human, to the risks and its evaluation as well as to the toxicological approaches, or on the bioaccumulation but also of pollutants of which arsenic. We can also notice the words related to health such as pulmonary and diseases linked to indoor/outdoor and environment. If we gather the words of the titles related to a "health" theme (such as food, diet, harmful, etc.; see Table 2), we will notice that it is the theme which gathers the most words on this first period of projects with 22 elements. The next two themes, very close, are those related to contamination and health with 20 and 19 words, respectively (Table 2). Finally, after the group related to the spatialization of the studies (Estarreja, Aveiro . . .) we find a group of words related to risk and its evaluation (rank 5; Table 2).

Table 2. Grouping of the words from the titles of funded projects by OHMi Estarreja in different general thematics over two periods: 2011/2015 and 2016/2020

Period	2011/2015 25 projects (321 words)			2016/2020 37 projects (333 words)			Evol
General thematics	Some words used in title	NP	RT	Some words used in title	NP	RT	
Spatial	Estarreja, d'Estarreja, OHM, ROHM…	17	4	Estarreja, ESTarreja, Estarreja/Aveiro, OHM	15	5	≈
	Aveiro, region, area, city, *ville*, municipal, municipality, Beduido-estarreja, district	9		Aveiro; city, region, area, Aveiro, Murtosa	10		≈
	Laguna, Vouga, Baixo, *Ria*, BioRia, wetlands	10		*Ria*; Baixo; lagunar; Vouga; saltmarsh	10		≈
	Complexo, quimico, fabricas, *complexe chimique*, *l'usine*	7		Industrialiezed, fabrica, factory, COPD, complex, *químico*, *complexo*	12		++
	Environnemental, ambient, ambiental, ecosystem, anthropoecosystem	12		Envionnement, ambient, *ambiental*, climate, environmental	8		-
Temporal	*Etude*, retro-observation, study, *années*, year/ans, *historique*, monitoring, progression, evolution, *archives*, *actuelle*, seasonal	12		Years, *vie*, *vida*, temporal, study, spatio-remporal, short-term, attenuate, resilience, past, long-term, life, *histoire*, decades, chronic, monitoring, journal, *narrativas*	19	2	++
Contamination	Contamination, contaminants, contaminated, *composés*, polluants, plume, dust	20	2	Contamination, contaminants, contaminated, levels, particulate, sources, elements	16	4	-
Typology of contaminants	Organic, organique, carbon	6		Organic, HAPs, carbon, lipid, *microplastique*	5		≈
	MTES (Hg, Ars, PTEs, inorganic, *mercure*…	7		Inorganic, As, Hg, metal,	4		-
Health	Health, diseases, *santé*, antibiotic, *alimentaire*, food, diet, dietary, harmful; home dust; neurodegenerative, maternal fetal, pulmonary, urinaires, reproductive, oral, inhalation	22	1	Respiratory, reproduction, pulmonary, placenta-fetal, patient, nutrition, microrespira, health, heavily, diseases, reproductive, lung, parturient, atherosclerosis, immunomodulation,	20	1	≈

(continued on next page)

Table 2. (continued)

Period	2011/2015 25 projects (321 words)			2016/2020 37 projects (333 words)			Evol
General thematics	Some words used in title	NP	RT	Some words used in title	NP	RT	
Human	Human (man/woman/humanized/families; education; social, women, female, *travailleurs*, *enfants*)	19	3	Human, citizen, civil, community, female, *homme*, men, population, portugueses, *trabalhadores*, social	15	5	-
Subjects of study	Soil (soils, *sol*, *sols*, regosol, anthroposols)	3		Soil, soils, regosol, anthroposol	17	3	+++
	Plant	1		Seagrass, zostera, salicornia, rice, ramossisima, plants, halophyte	7		++
	Animal, prey predator, vertebrate	4			0		
	Microbial	0		Microbial, microbiotA, microbial-laden, micro-organism	6		++
	Agricultural, *agriculture*, *agricole*	3		Agricultural, *agriculture*, No-tillage, meadow, cultivated, farming	8		+
	air, marsh, landscape	4		Air, Paddies, salt marsh	3		≈
	Eaux, wetlands, aquifer, rainwater, river, souterraines, sea, ground level	10		Irrigation, water, rainwater, sea, groundwater, freshwater	7		-
Risk assesment	Risk, *risque*, risks, assesment, *évaluation*	15	5	Assessment, risk, evaluation, effects, impact, *risques*	12		≈
Disciplinary/approaches	*écotoxicologie*, *toxicologie*, toxicity, ecotoxicological	3		Ecotoxicology, toxic	2		≈
	Bioaccessibility/transfer, mobility	5		Metabolic, bioavailability, remobilization	4		≈
	Geochemical, geochemistry, geostatistic, geophysic, mineralogical	5		Ecological, geochemical, genomic, physical	4		≈
	Interdisciplinary, multidisciplinary	1		Bioremediation, phytochemical based, phytoremediation, remediation, restoration,	5		+
	Total of words: 198 words/321			Total of words: 209/333			
	Percentage: 61.6%			Percentage: 62.8%			

(≈: no thematic evolution between the two periods; +, ++, +++: illustrates an increase in this thematic between the two periods; -: illustrates a reduction of the thematic between the two periods).

RT: Rank of the thematic; NP: Number of presences; Evol: Evolution; *In italic word in French or Portuguese.*

Therefore, during this first period, the projects and studies logically focused on the founding and/or structuring element of the observatory and therefore on the contamination of this region, the health of its inhabitants and the impact on society.

On the second period studied (2016–2020), the projects selected remain focused on the Observatory site (Estarreja, complex), but this time, contamination and soils stand out as strong elements (Figure 4b).

There is also a diversity and a focus on certain environments such as "laguna, Baixo and Vouga", as well as a temporal dimension in the studies with the words monitoring and temporal. Finally, microbiology and the study of microorganisms are apparent (Figure 4b). The grouping of words related to a health theme remains a priority, but with a greater and more precise diversity (in terms of disease or approaches taken, for example, with the words atherosclerosis or placenta-fetal; Table 2). Finally, the second most present theme is clearly that of temporality, particularly in terms of studies or research on the history of the complex or approaches on the scale of monitoring/measurement over several years (Table 2). Finally, the theme of "soil", particularly in terms of anthropic characterization, emerged during these years. Words related to contamination are also still present, although slightly decreasing in project titles with 16 words over this period, while those related to human decreased more noticeably with 15 words (Table 2).

During this second period, studies were more focused on contamination and health but were often more precise in terms of sites, or matrix study as soils, or also disease typologies for example, as well as considering time on effects, risks, and evolutions.

4.2.3. *Conclusion in the context of OHMi*

We can see both an evolution of certain themes or objects of study but also on the overall conservation of the human–environment interface.

As it can be expected, the projects are still very much focused on the site and its region, with the "health" theme in the first place for both periods, and well linked to the founding element (the chemical complex and the contamination that resulted from it). The human element is always at the heart of the studies. The strong points are often an integrative, and interdisciplinary approach and a time frame (long time frame that makes it possible to come back to several questions/studies over time periods of up to 10 years). The various annual calls for projects have been careful to highlight certain new approaches (for example, by focusing one year on the human sciences and the history of the chemical complex, or on microbiology or toxicology studies) but also by maintaining the link with the founding and structuring element of the observatory. These projects respond well to societal issues and illustrate good integration into the territory. The 62 projects funded highlight the desired interface between environmental sciences, health, and society.

5. Conclusions of OHMi Estarreja

The OHMi interdisciplinary approach to assess the health risks related to the ECC has benefited from a solid geochemical background on the area of Estarreja.

The participation of different teams from different universities with strong scientific background and field experience in Estarreja has been a major advantage, as well as the participation of representatives of civil society and industry. Another originality was to address health issues at the local level, under a double geochemical and socio-behavioural approach. The results already achieved show a complex relationship between industrial pollution and health risk. Further studies on PTEs speciation, as well as on biological individual responses to contamination or particularities of family lifestyles should be carried out in order to confirm or infirm this relationship between health, contamination and socio-ecosystem.

Looking to the data by period (before and after 2010), one can conclude that in the first 20 years research was focused on soil contamination and groundwater, whereas in the last decades several studies have been more dedicated to human health effects or matrixes that can directly affect it. In the two studies (bibliography and words in titles), different time periods were analyzed: while the first study, "bibliography", was conducted for the periods before and after 2010, the study of "word's titles" was focused only after 2011 due to the time after the creation (in march 2010) and the first call of project the year after (in 2011). Probably this can explain differences found but also that in the near future more

publications related to the topics identified in the last period will be expected.

Nevertheless, contamination is the structuring and unifying element of numerous studies and projects with a Portuguese specificity in terms of financing (few internal resources of its own) and the participation of laboratories or research centers that are often already multidisciplinary. The observatory thus becomes an object of study focused on the territory, its population (and therefore its health) and its contamination. The number of publications and projects submitted to each project call (some of which were financed by the Observatory) is an illustration of a dynamic and a desire to promote and communicate on this research.

Special attention should be paid to past and present evolution of the relationships between Estarreja population and its environment. The risk perception evolution, the economic crisis, the climate change, are likely to induce modifications in the local ecosystem, and consequently in the welfare of the population. The OHMi Estarreja gives the opportunity to catch, in a dynamic way, the adaptive pathways of the population–environment relationships.

Declaration of interests

The authors do not work for, advise, own shares in, or receive funds from any organization that could benefit from this article, and have declared no affiliations other than their research organizations.

Acknowledgments

This work was carried out within the framework of the OHMi Estarreja and (co)funded by the LabEx DRIIHM, French programme "Investissements d'Avenir" (ANR-11-LABX-0010), which is managed by the ANR". Further financial support was provided by the project on Joint Usage/Research Centre—Leading Academia in Marine and Environmental Research (LaMer), Ehime University, Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT). The authors acknowledge also the University of Aveiro, the Municipality of Estarreja, PACOPAR, and the Health Centre of Estarreja for all the support given to the OHMi Estarreja. This research was also supported by national funds through FCT—Foundation for Science and Technology within the scope of: UIDB/04423/2020 and UIDP/04423/2020; UIDB/05748/2020 and UIDP/05748/2020; UID/GEO/04035/2020 and through individual financing to AC (CEECIND/00058/2017). ND thanks also the Portuguese Foundation for Science and Technology (FCT) his working support by national funds (OE) in the scope of the framework contract foreseen in the numbers 4, 5 and 6 of the article 23, of the Decree-Law 57/2016, August 29, changed by Law 57/2017, July 19.

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