Mercury Management along the Chain from Remediation to Final Storage: Principles Facilitating the Development of Beneficial Mercury Thresholds

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Abstract

Beneficial management of mercury can be achieved by applying certain logical, technically oriented principles. Swiss environmental provisions for soil, site remediation and waste have been developed using these strategic, long-term-oriented rules. Describing the historical development of state-of-the-art techniques might help others shorten their own pathway to development. This paper reviews development of these rules and gives practical guidance and a comparison of data in the specific matter of mercury. Special attention is paid to the design of a threshold system within the chain starting from observation, proceeding to remediation, followed by treatment and ending with final storage. Thresholds are shown not necessarily to be rigid borders, but beneficial frameworks, which can even be flexible to local circumstances if designed well. The overall target of the demonstrated strategy is to keep the anthroposphere safe from hazards and at the same time maintain the lithosphere in a way that does not add new risks of environmental damage. Finally, for a few holistic cases, keeping in mind the targets of the Minamata Convention, practically proven elements of a transparent and lucid management system for mercury are suggested. As in many processes, involving all stakeholders in a transparent and integrative way is crucial for success.

Key words : clarke values, mercury, remediation, soil, threshold, waste

1. Introduction

Following preceding agreements such as the Aarhus Convention and the Genève Air Emission Reduction Protocol from 1972, the Minamata Convention entered into force in August 2017 with a clear mandate to protect human health and the environment from the adverse effects of mercury. Amongst other countries like Norway, Switzerland played the role of facilitator for the convention from the start. The basis for this engagement is twofold: on the one hand, the long-term engagement and rich experience of the Swiss in several international environmental treaties such as the Basel, Stockholm and Rotterdam conventions were brought in to facilitate the process to establish this new Convention. The other aspect is the historical and still growing expertise of Swiss industry in chemicals and pharmaceuticals, including their treatment and disposal. The latter have included the use and treatment of mercury in different processes, applications and products. Although most of these applications are no longer legal, the technical knowledge for treating mercury still exists and is used by several corporations. Last but not least, the uses of mercury in Switzerland described herein have resulted in contamination of sites. Most of these are undergoing remediation nowadays or have been cleaned up already, using a national fund to support these activities through effective funding from a landfilling fee. Again even these projects create expertise and capacity, as well as establish regulations in their specific domain. This paper reviews the milestones of Switzerland's development of a state-of-the-art mercury management system starting in the 1970s. Retracing this pathway may help others facilitate their own processes to improve environmental conditions. Additionally this paper aims to provide hints on specific challenges of the Convention, *e.g.*, the process of establishing thresholds for mercury waste.

2. Development of Environmental Law

Environmental law in Switzerland was not established on the basis of a uniform concept, but as a continuous reaction to specific problems that were occurring. Following changes to the country's economy from mainly agricultural to that with a strong second industrial leg in the last century, the first environmental

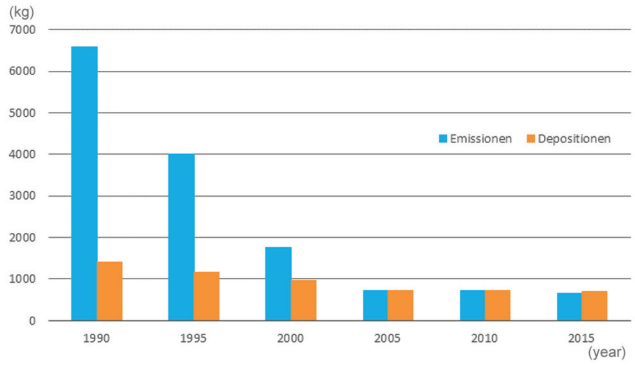

Fig. 1 Emission and Deposition of mercury in Switzerland (kg/year). (FOEN (2017), MSC-E (2017), Ritscher *et.al.* (2018))

regulations aimed to improve the quality of water and air (FOEN, 2013).

Today environmental regulation is still problem-oriented but much more specific (*e.g*., biogenetics, climate-change, mercury). As a cornerstone, the Swiss Environmental Protection Act (EPA) was established in 1985. The economic boom in these decades laid the ground for a strong increase in the amount of waste, which was then tackled by the internationally well-perceived Technical Ordinance on Waste (TOW), according to which waste had to be recycled after treatment or deposited in a suitable landfill site in an environmentally sound manner. The effectiveness of the TOW provisions had a positive environmental effect, for example, mercury emissions to air have shown a reduction by 90% since 1990. Side note: In the same period, deposition did not drop by the same percentage, suggesting global transport mechanisms for mercury and its compounds (Fig.1). TOW in its current format as the Ordinance on the Avoidance and the Disposal of Waste (ADWO, in force from 2016) is again a ground-breaking instrument as it settles new issues such as phosphorus recycling and handling waste from construction and demolition works (SFC, 2015) while still respecting proven basic principles.

Several additional Swiss environmental ordinances provide rules with respect to mercury (particularly for soil and remediation of contaminated sites). Nevertheless, if treatment of these materials is intended, the materials derived have to be handled as waste. This lays the ground for further consideration in this paper, examining the role and design of mercury waste management.

3. Basic Principles

In the years during development of TOW some basic principles were established as a beneficial framework and touchstone for this policy. These rules have proven helpful and are still applied today in updated versions of the environmental provisions. Chapter 2.2 of Bundesamt für Umweltschutz (BUS, 1986) gives a set of scientific and technical guidelines that can facilitate the setup of an environmentally safe management system for mercury.

3.1 Treatment and Recovery

Anticipating the now widespread rules of the circular economy, the following notion for tackling waste treatment was presented for the first time: “All waste treatment systems shall produce only two types of materials: reusable material and material suitable for final landfilling.” With respect to mercury, this principle today is a core consideration of the Minamata Convention. The now evident toxicity of mercury makes the removal of the element from economic cycles to the utmost degree desirable. As mentioned above in the Introduction, Swiss companies have developed vast experience in the treatment of mercury waste to fulfill this task. The diminishing usage of mercury products on the other hand generates a continuous flow of recovered mercury for stocks or landfilling, so clear rules and procedures for the final disposal of mercury will be crucial to fulfilling the goals of the Convention.

3.2 Landfilling

Both the aforementioned TOW and ADWO contain detailed provisions for landfilling of wastes, including thresholds for mercury. The main long-term consideration for landfilling is that landfilling shall be considered a transition of the material from the anthroposphere to the lithosphere without incurring environmental damage to either of them.

Substances are suitable for final storage if they can be kept long-term in an appropriate environment (in geo-physical and geo-chemical terms) in a way so that they are releasing only substances which cause no harm to air, water or soil. At the site, final disposal shall be substance specific: only solids are considered appropriate for final disposal.

3.3 Petrologic Approach

The petrological approach (Zeltner, 2002) was initially used to evaluate and project the behavior of residues derived from waste melting processes. It combines geological modelling, petrographic evaluation, material flux analysis and long-term experience with alteration and erosion. As a result, technical processes shall be steered in a dedicated way to gain materials of so-called "final storage quality," which in most cases result in a chemical and specifically mineralogical composition similar to the Earth's crust.

The guidance from BUS (1986) explains the same circumstances as follows: "Waste treatment processes shall be designed in a way that environmentally harmful substances, can be derived in a concentrated form, and environmentally harmless substances shall be in a form similar to soil or the Earth's crust." As Switzerland has a range of contaminated sites and some of the sites have to be remediated, the lesson learned was to avoid the buildup of new landfills that produce harmful emissions to the three environmental compartments of air, water and soil.

4. Threshold Systems and Levels

Prior to specification of a material as a waste, decisions have to be made, in most cases using chemical analyses and applying national legislation or guidelines. To facilitate triage of mercury contaminated sites or soils, ART (2013) recommends three levels of thresholds: background, observation and remediation levels (Fig.2).

The Swiss guideline to derive threshold values (if they are not present in existing policies) sets a stepwise approach to effectively assess the whole chain from investigation to remediation and finally, waste management procedures, keeping in mind human health

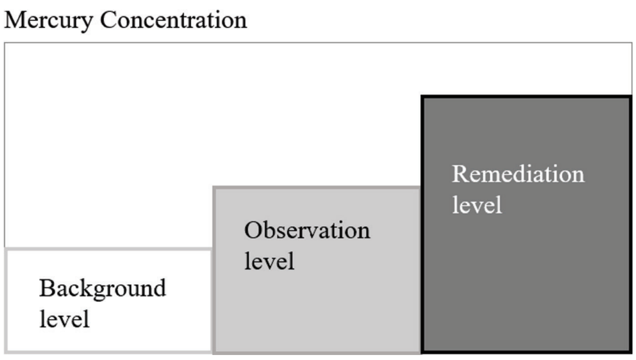

Fig. 2 Schematic diagram of levels to differentiate measures in management of mercury of soil or contaminated sites.

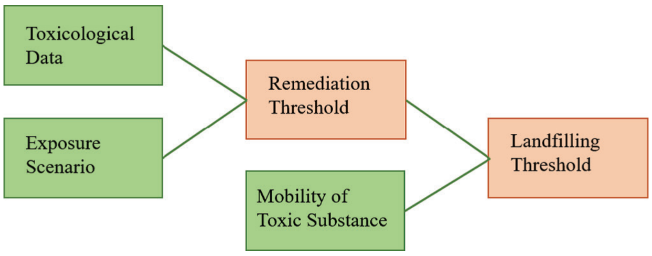

Fig. 3 Risk based scheme to derive threshold values for remediation and landfilling. (BAFU, 2013)

and environmental soundness. (BAFU, 2013)

The guideline uses US-EPA Regional Screening Levels (RSL) and the slope factor to evaluate toxicity and combines these with an exposure scenario defined by SFC (2017) to establish remediation thresholds. Derivation of landfilling thresholds starts with an assessment of the local geogenic background levels. This approach enables concentration levels to be differentiated depending on the genesis of the rock on-site, which can be remarkably different (*e.g.*, mining areas with high naturally occurring mercury minerals) (Fig.3).

As a core element for evaluating the landfilling threshold, a so-called virtual leaching test is used as represented in equation (1).

$$c_t = c_w \cdot (K_d + (W/S \cdot 1/\rho_w)) \cdot\cdot\cdot (1)$$

c_t = threshold (total solid concentration) [mg/kg]
c_w = conc. of constituent in pore solution (leachate) [mg/l]
K_d = solid/water distribution coefficient [liter/kg]
W/S = solid/water ratio [1]
ρ_w = density of pore-water [kg/liter]

4.1 Background Mercury Levels

As a definition, if the mercury concentration remains under the so-called background level, it produces no risk to human health, animals or the environment.

As deduced above in Section 3.3, Earth-crust values build a logical basis for development of background levels. Various sources in literature show the abundance of mercury in Earth's continental crust to be between 0.05 and 5 mg/kg, also known as clarke values. Table 1 (ART,

Table 1 Mercury background level in different countries. (ART, 2013, SFC 1998)

Country	Background mercury value (mg/kg)	Comments
Belgium (Flanders)	0.05	
China	0.15	Nature reserve area
China	0.3 – 1	Agriculture
China	1.5	Forestry
Czech Republic	0.4	
Germany	0.1 – 1	Dependent upon soil type
Finland	0.2	
France	3.5	
Netherlands	0.3 – 0.5	
Norway	0.05 – 0.2	
Slovakia	0.3	
Switzerland	0.5	Soil (SFC, 1998)
Sweden	2.5	Industry

4.2 Mercury Levels Necessitating Observation

Table 2 Range of mercury observation values for different land-use categories. (ART, 2013)

Land use category	Range: Mercury observation values (mg/kg)
Agricultural	0.362 – 10
Industry	2 – 4000
Gardening, sports, playgrounds	1 – 400
Housing	0.83 – 600
Other	0.1 – 36

Table 3 Mercury remediation values for different land-use categories. (ART, 2013, SFC, 2017)

Country	Category	Mercury remediation value (mg/kg)
Belgium	Agriculture	2.9
Canada	Agriculture	6.6
Finland	Industrial	5
Finland	None Industrial	2
Germany	Agriculture	2
Switzerland	Children's Playgrounds, Allotments	2

2013) summarizes background levels in terms of mg/kg.

A total of 82 literature sources provided the basis for the mercury observation levels in ART (2013). Table 2 displays these concentration values, recorded for several land-use categories. The rather wide range of values results from different definitions of the term "observation level" in most of the countries, for instance a given value can trigger restriction of an intended land use or, at a minimum, the obligation to observe or investigate the on-site situation in more detail. The recommendation is to apply the risk analysis approach.

4.3 Mercury Levels Necessitating Remediation

Similar to the sites under observation, regarding remediation there are a wide ranges of definitions and measures to take if a given threshold is exceeded, *e.g.*, decontamination (removal of the contaminant), containment (prevention, neutralization or reduction of contaminant diffusion) or restrictions on land use. The latter can be an option if remediation costs exceed the locally bearable level (Table3).

Switzerland applies a strict definition of its levels for remediation. SFC (2017) defines the obligation to remediate if the given values overshoot the limits. In the related ordinance, the mercury level requiring remediation of allotments (community gardens) and children's playgrounds was lowered from a value of 5 mg/kg to 2 mg/kg in 2015. The Swiss level for remediation of lands for agricultural use and horticulture was deduced in ART (2013) resulting in a level for mercury remediation of 20 mg/kg.

5. Mercury Waste Management

The chain of sustainable management of mercury ends with treatment and final disposal as a waste. In the past Swiss industry with its strong focus and vast experience on chemical and pharmaceutical products, caused mercury contamination incidents which have been remediated or still await remediation.

5.1 Mercury Waste Treatment

Swiss waste management firms play an important and innovative role in the field of mercury waste treatment.

One such firm, founded in 1989 with a state-co-funded operation, is dedicated to thermal treatment of batteries. The cooperative (which today is a subsidiary of an international waste management company) became a global player in the field of mercury treatment. Their scope of activities has expanded over time, so today, treatment of catalysts and activated carbon, in addition to batteries and various types of mercury-containing products, is their core expertise. Subsequently, in accordance with the aims of the Minamata Convention, the company developed a treatment plant to convert metallic mercury into its sulfidic form (HgS, cinnabar). This process provides the next-to-last step for producing the aforementioned final storage quality of mercury. Excavated salt mines provide the ultimate step today through their final disposal services for mercury in its stable, naturally occurring form as mercury sulfide (Fig.4).

To treat waste derived from mercury-contaminated sites in the Swiss canton Valais, another treatment facility started operation in 2019. The thermal process reclaims mercury and enriches it for further processing. The remaining matrix shall reach mercury levels suitable for a standard landfilling operation or even utilization in construction works.

5.2 Recovery and Landfilling Operations

As stated in Swiss Environmental Report 2018 (SFC, 2018), 84% of all waste falls under the categories of excavation and demolition materials. Efforts are being made to increase recovery of this high volume waste stream. In the same logic as discussed above in Section 4 to reach qualities similar to the composition of the Earth's crust (or background levels), the provisions of the Waste Ordinance (SFC, 2015) include mercury thresholds to be considered as unpolluted or to restrict landfilling with thresholds according to Table 4. Demolition and excavation materials with mercury levels below 0.5 mg/

kg are considered unpolluted and therefore can be kept in the economic cycle continuously to fulfil the goals of the circular economy.

5.3 State of Techniques and Procedures

Management of mercury has improved even in the short period of time since the Minamata Convention went into effect. Its guidelines, as well as corresponding corporate innovation, have brought progress in fulfilling national provisions as well as the Convention's objectives. Swiss best-practice in that respect is strong cooperation between industrial associations, enforcement bodies (cantons) and governmental authorities. Swiss authorities have had very positive experience establishing a state-of-the-art technique working group involving the aforementioned stakeholders. It enables exchanges and quick action in response to new provisions and acute environmental issues, as well as to reported issues. The following elements have helped to establish best practice, specifically in the field of mercury:

- Transparent, well-documented sampling procedures (if needed by third parties)
- Complete mercury mass balances of treatment facilities
- Tracking of the value chain
- Verification of intended mercury uses if not brought to final disposal

Fig. 4 Final storage of drums in a excavated salt mine deposit.

6. Conclusions

A holistic approach and view on the management of mercury is helpful toward achieving the goals of the Minamata Convention. Considering the whole value chain of this metal and its substances and applying best practice for each of the steps is important.

Mercury thresholds provide clarity and transparency along the cycle from products to waste and final disposal. This survey shows that thresholds can be handled with a certain flexibility, recognizing the needs of local circumstances, *e.g.*, the mining industry. As a first priority, treated materials should reach a degree of quality that will enable them to be used in the economic cycle without damaging human health or the environment. Secondly, residues from treatment should be brought back into the geological cycle in a form which is stable long-term and the degradation of which does not create the need for remediation.

Table 4 Mercury threshold in the Waste Ordinance (SFC, 2015)

Annex 3 (ref. in Article 17)	Criterion for demolition and excavation material (unpolluted)	0.5 mg mercury/kg dry matter
Annex 3 (ref. in Article 17)	Criterion for demolition and excavation material (subject to further use in construction materials)	1 mg mercury/kg dry matter
Annex 4 (ref. in Article 24)	Criteria for waste, used as raw material in cement and concrete production 1) Use of waste as raw material and raw mix corrective in cement clinker production 2) Use of waste as alternative fuel in cement clinker production	1 mg mercury/kg dry matter 1 mg mercury/kg dry matter
Annex 5	Criteria for landfilled waste 1) Type B landfills (inert waste) 2) Type C landfills (solidified MSWI1fly ash) 3) Type D landfills (MSWI slag) 4) Type E landfills (other wastes, slightly reactive)	2 mg mercury/kg dry matter 0.01 mg mercury/liter dry matter (leaching) The total content of mercury may not exceed 5 mg/kg dry matter for metal-containing, inorganic and badly soluble waste 5 mg mercury/kg dry matter 5 mg mercury/kg dry matter

Reference measurement methods for total mercury concentration: For sample preparation, microwave pressure digestion with aqua regia or conc. HNO3 or HNO3/H2O2 with a 0.5 to 2 g sample of material; measurement by cold-vapour AAS, AAS, ICP-MS, AFS (ISO 12846).

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