Euro Chlor
representing the chlor-alkali industry

# MANAGEMENT OF MERCURY CONTAMINATED SITES

## Env. Prot. 15

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# Euro Chlor

Euro Chlor is the European federation that represents the producers of chlorine and its primary derivatives.

Euro Chlor is working to:

- improve awareness and understanding of the contribution that chlorine chemistry has made to the thousands of products, which have improved our health, nutrition, standard of living and quality of life;
- maintain open and timely dialogue with regulators, politicians, scientists, the media and other interested stakeholders in the debate on chlorine;
- ensure our industry contributes actively to any public, regulatory or scientific debate and provides balanced and objective science-based information to help answer questions about chlorine and its derivatives;
- promote the best safety, health and environmental practices in the manufacture, handling and use of chlor-alkali products in order to assist our members in achieving continuous improvements (*Responsible Care).*

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Prior to 1990, Euro Chlor's technical activities took place under the name BITC (Bureau International Technique du Chlore). References to BITC documents may be assumed to be to Euro Chlor documents.

# RESPONSIBLE CARE IN ACTION

Chlorine is essential in the chemical industry and consequently there is a need for chlorine to be produced, stored, transported and used. The chlorine industry has co-operated over many years to ensure the well-being of its employees, local communities and the wider environment. This document is one in a series which the European producers, acting through Euro Chlor, have drawn up to promote continuous improvement in the general standards of health, safety and the environment associated with chlorine manufacture in the spirit of *Responsible Care.*

The voluntary recommendations, techniques and standards presented in these documents are based on the experiences and best practices adopted by member companies of Euro Chlor at their date of issue. They can be taken into account in full or partly, whenever companies decide it individually, in the operation of existing processes and in the design of new installations. They are in no way intended as a substitute for the relevant national or international regulations which should be fully complied with.

It has been assumed in the preparation of these publications that the users will ensure that the contents are relevant to the application selected and are correctly applied by appropriately qualified and experienced people for whose guidance they have been prepared. The contents are based on the most authoritative information available at the time of writing and on good engineering, medical or technical practice but it is essential to take account of appropriate subsequent developments or legislation. As a result, the text may be modified in the future to incorporate evolution of these and other factors.

This document has been drawn up by the Environmental Protection Working Group to whom all suggestions concerning possible revision should be addressed through the offices of Euro Chlor.

## Summary of the Main Modifications in this version

| Section | Nature |
|---|---|
| All | General update based on recent developments |
| 1.2 and 1.3 | Addition of information on risk assessment and possible actions |
| 2.1 and 2.2 | Addition of more details on the site characterisation steps |
| 5. | Addition of a short chapter on containment techniques |

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# FOREWORD

This document aims to give a state of the art of the management of mercury contaminated sites, showing which items have to be developed and/or confirmed in the practice. As actions are in progress in Europe and in North America (see USEPA study in ref. 0), this document will be periodically updated based on gained expertise.

It must be emphasized that the purpose of this document is not the creation of a contaminated site management guide, but that of providing managers with a relevant and updated review of techniques and trends in site characterisation, risk assessment and subsequent site management, including monitoring and remediation. Thereby the reader will encounter a useful toolbox from which to select, with the aid of qualified environmental professionals, and attending to specific local, regional and national regulations, the techniques and technologies which best suit each individual site, and which will undoubtedly be modulated by socio-economic and political considerations.

*To protect workers' health, it is necessary to control exposure to mercury.* For more detailed information about workers protection in case of exposure to mercury, it is advised to consult Euro Chlor document ***HEALTH 2 - Code of Practice: Control of Worker Exposure to Mercury in the Chlor-Alkali Industry*** (ref. 1)***.***

# 1 BACKGROUND

In Europe, about 38% of chlorine was still produced by the mercury process at the beginning of 2009. At some stage in the future, and in agreement with the Euro Chlor commitment, mercury cell chlor-alkali plants will be decommissioned by 2020 at the latest.

Efficient preventative provisions are at the present time taken at every plant to prevent soil contamination from spills and leaks and waste landfilling is implemented in a safe and traceable manner. Nevertheless, this has not always been the case in the past and, in some cases, historical mercury contamination in the subsoil is present. Additionally to the production unit area, consideration should also be taken for possible old and badly recognised (and investigated) waste landfills that may have occurred on some sites.

As far as contamination through atmospheric deposition is concerned, previous studies (Ref. 2 and 3), have shown that the levels of contamination within site limits are often rather low (generally less than 10 ppm) and limited to the superficial topsoil (~30 cm) in the surroundings of the mercury cells. This pathway is usually not expected to impact the groundwater quality, but should be confirmed case by case.

Moreover, experience has shown that concentration of mercury in the top soil 500 meters downwind from the cell room is typically less than 300 ppb and is usually of no concern (ref. 4), but also in this case the local situation should be assessed.

This paper deals only with subsoil contamination resulting from spills, leaks, waste disposal and not with the indirect pathway of soil contamination through atmospheric deposition.

Management of mercury contaminated sites will have to satisfy three main driving forces, according to the sustainability concept:

- Protection of human health and environment
- Responsible Care programme of the chemical industry
- Economics: an economic evaluation of the different possible solutions has to be set up to select the most cost effective solution.

This document is a general framework for the management of mercury contaminated sites. It is based on the three following points in order to support the decision making.

## 1.1 Define the current situation and the future use/development of the site

Knowing the actual use and any planned future use is necessary in order to develop relevant scenarios to be taken into account when evaluating the risk

## 1.2 Assess the risk associated with the contamination

In general, risk assessment consists in the determination of the potential consequence of a situation, and the probability that these consequences could occur.

In the context of contaminated land, a contamination (source) may represent a danger (toxicity, radioactivity, pathogenicity…). For any receptor (human, environmental), the probability of exposure to the danger represents the risk. In the case of chemical contamination, there must be an exposure pathway (link from the substance to the receptor) in order for a risk to occur.

At chlor-alkali plants, this means identifying

- the source (mercury concentration, localisation/-depth …)
- the pathway (exposure through vapour phase, groundwater, surface water migration …)
- the receptors (workers, residents, ecosystem …)

## *1.3 Decide the actions to be taken in case of risk*

The risk management strives to break the link mentioned above by:

- source control (remediation), removing the source or reducing the danger (toxicity) associated with the contaminant (e.g. changing the speciation for metals)
- pathway control: barriers or cut off screens (capping, containment, immobilisation …)
- receptors control: restriction of use.

Monitoring allows verifying the objective of the actions taken is kept.

To our understanding a risk management/fit for purpose approach, as stated in the CLARINET-NICOLE statement, will give the best results in terms of risk reduction, environmental merit and financial impact (see http://www.nicole.org/publications/NICOLEjoint2.PDF). This approach was incorporated in the Commission proposal for the directive 2006/0086 (ref. 5) on contaminated soils management, but the Council decided to temporarily put in hold the decision process (progress report 1019/09 of June 2009).

The activities to be undertaken during these steps are site specific and dependent on such issues as pollution intensity and extent, local hydrogeology, presence of potentially threatened targets.

There are a range of existing tools which may be directly applied to mercury contaminated sites, others may need some adaptations and some may be not widely applicable at present. New techniques may need to be developed to ensure cost effective management.

Sharing resources, experiences and cooperative development of techniques would be the first step in setting up an efficient, cost effective management of mercury contaminated sites.

# 2 SITE CHARACTERISATION

The site characterisation is dealing also with the exposure scenarios and includes three major steps with the following objectives:

- *Desk study*: to identify, from the available data, all relevant potential source, pathway and receptor scenario for a specific site, and then using conceptual models and a preliminary hazard assessment to select the relevant scenarios and to eliminate the implausible ones.
- *Screening survey*: to assess the presence of contaminated areas using rapid and cost effective screening methods.

- *Confirmation survey:* when contaminated areas are identified, to quantify their extent and intensity.

In some cases, other data could be required (e.g., to perform a detailed risk assessment) and further characterisation may be necessary.

Due to the physico-chemical properties of metallic mercury, care must be taken when carrying out engineering work during the characterisation of the site to avoid the formation of preferential pathways or the mobilisation of contamination:

- Separated phase (mercury droplets) tend to sink down the profile during soil sampling (liquid state, hydrophobicity, superficial tension and high density)
- Metallic mercury droplets render the contamination highly heterogeneous at a very small scale, so mass balance is difficult to estimate
- Volatility of the metallic phase should not be overlooked (losses, health and safety issues).

Therefore standard tools for site characterisation may not be suited for mercury contamination. However, there are a number of technologies that have been employed with varying levels of success. For the three steps presented above, the following tables highlight what is available and what developments/adaptations are needed (shaded) to be applicable to mercury contamination.

## 2.1 Desk Study

| | Comments | Status |
|---|---|---|
| Existing protocols and good practice manuals | Special attention needed to sewer and buried pipes as potential secondary point sources, past waste management and maintenance practices | Applicable to Hg contaminated sites |

The mercury contaminations in soils and in groundwater of chlor-alkali plants can basically be synthesised as follows:

- The areas of the production unit, and especially those of the cell room and the retorting unit, can be contaminated to a maximum depth of a few metres with concentrations that can reach some thousands of ppm
- The areas close to the production zone can be contaminated to a maximum depth of about 50 cm, with some tens of ppm, due to mercury deposition from cell room air emissions; the size of the contaminated area depends strongly of the direction and speed of the prevailing wind
- The walls, floors and structures in concrete of the cell room, and their surface covering, can absorb mercury to a depth of a few centimetres at concentration of a few hundred of ppm

- Sludge contaminated with mercury can accumulate in open and underground sewers
- The metallic structures and tanks can absorb mercury in their superficial layer
- The groundwater can be contaminated by mercury that leached from the surface soil layers by percolating water.

The mercury is mostly present as metallic component ($Hg^0$), but the pH and Redox potential of the soil can also influence the chemical form of the mercury; oxidising conditions can stabilise the ionic forms ($Hg^{2+}$), while slightly reducing conditions can favour the transformation of ionic and organic mercury into its metallic form. This form can be converted biologically into the toxic alkylate forms, representing usually less than 1% of the total, but potentially relevant in some scenarios due to their volatility and solubility into water.

Another important factor to consider for the choice of the treatment process is the grain structure/dimensions of the contaminated material, as mercury tends to accumulate in the finer fraction parts (< 100 microns) and to strongly bind with humic substances.

## *2.2 Screening Survey*

The screening survey should consider the characterisation of the sites geology, hydrology and hydrogeology as well as the nature and distribution of any potential contamination.

Soil gas survey measures the vapour content in the unsaturated soil. It could be applied to detect metallic mercury. The method would not detect contamination by mercuric or mercurous salts, possibly overlooking area with presence of mobile mercuric ions that could present a risk for underlying groundwater.

| | Comments | Status |
|---|---|---|
| Soil gas survey | Potentially applicable to metallic mercury contamination (volatility of $Hg^{\circ}$)<br>On site measurement possible (rapidity)<br>Measurement possible over a significantly greater volume than with discrete soil sampling<br>Low cost | Validation needed for Hg (for mercuric soils and metallic mercury) |

Geophysical methods work by measuring contrast of the measured physical parameter (electrical resistivity, gravitational field, sound …).The sensitivity is limited by the size of the object. It works well for groundwater, geological layers and buried metals, but the sensitivity is too low for many environmental applications, in particular in the case of mercury contamination because of

- The relative low level of contamination (< 10 mg/kg in general)
- The high heterogeneity of the contamination
- The presence of interfering factor on the industrial sites such as buried cable, pipe and chloride co-contamination.

| | Comments | Status |
|---|---|---|
| Geophysical methods (electrical resistivity, electromagnetic, georadar and gravimetric methods) | Non-intrusive, global<br>Each tool has its own limits<br>Measurement possible over a significant greater volume than with discrete soil sampling<br>Need to use a number of techniques in conjunction with other more traditional methods | Established methods for geological characterisation<br>No application to detect Hg contamination published<br>On industrial sites, interferences with pipes cables, chloride co-contamination |

X-ray fluorescence is a rapid method for the determination of chemical elements in a matrix. In the portable format, it has been applied to screening solid matrix for heavy metals. The sensitivity of the method varies from metal to metal; for mercury it is rather low compared to the environmental standards. It can be used for the identification of hot spots. XRF scans a very small section of the sample, so it is inherently unsuitable for determining the average mercury content of raw coarse, heterogeneous material in the field. Such material must be homogenised (e.g. by grinding of a representative sample) before a useful quantitative measurement can be made.

| | Comments | Status |
|---|---|---|
| Portable X ray fluorescence | Rapid, low cost, on site measurement<br>Poor sensitivity (detection limit~50 ppm total Hg)<br>Useful for screening potential hot spots<br>Measurement possible over a significant greater volume than with discrete soil sampling | Commercially available for Hg<br>Used for total mercury |

Some screening techniques are available in the field but have not been validated for mercury contamination or, in their present state of development, show serious limitations like a lack of sensitivity with regards to the environmental standards.

## *2.3 Confirmation Survey*

It is necessary to confirm the results of the screening survey by existing classical methods.

| | Comments | Status |
|---|---|---|
| Sampling protocol and strategy | Existing norms and guidelines for soil and water (ref. 6)<br>Care should be taken to avoid creation of pathways for contamination | Sampling : adaptation needed for metallic Hg droplets in soil<br>No strategy for highly heterogeneous contamination |
| Hg total assay | Existing norms for soil and water (ref. 7, 8 and 9) | Available |
| Hg speciation | No established norm | Methyl-Hg : debate over the protocols in the scientific literature (ref. 10 and 11)<br>Other forms : work done in the scientific community (ref. 12) |

In conclusions, tools already exist to characterise a site but, due to the physico-chemical properties of mercury, specific tools may be needed to improve the reliability of the characterisation of mercury contaminated sites. In other words, the uncertainties have to be reduced, therefore reducing some of the financial impact.

# 3 SITE RISK ASSESSMENT

Risk assessment is carried out with the objective of assessing the risk posed by the mercury contaminated soil for receptors over time and space and for the specific land use. In some cases, the food chain and eco-toxicity must be taken into account.

If mercury contamination of groundwater is suspected, attention must be given to the possible toxic effects of abstracted groundwater and to conformance with the EQS (environment quality standards) if groundwater discharges into a controlled water body.

Current models (ref. 13) used for risk assessment give a picture of the risk over space at a specific point in time (now) and assume steady state. They generally use total concentration input data and assume fixed coefficients for real impact on the receptor (human or environment). Neither site specific speciation nor substance specific bioavailability data are taken into account in the models, although the bioavailable fraction of the contaminant in the soil is a central concept in risk assessment. It can be defined (ref. 14) as "… the fraction of a compound in a matrix that, when released from the matrix, can be absorbed by an organism. This absorbed compound is then available to cause a biological effect."

To our knowledge, no model can estimate the evolution of the risk over time. Risk evolution over time will be controlled by changes in the bioavailability and the mobility of mercury (speciation, precipitation, adsorption and volatility).

To establish a high degree of confidence in the results generated during the risk assessment, consideration should be given to the use of experienced professionals/academics for peer review. The basis of selection is also their awareness of such issues as European and local legislations.

| | Comments | Status |
|---|---|---|
| Risk calculation/ models | Models account for some speciation / bioavailability effect but not necessarily from site specific data<br>TDI values provided by WHO | Selection of existing models is country specific<br>Models are not Hg specific<br>Based on total concentrations, no prediction of the evolution of the risk over time |
| Bioavailability | Important parameter in risk assessment<br>Biosensor for Hg and methyl-Hg could be developed | Norms exists: Soil quality -- Requirements and guidance for the selection and application of methods for the assessment of bioavailability of contaminants in soil and soil materials - ISO 17402:2008 |
| Leaching test/mobility | Existing leaching test for waste<br>Soil and sediment are considered as sinks for heavy metals including Hg<br>Sediments are identified as a major compartment where speciation reactions which are significant for risk assessment occur | Norm under development for soil (ref. 15)<br>Significant amount of data for mobility evaluation in the literature : adsorption coefficients on soil, humic material, sediments for ionic Hg and methyl Hg (ref. 16 and 17) |

In conclusion, models for risk assessment are available, but their applicability to mercury contaminated sites must be critically reviewed in the light of the available data on toxicity, speciation and mobility of mercury. Sensitivity analysis should be considered when assessing the parameters used within the risk assessment.

# 4 SITE MANAGEMENT

Site management options will be defined according to the defined future use of the site and the results of the risk assessment. Options include:

- *Monitoring* with the objective of judging the risk from the contamination over time and space.
- *Remediation technologies* with the objective of limiting the risk at an acceptable level to the receptors for the intended use of the land.

## 4.1 Monitoring

### 4.1.1 Vapour monitoring

Consideration should be given to the end use of the site and if necessary vapour monitoring, to assess the likelihood of vapour emissions from contaminated ground into buildings.

### 4.1.2 Groundwater monitoring

The sampling and analytical protocols may be identical to those used in site characterisation but the sampling strategy and analysis protocols must be specific to the monitoring required for each specific receptor as defined by the risk assessment. The frequency of the monitoring and the detection limit requirements will depend on the amount of knowledge of the long-term behaviour of the contamination.

| | Comments | Status |
|---|---|---|
| Sampling strategy | General protocol existing | To be develop specifically for Hg<br>Work on going (ref. 18) |
| Analysis | General methods exist for total mercury | Soil Quality ISO 16772 – 2004<br>Water Quality ISO 17852 - 2006 |

In conclusion, general tools already exist and specific approaches for mercury are currently being further developed.

## 4.2 Remediation Techniques

Two types of remediation techniques can be considered:

- On site and off site treatments which can be applied after excavation of the contaminated soil
- In situ treatment which can be performed without excavation of the soil.

Excavation can be technically difficult for contamination present at some depth in the soil and/or in the presence of an aquifer. Therefore, in situ techniques must be preferred to treat subsoil contamination related to spills, leaks, and waste disposal.

| | Comments | Status |
|---|---|---|
| Thermal treatment | On site or off site treatment after excavation<br>Gaseous effluents to be treated<br>Classical high temperature (>800℃) or lower T° (<500) + partial vacuum<br>Applicable to building and demolition materials<br>High cost, site and country dependant<br>Possibility of recovering Hg°<br>Impacts on the mechanical properties of the ground, load bearing capacity<br>Impacts on underground services (gas, electric etc.)<br>May affect the leaching potential<br>Health/hygiene issues to be monitored seriously on and around mobile units | Pilot + applications for Hg contaminated sites (ref. 19 and 20)<br>Mobile units mentioned (in the U.S.) for thermal desorption of Hg[20] |
| Alternative thermal treatment | In situ treatment<br>Electrical heating/vapour extraction and treatment<br>In situ vitrification (>1500℃), technology developed for radionuclides contamination. Considerations are soil subsidence and vapour which must be collected and treated. | TerraTherm Environmental Service Inc. (electrical heating) (ref. 21)<br>Applied to subsoil pesticide and Hg contamination (ref. 22) |
| Physical treatment<br>Dry and wet classification | On site or off site treatment after excavation<br>The Hg contamination is associated with the fine fraction ⇒ reduction in volume and mass of the residue to be disposed or treated<br>Applicable to soil with low fine and organic matter content<br>Health and safety issue not to be overlooked during excavation and treatment (Hg vapours, contaminated dust)<br>High cost, residue left | Pilot and commercial operation for Hg contaminated sites (ref. 19 and 20) |

| | Comments | Status |
|---|---|---|
| Hydro-metallurgical extraction | On site or off site treatment after excavation<br>Extraction with chemicals | No pilot or full scale data for Hg |
| Electro remediation | Applicable in situ or on/off site after excavation<br>Need for an extracting solution (oxidant/complexing agent)<br>Little in situ data; the electro-osmotic flow may yield to a significant risk of uncontrolled migration of the leaching solution<br>Applicability in heterogeneous soil (layer of varying conductivity)?<br>Applicability on industrial site with lots of conductive material in the soil (pipes, wires)? | Pilot applications to metals (ref. 23)<br>Data for Hg in the literature (ref. 24) |
| Immobilisation inertisation | For excavated material and waste<br>Does not avoid the cost of disposal | Available for excavated material and waste containing Hg |
| | In situ application : precipitation as sulphur compounds of soluble mercury<br>Evaluation of the long-term behaviour of the immobilised form | One commercial application in 2008 on a former batteries production site |
| Chemical extraction | Difficult to apply due to the heterogeneous contamination | |

In conclusion, solutions exist for remediation of excavated materials (on/off site). They rely on dry and wet classification, on thermal desorption or a combination thereof. The major limitation to their use is their relatively high cost. Deep contamination requires in situ techniques but none are currently available for industrial application.

# 5 CONTAINMENT TECHNIQUES

Containment techniques apply equally to mercury contamination as to any contamination. The principle is to cut off the exposure pathways. Possible techniques include hydraulic containment (pump & treat), mechanical containment like horizontal capping or vertical low permeability barrier installed in the subsurface.

# 6 HEALTH AND SAFETY

For all the activities related to mercury and contaminated materials or soils; it is necessary to follow the guidance in the ***Env Prot 3 – Guideline for Decommissioning of Mercury Chlor-Alkali Plants*** publications (ref. 25) about health protection and safety, request the use of suitable clothes and monitor the mercury exposure of workers.

# 7 BREF FOR THE CHLOR-ALKALI MANUFACTURING INDUSTRY

This document published in December 2000 gives information for the treatment of mercury containing wastes. The general recommendation about the treatment is related with the mercury content of the waste, selecting distillation, when possible, for high concentrations, and landfills for the others.

This document contains a review of all the European legislation for mercury containing wastes at the moment of redaction (1999) but its update has started in 2009 and could be completed by end 2011.

# 8 SUMMARY

To manage mercury contaminated subsoils as a result of historical spills, leaks or waste disposal, the following steps must be considered:

- Inventory
- Site characterisation
- Exposure scenarios definition
- Targeted risk assessment
- Site management (monitoring, remediation, etc.)

The activities to be undertaken during these steps depend on the site specificities (pollution intensity and extent, hydrogeology, potentially threatened targets etc.).

There is a range of existing tools which may be applied, and some of them may need to be adapted to the specificity of mercury. An economic evaluation of the different possible solutions has to be set up due to the fact that some tools could be too expensive.

Regarding site characterisation, tools already exist but, due to the physico-chemical properties of mercury, specific tools are needed to improve the reliability of the characterisation of mercury contaminated sites.

For targeted risk assessment, models are available but their applicability to mercury contaminated sites must be critically reviewed in the light of the available data on toxicity, speciation and mobility of mercury.

For site monitoring, general tools already exist and specific approaches for mercury are currently being developed.

Regarding remediation, solutions exist for excavated materials. They rely on dry and wet classification, on thermal desorption or combination thereof. The major limitation to their use is their high cost. Deep contamination requires in situ techniques but none is currently available for industrial application.

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Some additional information is available on the two following websites:

http://www.nicole.org/

http://www.umweltbundesamt.at/en/clarinet

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