Environmental background assessment: basic principles and practice

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Summary. Background assessment is crucial in setting remediation and management goals, in identifying contamination trends and in providing screening tools for the selection of contaminants of concern. Legal requirements and guideline documents place great emphasis onto the establishment of background levels. This contribution discusses background estimates for metals and organics in relation to bioavailability and to its potential impact onto living systems. Methods for defining background in Environmental Risk Assessment are provided by the US EPA and by the Dutch Added Risk Approach. Cited case studies, taken from projects carried out in Italy and abroad, show that practical approaches vary significantly. A standardised database of soil, sediment and water background levels for different contaminants, as well as a more comprehensive guideline of background assessment, are urgently needed.

Key words: background concentration, reference site, ecological risk assessment, soil/sediment contamination, screening level, remediation target.

Riassunto *(Valutazione dei valori di fondo: principi di base e applicazioni pratiche)*. La valutazione dei valori di fondo è cruciale per la definizione di obiettivi di ripristino e di gestione, per la definizione delle tendenze in atto della contaminazione e per fornire strumenti per la selezione di contaminanti che costituiscono un potenziale pericolo. Requisiti normativi e linee guida attribuiscono grande importanza alla definizione di valori di fondo. Il presente contributo discute stime di valutazione di valori di fondo per metalli e per organici in relazione alla biodisponibilità e al potenziale impatto sui sistemi viventi. Metodi per la definizione dei valori di fondo sono forniti dalla USEPA (agenzia statunitense per la protezione dell'ambiente) e dall'approccio olandese basato sul rischio incrementale. I casi studio citati, tratti da progetti condotti in Italia ed all'estero, dimostrano che le soluzioni pratiche variano considerevolmente. Si sottolinea l'urgente necessità di costituire una banca dati sui valori di fondo di suoli, sedimenti e acque dolci per diversi contaminanti, accanto ad una linea guida più completa sulla valutazione dei valori di fondo.

Parole chiave: concentrazione di fondo, sito di riferimento, analisi di rischio ecologico, contaminazione di suoli/sedimenti, livello di *screening*, obiettivo di ripristino.

INTRODUCTION

Issues related to background are critical when conducting environmental risk assessment (ERA; *Figure 1*) in sites characterised by contaminated sediments or soils. Background levels have to be considered right from the start, while laying down the problem formulation phase, which establishes preliminary management goals. During the risk characterisation phase, which is designed to determine the extent of environmental risk, background is compared to risk-based reference values. After the site remediation phase, which constitutes the core of risk management, background allows the evaluation of the achievement of remediation targets through a comparison to pre-impact conditions.

Throughout the ERA process, background assessment provides a focus for risk communication, by setting a scenario against which management options are iteratively re-evaluated and re-discussed by the team of risk assessors, site managers, local authorities, stakeholders and the general public. A large portion of the technical discussions and political confrontations about the destiny of the site to be restored and managed will focus on: 1) what is the desired ecological status to be achieved; 2) which levels of contamination should be considered safe; and 3) how such ecological status and concentration levels relate to pre-impact conditions.

The following discussion focuses on the concentration of polluting substances, both organics and metals, as these parameters are highly relevant for the remediation of contaminated sites, including both terrestrial and aquatic ecosystems. It should not be overlooked, however, that background conditions typically need to be established also for habitat characteristics and for biological parameters, to provide

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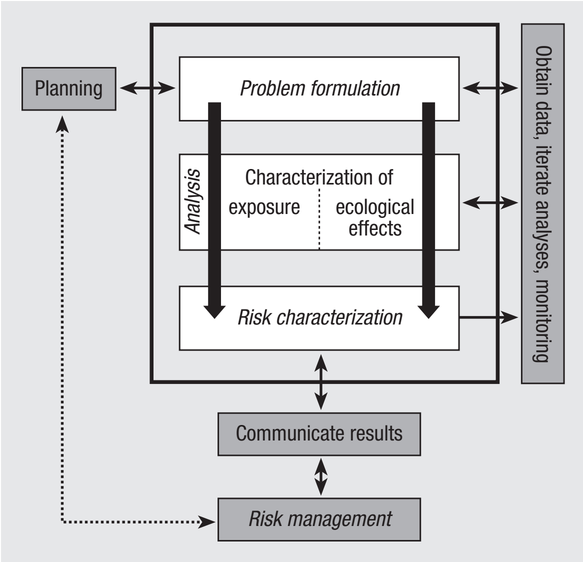

Fig. 1 | *Ecological risk assessment.*

an adequate characterisation of exposure and to ensure that remediation effectively translates into a restored ecological status.

THE ROLE OF NATURAL CONCENTRATION LEVELS

At low concentration levels, metals often play a significant positive role as essential constituents; their biological role is mainly to act as enzymatic catalysts in a number of energy-transfer systems. Among these, Mg and Fe are required in relatively plentiful amounts, while micronutrients such as Mn, Cu, Zn, B and Mo are required, but at low concentrations. A smaller number of organisms requires the presence of Cr, Co, Se and V due to highly specific physiological processes [1].

Even a theoretically pristine site, in the absence of human impact, is characterised by measurable amounts of metals and other substances which, beyond a minimal concentration, produce negative effects onto biological systems. Laboratory tests prove that seldom, natural concentrations result to be "poisonous" for a large number of organisms. However, even at very low concentrations, metals can cause negative impacts onto particularly sensitive organisms. When concentrations are high due to natural causes, such as the presence of igneous and highly mineralised rocks, their occurrence contributes to the selection of resistant varieties and species. In this way, metal concentrations have a direct impact on the natural distribution of populations and communities and contribute to regional biodiversity.

This consideration implies that the natural distribution of environmental concentrations needs to be taken into account during risk assessment. Critical threshold values, determined by means of ecotoxicological tests performed in the laboratory, cannot be directly transferred to the field without the due consideration of extant concentration levels. Remediation should aim at reducing the negative impacts caused by human activities, but it should not displace naturally occurring equilibria which modulate the distribution of populations and communities [2].

BACKGROUND DEFINITIONS

In a recent public consultation on ecological risk assessment [3], the United Kingdom Environment Agency proposed the following definition of *background concentration*:

"The concentration of an element or compound in an environmental compartment before significant anthropogenic additions. For practical purposes, it is taken to equal pre-industrial revolution concentrations".

According to the UK Environment Agency, the ideal background should be as close as possible to ambient concentrations which were common at the beginning of the 20th century. Stemming from pre-industrial concentrations, a "rural" and an "urban" background level can be defined, to provide reference conditions for the setting of remediation targets. In the United Kingdom, managers and regulators can rely on a substantial database of soil and water quality data, derived from early studies conducted in the turn of the XIX century, as well as from longterm monitoring programmes which have been able to follow and document the environmental history of single contaminants.

In The Netherlands, background values are defined on the basis of samples collected in rural sites devoid of contamination point sources. Aquatic background concentrations are established by means of mathematical models as it is considered that there are currently no sites that could adequately represent pre-industrial "aquatic" background values. Sediment background is derived from the aquatic background by means of partition coefficients. Soil background is established instead on the basis of "reference lines" produced by regression models relating contaminant concentrations in "pristine" soils to organic matter and clay content [2].

Following a similar principle, the Italian Marine Science and Technology Research Institute (Istituto Centrale per la Ricerca Scientifica e Tecnologica Applicata al Mare, ICRAM) proposed a set of regional reference background values for total metal concentrations in selected (no point sources) marine coastal sediments of the Tyrrhenian Sea. These reference values correspond to the upper limit of the 95% confidence interval of a regression between metal concentration and the percentage of the pelitic fraction (L. Tunesi, personal communication). For metallic species for which no positive correlation could be found, such as Cd and As, the background was defined as a concentration range corresponding to the average metal concentration in "clean" sites after subtracting the typical mean deviance (σ).

The approach taken in the United States distinguishes an entirely pristine background from an anthropogenic one. Taking into account the effect of diffuse contamination, the USEPA [4] defines background as “substances or locations that are not influenced by the releases from a site, and are usually described as naturally occurring or anthropogenic:

- naturally occurring: substances present in the environment in forms that have not been influenced by human activity;
- anthropogenic: natural and human-made substances present in the environment as a result of human activities”.

North America can rely on a number of pristine sites, with very little if any human interference; these can be handy in providing pre-industrial concentration data for a number of compounds. Such reference sites are much harder to find in Europe, where human settlements and activities are far more ancient and pervasive. Nevertheless, “pristine” sites are not devoid of the effects of diffuse anthropogenic contaminants commonly present in water, sediment and soil, which are able to reach even the most remote areas. Hence the need to define an anthropogenic background level: a concentration above the natural background which is independent from point sources.

On a similar note, the European Commission Joint Research Centre defines two background components [5]:

- natural background concentration: the concentration that is present due to natural causes only;
- ambient background concentration: the concentration that is present due to natural background plus the concentration due to contaminant release derived from diffuse sources of human origin.

BACKGROUND AT DIFFERENT SCALES

Considerations of scale impose the choice of an appropriate specific strategy in establishing a background reference. Background levels defined on the basis of regional, local and unit-specific concentration values have each their own advantages and disadvantages. In the US, regional and federal background concentrations are published and routinely updated by local and federal environmental protection authorities; such reference values are useful during desk studies and for setting preliminary risk estimates. Suter *et al.* [6] recommend the consideration of conservative reference values derived from regional background data which can be substituted (eventually the reference concentration can be increased) by locally determined backgrounds, if the ERA process warrants approaching a site-specific level of analysis. Factors that need to be taken into account to assess the relevance of regional background values to local situations include: sample homogeneity, knowledge of matrix characteristics to assess potential bioavailability and the local and regional variance of the parameter concerned.

The Ministry of Land, Water and Air Protection of the Government of British Columbia [7] publishes 7 regional background soil quality estimates (+1 for the Vancouver area) for 17 inorganic elements. Each background estimate is derived from the 95th percentile of the distribution of the concentrations obtained in a reference site chosen to represent the background for a given region. The values vary according to the mineralogy characterising different regions and to the presence of diffuse pollutants. For example, the mineralogy explains that the total Cr reference value is 150 µg/g in Region 3 “Southern Interior” against 100 µg/g in Greater Vancouver and 50 µg/g in Kootenay; on the other hand, the regional Pb reference concentration ranges from 15 µg/g to 300 µg/g in Greater Vancouver were road traffic is far heavier.

The European Commission took into consideration the definition of a background at a European scale, on the basis of available concentration databases. The Forum of European geological surveys (FOREGS) launched a Geochemical Baseline Mapping Programme with the aim of identifying changes in the levels of chemical elements on the Earth’s surface and providing a baseline for assessing the extent and consequences of their detrimental impact on the health of humans and on the environment [8]. A database was compiled, including soil, water and sediment samples, randomly selected on the basis of a 160x160 km grid, resulting in 807 selected stream water samples spread over Europe. This database, together with data provided by the OSPAR Commission for the Protection of the Marine Environment in the North West Atlantic and the HELCOM Baltic Marine Environment Protection Commission, gave support to the European Commission’s Expert Advisory Forum on background concentrations in proposing default background concentration ranges for dissolved and particulate metal phases in inland, transitional and coastal European waters. The reference values are based on the 90th percentile of the data distribution in the FOREGS baseline study. Each Member State is then requested to adopt the default value provided by the FOREGS study, or select a local background concentration applying a suitable methodology (sampling of local selected pristine sites, geochemical modelling, expert judgment) that should be notified to the Commission and described in detail, to reach consensus at the river basin scale.

Geochemical databases defined at a finer scale should be employed when available to establish regional baseline concentrations. This is the case of Piedmont and Emilia-Romagna regions in northern Italy, where monitoring networks have been set up at a 4 km x 4 km and 18 km x 18 km scale, respectively; these networks include data for both metals and organic contaminants [9]. At the national level, general information on the distribution of inorganic elements in soil/sediment, water and air was published by Italian Environment Agency [10].

THE LOST PARADISE

Very few truly pristine areas on the globe can be said to be devoid of significant human impact. Diffuse pollution and climate change caused by industrial, residential and transport emissions ensure that even wilderness frontiers, such as the few remaining primary forests and the sub-tropical and polar deserts, are affected by a number of anthropogenic contaminants and suffer from changes in their matter and energy flows. Diffuse contamination typically occurs in modest quantities and its effect on matrix properties, and on the bioavailability of matrix constituents, can be considered minor. Despite this, the concomitant impact of diffuse contamination and climate change can lead to large scale ecological effects which eventually bear a durable impact on the chemical composition of aquatic ecosystems and soils [11]. On land, diffuse pollution can be transferred through the hydrological network, from upper to lower sections of a river basin, or can migrate from one basin to the next by deep groundwater movement. In the sea, contaminant loads migrate along the coastline, in the direction of prevailing currents. On land, hydrological transport routes are considered to be fairly localised but have a quantitatively significant local impact. On the other hand, pollutants generated by vehicle, ship and airplane traffic represent lower concentrations, but far more diffuse loads, and can become distributed over great distances.

A large number of diffuse contaminants is distributed by means of atmospheric deposition. This concerns volatile as well as substances associated to air-borne particles; on a global scale this kind of contamination is steadily rising. Volatile organic contaminants (VOC) of common concern include: dioxins, dioxin-like furans, polychlorobyphenyls, and radionuclides such as ^{137}Cs, polycyclic aromatic hydrocarbons, DDT-DDE, $BrCH_3$; to these, we should add air-borne metals, attached to fine particles such as Hg from coal and waste incineration. Among organics, the number and concentration of xenobiotic compounds is under continuous change; while new compounds become part of the economic cycle, some others become regulated by government authorities and can be taken out of production through bans. Their concentration eventually decreases according to their specific degradation rate, migration route and migration velocity in the environment. To set a reasonable background level of a diffuse contaminant (following the definition of *ambient background concentration*), contaminant characteristics and history have to be well understood.

By definition, the natural background concentration for a xenobiotic compound could be assumed to be equal to zero; *i.e.* xenobiotic substances are not present in nature. However, the application of a natural background reference value equal to zero is of no practical relevance. Instead, the definition of an *ambient anthropogenic background concentration* is highly relevant to distinguish the portion of the pollution load detected at a given site which can be related to on-site activities from the portion which is due to diffuse pollution.

PRACTICAL BACKGROUND CONSIDERATION

Despite extensive anthropogenic impact and the worldwide distribution of diffuse contaminants, samples for establishing background levels of a number of parameters can, and indeed should, be collected in most cases. The selection of sampling sites and the interpretation of background concentrations require a specific strategy depending upon the behaviour of the contaminant and the nature of the matrix investigated. A specific location can represent an appropriate reference condition for a given case-study, although often it will not be considered "pristine" in absolute terms, implying that it will not be able to provide appropriate background values for all substances. In some cases, complex background sampling strategies may be required, including the selection of different locations for different parameters.

Selecting a reference site

An ideal reference site is characterised by the same distribution of contaminant concentrations that would have been observed in the impacted site if it had not been affected by anthropogenic contamination [4]. Apart for the contaminant concentration, samples from the background site should be perfectly comparable to those collected within the contaminated site. Both sets of samples should consist of the same matrix and mineralogical properties, to ensure that there is no difference in the bioavailability of relevant constituents which could interfere with the effects of contamination.

Sites characterised by local contamination may find a match with appropriate reference sites situated within the same river basin, upstream from the impacted area or in an adjacent river basin sharing common climatic conditions, geological origin and substrate type. Depending upon the characteristics of the chemical of concern identified during the preliminary problem formulation and scoping phases, an appropriate distance between the two sites will be chosen to avoid any chance of direct contaminant transport from the impacted site to the reference one. Selection of a reference site implies an in-depth knowledge of the history of anthropogenic impact in the surrounding area. In many cases, reference sites will be selected by mapping transport routes and local human activities. Useful information can be provided by local government authorities.

Operational guidelines

A stepwise approach for the collection of background data, on the lines of the one proposed by the USEPA [4], is provided in the following:

1) map the catchment surrounding the contaminated site and divide it into sub-units, separating upstream and downstream sites;

2) map potential sources of contamination;
3) collect background data that may be already available;
4) select data points which refer to locations which are sufficiently distant from point sources;
5) screen data for quality;
6) analyse available data and decide whether they are sufficient to perform meaningful statistical tests;
7) finally decide whether a new sampling campaign is needed to generate more background data.

Guidelines published by the Ministry of Water, Land and Air Protection of British Columbia [7] stress the criteria leading to a correct choice of a reference site. These include:

- geographical characteristics (location, topography, size/area, etc.);
- soil physical/chemical characteristics (soil maps, Geological Survey of Canada information, etc.);
- hydrology;
- soil sampling depth.

These last guidelines propose a stepwise contaminant screening approach of increasing refinement, starting from the consideration of toxicologically-based soil quality standards adjusted to provincial or lowest mainland background soil estimates (stage 1), than a comparison to regional reference background concentrations (stage 2) and finally the definition of a local background specific to the investigated site (stage 3). By means of a step by step procedure, a given site may be released as a contaminated site based on risk-based criteria, on a comparison with the regional background or on consideration of a locally assessed background concentration. The British Columbia administration does not recommend the definition of specific background levels for wide area sites which are large and complex, as their monitoring is expected to be prohibitively expensive.

Further recommendations can be found in a general overview of existing background assessment guidelines published by an Italian consortium of environmental protection agencies [12].

The upstream reference site

From many perspectives, river basins can be considered as distinctive ecoregional entities, sharing a relative homogeneity. Reference areas located upstream from impacted sites often constitute the first choice of risk assessors. They are within the same river basin and therefore share a common climate, a similar geological origin, and belong to the same biogeographical unit. On the other hand, upstream sites tend to be characterised by lower temperatures, higher precipitation, greater erosion and a soil composition which differs from the one characterising sites situated in lower floodplains. Exposure to greater erosional forces ensures that upper catchment locations have thinner soils, with a greater proportion of primary minerals while lower sites have deeper soils, rich in secondary minerals (*e.g.* clays) and products of rock denudation (*e.g.* precipitated carbonates). Such differences affect physical soil properties and the chemical environment which determines the bioavailability of chemical constituents of concern. The ACNA case study (see below) indicates that even within the same river basin, the natural distribution of background constituents can be affected by significant heterogeneity due to strong natural gradients.

ACNA case study: background arsenic in the upper Bormida river (Italy)

The Bormida is a second order stream, running for some 80 km before reaching the Po River, a 87 × 103 km^2 large basin, in northern Italy. A large-scale chemical factory started operating in the upper Bormida in the turn of the 19th century. The ACNA (Azienda Coloranti Nazionali ed Affini) produced more than 370 chemically active substances as ingredients of paints, pigments, pesticides and pharmaceuticals. Due to extensive contamination, the factory had to cease operation in the 1990s, and the site is currently undergoing remediation. Extensive sampling conducted during the risk characterisation phase revealed that high As concentrations were associated to the presence of organic substances typical of ACNA production which were used as tracers to detect the extent of the contamination caused by the industrial plant [13]. Consequently, As was included to the list of contaminants of concern considered in the remediation plan. To find suitable background levels, site managers selected 22 upstream reference areas devoid of obvious signs of anthropogenic contamination. Downstream areas were not considered as it was assumed that these areas would have been contaminated by hydrologic transport; the industrial site was built right within the Bormida riparian area. In reality, to the surprise of most, many of the upstream sites revealed higher As contamination than downstream ones, although these last ones were characterised by the presence of synthetic contaminants which clearly indicated impact due to ACNA effluents.Further studies revealed that within the upper Bormida Valley, surface As concentrations tend to naturally decrease along the slope due to the presence of a natural gradient. Mountain tops are characterised by a higher proportion of intrusive rocks and by a highly mineralised substrate, which becomes gradually submerged and mixed with alluvial deposits as the river proceeds downstream. Due to its geological origin, As concentrations in the upper Bormida are in excess of standard limits imposed by Italian national legislation. As a compromise solution, regulatory authorities signed an agreement with the site managers to set an indicative remediation target of 50 mg As/kg soil, despite the fact that such concentration is known to be well above generic no-effect levels. The distribution of As concentrations in reference areas located within the upstream portion of the upper Bormida Valley would have indicated a remediation target of 85.8 mg/kg.

Checking the soil/sediment profile

Whether a site is pristine or not may be checked by inspecting a sediment profile as a reflection of the history of the area (see Porto Marghera case study).

Metal background determination for Porto Marghera (Venice), Italy

Soil background concentrations were established during the remediation of Porto Marghera, one of the largest contaminated sites of national concern, situated within the immediate mainland surrounding the area of Venice in Italy [17]. This investigation was performed on realisation that some potentially harmful metals, such as As and Sn, were naturally present within the region at levels higher than the national concentration limits set by Ministerial Decree n. 471 of 1999, regulating contaminated site management. Sampling and data analysis were conducted following the ISO 19258 guidance on the determination of background values [18]. Twenty-seven sampling points were selected in an area adjacent to the contaminated site according to the following criteria: a) *wind direction* to prevent preferential atmospheric deposition; b) *soil layer structure* to distinguish intact profiles from disturbed soil layer sequences; c) *site history* to gain information about the past human activity on site; d) discarding the top 15 cm layer to reduce the influence of atmospheric deposition on soil mineralogical properties. Two samples, relative to the first two recognisable soil layers, were retained after extracting 120 cm long cores. Cores revealing an anomalous pedogenetic sequence or the presence of residues of anthropogenic origin were discarded. Values corresponding to the 90th percentile were chosen as background reference after a subjective assessment of the position of an inflection point in the quantile plot of each data distribution. The 90th percentile of the background concentration of Be and Zn coincided with the regulatory limit value, leaving several data points above the line; with As and Sn even the average was significantly higher, leaving the great majority of the data points above the limit. Contrary to expectations, no significant difference could be found between samples belonging to: 1) untouched profiles, 2) disturbed profiles and 3) agricultural plots. Comparing the concentration profile down the sediment core, it was noticed that while all other contaminants tend to decrease with depth (especially those characterised by significant atmospheric deposition such as Hg), the inverse is true for As, which appears to be "diluted" at the surface by illuviation and/or by additional soil/sediment deposition. Further research eventually confirmed that As has a natural pedogenetic origin, calling for a background level higher than the national regulatory limit.

Constant concentration with increasing depth is indicative of a pristine site; instead, changes in the sediment enrichment factor, defined by the ratio between modern and historical sediment concentrations, is indicative of a perturbed catchment. Element ratios found in stable deep sediments can be used to derive reference concentrations for water, sediment, soil and peat. Historical concentrations in the clay fraction of soils and river sediment can be derived by considering their normalisation to conservative inert elements such as total Al [14, 15], Si, Se, Li, Ti, as well as by analysing ancient sediment layers whose age is assessed by means of Pb dating techniques [16].

When ideal reference sites are lacking, risk assessors may decide to derive surrogate soil/sediment background values produced by geochemical modelling and by a recalculation of former concentrations based on the assumption of leaching, weathering and diagenetic process rates. Deeper groundwater can provide a reliable natural background, unless significantly disturbed by percolation. Sediment/water partitioning coefficients derived by pore water monitoring combined with experimental work can be applied to groundwater samples reflecting pristine conditions to produce estimated sediment background reference concentrations.

In some cases, no reasonable background level can be established forcing authorities to take into consideration all contaminants of concern for a full ERA process (see Anacostia river case study). Using risk-based threshold values not backed by background data widens the task of the ERA, increases cost, and exposes the overall assessment to the risk of wrongly indicating a target concentration which is actually too low to be realistically achieved.

The Anacostia river risk assessment study (District of Columbia, USA)

The lack of site-specific background concentration data may imply the need to consider the potential toxicity of a large number of compounds during initial screening assessment, as none can be singled out by comparing their concentration to background. In the tidal Anacostia River Watershed case study [19] no site-specific background data could be defined for the chemicals of concern selected during human health risk assessment (HHRA). All substances which exceeded risk-based concentration thresholds (RBC) set by regional USEPA authorities on the basis of chronic cancer risk, or applicable relevant and appropriate requirements (ARAR), defined according to a set of local, national and regional threshold values, were included among the contaminants of potential concern. The RBCs and ARARs were selected to be consistent with an industrial exposure scenario. When more than one screening value was provided for a given contaminant, the lowest was used. This process required the consultation of a number of databases for the selection of generic conservative thresholds. In contrast to HHRA, during ERA a more robust assessment is required to provide less uncertain concentration limits which can be demonstrably associated to potential damage to key elements of the ecosystem. Extant concentrations in water and sediment were compared to threshold effect levels (TEL) derived from the geometric mean of the lower 15th percentile concentration of the effects data and the 50th percentile of the no-effect data derived from freshwater exposure experiments with the amphipod *Hyalella azteca*. Regional background values provided by the National Oceanic and Atmospheric Administration [20] were used for trace elements for which TELs were not available. The greatest elements of uncertainty concerned the bioavailability of contaminants in river sediments, relative to their speciation, to the presence of acid volatile sulfides and of organic carbon.

DATA QUALITY REQUIREMENTS

Once an optimal reference site and an adequate sampling techniques are chosen, there is need to establish a strategy for determining representative statistical parameters and distributions. Performing data quality assessment [21] requires that parameters and distributions be of a sufficient quality to allow an adequate comparison between the background and the studied site. Typical difficulties are due to:

- the high variance which characterises concentrations in contaminated sites;
- the low number of data points sampled in the background site;
- the poor statistical rigour associated with parameters such as the variance, the mean, the median and other percentiles describing the distribution of background data;
- the poor observance of standard statistical requirements such as the normality of the distribution and the independence of the measured variables;
- incomplete datasets due to a large number of non-detects among background samples.

Such difficulties are often approached by creating inferred data distributions by means of modelling and by bootstrapping techniques. The comparison of data distributions by means of graphical plots is highly recommended to integrate the whole range of data values rather than few poorly representative synthetic parameters. The USEPA (2002) guideline [4] describes in detail a series of statistical requirements and tests specifically devoted to the analysis of background data.

One further issue is represented by pseudoreplication. As stressed by Suter *et al.*, comparing samples taken from a given contaminated study site to samples taken from a single selected reference site is not statistically rigorous in terms of replication [6]. Fundamental statistical requirements would recommend a randomised choice of samples taken from a number of reference sites. In practice, this is impossible. The pseudoreplication paradox underlines the need to mediate between common sense and rigorous statistical data treatment when performing background assessment. At the same time it is a warning: the choice of a reference site needs to be testable and transparent. Understanding the geochemical history and the environmental fate of contaminants may be crucial to be able to properly assess the validity of selected reference sites and values.

When background is "too low"

Both oligoelements and diffuse pollutants are detected at low concentrations. Not infrequently in both cases, extant concentrations are inferior to detection limits characterising sampling and analytical instruments (see the Campania Region case study).

<u>Organic contaminants in Campania Region (Italy)</u>

A recent monitoring campaign in Campania Region highlighted the presence of dioxins (PCDD), furans (PCDFs) and dioxin-like polychlorobiphenyls (PCB_{dl}) in soils, freshwater and coastal marine sediments, as well as in a number of dairy products and meat [22]. The presence of these contaminants was characterised by a diffuse concentration across the region and few hotspots, mostly around urban areas. Riverine sediments were sampled mainly along the course of the Sele, the Volturno and the Regi Lagni basins, including the river mouths. The concentrations found in sediments were generally coherent with the presence of significant point sources within their basin, in correspondence of contaminated soils, apart from a few cases were direct contaminant release into the river is suspected. Significantly lower concentrations were found at river mouths apart from the Sarno and Garigliano deltas, which exhibited concentration hotspots. Soil gradient analysis revealed the existence within the region of a less affected mountainous area which could provide indication of a reference background concentration associated with atmospheric deposition. In this area, most congeners resulted below mean detection concentration. Adequate data distributions for the estimation of a background value were limited to heavier, more persistent contaminants such as Octachlorodioxin (OCDD), Octachlorfuran (OCDF) and a couple of hepta-congeners, while in all other cases background values were assumed to be ½ of the median detection concentration. In this area, soils and sediments are low in organic carbon and the relationship between organic contaminant concentrations and total organic carbon, due to the well-known behaviour of lipophilous organic contaminants, here showed no consistent pattern. The ratio contaminant concentration/Total Organic Carbon was highest in riverine sediments and comparable to the one characterising soils belonging to the most densely urbanised area.

In practice, in such cases, background values may become equivalent to instrumental detection limits or are derived from surrogate data distributions generated by attributing a standard value to samples found to be under the limit of detection. The guidelines published by the Ministry of Land, Water and Air Protection of the Government of British Columbia [7] propose the use of estimates based on one half of the mean detection concentration.

When detection limits influence the value of the reference background, mean detection concentrations encompassing the instrumental detection limit as well as the potential error associated to overall monitoring procedures should be compared to risk-based thresholds, to determine whether these limits need to be challenged by seeking more sensitive techniques of analysis and monitoring.

THE ADDED RISK APPROACH (ARA)

Proposed by the Dutch Institute of Public Health and Environment (RIVM), the added risk approach (ARA) describes a conceptual scenario which facilitates the implementation of regulatory measures in the remediation of contaminated sites by integrating laboratory-derived effect concentrations and fieldwork-derived background values. The ARA has been endorsed by European Commission expert groups trying to define background reference levels for river-basin districts to assist the implementation of Water Framework Directive 2000/60/CE and has inspired the recommended policy for deriving "probable no-effect concentration" levels for hydrogen peroxide, chromates and zinc [3].

The ARA principle

The need to establish a specific methodology stems from the realization that concentrations measured in the field are characterised by a significantly lower bioavailability than those obtained during laboratory tests, which are typically bioavailable up to 100%. The ARA offers an operational solution designed to integrate background values in the calculation of concentration screening levels, to avoid that screening values determined by laboratory tests may end up being significantly lower than background. In the case of metals, such eventuality is common due to the significant difference in bioavailability between field and laboratory conditions. By disregarding the bioavailable portion of the natural background, regulators tend to impose overly conservative remediation targets, which lead to unrealistic risk assessments.

The concentration of a substance, detected at a given site, may be described as the sum of several compartments, divided into two major categories: background and anthropogenic contamination. According to the RIVM, the background includes:

- a quantity which is not able to exert toxic effects, as most of it is not bioavailable and strongly retained by the matrix;
- a quantity which is biologically active and which may interact with the ecosystem, with both positive (micronutrient) and negative (toxicity) effects.

Concentrations due to anthropogenic impact are portrayed as being added on top of the natural background. Remediation goals cannot be set below the natural background value, despite its potential activity onto biological systems. Only the portion of the contaminant concentration which is "added" due to human activities should be regulated and should warrant remediation initiatives. By precaution, its status is considered fully bioavailable and its impact is assumed to exert a significant effect on biological systems. The overall impact of anthropogenic additions is tolerated by the ecosystem up to a certain extent, beyond which remediation is warranted.

The ARA model

The bioavailability of substances present in the background is a site-specific factor. Four scenarios can be envisaged:

1) the background concentration of a given element is entirely inert and it engenders no exchange with the biotic community. The toxicity of bioavailable phases is irrelevant to the field situation;
2) the background level of the substance is zero. Extant concentrations are entirely due to contamination and for conservative reasons are assumed as entirely bioavailable. Screening levels can be derived directly from laboratory test data and compared to concentrations monitored in the field;
3) extant concentration levels can be represented as the sum of an inactive natural background and an added anthropogenic component, totally bioavailable. Low observed effect levels (LOEL) derived from chronic laboratory tests should be compared with the sole anthropogenic component, after establishing a conservative background value;
4) the most frequent and the most complex case. Background levels include:
   a. an inert quantity closely bound to the mineral lattice, to organic matter or to other substances which are not bioavailable under extant conditions;
   b. a biologically active component which interacts with the ecosystem causing both positive and negative effects.

The conceptual scheme shown in *Figure 2* illustrates why concentration limits derived from laboratory tests cannot be compared with concentrations from the field, as the observed effects cannot be attributed to the total concentration, but to the sum of the bioavailable portion of the background and the added anthropogenic component. The biological effect of the background concentration must be estimated to avoid the setting of over-conservative concentration limits, which would then cause the establishment of unrealistic remediation targets.

The ARA model provides a schematic simplification of the relationships between background and anthropogenic additions which stresses the need to correctly interpret the outcome of laboratory tests next to a realistic assessment of exposure to contaminants in the field. By combining no-observed effect-concentration (NOEC) values relative to at least four species belonging to different phyletic groups into a cumulative species sensitivity distribution (SSD) [23], the ecophysiological response of the ecosystem can be estimated and expressed as the maximum permissible concentration (MPC), *i.e.* the highest concentration which may be reached before significantly displacing

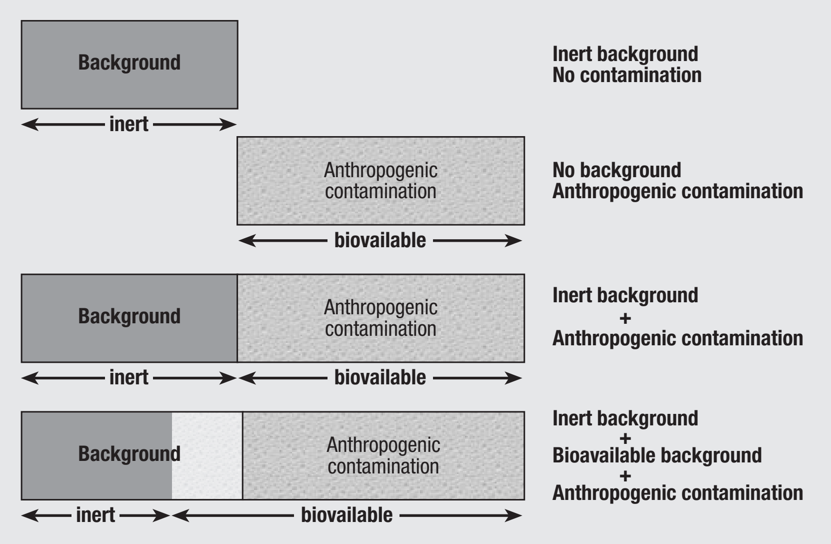

Fig. 2 | *The added risk approach (ARA) model.*

natural equilibria. A maximum permissible addition (MPA) can then be defined, representing the anthropogenic addition which is produced to reach the MPC; only beyond this point should be remediation warranted. The assessment of background bioavailability may be crucial for establishing the potential impact associated to anthropogenic addition.

CONCLUSIONS

Establishing a background concentration is a prerequisite for judging the evolutionary trend of a given substance in the environment and for setting realistic remediation targets. Both the steady natural background, which does not take into account the presence of diffuse anthropogenic sources, and the anthropogenic background, which varies with the environmental history of each contaminant, have a reason to be considered. For many organic contaminants, both background levels may vary due to the progressive refinement of technical instruments which tend to increasingly reduce the limit of detection.

Background reference values assessed from a selection and an elaboration of field data need to be compared and contrasted to risk-based values such as allowable concentration limits, environmental objectives and remediation targets, which are instead derived from laboratory experiments and field-based exposure assessment. Screening against background is safer and it prevents having to elaborate risk-based thresholds through experiments. Background reference levels can explain the natural distribution of species, constitute a relevant part of the characterisation of exposure and allow the monitoring of contamination trend-reversal policies.

The ARA provides an operational framework for developing practical tools. It has the merit of illustrating a theoretical basis for a reasoned interpretation and use of laboratory toxicity data, highlighting bioavailability issues. Its implementation requires a standardisation of the integration of laboratory data as well as of the estimation of background bioavailability. The method proposed remains open to criticism: 1) SSDs define a concentration without analysing the effective contribution of the selected species to ecological sustainability; 2) lab-derived NOECs are not random values of species sensitivity and therefore their cumulative statistical treatment is not as robust as would be required; 3) the ecophysiology of species bred in the laboratory may not accurately reflect the sensitivity of populations in the wild; 4) the applicability of the SSD method to complex ecosystems and highly inhomogeneous matrices needs to be assessed.

The ERA framework, in its current formulation, lacks a well developed strategy of Environmental Background Assessment. In the Mediterranean region, major impediments for developing an operational methodology of background assessment are due to the lack of an appropriate soil/sediment database, standardised at the national scale. As illustrated by several case studies, practical attempts to tackle the background issue are varied.

In Italy, current legislation dealing with the remediation of soils, sediments and waterbodies (DL.vo 152/2006), places much responsibility on Environmental Background Assessment. This calls for a strategy requiring:

- standardised statistical testing methods;
- bioavailability assessment protocols;
- a regulatory framework sensitive to issues related to background and bioavailability.

Regulators could benefit from the introduction of simple standardised bioavailability tests, such as $CaCl_2$ elution and/or the acute Microtox test using the luminescent bacteria *Vibrio fischeri*, applicable to a wide range of physical matrices.

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