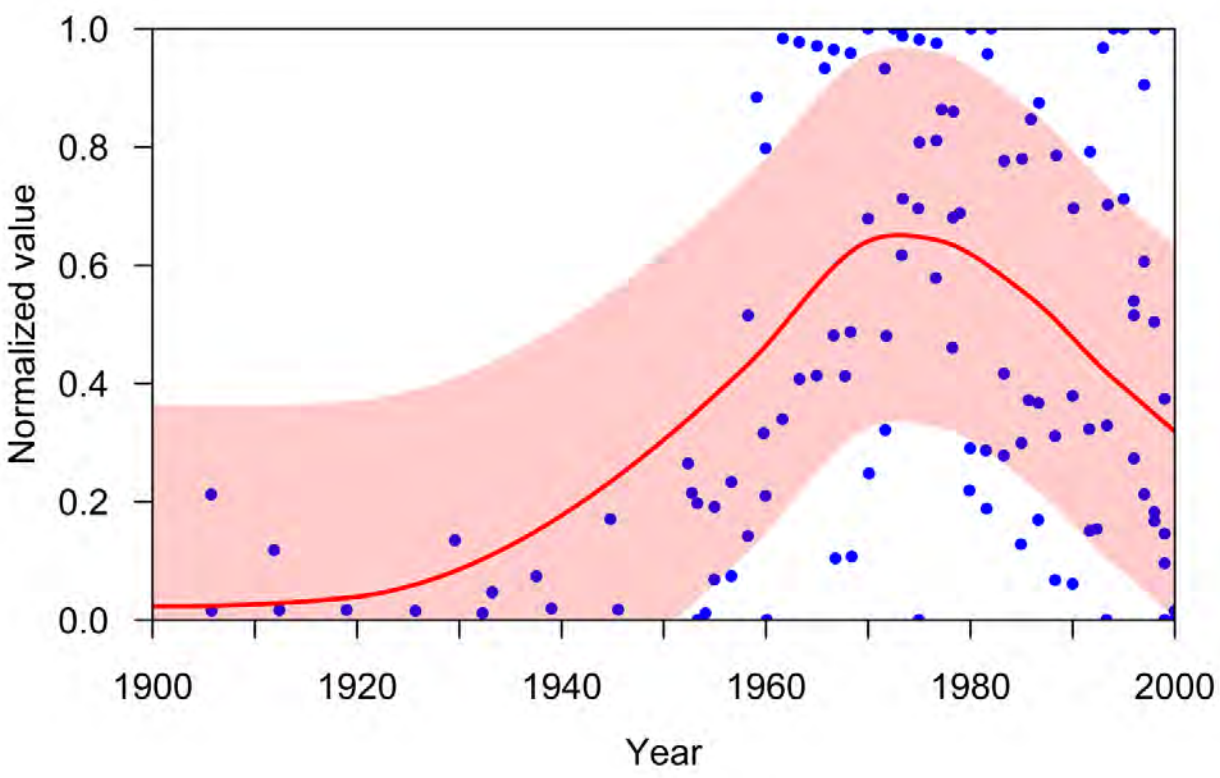
106

REPORTS AND STUDIES

GESAMP
Group of Experts on the Scientific Aspects of Marine Environmental Protection

GLOBAL POLLUTION TRENDS: COASTAL ECOSYSTEM ASSESSMENT FOR THE PAST CENTURY

GESAMP WORKING GROUP 39



IMO FAO UNESCO IOC WMO UNIDO IAEA UN UNDP ISA UN environment

Published by the
INTERNATIONAL ATOMIC ENERGY AGENCY
Vienna International Centre
PO Box 100
1400 Vienna, Austria

Printed by
Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
fax: +43 1 26007 22529, tel.: +43 1 2600 22417
email: sales.publications@iaea.org www.iaea.org/publications

ISSN 1020-4873

Cover image: Mercury in LME23 (Baltic Sea)
Joan-Albert Sanchez-Cabeza

Notes:

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ISSN 1020-4873 (GESAMP Reports & Studies Series)

For bibliographic purposes this document should be cited as:

GESAMP (2018). “Global Trends in Pollution of Coastal Ecosystems”, (Ruiz-Fernández A.C. and Sanchez-Cabeza J.A., eds) (IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP/ISA Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection). Rep. Stud. GESAMP No. 106, 101 p.

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EXECUTIVE SUMMARY

The world population has risen dramatically over the last century, as has humanity's influence on the natural environment. Worldwide urbanization, agriculture and industrialization cause a myriad of impacts on our ecosystems. Coastal environments provide a large number of ecosystem services, but are increasingly affected by multiple anthropogenic stressors, including pollution. Owing to the release of contaminants by many human activities, they can be good indicators of socio-economic development and the success of environmental policies at local, regional and global scales.

Coastal zone management and protection must be based on scientifically sound information. Since several decades ago, policy makers have designed and implemented national legislation and international agreements to protect coastal ecosystems. But have these policies really been effective? To answer this question, contaminants can be used to reveal if anthropogenic effects are stable, decreasing or increasing over time, which requires information to be collected over long time periods. Water and biota monitoring attempt to establish trends (e.g. the Mussel Watch Programme), but time series are usually short and geographically very sparse.

The only available strategy to quantify long term contamination trends in coastal zones worldwide is through the study of dated environmental archives, notably sediment cores. These also allow to set baseline values for each specific ecosystem, useful to design conservation strategies. GESAMP (Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection), with the support of the IAEA (International Atomic Energy Agency) and UNIDO (United Nations Industrial Development Organization), constituted Working Group 39 (WG39) on *Global Trends in Pollution of Coastal Ecosystems: Retrospective Ecosystem Assessment*, with the goal to provide stakeholders, scientists and society in general, with an objective and global assessment of pollution trends during the last century in coastal ecosystems.

The selected contaminants were grouped in metals and metalloids, radionuclides, pesticides, industrial organic chemicals, industrial organic byproducts, polycyclic aromatic hydrocarbons and nutrients. Both artificial and naturally present contaminants were released in keeping with socio-economic development in each region. Many of them started to be banned since the 1970s in developed countries, but are still produced and used in some developing countries.

This work was based on the compilation of the wealth of data already published in peer-reviewed journals on contamination time series from environmental archives (sediments and corals), and biota where available. Several uncertainties and limitations must be acknowledged when using data from sediment records, including possible post-depositional processes, and the use of data derived from cores collected in different moments, since sampling and analytical techniques have evolved with time. Our assessment is based on trend analysis of scaled contaminant data, avoiding the evaluation of absolute concentrations.

Trend analysis was performed on expert defined time periods and regionally assessed on the geographic regionalization of the NOAA (National Oceanic and Atmospheric Administration) Large Marine Ecosystems (LME). Although contaminant normalization by diverse techniques (such as grain size, Al and organic matter contents) is widely used, most collected data were not normalized, used non-standard normalization methods or information to normalize data was not reported. Therefore, trend analysis was performed on mathematically normalized (scaled) data, and no assessment of absolute concentrations was made.

Of over 1,000 papers screened, final trend analysis was based on 37,720 data points, included in 2,355 independent time series, obtained from 667 records and collected from the 272 peer reviewed papers. The database is available at UNINMAR – UNAM (Unidad de Informática Marina – Universidad Nacional Autónoma de México) geoportal. The key findings of this work were:

1. *Missing or poor information in 20 LMEs.* The Working Group could not find appropriate information from enough dated sediment cores, or could not analyze them because of their scarcity for 20 LMEs: East Bering Sea, Southeast U.S. Continental Shelf, Scotian Shelf, Newfoundland-Labrador Shelf, Guinea Current, Somali Coastal Current, Indonesian Sea, North Australian Shelf, Northwest Australian Shelf, Oyashio Current, Sea of Okhotsk, West Bering Sea, Laptev Sea, Kara Sea, Iceland Shelf and Sea, Faroe Plateau,

Hudson Bay Complex, Central Arctic Ocean, Aleutian islands and Canadian High Arctic-North Greenland. National and regional efforts should be engaged to obtain long-term contamination time series from dated sediments from these data poor regions. In some cases (e.g. the Arctic) there are large monitoring and assessment programs allowing the evaluation of pollution trends which should be carefully considered in future assessments.

2. *Unequal information on contaminants.* Most of the reported contaminants were metals. The less reported contaminants were pesticides, nutrients and polycyclic aromatic hydrocarbons (PAHs). Many records do not include nutrients, which are major contaminants and indicators of many contamination sources (notably urbanization and agriculture).

3. *Regional contamination histories are heterogeneous and reflect the socio-economic development of the riparian countries.* In some regions (e.g. the Baltic Sea LME), large and increasing contaminant trends were observed in the period 1950 – 1975, followed by substantial decreasing trends during the latter decades (1975 – 2015), owing to the effective implementation of international agreements and national policies and social awareness. Other regions (e.g. East China Sea LME) showed continuously increasing trends along the whole period. These two cases show that contamination trends are useful to understand the underlying causes of coastal deterioration and to provide scientifically sound information for decision-making, both locally and regionally.

4. *Some trends are already increasing in the early 20th century decades*. This reflects on-going regional development in some regions (e.g. Eastern and Southeastern Asia, Europe), likely since the industrial revolution in the 19th century.

5. *More and large increasing trends are observed in many regions during the 1950 – 1975 period.* This reflects the large worldwide development that occurred after World War II (WWII).

6. *During the latest decades (1975 – 2015) contaminant histories diverge.* Contaminant trends keep increasing in Eastern and Southern Asia, Africa and Central America, but the number of decreasing trends increase in the rest of the world, notably Europe, the Australian continent, South and North America and the Polar Regions. For most regulated contaminants the concentrations have decreased. This shows the interaction between regional development and the success of national policies, international agreements and social environmental awareness.

These findings led us to formulate some recommendations:

1. *Engage regional experts to provide regional assessments.* Case studies presented here could be used as examples for other regions. This report should be followed by a detailed analysis of all LMEs trends, by engaging local/regional experts and stakeholders to provide assessments and effective recommendations which could be published as an *Addendum* to this report.

2. *More and better data are needed.* Scientists should be encouraged to identify relevant geographical gaps and focus their attention on data-poor regions and lesser studied contaminants. This could be extended to regions visited some decades ago. Regional efforts are recommended to optimize resources and expand the number of study areas and contaminants. Scientists should make data more easily discoverable and available through data repositories. This might be ideally done by creating and properly funding a specific data repository of coastal contamination time series.

3. *Incorporate the study of environmental archives (especially sediment cores) in monitoring programmes.* Monitoring programmes likely visit relevant study areas on a regular basis. Scientists are encouraged to periodically (likely every 10 years) collect and analyze environmental archives from these areas to establish solid temporal trends of contamination and provide recommendations to decision makers.

4. *This effort should be maintained.* A mechanism should be setup to enlarge the existing database. Individual LME reports should be produced and updated, especially for data rich areas.

We suggest that world assessment could be produced regularly (e.g. each decade). This work should be maintained and funded in the long-term.

Temporal contamination records, notably from dated sediment cores, are rich sources of environmental information, and powerful tools to assess coastal ecosystem health and to establish protection and recovery targets for the world coastlines. This assessment provides decision-makers with predictive knowledge and a management tool to support decisions of socio-economic value at all scales. It is expected that this work will be widely disseminated amongst coastal environment stakeholders and society in general, and that it will have a positive impact on decision-making to better protect the coastal environments worldwide.

1. INTRODUCTION

Planet Earth is subject to a myriad of anthropogenic impacts, including soil erosion caused by land use change, climate change caused by the emission of greenhouse gases, and ecosystem degradation caused by, amongst many stressors, contamination. The transformation of our planet has been called *Global Change* (Vitousek, 1994). Indeed, humans have become a global geophysical force, especially after WWII, when the world population experienced a *Great Acceleration* (Steffen et al., 2007), causing what has been called *Anthropocene* (Crutzen and Stoermer, 2000).

Coastal ecosystems, the natural interface between land and sea, are very important environmental resources and provide a large number of ecosystem services, such as the maintenance of fisheries, nutrient cycling, carbon sequestration, coastal protection, erosion control, and recreation (Barbier et al., 2011). However, marine and coastal ecosystems are increasingly affected by multiple anthropogenic stressors, with the most relevant including physical alteration, sewage, nutrients, sediment mobilization, persistent organic pollutants, hydrocarbons, heavy metals, litter and radionuclides (GESAMP, 2001).

Many of these impacts are related to the release of contaminants to the environment. For example, agricultural activities contribute with nutrients and pesticides, mining contributes with heavy metals, industrial facilities discharge both chemical products (heavy metals, hydrocarbons, PCBs, PBDEs and pesticides) and by-products (such as dioxins and furans), and urban settlements release huge loads of nutrients and all kinds of contaminants. Contaminants reach the aquatic environment from the atmosphere and are also discharged with waste waters, associated with particles or in solution, and then attach to suspended particles, organic matter or bottom sediments. Many contaminants are transported in rivers and outfalls also in colloidal phase, which settle to the sediment after coagulation/flocculation upon reaching the sea, due to increased salinity. Therefore, the fate of at least a fraction of most contaminants are bottom sediments, where they can be immobilized for long time periods, creating a contamination archive in the aquatic environment.

If dated, unaltered sediments may become contamination records, i.e. the levels of sediment contamination with depth (and therefore age) reflect contamination levels of released particles and/or overlying waters. Other notable environmental archives in the coastal environment include corals and bivalve shells although, in these, contaminants are permeated through an organism and absolute levels are largely dependent on the biological cycling of each contaminant by each species and each individual.

One of the most useful dating approaches for environmental reconstruction through sediment archives is radiochronology, i.e. the derivation of time from the nuclear decay law. Owing to its suitable half-life (~22 years), geochronology of sediments using ^{210}Pb dating is the most used methodology, since it allows dating sediments up to 100 – 150 yr old, an appropriate period to study contamination records over the *Anthropocene* (a review can be found in Sanchez-Cabeza and Ruiz-Fernández, 2012). Similarly to ^{210}Pb, ^{226}Ra is a member of the ^{238}U natural decay series and decays into the noble gas ^{222}Rn ($T_{1/2}$ = 3.8 d). This exhales from surface soils and rocks, decays into ^{210}Pb, which is deposited as excess ^{210}Pb to surface soils and waters. Also, ^{210}Pb can be transported to aquatic ecosystems by runoff and be produced *in situ* in the water column from dissolved ^{226}Ra. As Pb is largely insoluble in water, ^{210}Pb attaches to suspended matter and is transported to bottom sediments, where it decays. Other radionuclides that may be used as chrono-stratigraphic markers in this period are bomb fallout radiocarbon, ^{137}Cs and plutonium isotopes. Dated sediment cores, and other environmental archives, allow the reconstruction of contamination levels and sedimentary processes in aquatic ecosystems. Therefore, they become valuable information sources of many of the relevant anthropogenic effects on coastal ecosystems, and in turn, indicators of their main stressors.

Increased awareness of coastal environmental values, services and degradation has led policymakers worldwide to develop and implement national legislation and international agreements aimed to protect coastal ecosystems, which started to be implemented in many developed countries in the 1970s. A fundamental question is whether the implementation of these policies is, in fact, reducing the impact of pollution on coastal ecosystems. A correct scientific approach should be based on the analysis of the time evolution of contamination levels in the ecosystems, in order to show with confidence whether the contamination levels are either stable, increasing or decreasing, requiring

measurements over prolonged periods (usually several decades).

A valuable approach is monitoring contaminants in water and biota, but long-term coastal pollution time series (i.e. age vs. concentration in an environmental matrix) of this kind are usually short (typically less than one decade) and geographically sparse. Therefore, the only available strategy to observe contamination trends in most coastal ecosystems is to reconstruct past contamination levels from dated environmental archives, such as bottom sediments, corals and bivalve shells.

The sedimentary record depends on the origin of contaminants. In general, artificial contaminants (such as most organic contaminants considered in this work) significantly dispersed in the environment since they were industrially manufactured, so baseline levels are zero and the record shows the contamination history in each location. On the other hand, natural contaminants (such as all metals considered in this work) show a baseline value different from zero and attribution to contamination sources requires their detection at somewhat elevated levels.

The use of sedimentary records also allows to provide information on real baseline values, usually pre-industrial, for each specific ecosystem. A well characterized sedimentary record, dated and analyzed for as many contaminants as possible, is undoubtedly a rich source of environmental information and a powerful tool to assess coastal ecosystem health and to define protection targets around the world coastlines.

GESAMP, with the support of the IAEA and UNIDO, constituted Working Group 39 (WG39; Table 1) on *Global Trends in Pollution of Coastal Ecosystems: Retrospective Ecosystem Assessment*. The goal of WG39 was to contribute to the reduction of coastal ecosystem stress globally by providing stakeholders, scientists and society in general an objective and global assessment of coastal ecosystem pollution trends during the last century (1900 – 2015). This was done through retrospective ecosystem analysis by using dated environmental archives and time series data, where available.

This work was achieved mainly by using the wealth of knowledge of dated coastal marine sediments, already published in peer-reviewed scientific documents up to 2015. They were assessed based on the geographic regionalization of the NOAA Large Marine Ecosystems (LME). LMEs are regions of ocean space of 200,000 km² or greater, that encompass coastal areas from river basins and estuaries to the outer margins of a continental shelf or the seaward extent of a predominant coastal current. LMEs are defined by ecological criteria including bathymetry, hydrography, productivity, and trophically linked populations.

This assessment was aimed to provide United Nations regulators, international, regional, national and local decision-makers with predictive knowledge and a management tool on contaminants trends worldwide to support decisions of socio-economic value at all scales.

WG39 members performed a large bibliographic review of the literature containing long-term records of pollutants in the coastal marine environment, classified by geographic location (LME) and contaminant type. They revised pollution indicators, dating methodologies, analytical techniques and trend analysis options and proposed a working methodology used to construct a database of contaminant levels in environmental records worldwide. It is emphasized that this assessment does not address specific contamination levels, as absolute concentrations may be affected by evolving sampling and analytical methodologies. To allow the comparison of diverse indicators and the way to report them, trend analysis was performed on mathematically normalized (scaled) values. The database was used to perform regional and global assessments of contamination trends.

WG39 met for the first time back-to-back with the International Symposium on Isotopes in Hydrology, Marine Ecosystems, and Climate Change Studies in Monaco, in April 2011. Members agreed to conduct a literature survey on contamination temporal records of coastal ecosystems, following a list of priority substances, and to classify them according to LMEs. Intersessionally, information was gathered in a bibliographic database, including a list of relevant information to be extracted from the scientific documents. A web platform was developed by UNINMAR-UNAM (Unidad de Informática Marina - Universidad Nacional Autónoma de México), with the aim to host and manage the information contained in the WG39 database. A second meeting was held at the Environment Laboratories of the IAEA, in Monaco, in May 2014, where the web-based platform was presented, including an analytical

tool to assess pollution trends in single temporal series, and a preliminary version of the LME 03 California Current report was produced. The WG39 methodology was revised and approved, as it was agreed that the tools and available data would warrant a high quality and impact product. Once funding was available, capture of information (mostly digitization of published figures) was performed. Available information includes full bibliographic information, location information (including LME), sample type, sampling date, chronology and contaminant information (age, concentration and unit). The database is available at the UNINMAR geoportal of Instituto de Ciencias del Mar y Limnología of UNAM. Trend analysis, performed with the open statistical software R, and report preparation was performed by two WG39 members (JASC and ACRF).

This report contains a detailed description of the established methodology, an overall description of the database (Results section), six case studies among the LMEs (selected based on geographic distribution and information availability), a global assessment, conclusions and recommendations. The Working Group expects that this report will be useful and will have an impact on decision-making to better protect the coastal environment worldwide.

2. METHODOLOGY

2.1. Environmental archives

Analytical time series of environmental contamination are scarce and usually very short, precluding quantitative and reliable estimates of temporal trends. Thus, the only alternative to evaluate the long-term evolution of contaminants in the aquatic environment is the use of environmental archives, which can provide suitable records to allow the identification of changes related to evolving socio-economic conditions and contamination sources. Time series derived from environmental archives may allow, if the number of data points are sufficient, to quantify significant temporal trends.

This work integrates the evaluation of contaminant data reported in scientific documents (published from 1973 to 2015). Based on availability and suitability, contaminant information was mainly obtained from sediment cores, although some data from coral cores and biota (e.g. fish tissue) were also included. Most records were collected from the continental shelf, typically from a water depth ≲ 200 m. When sedimentary records were collected from larger depths, they were included in the analysis if they were close to the coastline and could reflect coastal sedimentary processes (for example, the contamination recorded in deeper sediments close to Los Angeles, LME03).

2.2. Temporal framework

This work was focused on the study of environmental records that contained information within the period 1900 - 2015. Most sedimentary records were dated by using the ^{210}Pb method. ^{210}Pb is a natural radionuclide that belongs to the ^{238}U natural decay series, with a half-life of 22.23 ± 0.12 yr (Bé et al., 2008), thus perfectly suited to date sediment cores up to 100 – 150 yr. Owing to diverse geochemical disequilibria, ^{210}Pb is supplied to the sediment surface either from the atmosphere or from *in situ* decay of ^{226}Ra in the water column, named excess ^{210}Pb. Once deposited, several ^{210}Pb dating models can be used to date the sediment core, and the reader is referred to both the original papers (Appleby and Oldfield, 1978; Robbins, 1978) and recent review papers (Sanchez-Cabeza and Ruiz-Fernández, 2012).

As ^{210}Pb chronologies are established with the help of models, it is recommended that they are validated with the use of other stratigraphic markers. Where they can be measured, commonly used markers are ^{137}Cs and plutonium isotopes that, in addition, are considered to be contaminants in this work. These anthropogenic radionuclides showed maximum atmospheric concentrations near 1963, because of their release to the environment from nuclear weapons tests (the so-called global fallout) that constitutes a useful stratigraphic marker (Pennington, 1973; Goldberg et al., 1977). In the case of corals, dates are usually obtained from band counting of X-ray images (sclerochronology; Hudson et al., 1976) and, in the case of biota, time corresponds to the sampling date.

Although temporal trends can sometimes be visually identified from scatter plots of the couplets time – concentration (named here data points), the quantification of trends is needed to provide solid interpretations and conclusions. After expert discussion of several possible approaches, it was decided that the simplest and easiest tool to communicate trends was the use of linear regression during pre-defined temporal periods, and that trends would be reported as percentage of change per decade. It was acknowledged that the socio-economic

history of the world is very heterogeneous, but that during the 1970s many countries started to implement policies to control contaminant sources, and to ban the production of diverse toxic chemicals. The time limit of the expected records was 2015, when the bibliographic search was finished. Therefore, the common periods defined for this analysis were:

- 1900 – 1950: this is globally a low-industrialization period, with pollutant concentrations expected to be close to background. Although data previous to 1900 were available, they were not included in this analysis. The year 1950 is close to the beginning of the so-called *great acceleration* (Steffen et al., 2007), when many socio-economic indexes (e.g. population) and environmental indicators (e.g. contamination) started to show a rapid and unprecedented increase worldwide.
- 1950 – 1975: a period of important population and economic growth globally, characterized by increasing urbanization and industrialization.
- 1975 – present (2015): during this period environmental regulations to limit the use and production of hazardous substances (e.g. DDT, PCBs, tetraethyl Pb) started to be adopted, and new toxic compounds were introduced (e.g. tributyl-tin, PBDEs).

2.3. Target contaminants

Target contaminants were selected based on experts' knowledge, availability of historical data, suspected harmful effects and prevalence in coastal ecosystems. The target analyte list used in this work was broken up into seven contaminant families:

1. **Metals and metalloids** (for simplicity, named Metals in this work): Ag (silver), As (arsenic), Ba (barium), Cd (cadmium), Cr (chromium), Cu (copper), Hg (mercury), Ni (nickel), Pb (lead), Sn (tin), V (vanadium), Zn (zinc).
2. **Radionuclides:** Am (^{241}Am), Cs (^{137}Cs), Pb (^{210}Pb), Pu isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu), Ra (^{226}Ra), Th (^{232}Th), U (^{238}U).
3. **Pesticides:** aldrin, chlordane (CHL), DDT and its metabolites, dieldrin, endrin, heptachlor, hexachlorobenzene (HCB), lindane, mirex.
4. **Industrial organic chemicals:** polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs).
5. **Industrial organic byproducts:** polychlorinated dibenzo-p-dioxins (dioxins) and polychlorinated dibenzofurans (furans) (PCDD/PCDF).
6. **Polycyclic Aromatic Hydrocarbons (PAHs):** acenaphthene (Ace), acenaphthylene (Acy), anthracene (An), benzo(a)anthracene (B[a]An), benzo(a)pyrene (B[a]Py), benzo(b)fluoranthene (B[b]Flt), benzo(g,h,i)perylene (B[ghi]Pe), benzo(k)fluroanthene (B[k]Flt), chrysene (Ch), dibenzo(a,h)anthracene (D[a,h]An), fluoranthene (Flt), fluorene (Flu), indeno(1,2,3-cd)pyrene (Ipy), naphthalene (Na), phenanthrene (Phe), pyrene (Py).
7. **Nutrients:** N (nitrogen), P (phosphorus), Si (silicon).

Many of the target contaminants may have both a natural and artificial origin, which was taken into account in the final analysis through normalization (see section 2.8). In the case of sites affected by releases of naturally occurring radioactive material (NORM), including natural radionuclides from the ^{238}U decay series, dating was performed by other means such as the presence of ^{137}Cs peaks and/or other well-known events.

Although organic matter is a well-known contaminant of coastal ecosystems (usually associated with urban waste water discharges), carbon was not included in the list of nutrients because sediment depth profiles of organic carbon might be altered by post-depositional processes (e.g. aerobic and/or anaerobic degradation, or chemosynthesis). Although this might also influence other nutrients, there is large evidence that the sedimentary record reflects anthropogenic inputs (e.g. Eadie et al., 1994; Zimmermann and Canuel, 2002). Si is a widespread element but might also reflect the influence of catchment erosion and siltation.

Some of the target analytes are commonly reported as a sum of a variable number of chemicals (e.g., total PCBs) or their metabolites (e.g., total DDTs). Provided that the analyses are consistent through the environmental archive, this should not affect the trend analysis and were recorded as a single contaminant. Total PAHs were recorded only if they contained the sum of all individual PAHs listed above (i.e. priority PAHs; EPA - Environmental Protection Agency 2014).

Each contaminant family contained sub-families, i.e. the contaminants listed above, and each sub-family might include specific contaminants and/or different magnitudes related to the original concentration. For

example, for the sub-family *dioxins*, specific congeners were recorded. Papers reported contaminants in diverse forms, e.g. Hg might have been reported as concentration, concentration normalized to Al, concentration factor, enrichment factor and/or flux. Data were stored as reported in the original references.

2.4. Regions of interest

In this work, the Large Marine Ecosystems (www.lme.noaa.gov) approach was used as operational units to assess pollution trends, because they are rational units of ocean space where ecosystem-based management can be applied. Although when this work was defined the world coastal regions were divided into 64 LMEs, the final LMEs considered (including some changes in LME borders) were 66 (Figure 2.1).

2.5. Information sources

In order to collect data points and other information, only peer-reviewed scientific publications were used. It was assumed that the peer-review process guaranteed that the publication contained quality controlled scientific information, thus the data were collected from each document without further quality tests. Often the scientific documents used essential information that was reported in another publication (e.g. sedimentation rates), which was searched, and the information integrated to the database as part of the first paper.

Publications were searched through different web services, such as Scopus, Google Scholar, and EBSCOhost, and downloaded from publisher web platforms, such as ScienceDirect, SpringerLink and Taylor & Francis Online. Typical search keywords were: the LME name, large cities in the LME coastline, “sediment core”, “coral”, “shell”, “contaminant”, “pollutant”, contaminant family names and key contaminant names (such as “DDT”, “PCB” and “PAH”). The bibliographic database and papers were distributed among the WG39 members (Table 2.1).

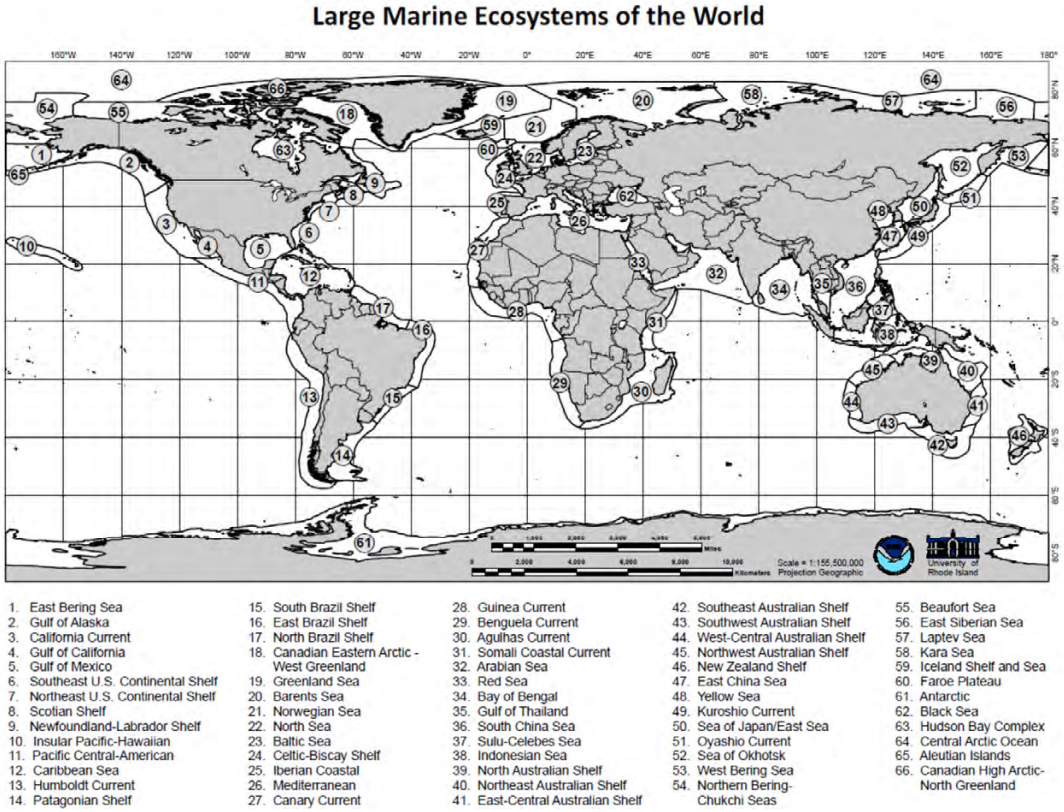


Figure 2.1. Large Marine Ecosystems of the world. Source: www.lme.noaa.gov

The publication should contain, at least, one dated environmental archive (e.g. ^{210}Pb and/or ^{137}Cs for sediment dating) with a description (or bibliographic reference) of how the measurements were carried out and the adopted dating model. In the case of biota derived time series, the publication should contain information from at least 5 sampling years. We considered that, if the data were consistently produced, such as in sediment cores or time series obtained by one laboratory, trend interpretation would be reliable. When the chronology for each section was not specified, sediment accumulation rates (e.g. cm yr^{-1}) were used to convert sediment depths to dates (the so-called age-model).

2.5.1. Bibliographic database

The bibliographic information of the collected scientific papers was first integrated in a Microsoft Excel spreadsheet, containing the following fields: LME number, short bibliographic reference (first author and publication year), full bibliographic reference, sampling year, collection depth (m), matrix type (sediment core, coral core, surface sediment survey, biota survey), contaminant family, and a detailed list of the figures, tables and other information needed for further data collection and analysis. All the information was initially gathered and stored by the GESAMP WG39 members in an online backup service (Dropbox).

2.6. Collection of information

The bibliographic database and the collected scientific papers were transferred to a web platform devoted to create and maintain biological and environmental databases related to aquatic ecosystems, developed by UNINMAR – UNAM. Papers were organized and were assigned unique catalogue numbers. Based on the information provided by WG39 members, also the relevant contaminant time series of each paper were identified, classified and catalogued. Each record contains:

- Record identification;
- Full bibliographic information (including DOI Digital Object Identifier);
- Location information: geographical coordinates, station name, collection depth;
- LME code and name;
- Sample type;
- Sampling date;
- Chronology information: dating method, analytical method, equipment;
- Contaminant information: name, analytical method, equipment; and
- Data point: age, contaminant value (concentration, enrichment factor or flux) and reported unit.

Full bibliographic information was retrieved from bibliographic search tools (such as Scopus and Google Scholar), or directly from the papers when not available from the web. Sample location information, sample type and sampling date were retrieved from the text, or from maps when coordinates were not provided. Coordinates were used to assign each record to a particular LME, following the LME boundaries, retrieved from NOAA website (NOAA, 2017a).

When both the chronology (age) and contaminant value were provided in tables, data points were directly captured into a database (MSExcel file). However, in most cases data point information only appeared as figures, thus data point collection was performed with the free software *PlotDigitizer*. To do this, several reference points (between 5 and 10) on both axes were chosen, and the figure data points were converted to paired data of contaminant value and age or depth (depending on the specific plot). All ages are stored in the database in ISO (International Organization for Standardization) format, which can be easily transformed into any desired format. During this process, each operator followed a quality control routine, in which a graphic with known coordinates was digitized. The quality test was approved only when the quadratically summed deviation was lower than 5%, although it was typically lower (of the order of 0 – 3%).

When both the age and contaminant value appeared in the same figure, digitization data points were directly entered into the database. When they appeared in different figures, first an age-model (age *versus* depth) was built, then the contaminant information was collected and, finally, the contaminant time series (i.e. final data points) was constructed and entered to the database. In some cases, only one or several mean sedimentation rates were reported, and were used to calculate a linear age-model. In some cases, papers referred to other papers for some information (e.g. sample coordinates, chronology). The paper was consulted to obtain the required information, but not included in the bibliographic database unless other useful records were found. The database contains mostly time series of about 100-150 years, however, when the time series were longer, the older data points were also kept in the database but not used in the analysis.

Table 2.1. List of LMEs and assigned WG39 member.

LME	LME name	Member	LME	LME name	Member
01	East Bering Sea	ACRF	34	Bay of Bengal	JASC
02	Gulf of Alaska	ACRF	35	Gulf of Thailand	ES
03	California Current	ACRF	36	South China Sea	ES
04	Gulf of California	ACRF	37	Sulu-Celebes Sea	ES
05	Gulf of Mexico	ACRF	38	Indonesian Sea	ES
06	SE U.S. Continental Shelf	JS	39	North Australian Shelf	ES
07	NE U.S. Continental Shelf	JS	40	NE Australian Shelf	ES
08	Scotian Shelf	JS	41	East-Central Australian Shelf	ES
09	Newfoundland-Labrador Shelf	JS	42	SE Australian Shelf	ES
10	Insular Pacific-Hawaiian	JS	43	SW Australian Shelf	EN
11	Pacific Central American	ACRF	44	West-Central Australian Shelf	EN
12	Caribbean Sea	ACRF	45	NW Australian Shelf	EN
13	Humboldt Current	JMG	46	New Zealand Shelf	EN
14	Patagonian Shelf	JMG	47	East China Sea	ES
15	South Brazil Shelf	JMG	48	Yellow Sea	ES
16	East Brazil Shelf	JMG	49	Kuroshio Current	ES
17	North Brazil Shelf	JMG	50	Sea of Japan / East Sea	ES
18	Canadian Eastern-Arctic West Greenland	RD	51	Oyashio Current	JASC
19	Greenland Sea	RD	52	Sea of Okhotsk	JASC
20	Barents Sea	JS	53	West Bering Sea	JASC
21	Norwegian Sea	RD	54	Northern Bering-Chukchi Seas	RD
22	North Sea	JASC	55	Beaufort Sea	RD
23	Baltic Sea	NT	56	East Siberian Sea	RD
24	Celtic-Biscay Shelf	JMG	57	Laptev Sea	RD
25	Iberian Coastal	JASC	58	Kara Sea	RD
26	Mediterranean	JASC	59	Iceland Shelf and Sea	RD
27	Canary Current	EN	60	Faroe Plateau	RD
28	Guinea Current	EN	61	Antarctic	JS
29	Benguela Current	EN	62	Black Sea	JASC
30	Agulhas Current	EN	63	Hudson Bay Complex	JS
31	Somali Coastal Current	EN	64	Arctic Ocean	JS
32	Arabian Sea	JS	65	Aleutian Islands	JASC
33	Red Sea	JASC	66	Canadian High Arctic-North Greenland	JASC

ACRF: Ana Carolina Ruiz-Fernández, EN: Elvis Nyarko, ES: Elvira Sombrito, JASC: Joan-Albert Sanchez-Cabeza, JMG: José Marcus Godoy, JS: Jose Sericano, NT: Norbert Theobald, RD: Roberta Delfanti.

2.6.1. Data quality and transformations

Several quality tests were performed before analysis. First, a search for duplicates was made. The correct LME assignment of the digitized records was visually revised by comparison of published maps and LME borders displayed with the software Google Earth. This process was hindered by the change from 64 to 66 LMEs and the redefinition of some LME limits during the analysis process. Bibliographic information was verified against a master table of journal names and DOIs were confirmed.

All contaminant records (i.e. time series) were plotted visually compared to the published profiles, and the lowest and highest values were confirmed with the original papers. Contaminant family and individual contaminant names were checked against a master table. This was particularly complex in the case of organic contaminants, as there is not a standard single nomenclature (several names are used for a single compound), so homonym compound names were identified, names were homogenized and were assigned to a sub-family and family (e.g. BHCs were renamed HCBs, and assigned to sub-family hexachloro-benzenes, family Pesticides).

Systematically, all values were transformed to the same units as follows: Metals in µg g^{-1}, Radionuclides in Bq kg^{-1}, Pesticides, Industrial organic chemicals, Industrial organic byproducts and PAHs in ng g^{-1}, and Nutrients in %. Similar rules were used for fluxes and, if needed, enrichment and concentration factors were transformed to a dimensionless value. When the same contaminant was reported in diverse forms (e.g. concentrations, normalized concentrations, fluxes, etc.) only one form was used in the following preference order: concentration > flux > enrichment factor > concentration factor > normalized value.

All detected errors were corrected by using routines programmed in the software R (R Core Team, 2017), so all changes could be traceable. For the final analysis, data points earlier than 1900 were not considered.

2.7. Storage, display and preliminary analysis

The GESAMP WG39 database is stored and available in the UNINMAR web platform (www.icmyl.unam.mx/uninmar) under the keyword *GESAMP* and the collection name *Global trends in pollution of coastal ecosystems*, and all records are flagged as *LME pollutants*. Each bibliographic record has its own web page (Figure 2.2) which is cited and can be directly reached through a link. The platform allows to consult (Figure 2.3), visualize on a map (Figure 2.4) and explore the contents (Figure 2.5) of each record.

2.7.1. Preliminary analysis

Each single time series can be analyzed in UNINMAR through a routine specifically designed to evaluate temporal trends, based on correlation and linear regression analysis (Figure 2.6), which provides correlation coefficient, linear regression slope (trend) and uncertainty. These results were used to evaluate the pollution trends of selected records: increasing (positive trend) or decreasing (negative trend), and statistically significant ($p < 0.05$) or not ($p > 0.05$) (Figure 2.6). Information was then transferred to an Excel spreadsheet to perform a preliminary LME assessment.

Data were carefully explored for the detection of errors and inconsistencies, produced during digitization or present in publications, and were corrected. Although some substances (e.g. radionuclides, PCBs and DDTs) were only produced after 1900 (e.g. ^{137}Cs releases were significant at a global scale only since the 1950s), they were sometimes detected before production, usually at very low concentrations. This might be caused by some degree of sediment mixing, substance mobility and/or imprecise dating. As this work was based on peer reviewed scientific publications, data were included in the analysis, howevwer trend interpretation during the period 1900 – 1950 must be cautiously taken and inconsistencies should be studied in each case.

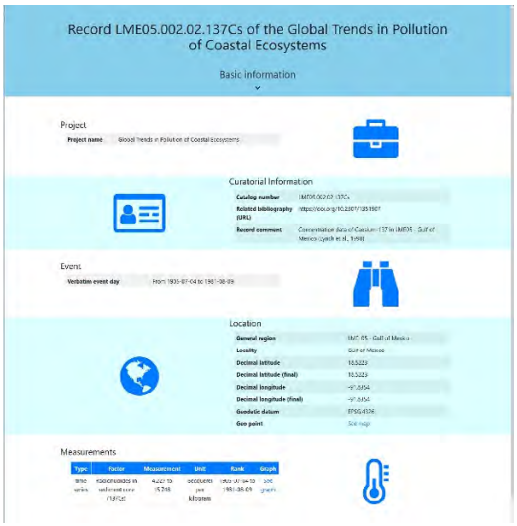


Figure 2.2. Example of basic information for a contaminant record in the GESAMP WG39 database in UNINMAR.

Figure 2.3. Example of bibliographic information for a contaminant record in the GESAMP WG39 database in UNINMAR.

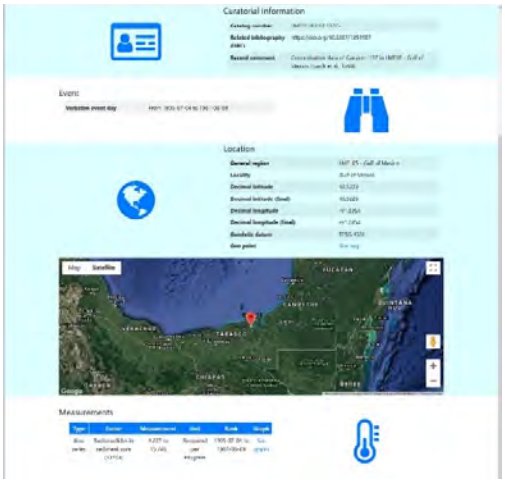


Figure 2.4. Example of contents (including map) of a single record in the UNINMAR database.

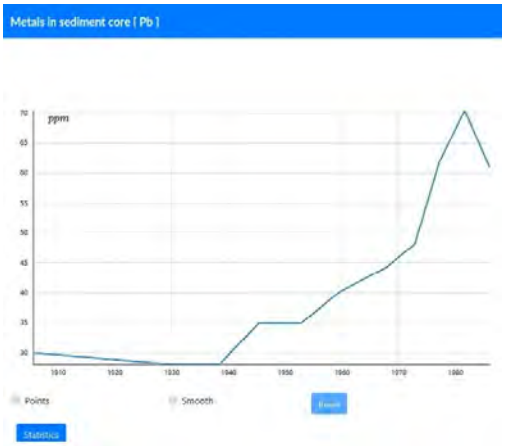


Figure 2.5. Example of a time series as displayed in the UNINMAR database.

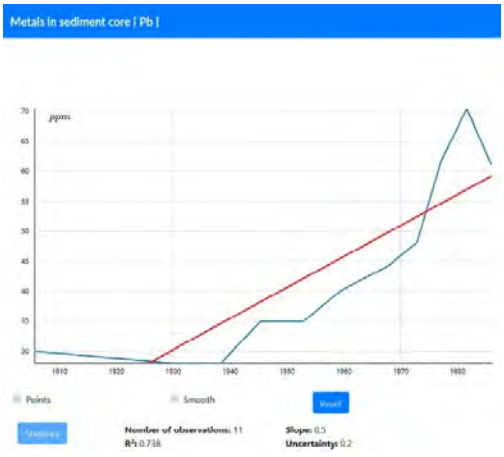


Figure 2.6. Example of a linear regression in the UNINMAR database.

2.8. Final analysis

Data manipulation, aggregation and analysis were performed with the statistical package R (R Core Team, 2017), so all analyses were traceable. Although contaminant normalization by diverse techniques (such as grain size, Al and organic matter) is recommended and widely used (e.g. Loring, 1990; Daskalis and O'Connor, 1995; Chapman and Wang, 2001), most data points included in the GESAMP WG39 database were neither normalized, nor used non-standard normalization methods, nor information to normalize data points was not reported. Therefore, a homogenous geochemical normalization of the database was not possible.

To perform trend analysis of several records, time series need to be comparable. Absolute contaminant concentrations, or some derived magnitudes (such as flux) cannot be directly compared, as they can be different by orders of magnitude. Since the range of values contained in the database was very large, we opted for feature scaling, by rescaling the values range to the [0,1] interval using the equation:

$$y_n = \frac{y - y_{min}}{y_{max} - y_{min}}$$

where y, y_{min}, y_{max} and y_n are the measured, minimum, maximum and normalized values, respectively. As such, for all time series minima were zero and maxima were 1, and data points could be compared.

The experts considered that, for the range of contaminant profiles expected, the most robust trend analysis possible was linear regression for the proposed time periods. Regression analysis

was performed only when at least 3 time series were found and periods when at least 5 data points were available. The R function *linear model* (lm) was used and information on the regression slope (temporal trend), uncertainty and statistical significance (p value) were extracted. Linear regression is a simple and robust tool to analyze trends and therefore more complex approaches were not considered. By fixing the periods, trends can be easily compared for different regions. Statistical significance depends heavily on the number of data points for each LME and period considered.

For an easier interpretation, the regression slope was reported as the percentage of variation per decade. In the trends reports, the following nomenclature was used:

- n.d. (no data): when no data points were available for the specific period considered.
- n.a. (not analyzed): when the number of data points available in the specific period was not enough for analysis (< 5).

In figures, these categories were assigned a white color, indicating the lack of enough data points for analysis. On the other hand, calculated regressions were considered statistically significant if $p < 0.05$, i.e. a 95% statistical confidence. Trends were considered to be:

- Decreasing: when the slope was negative and $p < 0.05$.
- Neutral: when $p > 0.05$.
- Increasing: when the slope was positive and $p < 0.05$.

The neutral qualification is relevant in this analysis, as it means that no significant trend could be calculated. This qualification must be interpreted with caution, because it could be due to a real lack of trend, or to the small number of data points for the analyzed period.

In figures, we followed a recommended set for 3 diverging levels (Brewer, 2019). We assigned a “brown” colour to statistically significant increasing trends, light-yellow to neutral trends, and green to statistically significant decreasing trends. Regression analysis was performed on the dataset grouped by LMEs, families, subfamilies and periods which can be found in the case studies.

2.9. LME reports

Once the full trend analysis had been performed, specific reports for selected LMEs were produced. Six cases were chosen based on their geographical distribution and number of time series available for analysis. Each report contains an introduction, with an overall description of the LME, largely based on the NOAA LME briefs (NOAA, 2017b) and Transboundary Waters Assessment Programme (TWAP, 2017). The section *Database information* contains a synthesis of the number of papers reviewed, including the total number of time series used in the analysis by contaminants and contaminant families.

Barplots were used to graphically describe the number of time series by contaminant and contaminant family, allowing to easily identify the most reported contaminants and contaminant families in each LME. Some examples of the time series analyzed (normalized value *versus* time) are shown in scatter plot figures. The smooth line and 95% confidence interval band were calculated with a local regression technique (Cleveland et al., 1992).

The trend analysis section synthesizes the main findings by contaminant and contaminant family, summarized in tables. For easier interpretation, these trends are translated into a trends chart, by contaminant and period. A final figure integrates the locations of LME records and the trends chart by family. This assessment is largely qualitative, as it includes contaminants with diverse geochemical behavior.

Finally, an assessment of the observed trends in each LME was produced, paying special attention to the most important contaminants and trends observed, and likely contamination sources. Specific LME recommendations were also provided, such as missing relevant areas and contaminant time series. Finally, the references and the bibliographic database for each LME were reported.

2.10. Uncertainties and limitations

As for any regional and global assessment, this work is subject to uncertainties and limitations. Both point and non-point contamination sources are neither regularly distributed in each LME nor, obviously, globally. Most published data are focused on contaminated areas and in multi-national LME’s the coastline of more developed countries are usually better covered. Therefore, a bias towards contaminated regions

of developed countries is present. Also, the development histories at each LME will largely depend on the riparian countries, which must be considered when analyzing the results. Although a major effort was made during the collection of information, it is possible that not all papers relevant to this review were identified during the data capture process.

Sampling, analytical techniques and dating methods have evolved during the past decades. Owing to post-depositional processes (notably mixing), sedimentary records do not always provide unambiguous histories of sediment accretion, and this is assumed to have been identified by the authors and peer reviewers of the analyzed papers. Furthermore, the mathematical normalization used cannot capture along-core changes in geochemical properties such as grain size and sedimentary sources.

Taking into account all these caveats, it is considered that the methodology was rigorous enough to reach valid results. These must be carefully analyzed on a case-by-case basis and more information in future compilations will certainly yield more precise regional and global assessments.

3. RESULTS

During the bibliographic research phase, over 1,000 papers were screened for information. Of these, experts critically selected 418 papers. During the compilation of information some papers were discarded because some basic information, such as core locations and age-models was missing, unclear or referred to inaccessible documents. Finally, information was analyzed from 272 papers containing contaminant information from 667 records.

3.1. Database description

The initial database contained a total of 56,499 data points (paired age - contaminant value), together with all its supporting information (over 4 million fields). After selection of contaminants of interest, restriction of ages to the analyzed period, and selection of time series with at least 5 data points by time period, a total of 37,720 data points were used for analysis. A summary of the database contents is shown in Table 3.1. It is remarkable that in several LMEs (California Current, South Brazil Shelf, Baltic Sea, Iberian Coastal, Mediterranean, Arabian Sea, South China Sea, East China Sea, and Kuroshio Current) the number of data points available for analysis was over 1,000. Of these, the South China Sea and the Baltic Sea contained over 5,000 data points for analysis.

Table 3.1. Summary contents of the analyzed database.
Non-listed LMEs contained no data points, or they were not enough for analysis.

LME	Name	References	Time series	Families	Contaminants	Data points
LME02	Gulf of Alaska	1	2	1	1	25
LME03	California Current	12	160	6	32	1,972
LME04	Gulf of California	8	37	3	10	669
LME05	Gulf of Mexico	6	37	3	13	677
LME07	Northeast U.S. Continental Shelf	6	42	4	14	566
LME10	Insular Pacific-Hawaiian	1	2	1	1	78
LME11	Pacific Central-American	4	7	2	7	121
LME12	Caribbean Sea	5	28	3	12	428
LME13	Humboldt Current	2	4	2	2	41
LME14	Patagonian Shelf	1	4	1	2	24
LME15	South Brazil Shelf	16	70	4	19	1,104
LME16	East Brazil Shelf	3	14	2	5	183
LME17	North Brazil Shelf	2	7	1	2	111
LME18	Canadian Eastern – Arctic West Greenland	8	66	2	9	813
LME19	Greenland Sea	3	9	2	3	105
LME20	Barents Sea	7	20	4	4	300
LME21	Norwegian Sea	2	20	2	8	110

LME	Name	References	Time series	Families	Contaminants	Data points
LME22	North Sea	6	25	5	10	294
LME23	Baltic Sea	33	422	7	32	5,651
LME24	Celtic-Biscay Shelf	1	4	2	4	108
LME25	Iberian Coastal	15	157	4	15	3,290
LME26	Mediterranean	31	304	5	20	4,366
LME27	Canary Current	5	30	2	8	212
LME29	Benguela Current	1	1	1	1	23
LME30	Agulhas Current	1	1	1	1	14
LME32	Arabian Sea	7	63	3	9	1,462
LME33	Red Sea	1	4	1	4	64
LME34	Bay of Bengal	3	71	2	7	650
LME35	Gulf of Thailand	3	11	3	7	877
LME36	South China Sea	24	253	6	27	6,939
LME37	Sulu-Celebes Sea	3	21	3	7	429
LME40	Northeast Australian Shelf	2	6	2	3	56
LME41	East-Central Australian Shelf	2	32	1	5	236
LME42	Southeast Australian Shelf	2	10	1	7	102
LME43	Southwest Australian Shelf	1	2	1	2	20
LME44	West-Central Australian Shelf	2	7	2	3	36
LME46	New Zealand Shelf	3	15	2	6	163
LME47	East China Sea	16	156	7	33	2,173
LME48	Yellow Sea	10	47	4	13	973
LME49	Kuroshio Current	9	116	6	15	1,478
LME50	Sea of Japan - East Sea	1	6	2	3	118
LME54	Northern Bering – Chukchi Seas	4	16	1	3	126
LME55	Beaufort Sea	1	18	1	2	181
LME56	East Siberian Sea	1	6	2	5	59
LME61	Antarctic	3	14	4	8	114
LME62	Black Sea	3	8	1	4	179

The data used for trend analysis belong to 46 LMEs. We could not find appropriate data points (mainly from environmental records), or could not analyze them because of information scarcity for 20 LMEs (30% of all LMEs), namely: East Bering Sea, Southeast U.S. Continental Shelf, Scotian Shelf, Newfoundland-Labrador Shelf, Guinea Current, Somali Coastal Current, Indonesian Sea, North Australian Shelf, Northwest Australian Shelf, Oyashio Current, Sea of Okhotsk, West Bering Sea, Laptev Sea, Kara Sea, Iceland Shelf and Sea, Faroe Plateau, Hudson Bay Complex, Central Arctic Ocean, Aleutian islands and Canadian High Arctic-North Greenland. Many of these LMEs correspond to Arctic environments with low population density, and two of them belong to the African continent. Despite a large monitoring program and periodic assessments carried out in the Arctic area (Hung et al., 2016), the regional analyses do not consider sediment or ice cores (AMAP - Arctic Monitoring and Assessment Programme, 2016). In general, the analysis of dated environmental records would complement these efforts by i) expanding the time interval, as regular monitoring programmes usually do not include more than 3 – 4 decades, ii) defining background concentrations, and iii) providing integrated long-term information from poorly monitored and studied areas.

After the analysis was completed, we found new papers (published during or after 2015) related to some of these areas. We propose that this first compilation and analysis effort should be maintained and expanded to periodically produce similar products (e.g. every ten years). In any case, this first compilation and assessment clearly indicated that future efforts in coastal pollution reconstructions should include these data poor areas.

The analyzed dataset included a total of 2,355 time series (i.e. a specific contaminant *versus* time), with a mean of 16 points per time series. The number of time series per LME is shown in Figure 3.1. The LMEs with over 100 time series collected were Baltic Sea, Mediterranean,

South China Sea, California Current, Iberian Coastal, East China Sea and Kuroshio Current.

Time series were collected from several environmental matrices (sediment cores, coral cores and biota), although sediment cores were by far the most abundant in the database (> 90% of the time series; Figure 3.2). The contaminant family most commonly found in the database was, by far, Metals (1,343 time series). If considered together, organic contaminants contained 536 time series (Figure 3.3). In total, 51 contaminants were included in the database, of which the most abundant were the metals Pb, Zn and Cu (Figure 3.4).

As expected, dates were not homogeneously distributed (Figure 3.5). The period covered in the initial database starts in year 167 A.D. until 2014, with a median value of 1970. Usually, ages before 1850 corresponded to radiocarbon dated cores or, in some cases, extrapolated from constant sediment accumulation rates. As most researchers subsample sediment cores with a constant width (typically 1 or 2 cm), owing to compaction deeper sections span longer time periods, so the density of data points diminishes with depth, as observed in Figure 3.5. The maximum number of data points occurs in the interval 1980 – 2000, with up to 700 data points per year.

The decreasing number of data points in recent years (since 2000) is mainly due to i) the lag between core collection and publication, and ii) the fact that, owing to radiometric dating methodological reasons and depending on the sediment accumulation rate, the age of the first section of a sediment core might be several years older than the sampling (and consequently the publication) date. It is expected that, in new assessments, these years should be better represented.

In the case studies, it was considered useful to plot examples of normalized data points for some contaminants, including a smoothed mean and the 95% confidence band. As the number of data points after year 2000 was very small and confidence bands become unrealistic, the plots concluded in year 2000.

3.2. Trend analysis

Trend analysis was performed at the level of contaminants and contaminant families. For contaminants, a total of 548 trend analyses were performed, of which 37 were decreasing, 353 were neutral, and 158 were increasing (Table 3.2). For contaminant families, the total number of calculated trends was 238, of which 20 were decreasing, 122 were neutral and 96 were increasing (Table 3.3).

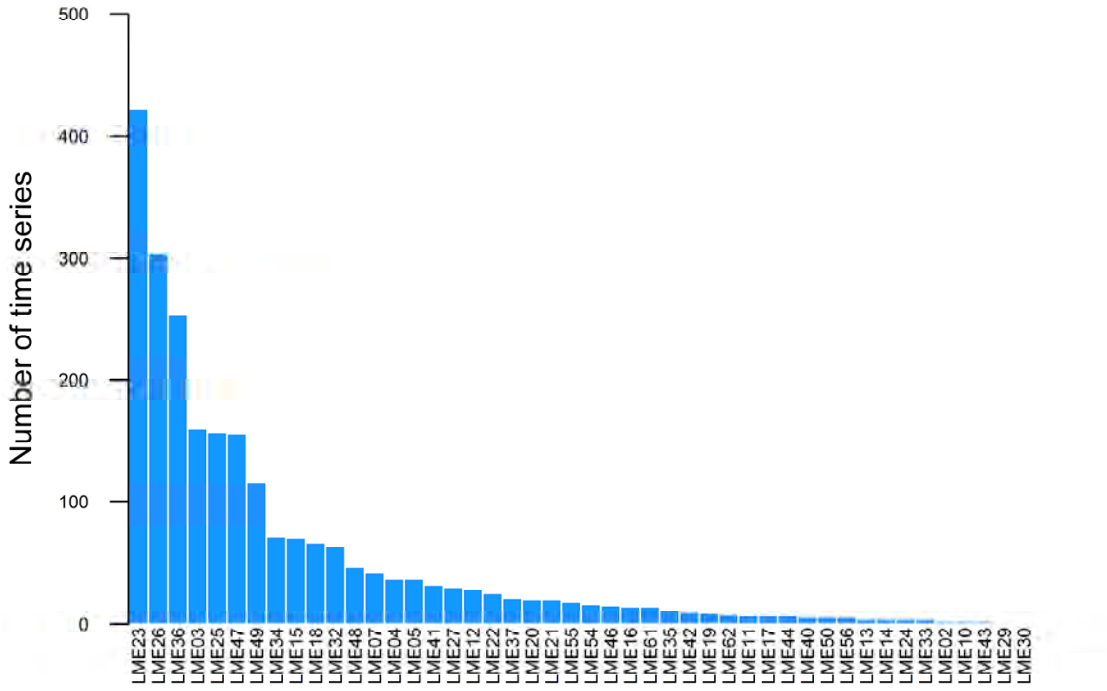


Figure 3.1. Number of time series per LME.

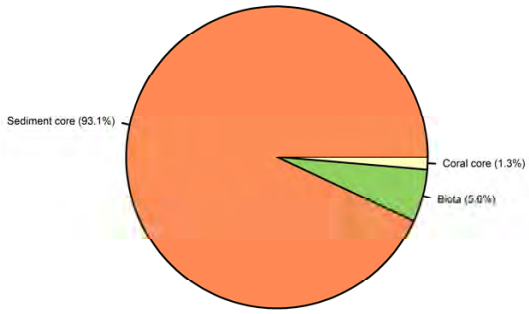


Figure 3.2. Time series (%) by environmental matrix.

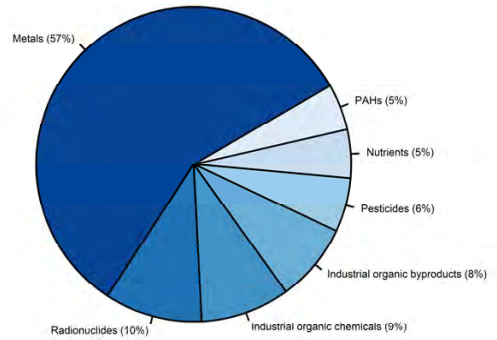


Figure 3.3. Time series (%) by contaminant family.

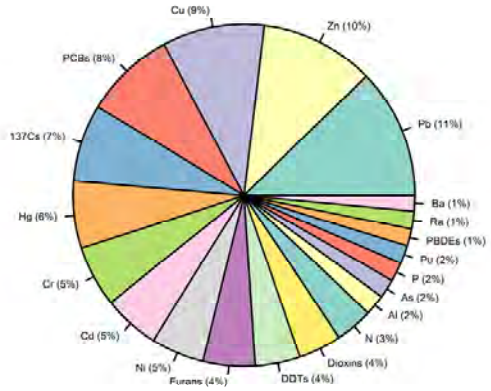


Figure 3.4. Time series (%) by contaminant sub-family. Only the 20 more abundant subfamilies are plotted.

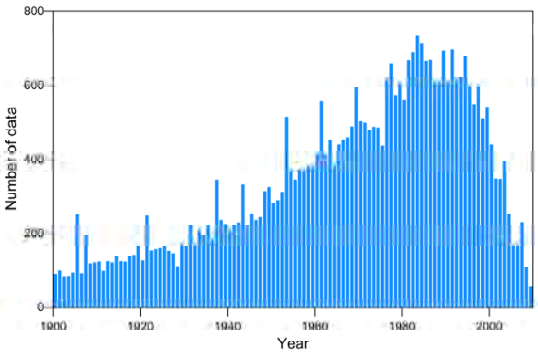


Figure 3.5. Number of data points per year.

Table 3.2. Summary of trend analysis by contaminant for all time periods.

LME	Name	Calculated	Decreasing	Neutral	Increasing
LME03	California Current	34	2	19	13
LME04	Gulf of California	18	2	11	5
LME05	Gulf of Mexico	9	0	6	3
LME07	Northeast U.S. Continental Shelf	7	1	4	2
LME12	Caribbean Sea	6	0	4	2
LME13	Humboldt Current	3	0	3	0
LME15	South Brazil Shelf	27	0	21	6
LME16	East Brazil Shelf	3	0	2	1
LME17	North Brazil Shelf	6	1	4	1
LME18	Canadian Eastern-Arctic West Greenland	18	0	14	4
LME19	Greenland Sea	3	0	3	0
LME20	Barents Sea	3	0	1	2
LME21	Norwegian Sea	6	0	5	1

LME	Name	Calculated	Decreasing	Neutral	Increasing
LME22	North Sea	3	1	1	1
LME23	Baltic Sea	68	16	30	22
LME25	Iberian Coastal	32	4	19	9
LME26	Mediterranean	41	1	29	11
LME27	Canary Current	21	0	19	2
LME32	Arabian Sea	21	2	16	3
LME34	Bay of Bengal	21	1	19	1
LME35	Gulf of Thailand	3	0	2	1
LME36	South China Sea	46	0	26	20
LME37	Sulu-Celebes Sea	12	1	8	3
LME40	Northeast Australian Shelf	3	1	2	0
LME41	East-Central Australian Shelf	15	0	15	0
LME44	West-Central Australian Shelf	2	0	2	0
LME46	New Zealand Shelf	9	0	9	0
LME47	East China Sea	46	0	26	20
LME48	Yellow Sea	24	0	15	9
LME49	Kuroshio Current	18	4	2	12
LME54	Northern Bering-Chukchi Seas	9	0	6	3
LME55	Beaufort Sea	6	0	6	0
LME61	Antarctic	3	0	3	0
LME62	Black Sea	2	0	1	1
Totals		*548*	*37*	*353*	*158*

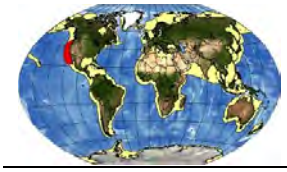
Table 3.3. Summary of trend analysis by family for all time periods.

LME	Name	Calculated	Decreasing	Neutral	Increasing
LME03	California Current	13	2	7	4
LME04	Gulf of California	6	0	3	3
LME05	Gulf of Mexico	6	0	4	2
LME07	Northeast U.S. Continental Shelf	5	1	3	1
LME12	Caribbean Sea	6	0	5	1
LME13	Humboldt Current	3	0	3	0
LME15	South Brazil Shelf	12	2	5	5
LME16	East Brazil Shelf	6	0	5	1
LME17	North Brazil Shelf	3	1	1	1
LME18	Canadian Eastern-Arctic West Greenland	6	0	5	1
LME19	Greenland Sea	4	0	4	0
LME20	Barents Sea	3	0	1	2
LME21	Norwegian Sea	6	0	5	1
LME22	North Sea	5	1	2	2
LME23	Baltic Sea	21	5	2	14
LME25	Iberian Coastal	10	1	3	6
LME26	Mediterranean	12	1	7	4
LME27	Canary Current	3	0	0	3
LME32	Arabian Sea	3	1	1	1
LME34	Bay of Bengal	6	0	5	1

LME35	Gulf of Thailand	4	0	3	1
LME36	South China Sea	18	0	6	12
LME37	Sulu-Celebes Sea	6	0	4	2
LME40	Northeast Australian Shelf	3	1	2	0
LME41	East-Central Australian Shelf	3	0	3	0
LME44	West-Central Australian Shelf	4	0	4	0
LME46	New Zealand Shelf	6	0	4	2
LME47	East China Sea	20	1	6	13
LME48	Yellow Sea	12	0	6	6
LME49	Kuroshio Current	9	3	0	6
LME54	Northern Bering-Chukchi Seas	3	0	2	1
LME55	Beaufort Sea	3	0	3	0
LME61	Antarctic	5	0	5	0
LME62	Black Sea	3	0	3	0
Totals		*238*	*20*	*122*	*96*

4. CASE STUDIES

In the following sections, the analysis of six regions are included as case studies. These are selected on the basis of geographic distribution and information availability. Local and regional experts are encouraged to provide analysis of the other LMEs that could be published as an addendum to this report.



LME03
California Current

1. INTRODUCTION

The California Current LME, in the coastal area of the Pacific Ocean, encompasses the region between Washington State in the USA and the Baja California peninsula in Mexico. The largest coastal cities are San Francisco, Los Angeles and San Diego, in the USA, and Tijuana in Mexico. Climate ranges from temperate (California) to arid (Baja California peninsula) and is classified as a low productivity ecosystem. However, there can be strong seasonal coastal upwelling that generates localized areas of high primary productivity that support important commercial fisheries (e.g. sardines, anchovy). It has a shoreline longer than 3,200 km and contains more than 400 estuaries and bays, many of those located in the USA territory (West Coast estuaries, Columbia River, San Francisco Bay and Puget Sound) are impaired by some form of pollution or habitat degradation, specially affecting sediment quality (EPA, 2004).

2. DATABASE INFORMATION

In LME03, 13 papers were selected and 12 were used for analysis after screening of their full contents. Information on 160 time series (all obtained from sediment cores) and 1972 data-points was extracted, meeting the requirements of the methodology. The selected papers contained information on 32 contaminants (Figure LME03.1) included in 6 families (Figure LME03.2).

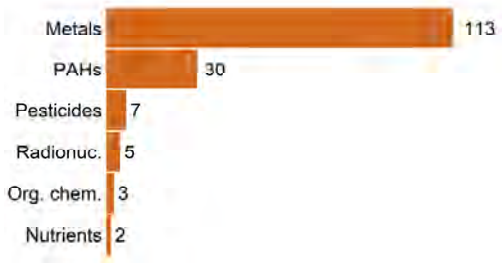


Figure LME03.2: Number of time series by contaminant family.

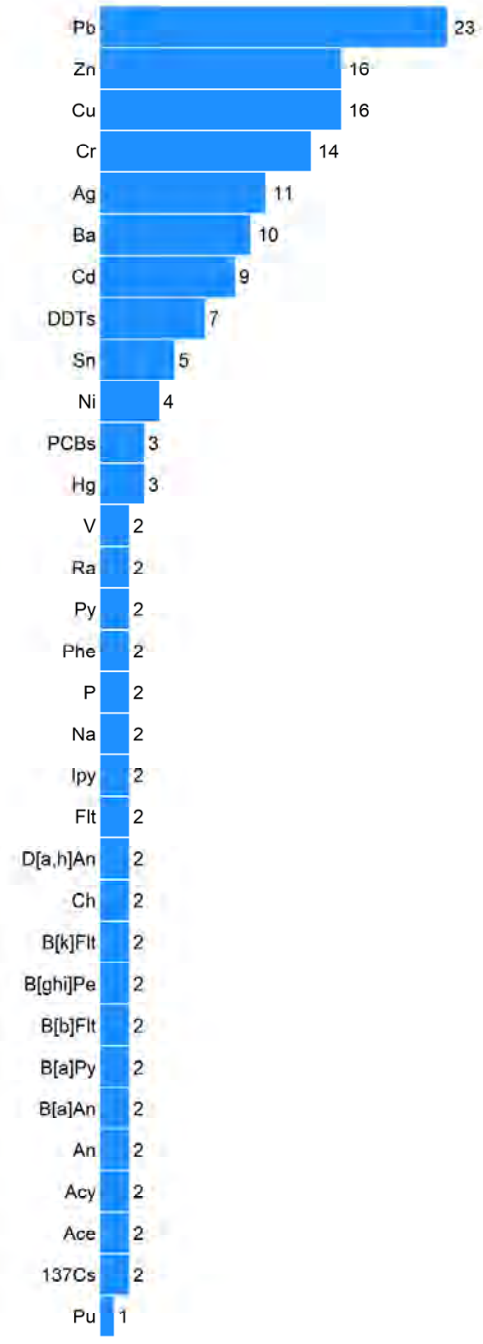


Figure LME03.1: Number of time series by contaminant.

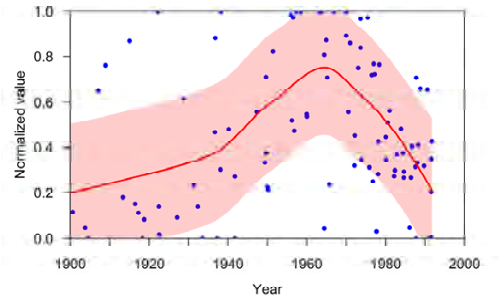


Figure LME03.3: Cd time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

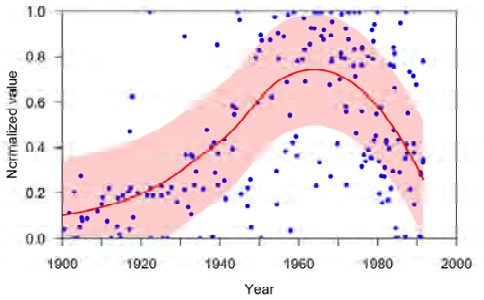


Figure LME03.4: Zn time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

As examples, two time series are shown in Figures LME03.3 and LME03.4.

3. TREND ANALYSIS

Trends of the selected time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found, and periods were analyzed when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminants found in this LME is shown in Table LME03.1 and Figure LME03.5. In this case, 34 trends were analyzed and 15 were found to be statistically significant ($p<0.05$).

The trends are expressed as percentage of variation per decade. During the period 1900-1950, neutral trends were observed for Cd, Hg and DDTs, and increasing trends were observed for Ag, Ba, Cr, Cu, Ni, Pb, Sn and Zn. During the period 1950-1975, neutral trends were observed for Ba, Cd, Cr, Ni, Zn and DDTs, and increasing trends were observed for Ag, Cu, Hg, Pb and Sn. During the period 1975-2015, decreasing trends were calculated for Cd and Zn, neutral trends were observed for Ag, Ba, Cr, Cu, Hg, Ni, Pb, Sn and DDTs and no increasing trends were observed.

Trend analysis was also carried out by aggregation of all contaminants in the same family during the same periods. Families were analyzed if at least 3 time series were found and periods when at least 5 data points were available. A summary of the analyzed trends during each period and for all contaminant families found in this LME is shown in Figure LME03.6. In this case, 13 trends were analyzed and 6 were found to be statistically significant ($p<0.05$).

During the period 1900-1950, decreasing trends were calculated for PAHs, neutral trends were observed for Radionuclides and Pesticides and increasing trends were observed for Metals. During the period 1950-1975, neutral trends were observed for Radionuclides and Pesticides and increasing trends were observed for Metals and PAHs. During the period 1975-2015, decreasing trends were calculated for Metals, neutral trends were observed for Radionuclides and Pesticides and increasing trends were observed for PAHs.

Table LME03.1: Summary of temporal trends (percentage of variation per decade) by contaminant and period analyzed. Only contaminants with at least one significant trend are reported.

Contaminant	1900-1950	1950-1975	1975-2015
Hg	neutral	+28.4	neutral
Sn	+5.9	+27.8	neutral
Ag	+6.1	+16.7	neutral
Pb	+9.4	+15.6	neutral
Cu	+7.9	+12.3	neutral
Ni	+9.2	neutral	neutral
Cr	+8.2	neutral	neutral
Ba	+5.5	neutral	neutral
DDTs	neutral	neutral	neutral
PCBs	n.d.	n.d.	neutral
Cd	neutral	neutral	-16.7
Zn	+9.3	neutral	-18.2

n.d. = not enough data points for trend analysis

4. ASSESSMENT

In LM03, the 12 analyzed papers mainly study the areas of Los Angeles and San Francisco. This is not surprising, as Los Angeles is one of the largest cities in the world, and the San Francisco metropolitan area i) ranks amongst the 100 largest populated areas in the world, and ii) is the largest urbanized estuary on the west coast of the United States (Pereira et al., 1999). Both areas had a rapid expansion of urbanization and industrialization since the gold rush in the 1850s, intensified since WWII. A Mexican record was included in the database, but the analyzed contaminants (dioxins and furans) were not contained in the other records. As only contaminants with at least 3 time series were assessed, these data were not reflected in the final assessment. The number of analyzed contaminants is large (32) and includes 6 of the 7 contaminant families. This shows the interest of the scientific community in Los Angeles and San Francisco areas, as early as the 1970s (Chow et al., 1973), owing to the intensity of the pollution sources (such as large water treatment plants and some military facilities) and the presence of basins with anoxic conditions near Los Angeles, favoring the preservation of undisturbed sediments. Special attention has been paid to metals, although other contaminants (notably PAHs) have been thoroughly studied.

Sn, Ba, Ag, Cu, Pb, Cr, Zn and Ni show increasing trends already during the period 1900 - 1950 (notably Pb, Zn and Ni, with 9.4, 9.3 and 9.2% increase per decade, respectively) owing to the early urbanization and industrialization of this region, and likely associated with the operation of metal smelters. Ba contamination is attributed to offshore barite dumping from oil-drilling operations (Huh, 1996).

The largest increments in metal concentrations were observed during the 1950 – 1975 period (Hg, Sn, Ag, Pb and Cu, with 28.4, 27.8, 16.7, 15.6 and 12.3% increase per decade, respectively). Although observations are mainly focused on point sources (such as waste water treatment plants and military installations), the large contamination increasing trends are clearly related to the parallel increase in population and industrialization, including sources associated with i) urbanization, such as municipal wastewaters, urban runoff and gasoline combustion, ii) industrial development, such as metal smelters, power plants and production of fertilizers and pesticides, iii) oil exploitation, including oil platforms, refineries and oil spills, iv) port activities, such as dredge spoil dumping and ship building facilities, and v) mining activities in the catchments.

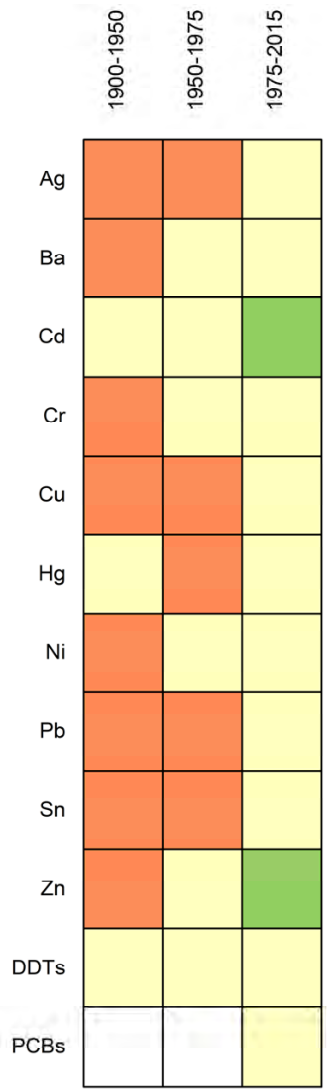


Figure LME03.5: Temporal trends by contaminant and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

In the recent period (1975 – 2015) most contaminants showed no significant trends, except for large decreasing trends for Zn and Cd (18.2 and 16.7 % decrease per decade, respectively: Figures LME03.3 and 4). Although data points extend to 2001, the actual period covered for each contaminant depends on the statistical constraints of the analysis (i.e. at least 3 times series, and at least 5 data points per period). This overall decrease is clearly attributed to the improvement in waste water treatment processes, interdiction of specific practices (e.g. production of leaded gasoline and DDTs) and enhanced source controls, such as urban and industrial waste management. Overall, this LME shows clear signs of recovery, in general attributed to better environmental and coastal zone management practices.

The issue of non-point sources deserves attention. On one hand, as soils in the region are contaminated, watershed erosion and runoff become a diffuse contamination source, so contaminants reach the coastal zone even if the original contamination sources cease releases. Therefore, environmental records register delayed contamination signals, most likely reflecting a slow transport of contaminated particles from the catchments. On the other hand, some coastal sites close to Los Angeles and San Francisco are “hot spots” of contamination, and become sources of contamination to more distant sites, by remobilization/resuspension and transport/-dispersion of older residues. This is a typical “legacy” problem and attention must be maintained in the future.

Although this is a well-studied LME, it is evident that efforts are needed to better represent its coastline, notably to the south (e.g. Tijuana and the Baja California Peninsula) and the north (e.g. Seattle). It is interesting that, in our review, the latest reported core was collected in the late 1990s. Obviously, newer studies are needed to update the state of the more contaminated areas and to learn about remobilization and dispersion processes of contaminants. More contaminants, now analyzed with better techniques, should also be included in new studies.

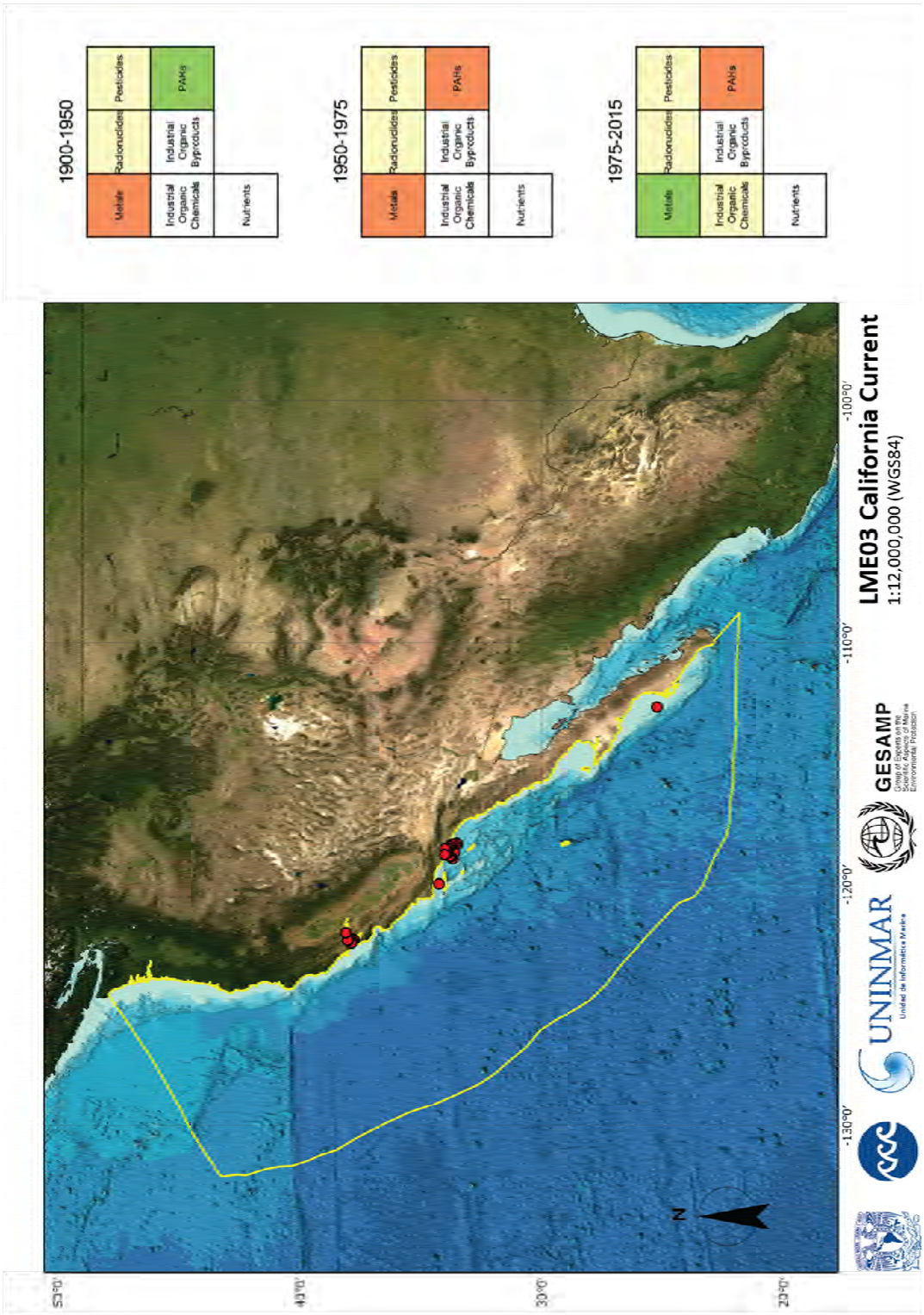
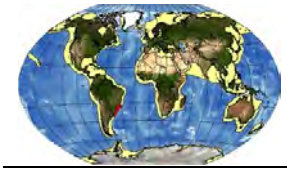


Figure LME03.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.



LME15
South Brazil Shelf

1. INTRODUCTION

The South Brazil Shelf LME extends from 22°-34°S along the South American Atlantic coast, from the Brazilian states of Rio de Janeiro in the north, up to Rio Grande do Sul in the south. This LME has a surface area of about 565,500 km^2, and 1.47% is protected. Major rivers and estuaries include Patos - Mirim and Cananeia - Paranaguá Lagoon systems, Ribeira de Iguape and Paraiba do Sul rivers, and the Santos/São Vicente estuarine complex. The South Brazil Shelf LME has a mixed warm-temperate climate, and a moderately high productivity, sustained by a relatively intense coastal shelf-edge and offshore upwelling, and various terrigenous sources such as the Patos-Mirim Lagoon system and La Plata River plume.

Artisanal and commercial fishing, agriculture, tourism and shipping are important activities in this LME. It sustains about half of Brazil's commercial fisheries yield: sardines is the most important group in shelf catches, and also includes important demersal species such as the whitemouth croaker, the argentine croaker and other *sciaenids*, the skipjack and yellowfin tuna, *penaied* shrimps, and aquaculture produced shrimp, oysters, mussels and clams (Heileman and Gasalla, 2017).

The littoral area of the LME covers a total of 214,895 km^2. In 2010 this area accounted for 57 million population, a population density of 264 persons per km^2, and about 10% of coastal population lives in rural areas (TWAP, 2015). The major contamination issues are usually associated with the development of coastal urban centers, industries, tourism, agriculture and shipping. The LME is influenced by the presence of two large metropolitan areas, located in or close to the coastal area: São Paulo (12 million inhabitants) and Rio de Janeiro (6 million inhabitants).

Coastal areas downstream of urban centers are affected by untreated domestic sewage and industrial effluents, carrying heavy metals, organic compounds and nutrients. Eutrophication and occurrence of dead zones are reported in four regions of the South Brazil Shelf LME (Patos Lagoon, Guanabara Bay, Rodrigo de Freitas and Conceição lagoons). The country's main sea terminal, accounting for around 55% of all oil transported in Brazil, is situated on the São Paulo coast, promoting chronic oil contamination in nearby areas. Dredging (for shipping or rice culture) and deforestation have resulted in increased soil erosion and siltation of coastal zones.

Guanabara and Sepetiba bays, in Rio de Janeiro, are highly polluted by trace elements owing to industrial activities, and the changes in sediment loading generated by river basin activities and port operation. Agricultural run-off is a significant cause of pollution in some areas such as Guanabara, Santos, Paranaguá Bays and Patos Lagoon. Several beaches are not recommended for swimming (Heileman and Gasalla, 2017 and references therein).

2. DATABASE INFORMATION

In this LME, from 20 papers selected, 16 were used for analysis after full content screening. Information on 70 time series was extracted, meeting the requirements of the methodology. The selected papers contained information on 4 families (Figure LME15.1), including 19 contaminants (Figure LME15.2). As examples, two time series are shown in Figures LME15.3 and LME15.4.

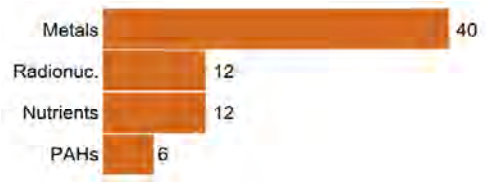


Figure LME15.1: Number of time series by contaminant family.

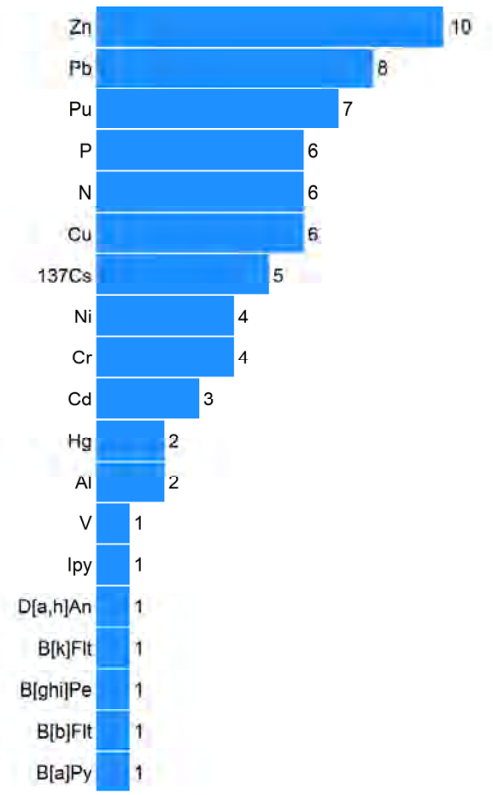


Figure LME15.2: Number of time series by contaminant.

3. TREND ANALYSIS

Trends of the time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present. A summary of the trends for each period and for the contaminants found in this LME is shown in Table LME15.1 and Figure LME15.5. In this case, 27 trends were analyzed, and 6 were statistically significant ($p<0.05$).

The trends are expressed in units of percentage of variation per decade. During the period 1900-1950, trends were neutral for all contaminants. During the period 1950-1975, trends were neutral for most elements, but increasing for Cr and Pu. During the period 1975-2015, trends were increasing for N, ^{137}Cs, Pu and P, but neutral for the rest of the contaminants

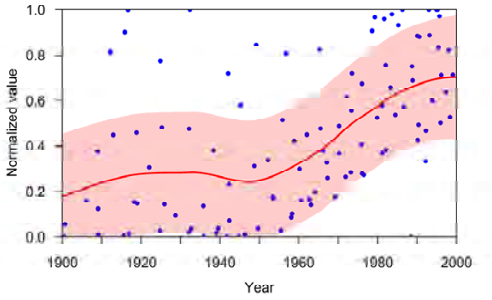


Figure LME15.3: Cu time series. The brown line is a smoothed mean, and the shaded area is the 95% confidence band.

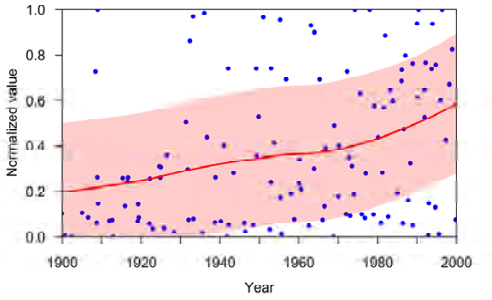


Figure LME15.4: Pb time series. The brown line is a smoothed mean, and the shaded area is the 95% confidence band.

Table LME15.1: Summary of temporal trends by contaminant and period analyzed.

Contaminant	1900-1950	1950-1975	1975-2015
N	neutral	neutral	+20.9
^{137}Cs	n.d.	neutral	+13.9
Pu	neutral	+21.3	+8.2
P	neutral	neutral	+8.1
Cd	n.d.	n.d.	neutral
Cr	neutral	+21.9	neutral
Cu	neutral	neutral	neutral
Ni	neutral	neutral	neutral
Pb	neutral	neutral	neutral
Zn	neutral	neutral	neutral

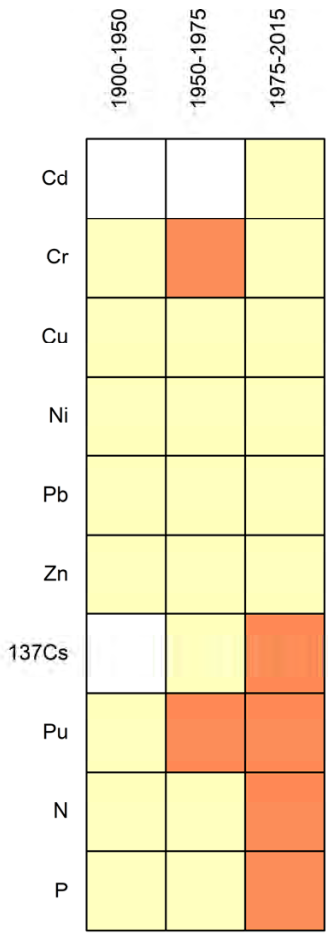


Figure LME15.5: Temporal trends by contaminant and period. Brown= increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

Trend analysis was also carried out by aggregation of all contaminants in the same family for the same periods. Families were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminant families found in this LME is shown in Figure LME15.6. In this case, 12 trends were analyzed and 7 were found to be statistically significant ($p<0.05$). Although not enough data points were available for trend analysis of individual PAHs, their aggregation allowed trend analysis of the PAHs family.

During the period 1900-1950, neutral trends were observed for Metals and Radionuclides, and increasing trends were observed for PAHs and Nutrients. During the period 1950-1975, decreasing trends were obtained for PAHs, neutral trends were observed for Metals and Nutrients, and increasing trends were observed for Radionuclides. During the period 1975-2015, decreasing trends were calculated for PAHs, neutral trends were observed for Metals and increasing trends were observed for Radionuclides and Nutrients.

4. ASSESSMENT

Information available on this LME was mostly derived from the two largest Brazilian cities (São Paulo and Rio de Janeiro), and the Patos lagoon in the southern coast, but calculated trends were mostly based on publications from the Rio de Janeiro coastline. Therefore, results are not representative of the whole LME. Rio de Janeiro City is the second most important metropolitan area of the country and contains large sources of contaminants owing to urbanization and industrialization, including

untreated sewage, gasoline combustion, smelting operations, mining and agricultural runoff (Baptista-Neto et al., 2013).

Guanabara Bay, next to Rio de Janeiro City, is a highly impacted estuary that receives a large fraction of these contaminants from thousands of industries in the bay catchment through more than 100 rivers and channels (Godoy et al., 1998). The main contamination sources are i) the petroleum industry, including a large refinery and several oil terminals, one of which caused a large oil spill in 2000, ii) a chlor-alkaline plant with Hg cells, closed in 2001, iii) a tannery and a chromium oxide plant, both closed in 1998, iv) non-treated sewage, v) a naval metal-mechanic industry (shipyard), and vi) the harbor. To the south, Sepetiba Bay receives waste from hundreds of industries, specially a metallurgical plant and steel production facilities (Gomes et al., 2009).

The largest number of time series analyzed corresponded to metals, followed by nutrients and radionuclides. Little information was found on organic contaminants and only trends of the PAHs family could be determined. Regarding metals, the attention has been focused on Pb and Zn, owing to the industrial impact in Sepetiba and Guanabara bays. The most general conclusion of trend analysis in this LME is that, except for PAHs, all trends are either increasing or neutral, reflecting the on-going socio-economic development of this region. Increasing trends for P were already observed during the periods 1900-1950 and 1975-2015, likely reflecting the increase of urban and agricultural waste inputs. The largest increasing trends were observed during 1950-1975 for Pu (from global fallout) and Cr (+21.9 %), the latter likely reflecting the development of the metallurgy industry in Sepetiba Bay (especially Ingá Metals, operating during 1958 – 2008; Gomes et al., 2009).

When contaminant families are considered, it is interesting to note that PAHs trends are increasing only for the period 1900-1950, but decreasing for the other periods. This is attributed by Mauad et al. (2013) to the effect of the oil crisis in the 70s, the modernization of fuel combustion in light vehicles and increasing environmental policies in Brazil, but further research on the issue is needed. Regarding radionuclides, global fallout reached a maximum in the 1960s. The persistence of increasing trends in the period 1975 – 2015, when atmospheric fallout was decreasing worldwide, is caused by the input of contaminated surface soils from the catchment by runoff. The unexpected different trends of the bomb fallout derived ^{137}Cs and Pu, in the 1950 – 1975 period, was likely due to the limited amount of data points available for both radionuclides, and their different geochemical behavior in the marine environment and the related catchments (e.g. higher solubility of ^{137}Cs in seawater).

During this study, little information was found on the reconstruction of coastal contamination close to São Paulo City. This is not surprising, as São Paulo is crossed by the Tietê River, a tributary of the Paraná River, flowing into the Plata River. The most contaminated region from São Paulo State is the area around Santos harbor and city, also influenced by the Cubatão industrial area. Also, we found no information on contamination from Pesticides, Industrial organic chemicals and Industrial organic byproducts. Further research should be focused on unexplored regions and/or contaminants.

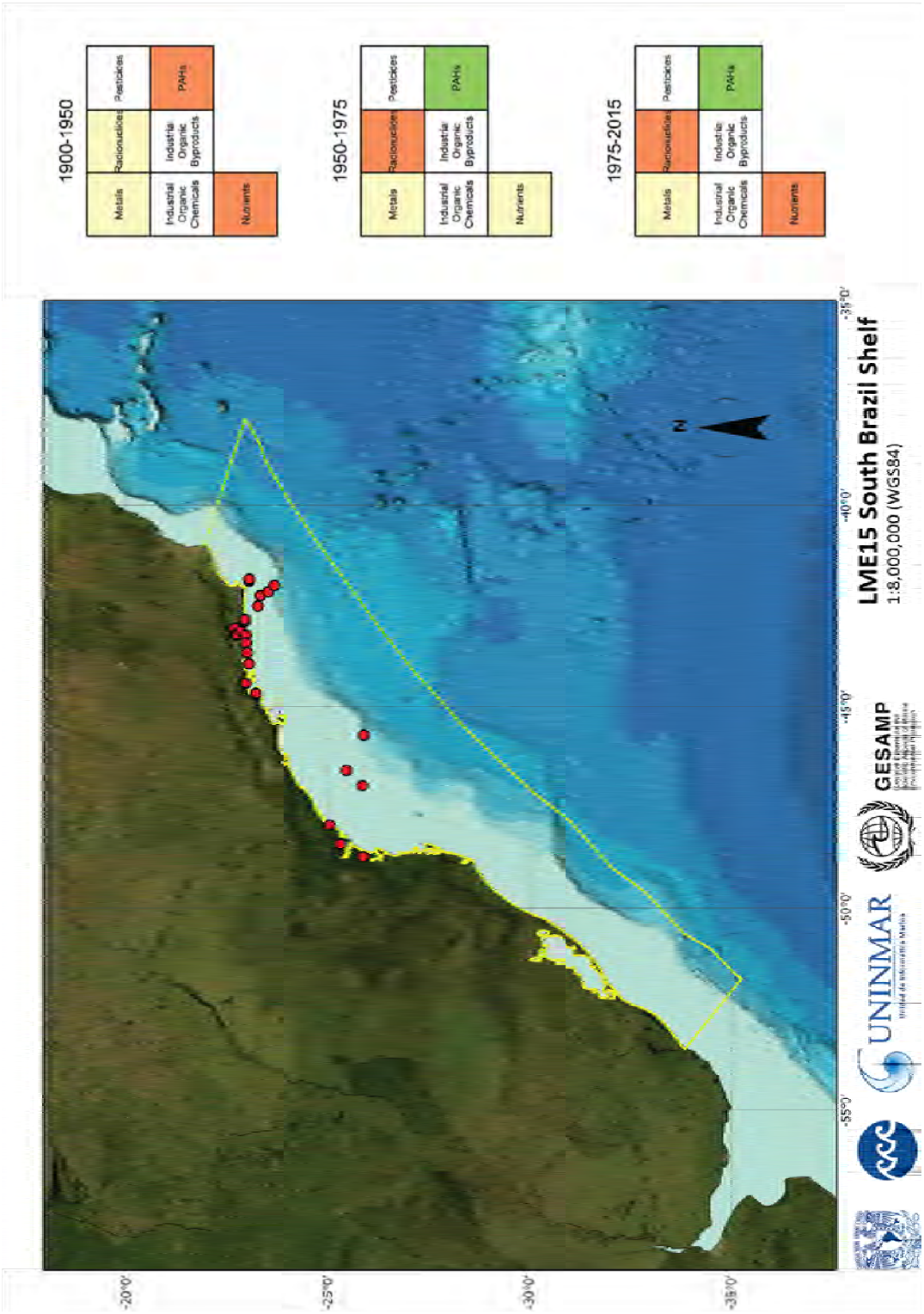
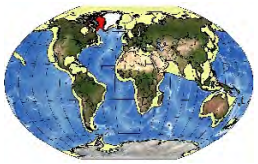


Figure LME15.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.



LME18
Canadian Eastern Arctic - West Greenland

1. INTRODUCTION

The Canadian Eastern Arctic – West Greenland LME extends along the coast of West Greenland and eastern Baffin Island. It includes the northern part of the Labrador shelf, Davis Strait, Baffin Bay, Lancaster Sound and adjacent inlets such as Peel Sound and Gulf of Boothia. The reported extent of the LME ranges between 359,422 km^2 (TWAP, 2015) and 1.4 million km^2 (PAME - Protection of the Arctic Marine Environment, 2016). Most of the LME is ice-covered in winter and clears of ice in summer, excepting along southwestern Greenland and some inner parts of eastern Canadian archipelago and northern Baffin Bay (PAME 2013, 2016). Atlantic waters reach the LME through Baffin Bay and Lancaster Sound, and nutrient rich Pacific waters through the Canadian archipelago and Nares Straight, contributing to the relatively high primary production in the LME.

Important fish species in this LME includes polar cod, Atlantic cod, Greenland halibut, capelin, and the squid *Gonatus fabricii*, although the most important species group in terms of shelf catches for recent years is the northern prawn (*Pandalus borealis*), representing more than two-thirds of the total catch (TWAP, 2015).

The coastal area includes the north and eastern shores of Nunavut and western Greenland, stretching over 743,645 km^2. A population of 70,000 inhabitants with density of 9 persons per 100 km^2 in 2010, and about 81% of coastal population lives in rural areas (TWAP, 2015).

The waters of the LME are slightly contaminated. Higher levels of lead and zinc have been reported in sediments and biota in the fjord at Maarmorilik, West Greenland, a legacy from past mining, and lead in Søndre Strømfjord sediments, from both European and Russian sources. The industries of West Greenland include fish processing, gold, uranium, iron and diamond mining, handcrafts, hides and skins, and small shipyards (Aquarone et al., 2017). Key stressors include ocean-based pollution and long-range atmospheric transport (TWAP, 2015). The cold Arctic climate creates a sink for long range transported pollutants (such as Hg and POPs), and the already high levels of mercury in the Arctic are not declining despite significant emissions reductions in Europe and North America.

2. DATABASE INFORMATION

In this LME, 9 papers were selected and were used for analysis after full content screening. Information on 66 time series was extracted, meeting the requirements of the methodology. The selected papers contained information on 9 contaminants (Figure LME18.1) included in only 2 families (Figure LME18.2: Metals and Radionuclides). As examples, two time series are shown in Figures LME18.3 and LME18.4.

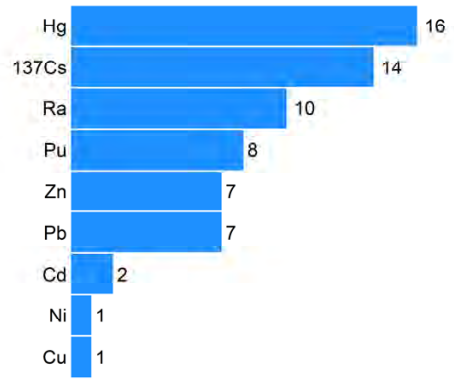


Figure LME18.1: Number of time series by contaminant.

Figure LME18.2: Number of time series by contaminant family.

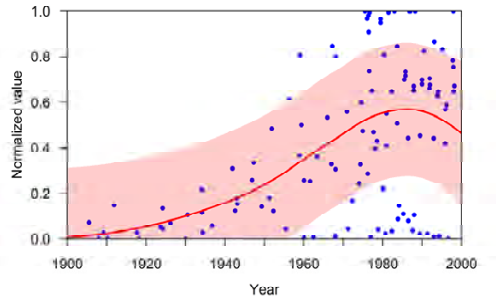


Figure LME18.3: Zn time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

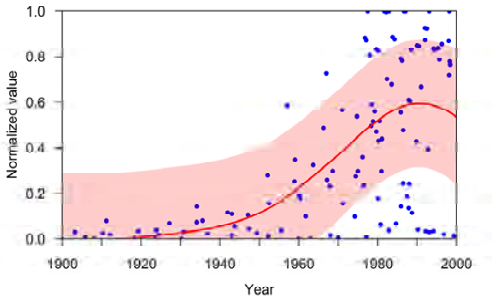


Figure LME18.4: Pb time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

3. TREND ANALYSIS

Trends of the selected time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminants found in this LME is shown in Table LME18.1 and Figure LME18.5. In this case, 18 trends were analyzed, and 4 were found to be statistically significant ($p<0.05$).

The trends are expressed in units of percentage of variation per decade. During the period 1900-1950, trends were neutral for Hg, ^{137}Cs and Ra, and increasing for Pb, Zn and Pu. During the period 1950-1975, neutral trends were observed for Hg, Pb, Zn and ^{137}Cs, and increasing trends were observed for Pu and Ra. During the period 1975-2015, trends were neutral for Hg, Pb, Zn, ^{137}Cs, Pu and Ra.

Trend analysis was also carried out by aggregation of all contaminants in the same family during the same periods. Families were analyzed if at least 3 time series were found, and for periods with at least 5 data points. A summary of the analyzed trends during each period and for all contaminant families found in this LME is shown in Figure LME18.6. In this LME, 6 family trends were analyzed, of which only 1 was found to be statistically significant ($p<0.05$).

During the period 1900-1950, neutral trends were observed for both Metals and Radionuclides. During the period 1950-1975, neutral trends were observed for Metals, and increasing trends were observed for Radionuclides. During the period 1975-2015, neutral trends were observed for both Metals and Radionuclides.

4. ASSESSMENT

Seven of the eight analyzed papers reported contamination from West Greenland, whereas information for other areas only included ^{137}Cs. Although there are few relevant point contamination sources in the LME, special attention has been paid to an abandoned Pb - Zn mine (1973-1990) in the central coast of West Greenland (Black Angel Mine, Maarmorilik), and a site where plutonium was released by a nuclear weapons accident (Thule, northwest Greenland). Analysis indicates a mixture of both local and global contamination sources (long-range atmospheric transport). Except for the sediment cores collected close to Thule and Maarmorilik, the articles concluded that the main contamination source in the region is mainly atmospheric, such as global fallout for ^{137}Cs.

Table LME18.1: Summary of temporal trends by contaminant and period analyzed.

Contaminant	1900-1950	1950-1975	1975-2015
Hg	neutral	neutral	neutral
Pb	+1.2	neutral	neutral
Zn	+4.6	neutral	neutral
^{137}Cs	neutral	neutral	neutral
Pu	neutral	+18.5	neutral
Ra	neutral	+27.3	neutral

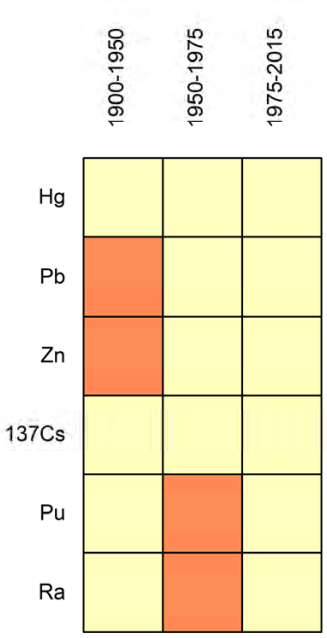


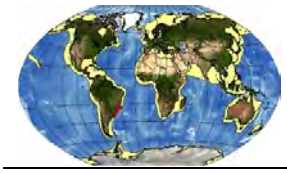
Figure LME18.5: Temporal trends by contaminant and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

Significant increasing trends were observed for Pb (+1.2%) and Zn (+4.6%) during the period 1900 – 1950, before the operation of the Black Angel mine (Figures LME18.3 and 4). This suggests that some operations were taking place previously, although atmospheric transport cannot be discarded. A large Pu increasing trend (+18.5%) occurred, as expected, during the period 1950 – 1975 owing to nuclear weapons fallout, and levels remained stable afterwards. In keeping with the absence of relevant sources in the region. ^{226}Ra concentrations ranged from 10 – 53 Bq kg^{-1}, which is a typical range of natural levels. Therefore, the large increasing trend calculated for 1950 – 1975 period (27.3%) can only be attributed to a statistical false positive, as was confirmed from a visual inspection of the data.

The neutral trends observed for all contaminants during the period 1975 – 2015 are indicative of the stabilization of the contamination processes. Indeed, although some contamination sources ceased (such as ^{137}Cs from global fallout, and the ending of the mining operations in Maarmorilik), both catchment soils and remobilized contaminated sediments continued to be dispersed and contribute to the sedimentary record of contamination. Although the smoothed mean shows slightly decreasing trends for Zn and Pb (Figures LME18.3 and 4), these were not significant, likely because there is a maximum within the period 1975 – 2015 and therefore it includes both an increasing and a decreasing time interval. A more detailed analysis should be needed in this LME.

This area is part of AMAP where there is regular monitoring of biota and air. The analysis of selected sedimentary archives could effectively complement the on-going monitoring programmes. As it is likely visited by oceanographic research cruises periodically, it is recommended to extend the study of contamination records to non-explored areas, especially in the Canadian coast of the Davis Strait and Baffin Bay, and the Gulf of Boothia (northern Canada). As the accumulation of relatively volatile organic pollutants in polar ecosystems through global fractionation is well-known, on-going monitoring should be complemented with environmental records of Pesticides, Industrial organic chemicals, PAHs and Industrial organic byproducts.

Figure LME18.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.



LME23
Baltic Sea

1. INTRODUCTION

The Baltic Sea LME is the world's largest brackish water body, covering an area of about 390,000 km^2 and 2.21% is protected. The LME catchment area of about 1.7 million km^2 (four times larger than its surface area) belongs to the riparian countries of Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Poland, Russia and Sweden; and the non-coastal countries of Belarus, Czech Republic, Slovakia and Ukraine. The LME receives freshwater from several ,rivers while saltwater enters from the North Sea along the bottom of the narrow straits between Denmark and Sweden.

The LME is characterized by a temperate climate and a high productivity. The biomass of the fish community in the Baltic Sea is dominated by cod, herring and sprat; commercially important marine species are sprat, herring, cod, various flatfish and salmon; and other important target species are sea trout, pike-perch, whitefish, eel, bream, perch and pike (Heileman and Thulin, 2017).

The coastal area stretches over 567,489 km^2. In 2010, it was reported to have a population of 34.3 million, with a density of 60 persons per km^2, with about 29% of coastal population living in rural areas (TWAP, 2015). Contamination in the Baltic Sea LME is generally severe because of limited water exchange, and run-off from its vast, highly populated and industrialized catchment area. The most relevant environmental issue is eutrophication, mostly due to non-point source agricultural pollutants and direct discharges by industries and municipalities. This has been associated with hypoxic conditions and occurrence of harmful algal blooms over widespread areas of the LME.

Reports of pollution in the Baltic Sea LME include (a) suspended solids, resulting from the increased amounts of phytoplankton in eutrophic areas and increased coastal erosion in southern and eastern areas of the Baltic Sea LME; (b) heavy metals such as mercury, cadmium, zinc, copper and lead in various areas (e.g. the Bay of Bothnia, the eastern Gulf of Finland, the Kattegat, the Gulf of Riga and the central basin of the Baltic Sea); and (c) persistent organic pollutants, such as pesticides, polychlorinated compounds and dioxins. Although most of the oil input into the Baltic Sea comes from dilute but persistent land-based sources, acute oil spills associated with vessels and offshore platforms are frequent (an average of about three accidents each year; Heileman and Thulin, 2017) and have the most important ecological effects.

2. DATABASE INFORMATION

In this LME, 38 papers were selected, of which 33 could be used for analysis after full content screening. Information on 422 time series was extracted, meeting the requirements of the methodology. The selected papers contained information on the 7 contaminant families (Figure LME23.1), including 32 contaminants (Figure LME23.2). As examples, two time series are shown in Figures LME23.3 and LME23.4.

3. TREND ANALYSIS

Trends of the selected time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminants is shown in Table LME23.1 and Figure LME23.5. In this case, 68 trends were analyzed and 38 were found to be statistically significant ($p<0.05$).

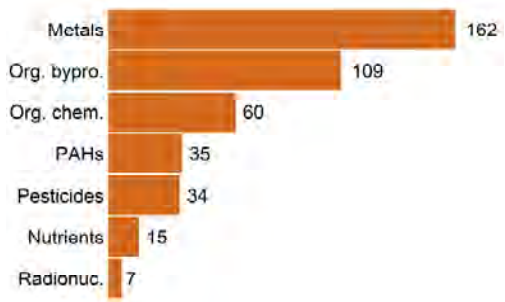


Figure LME23.1: Number of time series by contaminant family.

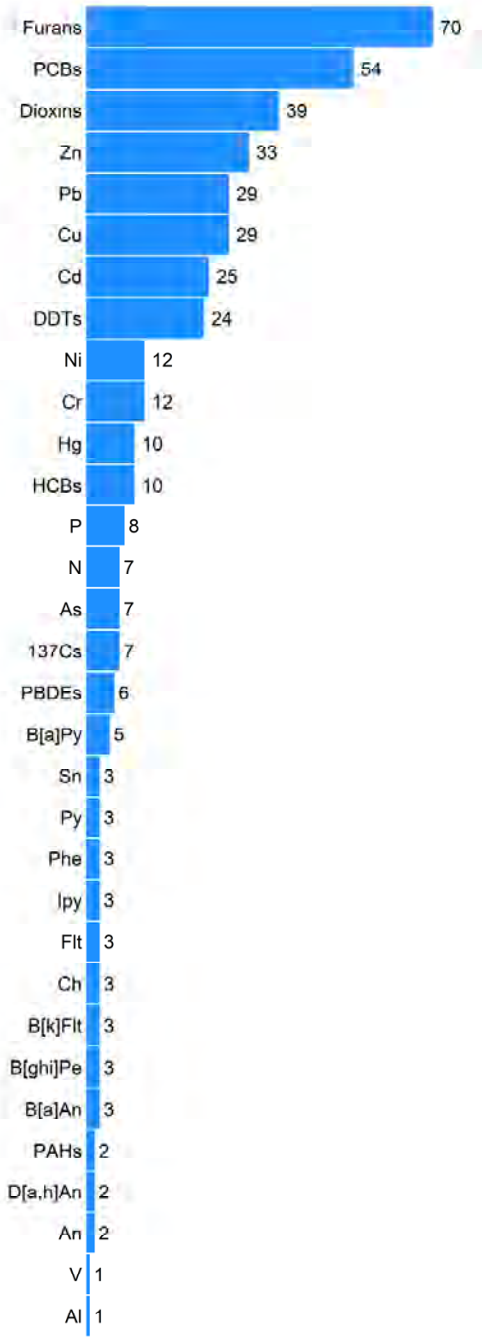


Figure LME23.2: Number of time series by contaminant.

The trends are expressed in units of percentage of variation per decade. During the period 1900-1950, neutral trends were observed for N, Ni, B[a]Py, Phe, Cd, Hg and DDTs, and increasing trends were observed for P, ^{137}Cs, Dioxins, Cu, Zn, Pb, furans and PCBs. During the period 1950-1975, neutral trends were observed for As, Cr, Ni, PBDEs, B[a]Py, Phe, Cd, Flt, Py, B[k]Flt, B[a]An, Ch, Ipy and B[ghi]Pe, and increasing trends were observed for N, P, ^{137}Cs, dioxins, Cu, Zn, Pb, Hg, DDTs, furans, PCBs and HCBs. During the period 1975-2015, decreasing trends were calculated for Cu, Cd, Zn, Pb, Hg, DDTs, Furans, PCBs, Flt, Py, HCBs, B[k]Flt, B[a]An, Ch, Ipy and B[ghi]Pe, neutral trends were observed for As, Cr, Ni, Sn, ^{137}Cs, PBDEs, dioxins, B[a]Py and Phe, and increasing trends were observed for the nutrients N and P.

According to the methodology, trend analysis was also carried out by aggregation of all contaminants in the same family during the same periods. Families were analyzed if at least 3 time series were found and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminant families is shown in Figure LME23.6. In this case, of the 21 trends analyzed, 19 were found to be statistically significant ($p<0.05$).

During the period 1900-1950, neutral trends were observed for Pesticides and increasing trends were observed for the other contaminant families. For the 1950-1975 period, increasing trends were observed for all contaminant families and for 1975-2015, increasing trends were observed for Nutrients, neutral trends were observed for Radionuclides, and decreasing trends were observed for the other families.

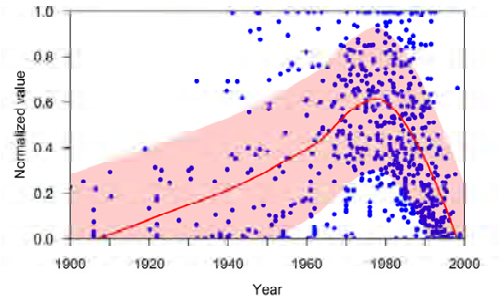


Figure LME23.3: PCBs time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

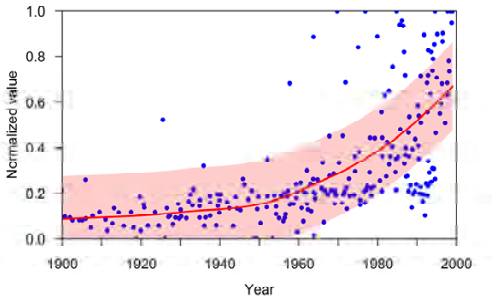


Figure LME23.4: P time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

Table LME23.1: Summary of temporal trends by contaminant and period analyzed.

Contaminant	1900-1950	1950-1975	1975-2015
N	neutral	+9.2	+22.8
P	+1.4	+7.3	+12.4
As	n.d.	neutral	neutral
Cr	n.d.	neutral	neutral
Ni	neutral	neutral	neutral
Sn	+n.d.	n.d.	neutral
^{137}Cs	+6.5	+18.0	neutral
PBDEs	n.d.	neutral	neutral
Dioxins	+1.4	+7.2	neutral
B[a]Py	neutral	neutral	neutral
Phe	neutral	neutral	neutral
Cu	+5.0	+8.5	-10.4
Cd	neutral	neutral	-11.7
Zn	+6.2	+9.3	-11.8
Pb	+4.9	+15.6	-15.4
Hg	neutral	+24.9	-16.2
DDTs	neutral	+16.9	-24.0
Furans	+1.3	+10.8	-24.6
PCBs	+6.7	+14.7	-27.4
Flt	n.d.	neutral	-43.3
Py	n.d.	neutral	-44.0
HCBs	n.d.	+17.0	-46.6
B[k]Flt	n.d.	neutral	-48.6
B[a]An	n.d.	neutral	-51.2
Ch	n.d.	neutral	-51.3
Ipy	n.d.	neutral	-53.2
B[ghi]Pe	n.d.	neutral	-54.1

n.d. = not enough data for trend analysis

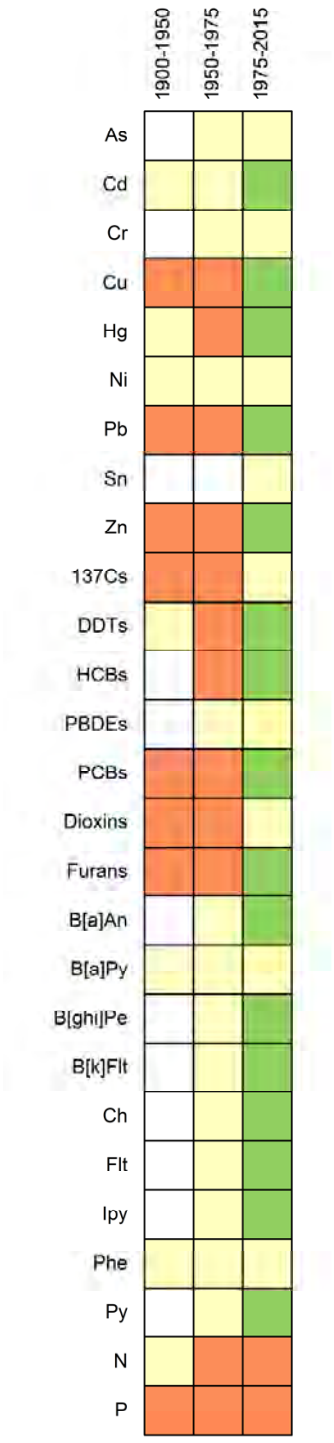


Figure LME23.5: Temporal trends by contaminant and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

4. ASSESSMENT

The Baltic Sea is a paradigmatic LME because of its deterioration during the early 20th century and of the mainly successful national efforts and international agreements to improve its environmental quality (notably the Baltic Marine Environment Protection Commission - Helsinki Commission - HELCOM). The scientific interest in the riparian countries, mostly well developed and very likely encouraged by national legislation and international agreements, makes this one of the best studied LMEs, allowing to draw solid regional conclusions. Overall, 422 time series included information on 47 of the 51 contaminants considered and the 7 contaminant families. In total, 5,651 data points were analyzed. Although special attention has been paid to large urban and industrial areas, studies have also been carried out in open sea waters from most areas.

This LME has undergone profound changes during the 20th century, transformed from a clean to a severely polluted water body. Although naturally occurring, anoxic waters expanded and promoted the presence of laminated sediments (Borg and Jonsson, 1996), facilitating the establishment of robust chronologies and the detailed discussion of sedimentary processes. Results confirmed the usefulness of dated sediment cores to establish baseline values (e.g. Pempkowiak, 1991), and the importance of resuspension and transport of contaminated sediments in the basins (e.g. Vaalgamaa and Korhola, 2004). Although several papers presenting contaminant data in biota were included in the analysis, their usefulness was limited because of the usually short time span of these studies in comparison to dated sediment cores.

As in other LMEs, the Metals family captured the largest attention with 162 time series. Most reported metals are Zn (33 time series), Pb (29), Cu (29) and Cd (25). Metal contamination is largely attributed to traffic (mainly leaded gasoline), corrosion of constructions, sewage, waste incineration and coal combustion. Other specific sources include the timber industry, because of the use of Cr as a preservative (Vaalgamaa and Korhola, 2004). Zn, Cu and Pb showed increasing trends already in the period 1900 – 1950 (6.2, 5.0 and 4.9 % increment per decade respectively), but largest increases were observed for Hg and Pb during the 1950 – 1975 period (24.9 and 15.6 % increment per decade, respectively). However, during the most recent period 1975 – 2015, the contamination trends for the metals have been

markedly decreasing (more than 10% decrease per decade for Cd, Cu, Hg, Pb and Zn) or neutral (As, Cr, Ni and Sn). Large metal contamination occurred after WWII, mainly through atmospheric deposition, but since the 1970s, atmospheric releases diminished because of pollution prevention policies and improvements in industrial processes and urban sewage plants (Sternbeck and Östlund, 2001).

A similar picture arises from the analysis of organic contaminants. Many time series belong both to the Organic byproducts (109 time series) and Organic Chemicals (60 time series) families. In fact, when individual contaminants are inspected, the three most reported contaminants in this LME are organic contaminants (furans, PCBs and dioxins).

During the 1900 – 1950 period, trends were neutral (DDTs, and the PAHs B[a]Py and Phe) or slowly increasing for dioxins and furans, although the PCBs trend was already increasing at 6.7 % per decade. Large increases (> 10% per decade) occurred during the 1950 -1975 period for HCBs, DDTs, PCBs and furans. However, during the subsequent 1975 – 2015 period, the decreasing trends of these contaminants are striking, for example, 46.6 % decrease per decade for HCBs and more than 50% decrease per decade for many PAHs, indicating the effectiveness of reduction strategies of these contaminants in the region. However, trends are still neutral for PBDEs, dioxins and the PAHs B[a]Py and Phe.

The described temporal trends can be well understood from the development history in the region. For example, PCBs were largely used as coolants, insulating fluids, lubricating fluids, plasticizers, pesticides, reactive flame retardants and sealants in numerous industries. Their worldwide production reached a peak in the 1970s (Blanz et al., 1999), but several countries banned their production and use since this same decade. In Sweden they were mainly used as fungicides for wood protection, for textile treatment, and to control slime in pulp mills, but their production and use was partially banned in 1973 and fully banned in 1978 (Kjeller and Rappe, 1995). Since then, environmental levels decreased, as shown by the evolution of PCBs normalized concentrations in sediments, with a peak value in the late 1970s (Figure 23.3).

One of the main problems of the LME is eutrophication, largely aggravated by the increased use of mineral fertilizers in intensive agriculture and farming and by urban sewage, including P compounds used in detergents, although agriculture could still be the largest contributor (Vaalgamaa and Conley, 2008; Müller et al., 1980). Unlike other contaminants, nutrients show increasing trends along the whole period (Figure LME23.4). Trends during the first two periods (1900 – 1950 and 1950 – 1975) are neutral or below 10 % increase per decade for N and P, but large increases are observed for the most recent period (1975 – 2015: 22.8 and 12.4 % increase per decade, respectively).

This information is in good agreement and complements the latest HELCOM assessment (HELCOM, 2017), showing that contaminant levels are stable or decreasing, with only a few time series displaying an upward trend. However, although signs of improvement in the state of the Baltic Sea are seen in some cases, contamination levels are still of concern and the goals set by the Baltic Sea Action Plan have not yet been reached. HELCOM concludes that “some measures already put into operation have not been in place long enough to have an effect. For measures such as reduction of nutrient loads, it will take several decades before full effects can be measured in the environment”.

In general, the evolution of contaminant trends in the region is encouraging, as most contaminants peaked in the late 1970s and decreased during the recent decades, when trends were either decreasing or neutral, except for N and P. This information should be useful for decision makers in the region, and might help to focus future policies and actions, such as enforcing more and better sewage and industrial treatment plants, very effective in the Neva River (Vallius and Leivuori, 1999) and the Stockholm region (Sternbeck and Östlund, 2001).

In any case, the overall message is promising: the combination of environmental policies and public awareness can, indeed, reduce contamination at the ecosystem level. No doubt many problems persist, but in general, pollution is decreasing and the effort should be maintained to protect the environment. In the papers reviewed in this work, still some regions show limited (Gulf of Bothnia) or lacking (Gulf o Riga) information, and further research is recommended. Some of the studies were performed a few decades ago, and revisiting sites might shed more light on the on-going trends.

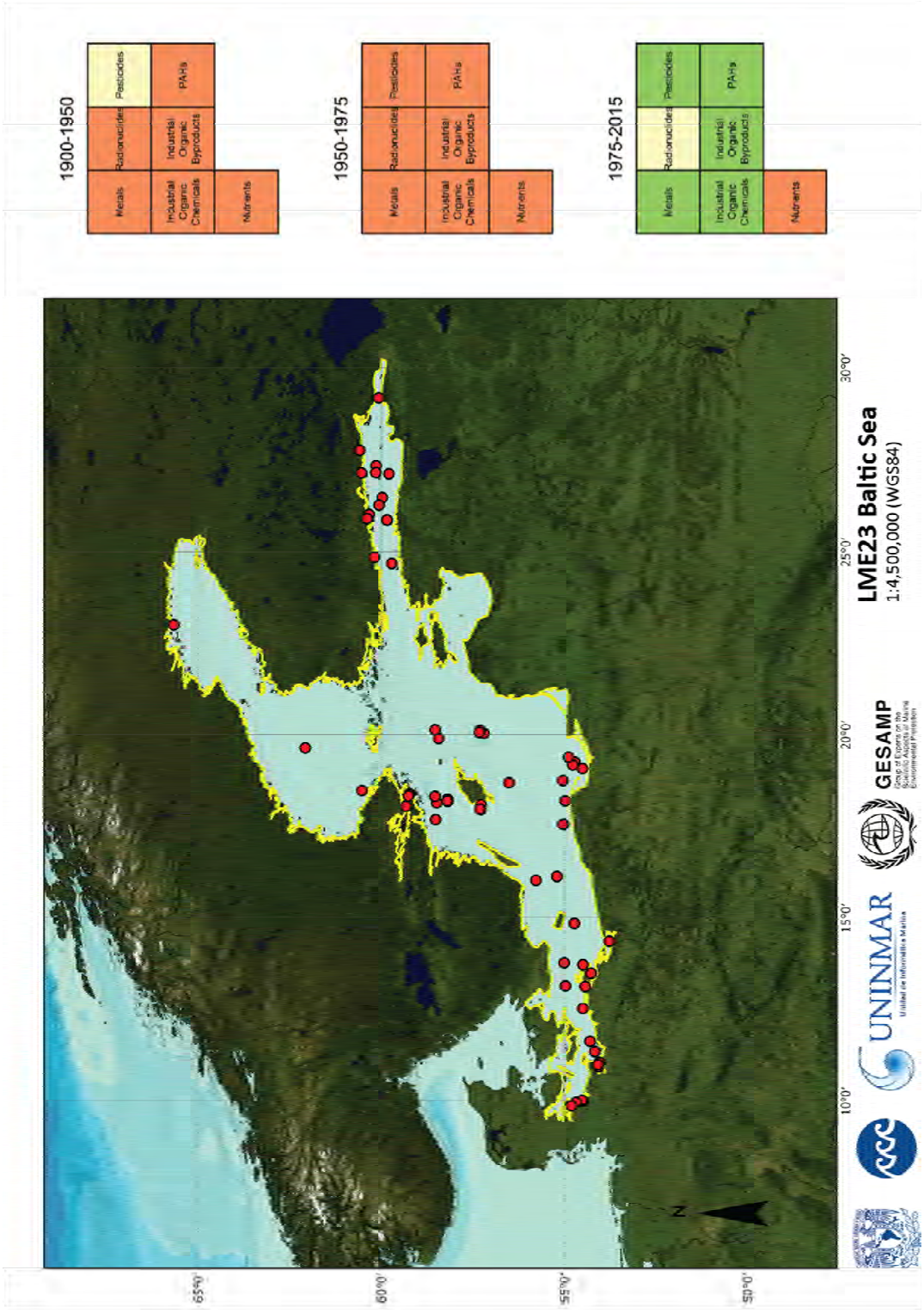
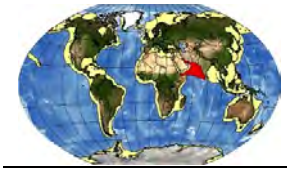


Figure LME23.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.



LME32
Arabian Sea

1. INTRODUCTION

The Arabian Sea LME lies in the northwestern Indian Ocean, between the Arabian Peninsula and India, and is bounded by Bahrain, India, Iran, Iraq, Kuwait, Oman, Pakistan, Qatar, Saudi Arabia, Somalia, United Arab Emirates and Yemen. It covers an area of about 3.9 million km^2, of which 0.21% is protected, contains 1.84% of the world's coral reefs, and receives freshwater run-off from the Indus, the Arvand (Shatt al-Arab) and Tigris rivers.

The Arabian Sea LME is a highly productive ecosystem, influenced by the intense and large-scale seasonal upwelling along the Oman and Somalia coasts, making the Arabian Sea one of the most productive regions of the world's ocean. Despite its high primary productivity, the abundance of coastal pelagic fish is anomalously low and catch of this group is not consistent with other similar world regions. Among the major exploited groups are Indian oil sardine, drums and croakers (Heileman et al., 2017 and references therein).

The coastal area of the Arabian Sea LME stretches over 513,873 km^2. In 2010 the area had a population of 28 million, with a density of 54 persons per km^2, and about 58% of coastal population living in rural areas (TWAP, 2015). Overall, contamination in the Arabian Sea LME is moderate, but severe in hotspots located in the mouths of some rivers (e.g., Tigris, Euphrates, Karun, Hileh and Monds Rivers) coastal areas where domestic and industrial outfalls release vast quantities of untreated sewage and industrial wastes into the sea. Coastal areas are affected by eutrophication and persistent toxic substances due to sewage, industrial effluents and agricultural runoff, carrying fertilizers, heavy metals, chlorinated pesticides, PAHs and organometallic compounds. Throughout much of the LME, the coastal zone is a repository for solid wastes from both land-based sources and ships.

The LME contains one of the world's busiest oil tanker routes, with more than 70% of the oil produced in the northern areas transported through the Arabian Sea. Significant levels of marine hydrocarbon contamination associated with routine operations have been detected around coastal petroleum refineries, offshore oil and gas platforms, tanker loading terminals and shipping localities (Heileman et al., 2017 and references therein).

2. DATABASE INFORMATION

In LME32, the 7 papers available were used for analysis. Information on 63 time series was extracted, meeting the requirements of the methodology. The selected papers contained information on 9 contaminants (Al, Cr, Cu, Hg, Ni, Pb, Zn, ^{137}Cs and PAHs; Figure LME32.1) included in 3 families (Figure LME32.2: Metals, Radionuclides and PAHs). As examples, two time series are shown in Figures LME32.3 and LME32.4.

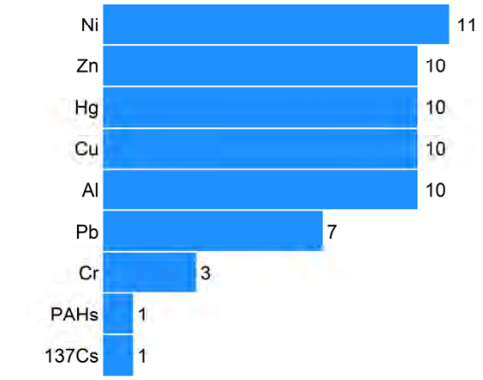


Figure LME32.1: Number of time series by contaminant.

Figure LME32.2: Number of time series by contaminant family.

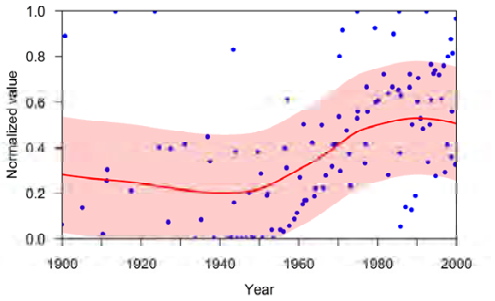


Figure LME32.3: Pb time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

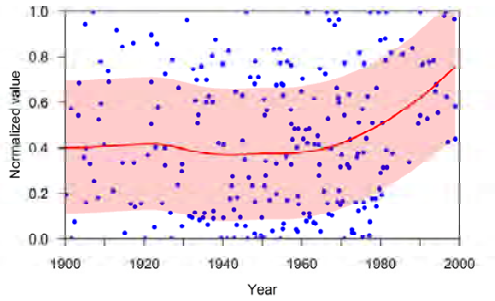


Figure LME32.4: Ni time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

Table LME32.1: Summary of temporal trends by contaminant and period analyzed.

Contaminant	1900-1950	1950-1975	1975-2015
Ni	neutral	neutral	+15.7
Al	neutral	neutral	neutral
Cr	neutral	neutral	neutral
Cu	neutral	neutral	neutral
Hg	neutral	+18.2	neutral
Zn	neutral	neutral	neutral
Pb	-8.2	+21.1	-6.3

3. TREND ANALYSIS

Trends of the selected time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminants found in this LME is shown in Table LME32.1 and Figure LME32.5. In this case, 21 trends were analyzed and 5 were found to be statistically significant ($p<0.05$).

The trends are expressed in percentage of variation per decade. During the period 1900-1950, the trends were decreasing for Pb, and neutral for the other contaminants. Increasing trends were observed for Hg and Pb, while those for the other contaminants were neutral, between 1950 and 1975. During the period 1975-2015, increasing trends were observed for Ni, decreasing trends were observed for Pb and the other contaminants showed neutral trends.

Trend analysis was also carried out by aggregation of all contaminants in the same family during the same periods. Families were analyzed if at least 3 time series were found and for periods when at least 5 data points were present. From the 3 trends analyzed, 2 were found to be statistically significant ($p<0.05$). A summary of the analyzed trends during each period and for all contaminant families found in this LME is shown in Figure LME32.6.

Family trends were only significant for Metals, and they were slowly decreasing during the period 1900-1950, increasing during the period 1950-1975 and were neutral during the period 1975-2015.

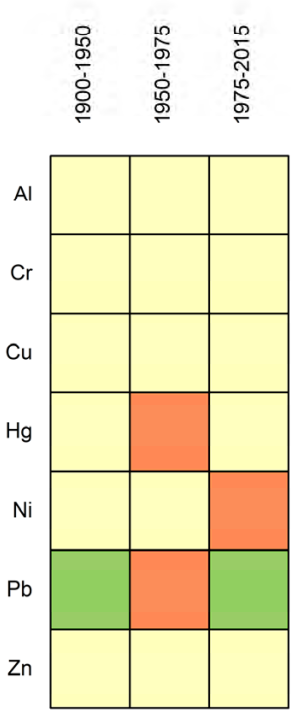


Figure LME32.5: Temporal trends by contaminant and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

4. ASSESSMENT

In LME32, all the analyzed papers reported coastal pollution in West India. Therefore, conclusions drawn from this analysis cannot be generalized to the whole region. In fact, 6 of the 7 papers study the great Mumbai (Bombay) area, one of the largest cities in the world and must be considered mostly representative of this area. This region is characterized by large urban loads and discharges from a great variety of industries, with specific mention of coal fired power plants, refineries, chlor-alkali plants, electroplating, alloy-making and petrochemical complexes. Although many metals are reported, Hg is of special concern because of the contamination from chlor-alkali plants. Very little attention has been paid to the study of contaminant families other than metals. One paper covers a large coastal area from West India, where no large industrial complexes are present, but mining started by the 1950s.

Largest and increasing contamination trends were observed for Pb (+21.1% increase per decade) and Hg (+18.2%) during the period 1950-1975. This is consistent with the “great acceleration” of population growth in the 1950s (Steffen et al., 2015) and the associated economic development, for which India is a good example. In fact, Mumbai is the largest Indian city and port and is the financial and commercial capital of India. It is therefore not surprising that industrial and urban development caused large pollution increases during this period. Interestingly, after 1975, Pb shows a decreasing trend (-6.3% per decade) and Hg shows a neutral trend, but Ni shows a large increasing trend (+15.7%), possibly reflecting higher metal production activities in the region, since India is nowadays among the most important steel producers in the world. Contaminant family trends could only be estimated for metals, showing decreasing trends for the first half of the 20th century, then increasing during 1950-1975 (+4.9% increase per decade) and neutral since then.

Although it is recognized that this review is not exhaustive and some papers might not have been detected during the bibliographic search, it is recommended that coastal sedimentary records are analyzed from largely unexplored areas such as Pakistan, The Gulf, Oman and the Gulf of Aden. A new search focused on this specific LME should provide more useful information in this area. From a technical point of view, it was noted that in all studies, ^{210}Pb dating was performed by using the Constant Flux Constant Sedimentation model; it is recommended that more precise dating methods, such as the Constant Flux model (Sanchez-Cabeza and Ruiz-Fernández, 2012; Sanchez-Cabeza et al., 2014) be explored for ^{210}Pb sediment dating. Dated sediment cores should be used to study as many contaminants as possible, including PAHs, Pesticides, Industrial organic chemicals, Industrial organic byproducts and Nutrients.

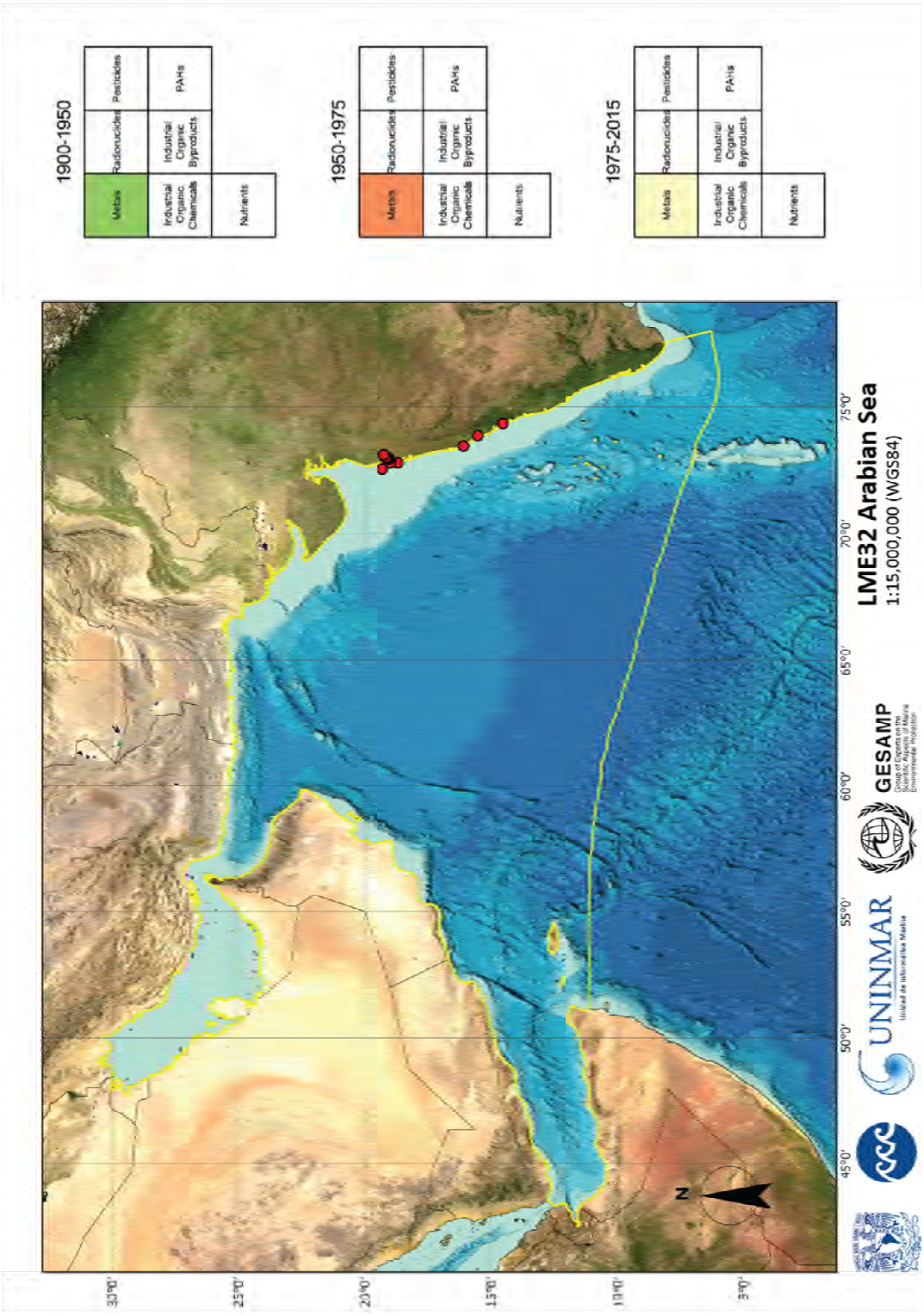
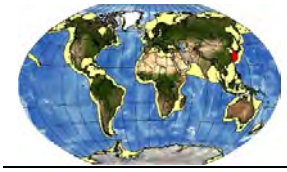


Figure LME32.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.



LME47
East China Sea

1. INTRODUCTION

The East China Sea LME is bordered by mainland China, the northern coast of Taiwan, the Japanese Archipelago, and the southern coast of the Korean Peninsula. It has a surface area of about 775,000 km^2, of which 0.09% is protected and contains 0.34% of the world's coral. The East China Sea LME has a monsoonal climate, and depending on the source of information, it can be classified as moderately to highly productive ecosystem.

The fishing resources are mainly composed of yellow croaker, edible jellyfish, cuttlefish, green filefish, hairtail, mackerel, scad, Spanish mackerel, pomfrets, Chinese herring, shrimp and crabs (Chen et al., 1997).

The coastal fringe of the East China Sea LME stretches over 200,474 km^2. The reported population in 2010 was 137 million, with a density of 681 persons per km^2 and about 30% of coastal population living in rural areas (TWAP, 2015). Rapid economic development and a growing population in eastern China have led to significant increases in the discharge of inadequately treated industrial and domestic wastewaters into the LME. The main contaminants carried by the Changjiang, Mingjiang and Jiulongjiang rivers include organic matter, nutrients, petroleum hydrocarbons and heavy metals.
Aquaculture has also become one of the primary sources of pollution in localized coastal areas. Excessive nitrogen input from sewage and runoff of chemical fertilizers is causing eutrophication and HABs (Harmful Algal Blooms), ubiquitous in coastal areas. Soil erosion, deforestation and intensive cultivation are the main sources of high levels of suspended solids in coastal waters (e.g. the Changjiang drainage basin). Other activities such as dredging of waterways, building of bridges and dams, sand mining and reclamation increase the concentration of suspended solids in the coastal areas.

Accidental oil spills, offshore oil fields and marine transportation, especially ballast water from oil tankers, are major sources of coastal and marine area pollution, particularly in estuaries. Pollutant residues such as petroleum hydrocarbon, arsenic, DDTs and PCBs were found in some commercially produced mussels and oysters (Heileman and Tang, 2017 and references therein).

2. DATABASE INFORMATION

In this LME, 17 papers were selected and 16 could be used for analysis after full content screening. Information on 156 time series was extracted, meeting the requirements of the methodology. The selected papers contained information on 33 contaminants (Figure LME47.1) included in the 7 contaminant families (Figure LME47.2). As examples, two time series are shown in Figures LME47.3 and LME47.4.

3. TREND ANALYSIS

Trends of the selected time series were calculated for the periods 1900-1950, 1950-1975 and 1975-2015. Contaminants were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present.

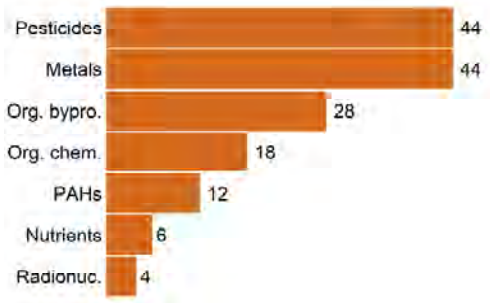


Figure LME47.2: Number of time series by contaminant family.

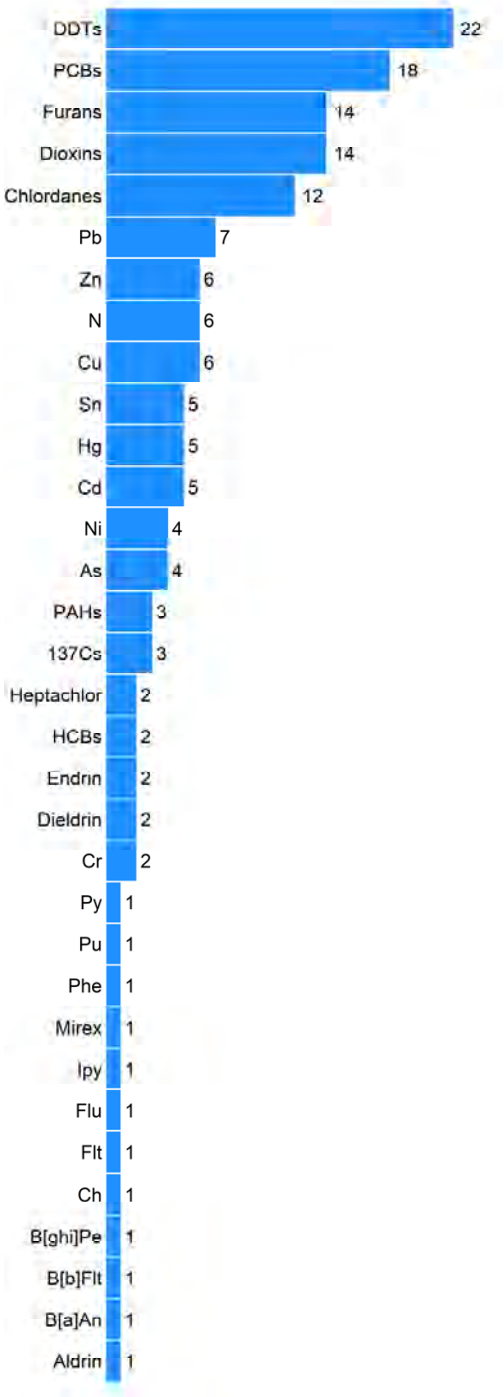


Figure LME47.1: Number of time series by contaminant.

A summary of the analyzed trends during each period and for all contaminants found in this LME is shown in Table LME47.1 and Figure LME47.5. In this case, 46 trends were analyzed, and 20 were found to be statistically significant ($p<0.05$).

The trends are expressed in units of percentage of variation per decade. During the period 1900-1950, neutral trends were observed for Cd, Cu, Hg, Ni, Sn, chlordanes, DDTs, dioxins and PAHs, and increasing trends for Pb, Zn, PCBs, furans and N. During the period 1950-1975, increasing trends were observed for furans, dioxins, ^{137}Cs, DDTs and chlordanes, and neutral trends were observed for the other contaminants. During the period 1975-2015, increasing trends were observed for PCBs, PAHs, N, Cu, Dioxins, Zn, Pb, DDTs, Chlordanes and As, and neutral trends were observed for the other contaminants.

Trend analysis was also carried out by aggregation of all contaminants in the same family during the same periods. Families were analyzed if at least 3 time series were found, and for periods when at least 5 data points were present. A summary of the analyzed trends during each period and for all contaminant families is shown in Figure LME47.6. In this case, 20 trends were analyzed, and 14 were found to be statistically significant ($p<0.05$).

During the period 1900-1950, neutral trends were observed for Pesticides and PAHs, and increasing trends were observed for Metals, Industrial organic chemicals, Industrial organic byproducts and Nutrients. During the period 1950-1975, neutral trends were observed for Metals, Radionuclides, Industrial organic chemicals and Nutrients, and increasing trends were observed for Pesticides, Industrial organic byproducts and PAHs. During the period 1975-2015, increasing trends were observed for all contaminant families except Radionuclides, for which the trend was decreasing.

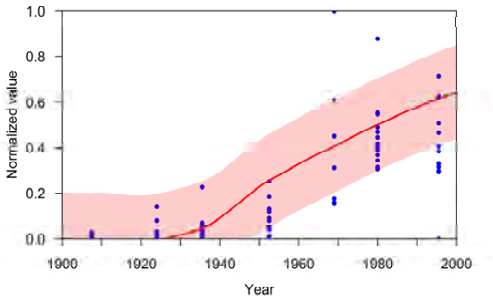


Figure LME47.3: Dioxins time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

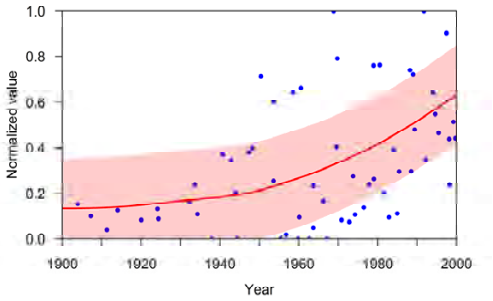


Figure LME47.4: N time series. The red line is a smoothed mean, and the shaded area is the 95% confidence band.

Table LME47.1: Summary of temporal trends by contaminant and period analyzed.

Contaminant	1900-1950	1950-1975	1975-2015
PCBs	+2.0	neutral	+23.0
PAHs	neutral	neutral	+22.5
N	+4.6	neutral	+15.7
Cu	neutral	neutral	+14.6
Dioxins	neutral	+35.0	+11.8
Zn	+12.0	neutral	+11.5
Pb	+9.0	neutral	+11.3
DDTs	neutral	+13.3	+11.1
Chlordanes	neutral	+11.1	+9.1
As	n.d.	neutral	+7.1
Furans	+2.4	+43.4	neutral
137Cs	n.d.	+21.0	neutral
Cd	neutral	neutral	neutral
Hg	neutral	neutral	neutral
Ni	neutral	neutral	neutral
Sn	neutral	neutral	neutral

n.d. = not enough data points for trend analysis

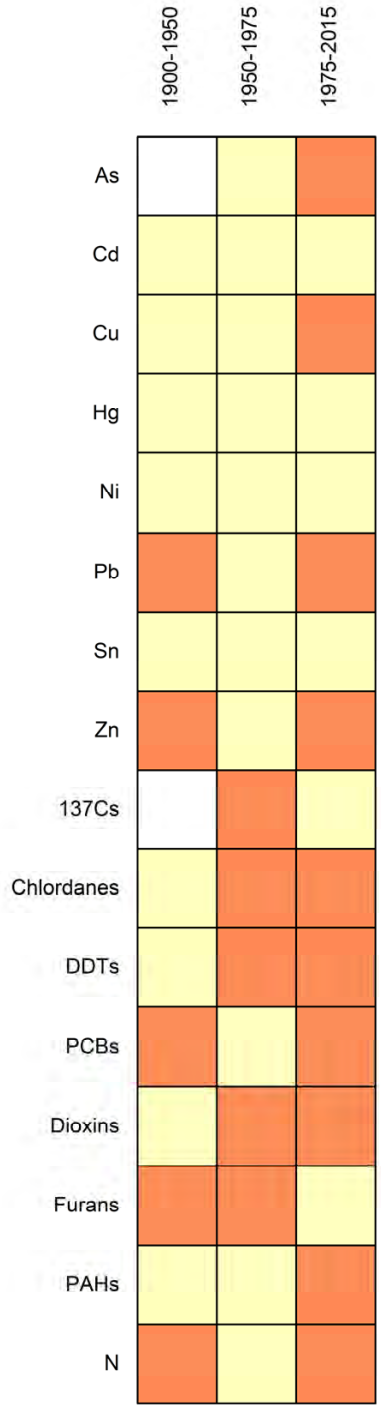


Figure LME47.5: Temporal trends by contaminant and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis. Only contaminants with at least one calculated trend are shown.

4. ASSESSMENT

This LME is bordered by 3 highly industrialized regions (China, Japan and South Korea) with different development histories. Not surprisingly, most papers focus on the Shanghai metropolis, one of the largest coastal cities worldwide. The metropolis is located on the Yangtze (Changjiang) River Delta. The Yangtze River is the longest river in Asia, and the world's fourth largest river in sediment discharge. Over the past decades there has been a significant decrease in the sediment discharge due to dam construction, and soil conservation practices since 1989. The river drainage basin is populated by over 400 million people and contains over 40,000 reservoirs, estimated to retain 90% of the sediment load, before even considering the construction of the Three Gorges dam (Li et al., 2011). During the first third of the 20th century, China's economy was not affected by the Great Depression and large cities (such as Shanghai) started to grow. After WWII and the Chinese Civil War, China entered a period of reconstruction and rapid economic development. With the Reform and Opening-up Policy in 1978, China entered the fastest development period, sustained until today (Guo et al., 2006). Nowadays, the Yangtze River Delta region, with 10% of China's population, accounts for a quarter of China's production (Yang et al., 2012). This economic development causes large coastal effects owing to the discharges of urban, industrial and agricultural wastes.

In South Korea, most of the research is focused on Masan Bay, a semi-enclosed bay with a slow rate of water exchange. The South Korean coastal zones underwent accelerated urbanization in the 1910s - 1920s, based on fossil fuel (especially coal) consumption. After WWII, the area became densely industrialized, particularly by the chemical sector, although until the 1970s, the areas surrounding Masan Bay had been used for agriculture, primarily for farming.

Since 1990s, the South Korean government has attempted to improve water and sediment quality in Masan Bay through a "Total Pollutant Load Management System" (Lim et al., 2013). Nowadays, it hosts about 1300 industrial complexes, including petrochemical, heavy metal, electrical and plastic industries (Moon et al., 2009). South Korea is still one of the fastest growing economies in the world.

Japan became one of the largest world economies in the 19th century and largely

expanded after WWII. In southern Japan, two papers focus on Ariake Bay, described as an inland sea, with many fishery resources and surrounded by several cities and industrial complexes.

For several contaminants, some authors mention the coastal zone as a sink of anthropogenic pollutants (e.g. Yang et al., 2012; Liu et al., 2013). Owing to the large economies of the riparian countries, the contaminant sources in the region are numerous and are associated with urban, industrial, mining and agricultural activities. Overall, the largest contamination source is related with the Shanghai region, in particular the high loading of wastes transported by the Yangtze River and direct releases to the coastal areas. In some cases, the high river sediment yield may dilute the contaminant signal from the catchment (Liu et al., 2013).

Long-range atmospheric (Gio et al., 2006; Lim et al., 2013) and marine (Kim et al., 2013) transport are also described, although rivers are the largest sources for most contaminants (e.g. Guo et al., 2007; Kim et al., 2008a), as reflected in some sediment layers caused by large floods, which transported contaminated soils to the coastal zone (Hao et al., 2008). In the case of lead, coal combustion appears to be the most important regional source (Lim et al., 2013).

In South Korea, the decrease of some metals' pollution from the 1980s onwards is related to the implementation of policies for the preservation of coastal environments, such as the establishment of appropriate wastewater treatment practices and later the Total Pollutant Load Management System in 1994.

The Duckdong waste water treatment plant is still considered to be a major source of dioxins to Masan Bay, South Korea (Moon et al., 2009). Although some organic contaminants have been banned in the LME, PCBs are still contained in domestic and industrial devices (Yang et al., 2012) and organochlorine pesticides are still produced and used in China rural areas (Chen et al., 2002), for example in cotton crops as insecticides (Zhang et al., 2011). This cannot be extended to the whole region, as Japan stopped production and use of some organochlorine pesticides in the early 1970s, also reflected in decreasing dioxin releases (Kim et al., 2007; 2008a). In South Korea, although the use of organochlorine pesticides decreased after the 1970s, the signs of recent accumulation in the sediments are attributed to long-range transport, either from recent use and/or redistribution from contaminated regions (Kim et al., 2008b).

A relatively new source of contamination is the treatment of e-wastes, as two of the world largest e-waste recycling and processing centers in the world (Taizhou, Zhejiang; Guiyu, Guangdong) are in the eastern coastline of China. Regarding radionuclides, plutonium from the low-yield 1945 Nagasaki nuclear bomb and long-range transport from atmospheric thermonuclear tests were observed in coral cores (Lindahl et al., 2012).

Both pesticides and metals deserve similar attention in the reviewed literature, with 44 time series each, followed by organic industrial byproducts (28 time series). In fact, the most reported contaminants are organic compounds (DDTs, PCBs, furans, dioxins and chlordanes). The trend analysis in this LME is particularly difficult, as the economic development in the region is large but asynchronous.

Some contaminants already showed increasing trends during the period 1900 – 1950, notably Pb and Zn (12.0 and 9.0% increase per decade, respectively), in good agreement with the urbanization and industrialization trends in the LME. Very large increasing trends are observed for the organic industrial byproducts furans and dioxins during the period 1950 – 1975 (43.4 and 35.0% increase per decade, respectively). These compounds are produced, for example, as byproducts during the production of pesticides and during coal combustion in incinerators and smelters, and they reflect the rapid urbanization and industrialization of the whole region.

Unlike other developed world regions (such as Europe and USA), trends of many contaminants are still rapidly increasing during the recent period 1975 – 2015, notably for PCBs, PAHs, N (Figure LME47.4) and Cu (23.0, 22.5, 15.7 and 14.6% increase per decade). As this analysis was designed to discuss overall LME trends, no distinction was done per country, but likely some of the recently increasing trends are heavily weighed by the recent large economic development of China. A specific sub-regional study might give a better regional perspective in this socio-economically complex LME.

Although this is a relatively well studied LME, several knowledge gaps still exist, mainly in northern Taiwan and the coastline south of Shanghai. Also, some better regional perspective might be gained from the study of sediment cores from the open sea platform

through regional collaboration efforts. Although time series from all contaminant families were calculated, relatively less information was found on nutrients, likely one of the most relevant and increasing contaminants in the region (e.g. N, figure LME47.4).

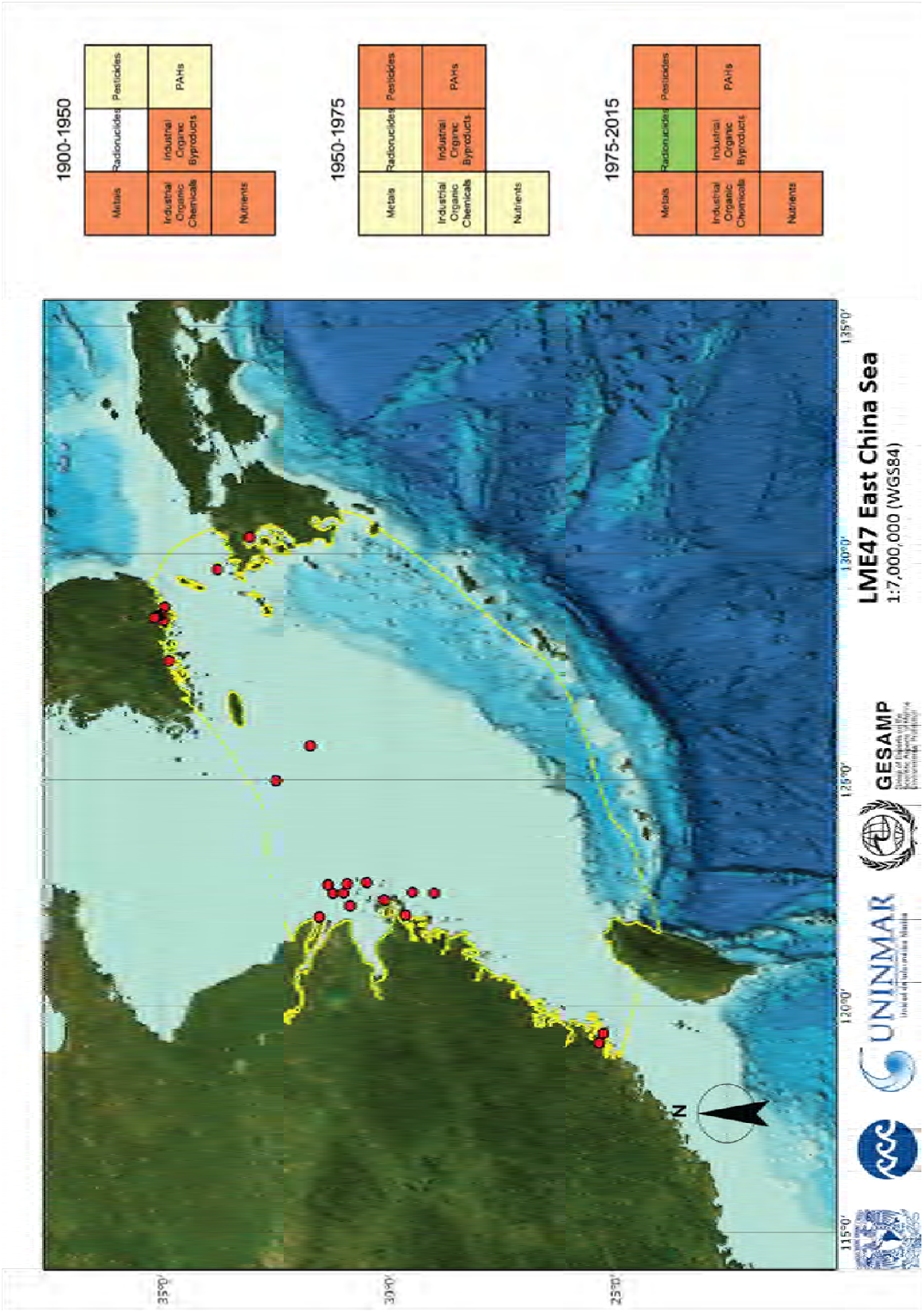


Figure LME47.6: Sampling locations and temporal trends by contaminant family and period. Brown = increasing, yellow = neutral, and green = decreasing trend; blank = not enough data points for analysis.

5. GLOBAL ASSESSMENT

The reported case studies highlight the complexity of the distribution, scale, and rate of the development history worldwide, and the multiplicity of contamination sources (even in single LMEs, such as the East China Sea LME). Contamination sources are globally widespread and rather similar, and the general description of their common causes, including urbanization, increase of agriculture activities, mining and industrialization, are all mainly driven by the worldwide population increase during the 20th century. In this attempt to provide a global assessment of contamination trends, the geographical distribution of the calculated trends per each analyzed period are qualitatively described and some examples of global trends are discussed.

5.1 Geographical Distribution

To provide the basis for a qualitative global assessment, the trend analysis results per family at each LME are displayed in world maps and are shown in Figures 5.1 – 5.3. In some cases, study areas are representative of only one or two locations (such as large cities). This is especially true for LMEs with very few study sites. In these cases, trends are mostly related to the socio-economic development of these locations (Table 5.1).

Table 5.1. Main areas of study per LME.

LME	Name	Most studied sites
LME03	California Current	San Francisco (USA) Los Angeles (USA)
LME10	Insular Pacific-Hawaiian	Honolulu (USA)
LME11	Pacific Central-American	Gulf of Tehuantepec (Mexico)
LME13	Humboldt Current	Concepción (Chile)
LME14	Patagonian Shelf	La Plata (Argentina)
LME15	South Brazil Shelf	Rio de Janeiro (Brazil)
LME16	East Brazil Shelf	Salvador de Bahía (Brazil)
LME17	North Brazil Shelf	Belen (Brazil)
LME24	Celtic-Biscay Shelf	Southampton (UK)
LME29	Benguela Current	Berg river
LME30	Agulhas Current	Soetendalsvlei Lake
LME32	Arabian Sea	Mumbai
LME33	Red Sea	Gulf of Aqaba (Jordan)
LME34	Bay of Bengal	Chidambaram (India) Calcuta (India)
LME35	Gulf of Thailand	Bangkok (Thailand)
LME36	South China Sea	Pearl River (China) Hong-Kong (China)
LME37	Sulu-Celebes Sea	Manila (Philippines)
LME41	East-Central Australian Shelf	Brisbane (Australia) Sidney (Australia)
LME42	Southeast Australian Shelf	Tasmania (Australia)
LME43	Southwest Australian Shelf	Stokes Inlet (Australia)
LME44	West-Central Australian Shelf	Perth (Australia)
LME46	New Zealand Shelf	Hawke Bay (New Zealand) Waipaoa River (New Zealand)
LME50	Sea of Japan - East Sea	Ulsan (Korea)
LME61	Antarctic	King George Island (Antarctica)

A first inspection reveals that limited information exists in some world regions, such as non-Mediterranean Africa (LMEs 27-33), the Australian continent and bordering regions (LMEs 38 – 46) and most of the polar regions (both the Arctic and Antarctica).

During the initial period 1900-1950, although many trends are neutral, increasing trends are

already observed in Eastern Asia, Southeastern Asia, Europe, and to a lesser degree North America, South America, Arctic and Antarctica.

As expected, during the subsequent period 1950 – 1975, more increasing trends are observed worldwide: the number of increasing trends increase in the Australian region, Africa, Northern and Southern America, and remain similar in Eastern Asia, Southern Asia and Europe.

Finally, during the recent decades (1975 – 2015), the overall picture diverges: trends are still increasing in Eastern Asia, Southern Asia, Africa and Central America, but more decreasing trends are observed in the rest of the world, notably Europe, the Australian continent, North and South America and the Polar Regions. This is likely the result of the combination of regional development during the last few decades, and the varying success of national and international environmental policies and social environmental awareness.

5.2 Contaminant Trends

Worldwide contaminant trends were calculated but should be interpreted with caution owing to the spatio-temporal heterogeneity of contamination sources and drivers and the uncertainties and limitations of sedimentary records (section 2.10). Global contamination trends of two contaminants (PCBs and Hg, Figures 5.4 and 5.5) and two families (Industrial organic byproducts and Nutrients, Figures 5.6 and 5.7) are shown as examples.

PCBs are organic chlorine compounds that belong to the family of Industrial organic chemicals. They were first synthesized in 1881 (Myers, 2007) and used industrially since 1929 e.g. under the commercial name of Aroclors. Monsanto stopped the production of this product in the late 1970s. Amongst many applications, PCBs were used as dielectric and coolant fluids in electrical equipment, motors and hydraulic systems and as pesticides. They are environmentally persistent and have both carcinogenic and non-carcinogenic effects on humans, including the immune, reproductive, nervous and endocrine system (Carpenter, 2011). They were banned in many developed countries in the late 1970s, and by the Stockholm Convention on Persistent Organic Pollutants in 2001, but many products still contain PCBs.

Some studies reported detectable concentrations of PCBs in sediments older than the onset of their industrial production, which has been reported in many other studies elsewhere (Ruiz-Fernández et al., 2012 and references therein) and explained as the result of diverse potential mechanisms, including sediment resuspension, diagenetic downward mobility in the sediment column and the pyrolysis of natural organic matter. Nonetheless, the world trend of PCBs (Figure 5.4) is a good example of the effectiveness of national policies and international agreements, as mean levels rise steadily from the 1920s and reach a broad maximum around 1980, in agreement with the PCB banning in many developed countries. Since then, mean world values have been decreasing slowly, but they have not disappeared. Likely causes of the recent presence of PCBs in the environment are their persistence in contaminated soils and waters, long-range transport, improper disposal or leakage from older electrical equipment, the incineration of PCB-containing wastes, treatment of electronic wastes, vehicular emissions and storm-water runoff (ATSDR, 2011).

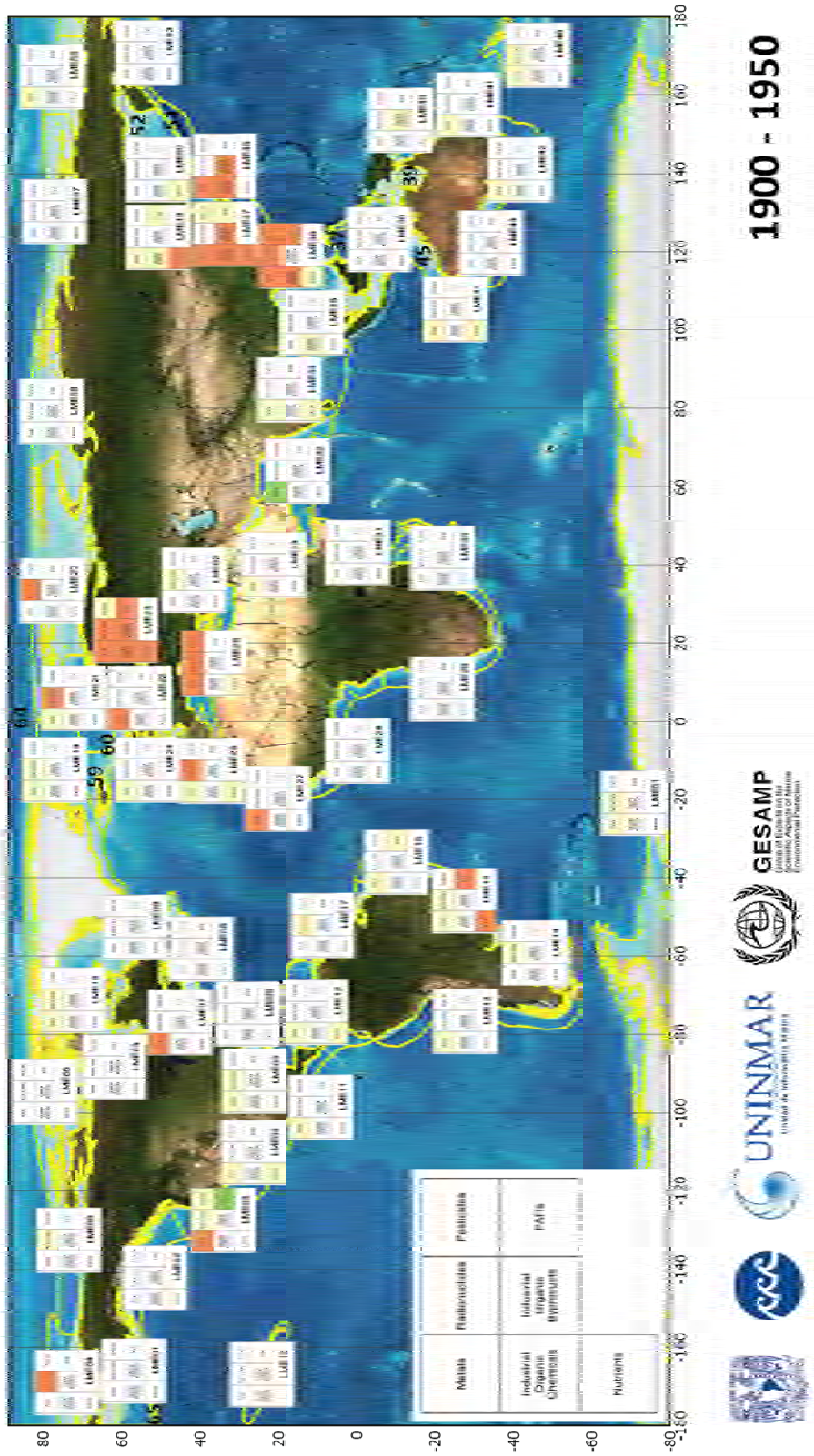


Figure 5.1. Trends of contaminant families for the period 1900-1950. The trend color codes are: green = decreasing, yellow = neutral, brown = increasing, white = not enough data points for analysis. See Table 5.1. for LME's with limited area coverage.

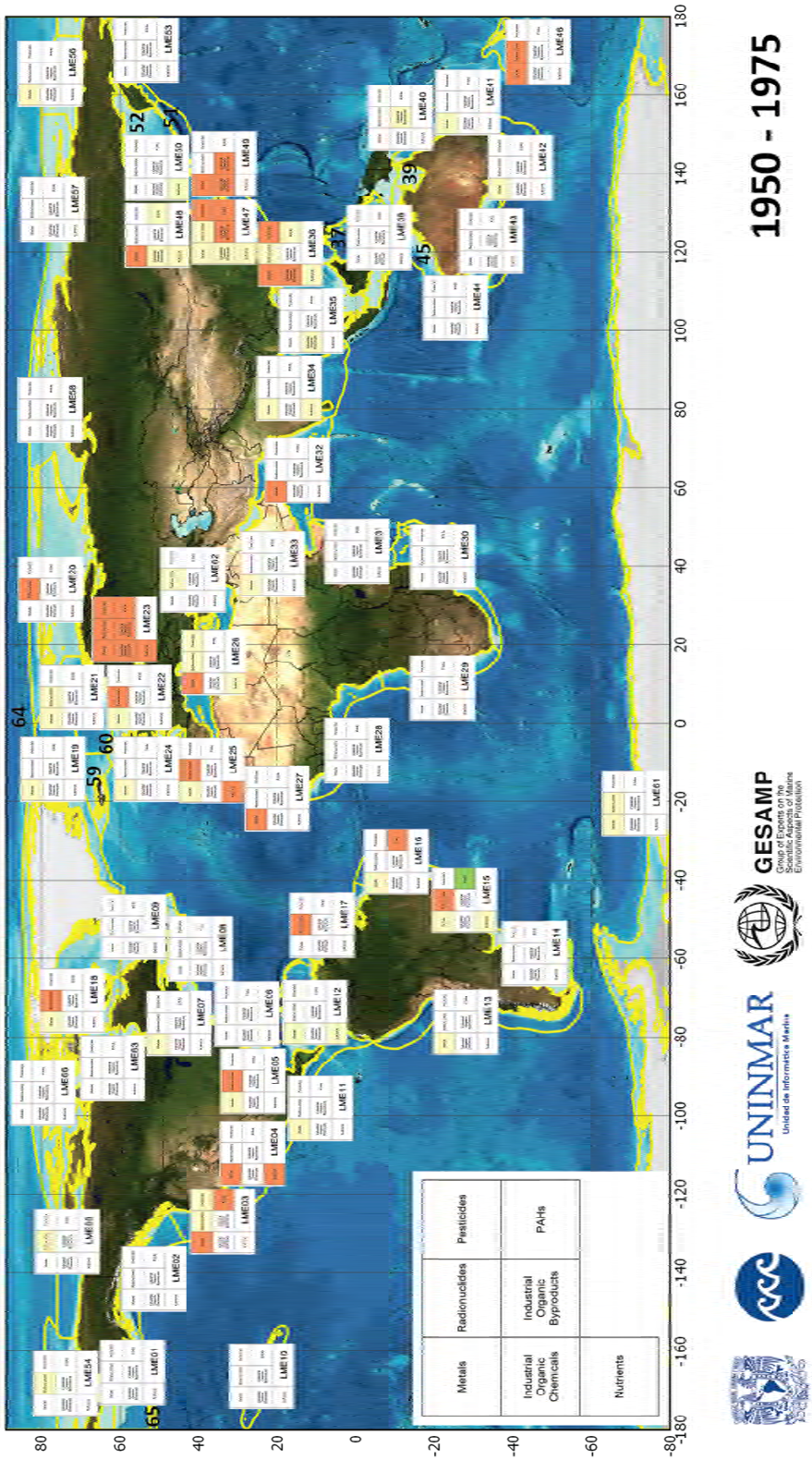


Figure 5.2. Trends of contaminant families for the period 1950-1975. The trend color codes are: green = decreasing, yellow = neutral, brown = increasing, white = not enough data points for analysis. See Table 5.1. for LME's with limited area coverage.

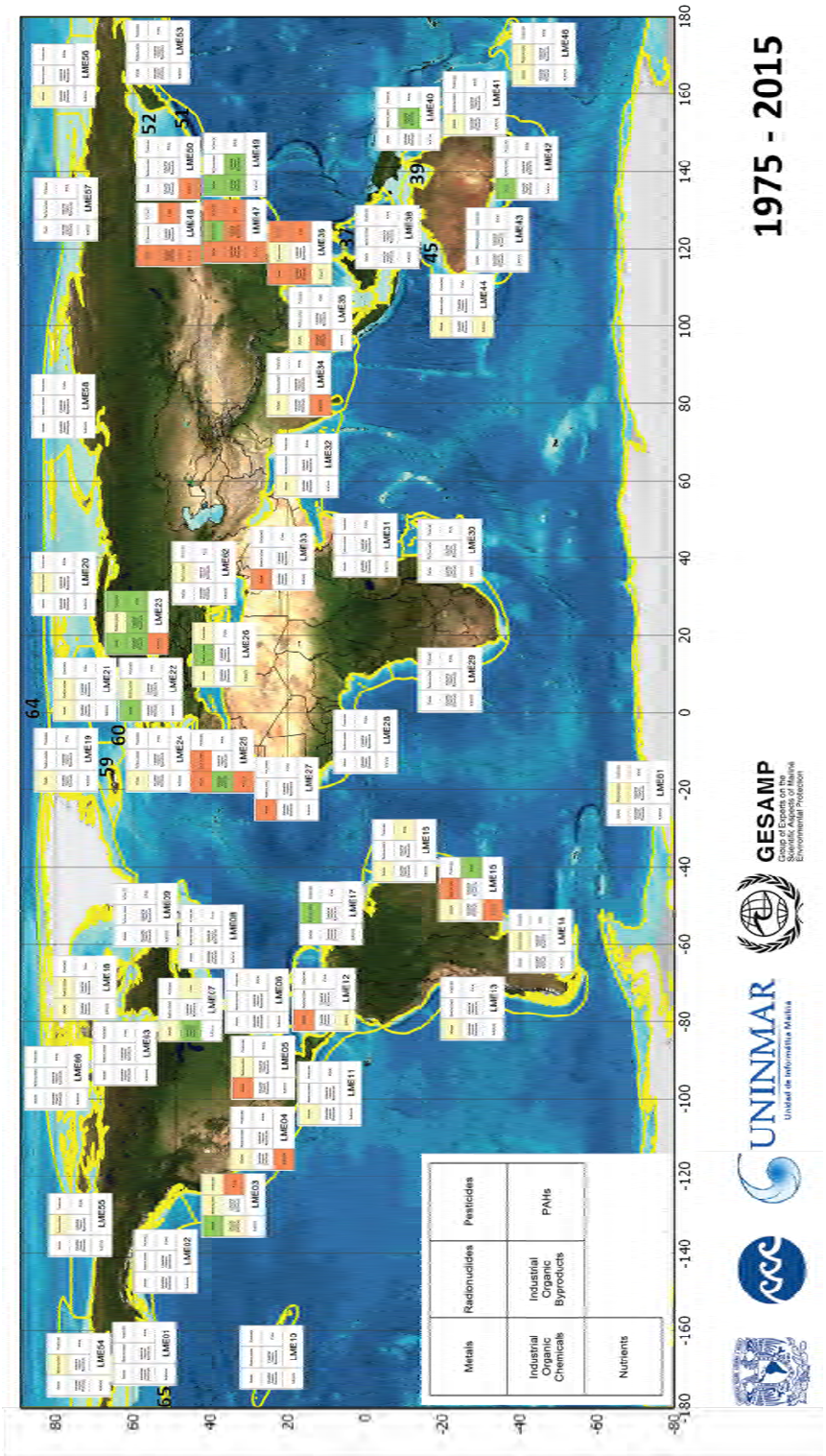


Figure 5.3. Trends of contaminant families for the period 1975-2015. The trend color codes are: green = decreasing, yellow = neutral, brown = increasing, white = not enough data points for analysis. See Table 5.1. for LME's with limited area coverage.

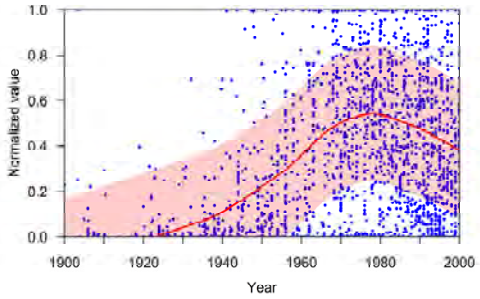


Figure 5.4. World time series of the contaminant PCBs.

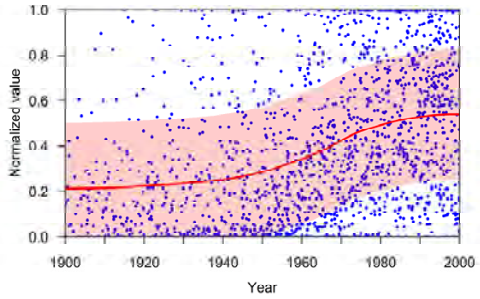


Figure 5.5. World time series of the contaminant Hg.

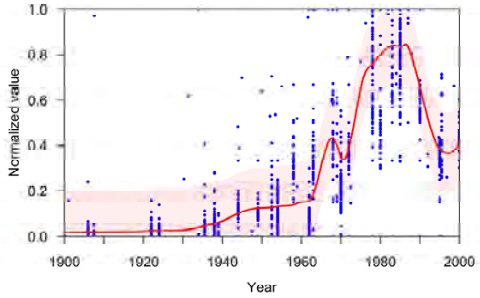


Figure 5.6. World time series of the contaminant family Industrial organic byproducts.

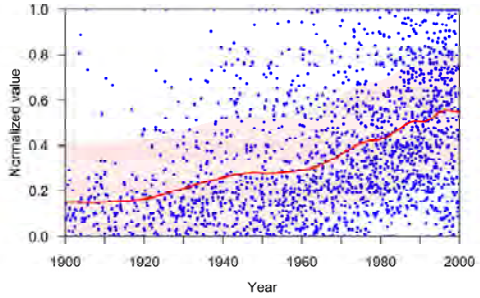


Figure 5.7. World time series of the contaminant family Nutrients.

Mercury is a metal naturally present in the Earth's rocks and coal deposits. It has a complex speciation and some Hg forms are volatile. It is toxic in relatively low concentrations and affects the central and peripheral nervous system (WHO - World Health Organization, 2007). It is produced in many domestic (such as coal boilers), urban (such as municipal waste treatment plants) and industrial applications (such as energy production by coal burning and chlor-alkali plants) (e.g. Boening, 2000); it is also widely used in artisanal and small-scale gold mining around the world (WHO, 2016). As for other toxic metals, regulation has been progressive, and has led to the international Minamata Convention on Mercury, signed in 2013. The world time trend of mercury (Figure 5.5) does not show a very clear pattern, likely because of the highly diverse Hg contamination history worldwide. However, the smoothed mean reveals a slow growth since the beginning of the 20th century (obviously starting before 1900), an acceleration in the 1950s, and an apparent stabilization in the 2000s.

Regarding Industrial organic byproducts, the database included 1,744 data points from 8% of the total number of time series, from 4 LMEs: LME23 (Baltic Sea), LME40 (Northeast Australian Shelf), LME47 (East China Sea) and LME49 (Kuroshio Current). Although the number of LMEs is small, they are quite representative of a large world area, except America. The Industrial organic by-products included in this report are dioxins and furans, toxic substances that have similar chemical forms. They are mostly byproducts of other processes, such as combustion, metal smelting, production of certain chemicals (such as PCBs, PVCs and some pesticides) and bleaching in pulp mills (EPA, 2006). They are carcinogenic, and affect the immune and reproductive systems (Birnbaum, 1994). As for other organic pollutants, environmental controls started to be included in national legislations in the 1970s and were included in the Stockholm Convention on Persistent Organic Pollutants, signed on 2004. The worldwide trend (Figure 5.6) shows little effects during the early decades of the 20th century, and a gradual increase since the 1930s. Maximum concentrations were reached in the 1980s, and the worldwide mean appears to have decreased since then to similar levels to those observed in the early 1970s.

The elements considered in the Nutrients family are N, P and Si. Of the 118 time series

corresponding to this contaminant family, 5 refer to Si, 38 to P and 75 to N, so this discussion largely reflects contamination by N and P. These two elements are naturally widespread and essential for the development of life, including the human species. Also, they are present as contaminants in many anthropogenic sources, notably urban sewage and agricultural activities, including fertilizers. Global trends of the Nutrients family (Figure 5.7) increased slowly and steadily since the early 20th century, and an acceleration was observed in the 1950s. Nutrient releases to aquatic ecosystems are associated with eutrophication and hypoxia. Although many national and international policies have addressed the reduction of nutrient releases, it appears that present efforts are insufficient, as recent trends are increasing. For example, in the Baltic Sea LME, where national and international efforts have proven to be effective in the reduction of levels of many contaminants, the trends of N and P are still increasing. Further efforts are needed to control the release of these elements to aquatic ecosystems.

5.3 Assessment

Globally, the best studied contaminant family is metals, with 1,353 time series (Figure 5.8). This is clearly related to the availability of simpler and well-known techniques for their analysis. In fact, the three contaminants better studied at the global scale are Pb (260 time series), Zn (228) and Cu (204). It is also not surprising that the second family considered is that of Radionuclides, as most publications rely on radiometric methods for sediment dating. The main radionuclides reported are ^{137}Cs (155 time series) and Pu (37). Other contaminants with large numbers of time series are PCBs (181 time series), furans (104) and DDTs (90).

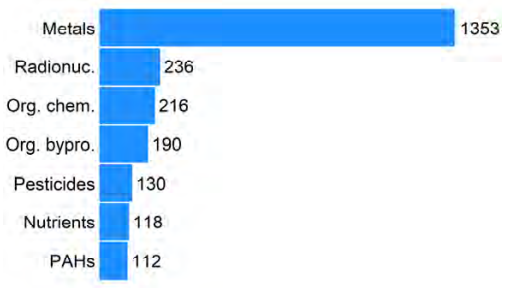


Figure 5.8. Global Number of time series per contaminant family.

As an example, trend anlysis of all the time series for some contaminants are shown in Table 5.2. The two Industrial organic by-products, furans and dioxins show large increasing trends (> 10% per decade) during the period 1950 – 1975 and they also show large decreasing trends in the recent period (1975 – 2015). A similar behavior is observed for the PAHs Ipy, B[ghi]Pe and the metals Ag, Sn, Cd and Pb, indicating that national and international policies have been effective to reduce environmental levels of many contaminants in the recent decades.

In some cases, increases during the 1950 – 1975 period are followed by neutral (Ni, Cu, Hg, Py, Ch and B[a]An) or even increasing (Zn, DDTs, Pu, P and Chlordanes) trends. Although in some cases this may be due to the remobilization and transport of surface contaminated soils to the coastal zone (such as radionuclides, as global fallout has decreased since the mid-1960s), special attention needs to be paid in the future to releases of these substances in order to further reduce their levels in the coastal ecosystems. This appears to be particularly important for N and P, as urban and agricultural releases are clearly still increasing owing to the growth of global population and agriculture.

To summarize the trend analysis at a global scale, contaminant family trends for all LMEs are shown in Table 5.3. Large increasing trends during the 1950 – 1975 period were observed for pesticides (+16.6% increase per decade), industrial organic byproducts (+13.9%), industrial organic chemicals (+13.8%) and PAHs (+10.7%). During the recent decades, decreases were only observed for Industrial organic byproducts (-17.6%), Industrial organic chemicals (-5.0%) and barely Metals (-0.8%).

Although for some contaminants and contaminant families (Table 5.3) decreasing or neutral trends are observed in the recent decades (1975 – 2015), it is evident that many increasing trends are still observed globally. Although much more research and information are needed to have a more precise diagnostic, especially at the LME scale, it is quite clear from this analysis that, with different time and regional patterns, the coastal environments show significant increasing trends for many contaminants.

Table 5.2. Summary of global trends (percentage of variation per decade) for selected contaminants, ordered by recent trend value (period 1975 – 2015).

Contaminant	Family	1900-1950	1950-1975	1975-2015
Furans	Industrial organic byproducts	+1.5	+14.3	-21.8
Ipy	PAHs	neutral	+18.6	-15.2
Dioxins	Industrial organic byproducts	+2.1	+13.4	-13.7
B[ghi]Pe	PAHs	neutral	+18.3	-13.4
Ag	Metals	+5.3	+12.3	-13.1
HCBs	Pesticides	+6.0	neutral	-11.3
PCBs	Industrial organic chemicals	+3.0	+17.3	-8.7
Sn	Metals	neutral	+13.7	-8.5
Cd	Metals	neutral	+5.0	-2.8
Pb	Metals	+2.6	+7.4	-2.4
Ni	Metals	+2.0	+4.1	neutral
Cu	Metals	+2.7	+4.7	neutral
Hg	Metals	neutral	+9.3	neutral
Py	PAHs	neutral	+17.7	neutral
Ch	PAHs	neutral	+21.4	neutral
B[a]An	PAHs	neutral	+23.5	neutral
Zn	Metals	+3.2	+5.2	+1.8
DDTs	Pesticides	neutral	+17.9	+2.5
N	Nutrients	+3.0	neutral	+5.1
Pu	Radionuclides	neutral	+11.7	+7.1
P	Nutrients	+1.7	+6.9	+7.9
PAHs	PAHs	neutral	neutral	+8.0
Chlordanes	Pesticides	neutral	+8.1	+10.3
An	PAHs	neutral	neutral	+28.1

Table 5.3. Summary of global trends (percentage of variation per decade) per family of contaminants, ordered by the trend value during the period 1975 – 2015.

Family	1900-1950	1950-1975	1975-2015
Industrial organic byproducts	+1.9	+13.9	-17.6
Industrial organic chemicals	+2.4	+13.8	-5.0
Metals	+2.1	+5.2	-0.8
PAHs	neutral	+10.7	neutral
Radionuclides	+1.9	+7.9	neutral
Pesticides	neutral	+16.6	+2.7
Nutrients	+3.0	+4.8	+6.1

6. SUMMARY AND CONCLUSIONS

Data published in peer-reviewed scientific documents were collected and used to assess coastal contamination trends around the world, based on the Large Marine Ecosystems geographic regionalization. Environmental matrices used in this review were sediment and coral cores, as well as biota, although most data points (> 90%) were obtained from the analysis of sediment cores. The use of environmental archives, especially of sediment cores, is particularly useful as they can provide consistent reconstructions of many contaminants simultaneously and allow the establishment of background / reference values for each studied site. On the other hand, results must be interpreted with caution owing to the spatio-temporal heterogeneity of contamination sources and drivers and the uncertainties and limitations of sedimentary records (section 2.10).

Final information was analyzed from 272 published and refereed papers, containing information on 667 records, including 2,355 time series (contaminant versus time) and 37,720 data points (paired contaminant concentrations and age). Main problems encountered during the collection of information were deficient data reporting and a simplistic use of dating techniques. The contamination trends were assessed for 3 time periods: 1900-1950, 1950-1975 and 1975-2015.

In this review we could not find appropriate information, or could not analyze it because of insufficient data points, for 20 LMEs, namely: East Bering Sea, Southeast U.S. Continental Shelf, Scotian Shelf, Newfoundland-Labrador Shelf, Guinea Current, Somali Coastal Current, Indonesian Sea, North Australian Shelf, Northwest Australian Shelf, Oyashio Current, Sea of Okhotsk, West Bering Sea, Laptev Sea, Kara Sea, Iceland Shelf and Sea, Faroe Plateau, Hudson Bay Complex, Central Arctic Ocean, Aleutian islands and Canadian High Arctic-North Greenland. In some of these LMEs (e.g. the Arctic area) and several other areas (e.g. the Baltic Sea) monitoring programs are on-going, and data and assessment reports are available on a regular basis, although time series are often relatively short and not appropriate to define background levels. In these cases, retrospective analysis can effectively complement the monitoring efforts by deriving pollution trends from long time periods, as required for the evaluation of the effects of environmental policies. On the other hand, several LMEs contained over 1,000 data points, namely California Current, South Brazil Shelf, Baltic Sea, Iberian Coastal, Mediterranean, Arabian Sea, South China Sea, East China Sea, and Kuroshio Current.

Most of the analyzed time series corresponded to the Metals contaminant family, and the least abundant contaminant family was PAHs. The most reported contaminants were Pb, Zn, Cu, PCBs and ^{137}Cs. The period with more abundant data points contained in the database was 1980s – 1990s. The lower number of data points towards the past can be explained because of i) the lower age resolution in the older sediments of a sediment core, owing to usually lower sediment accumulation rates and/or lower sampling resolution towards the sediment cores bottom and ii) the low availability of consistent data from biota time series before the decade 1970s. The reduction of data points in recent years is due to i) the lag between sample collection and publication and ii) the lag between the sampling date and the age of the surface sediment section (usually several years older). Recent years will be better represented in future compilations.

Regarding human impacts, the world coastal regions are highly heterogeneous, owing to diverse socio-economic histories. In the case studies included in this work, two paradigmatic and well-studied examples are described: the Baltic Sea and the East China Sea. The Baltic Sea LME suffered profound transformations and deterioration during the 20th century, reflected in large increasing trends of many contaminants. However, international agreements (HELCOM), national policies and social awareness led to relevant improvements during the recent decades, causing the reduction of many (but not all) contaminant trends. On the other hand, contamination in the East China Sea, although bordered by countries with rather different socio-economic development histories, is nowadays largely driven by Chinese development, clearly reflected in ongoing and continuously increasing trends for most contaminants. These two cases show that, when enough retrospective data are available, contamination trends are a useful tool to understand the underlying causes of coastal deterioration and to provide scientifically sound information for decision-making, both locally and regionally.

When all LMEs trends are considered globally and with all due caveats regarding local and regional differences, some global conclusions may be drawn. Some trends are already increasing in the early 20th century decades

(e.g. Eastern and Southeastern Asia, Europe), likely reflecting on-going development since the industrial revolution in the 19th century. During the 1950 – 1975 period, of large development worldwide, more increasing trends (and some of the largest observed) occurred in many regions. And during the recent decades (1975 – 2015), worldwide contamination trends diverge: they keep increasing in Eastern and Southern Asia, Africa and Central America, but the number of decreasing trends increase in the rest of the world, notably Europe, the Australian continent, North and South America and the Polar Regions. This shows that for most contaminants that have been regulated the concentrations have decreased. This also reflects the interaction between regional development and the success of national policies, international agreements and social environmental awareness in the latter regions. This result emphasizes the importance of international efforts to reduce coastal deterioration at the regional level, which should be translated into effective national policies.

Although regional contamination trends are not homogenous and are highly dependent on national and regional development histories, an effort was made to cautiously provide a worldwide assessment, based on the aggregation of contaminant data. As in the LME case studies, this analysis is biased towards the regions with more abundant data and, within these, the areas studied, usually near clear contamination sources. The overall picture that emerges is, as expected, complex. Neutral or small increasing trends are observed in the early 20th century, followed by large increases in the period 1950 – 1975, > 10% increase per decade for the PAHs congeners Ch and Flt, the pesticide DDT, the Industrial organic chemical PCB, the Industrial organic by-products furans and dioxins, the Metal As, and the Radionuclide Pu. During the recent decades (1975 – 2015) it is noticeable that i) some trends are largely decreasing (> 10% decrease per decade), such as the Industrial organic by-products furans and dioxins, the Metal Ag, and the pesticide HCB, reflecting the effective banning of many compounds, and better control of urban releases, and ii) still some contaminants show increasing trends, some of them large (e.g. the pesticide chlordane).

Despite the improvement in several contamination trends in recent decades, in most cases the contamination levels reached are not yet comparable to pre-industrial levels and continuous efforts at national and international levels are still needed globally to protect the coastal environment worldwide.

7. RECOMMENDATIONS

To the best of our knowledge, this is the first effort to compile and analyze contamination trends on a global basis, largely based on time series from ^{210}Pb dated sediment cores. Although many aspects of the study can be revisited and improved, this work will be the basis of further efforts at the national, regional and global level and should be both maintained and improved.

Methodological recommendations

More data are needed. During the collection of information, data from many papers could not be used because of insufficient or wrong reporting. Although this might change soon, because of new journal policies to make information available in data repositories, still lack of information is possible, as no specific recommendations exist on the required information for environmental time series. It is recommended that an expert group provides full guidance on reporting environmental time series, especially from environmental archives. From our findings, we propose the following recommendations (sometimes specifically related to sediment cores):

- Data points: although nowadays graphical representation of results (e.g. in form of sediment profiles or time series plots) is preferred, we encourage scientists to provide full tables in electronic form, with easily extractable information to reduce errors and workload of collection.
- Units: these largely depend on the observed levels and the research field praxis. We recommend using either SI (International System) units (in scientific notation) or multiples (such as mg kg^{-1} instead of ppm). When the concentration is referred to a specific sediment fraction (e.g. total organic carbon), the concentration of this fraction should also be provided (e.g. total organic carbon / sediment weight).
- Contaminant: especially for organic contaminants, an effort must be made to follow international recommendations on nomenclature. When several contaminants are reported together (such as PAHs), not only the concentration of individual contaminants should be provided, but also attention should be paid to the meaning of the reported aggregate when data are used for comparison among studies. For total

PAHs, we suggest making the effort to analyze the whole list of priority PAHs for meaningful comparisons.

- Variables: for sound scientific reasons, many authors prefer to report and discuss derived variables such as enrichment factors. When this is done, we recommend providing (likely as supporting information) a clear description of the derived variable, and concentrations of both the contaminant and the variable used for normalization (commonly Al).
- Age-model: irrespective of specific recommendations on dating, papers should always include an age-model (age *versus* depth), preferably as electronic supporting information. If the age-model is constructed from a constant sediment accumulation rate, this value should be clearly mentioned in the paper.
- Site information: it is common to provide maps or referring sampling locations to other papers or technical reports (many times unavailable). It is recommended that geographical coordinates are also provided in the form of a table, or as electronic supporting information. The sample code should also be provided, so several publications referring to the same environmental archive can be identified.
- Region: to facilitate compilation efforts (such as this), it is recommended that authors identify in the text the ocean/sea and coastline studied, and ideally the LME region of each time series.
- Other variables that should always be provided are sampling date and sample collection depth.

Some papers with valuable information might not have been found during this review. This is usually caused by lack of informative keywords. Sometimes, papers are not addressing contamination issues, but may contain useful information on these. In case they are not used in the title nor the abstract, we recommend always including keywords such as “sediment” (or “coral”, “biota”) and “contamination”. Especially in these cases, the use of informative keywords is essential to find relevant published data.

One possible way to overcome these difficulties and to allow much easier reviews, is the creation of a specifically designed data repository, with clear and complete templates. To make this sustainable, the repository must be well-known, the database must be continuously populated with new information (either from old and new papers) and it must be institutionally supported. Alternatively, information can be gathered at national / LME level, and periodically merged in a single repository.

Alternative information sources should be considered during new assessments. For example, the chemistry portal of the European Marine Observation and Data Network (EMODnet) includes valuable information of contaminants in surface sediments.

When addressing new projects that include the collection and analysis of sediment cores, a strong collaboration between geochronologists and scientists from other disciplines (such as physical oceanography, geophysics, sedimentology, benthic ecology, …) is strongly encouraged, as this enhances project success by the selection of optimal collection sites, helps to constrain uncertainty sources and validity of the record, as well as the interpretation of the core profiles.

For sampling, one must consider the sediment dynamic processes affecting each site, i.e. transport, resuspension and accumulation processes. Understanding these processes may help to find where centers of deposition are and consequently where contaminants accumulate at higher rates. Areas with low sediment accumulation rates do not show the contamination record and should be avoided.

The issue of dating deserves a special mention. In many cases, dating is simply based on stratigraphic markers (such as ^{137}Cs) or constant sedimentation rates. When possible, ^{210}Pb dating based on variable sedimentation rates models should be used (e.g. the Constant Flux model, Robbins, 1978; Appleby and Oldfield, 1978; Sanchez-Cabeza and Ruiz-Fernández, 2012). Age-models should be validated, either by radiometric methods (such as ^{137}Cs or plutonium isotopes), or other stratigraphic markers such as volcanic ash layers, disturbances caused by coastal works or large sedimentary events and the onset of specific contaminants.

National and regional (LME) recommendations

This work does not include a review of the available data from all LMEs. A first and evident recommendation is to engage local / regional experts to provide specific LME assessments, and publish this as an Addendum to this report. Taking advantage of this, experts could also recover new papers to be included in future reports of this kind.

In some LMEs, dated sediment cores are only available from a specific area and are not representative of the whole region. LME scientists could participate in national and international collaborations to identify relevant geographical gaps, and little studied contamination “hot-spots” and contaminants. Priority should be given to the generation of adequate dated core data through the activities of national and regional stakeholders (Regional Seas Conventions, the EU, etc.).

In the case that samples were properly preserved, it would be useful to complement existing datasets with the analysis of more contaminants. This might require collaboration with other LME scientists, in case regional analytical capabilities were not available. In some cases, polluted areas were visited a few decades ago, and revisiting them might shed more light on recent contamination trends and the sedimentary processes contributing to the record.

It is recommended to develop new studies in little or non-contaminated sites that may provide information on long-range contaminant transport. Contaminant data from pre-anthropogenic or low industrialization periods are useful to identify regional background information that could be used as concentration targets for remediation or conservation programs. A possible mechanism to foster LME efforts could be to setup small and long-term LME working groups, to address these and other issues of their interest.

The information contained in this report should be useful to policy-makers worldwide, as contamination trends are excellent indicators of environmental degradation and, at the same time, may reveal the effectiveness of environmental protection policies. Indeed, it was observed that for most contaminants that have been regulated the concentrations have decreased. Specific regional recommendations from this study can only be done from specific LME assessments, preferably in collaboration with local scientists and stakeholders. In general terms, it is evident that all regions share similar (though with different intensity) impacts, from urbanization, agricultural activities, mining and industrialization. In many areas, urbanization and industrialization progress in parallel, so large metropolitan areas are important sites to study.

Several international organizations (e.g. HELCOM, OSPAR and MED POL - Programme for the assessment and control of marine pollution in the Mediterranean) and national authorities, maintain large and relevant programmes on marine pollution monitoring and assessment, but in general they are based on water, biota and surface sediment monitoring. The analysis of dated environmental records would complement these efforts by i) expanding the time interval, as regular monitoring programmes usually do not span more than 3 – 4 decades, and ii) allowing the definition of background concentrations. Moreover, the analysis of sediment cores may help to obtain information from poorly monitored and studied areas. On the other hand, retrospective analysis is less precise in defining recent trends, as sediment accumulation rates are usually not fast enough to provide annual resolution.

The adoption of this methodology in the framework of regional pollution assessments needs a careful selection of representative sampling sites and high-quality sediment dating protocols. The IAEA has carried out several actions in this direction, for example the IAEA Technical Cooperation Project RLA7012 Use of Nuclear Techniques to Address the Management Problems of Coastal Zones in the Caribbean Region, the book Radiochronology of Coastal Sediments Using ^{210}Pb: Models, Validation and Applications (Sanchez-Cabeza et al., 2012), and the ongoing Coordinated Research Project CRP K41016 Study of Temporal Trends of Pollution in Selected Coastal Areas by the Application of Isotopic and Nuclear Tools, expected to provide revised and useful recommendations.

Further work

This effort should be maintained. To the best of our knowledge, existing regional and global assessments (such as Regional Seas, HELCOM, OSPAR and TWAP) do not include the analysis of sediment cores in their protocols. This work shows that contaminants reconstruction from environmental archives provides information on both historical and recent trends. This may allow decision-makers to assess the effectiveness of regulation of some contaminants and to focus attention on contaminants with increasing trends. Both regional and global assessments (such as the GEF-funded project TWAP, and the UN General Assembly funded World Ocean Assessment) could benefit of this approach.

A permanent mechanism should be setup to enlarge the existing database with papers not included in this review and those published

since 2015. Individual LME reports should be produced and updated, especially for data rich areas. We suggest that world assessment could be produced regularly (e.g. each decade). The potential use of other information sources (such as technical reports) must be debated, as a peer review procedure should be performed before information is collected and archived in a repository.

New assessments should be made by small LME working groups, knowledgeable of the scientific issues reported here, and periodically coordinated by a small leading group. The organization of specialized workshops (maybe as special sessions of large international congresses) could be an effective way to periodically share data and experiences worldwide.

Some suggested improvements for future reviews are:

- Carry out new and intensive bibliographic searches in LMEs with little or no information.
- Perform analysis on specific contamination problems, for example siltation, by using accumulation rates and geochemical indicators.
- Revise and expand the list of contaminants considered. This could include the so-called emerging contaminants, such as micro-plastics. In many regions, large emphasis must be made on the analysis of organic contaminants. It is surprising that many records do not include relatively simple to measure nutrient concentrations (N and P), as nutrient enrichment might indicate diverse contaminant sources, such as agricultural and urban waste, and several industrial processes. Also, the possibility of including organic C in these analyses might be explored, although it should be considered that, besides the anthropogenic disturbances, organic carbon concentrations might be affected by aerobic and anaerobic degradation (and the release from the sediments in the form of CO_2 or CH_4) as well as chemosynthesis.
- Expand the matrices to be included in the analysis, in case the time series cover several decades (such as mussel watch time series).
- Complement LME analyses with assessments in larger regions (e.g. Southern and East Asia, Northern Europe, South America).
- Critically revise and improve statistical techniques to provide robust and continuous time trend analysis. This could be best achieved through an experts' meeting, gathering the knowledge and experiences in data rich areas (such as the Baltic Sea).

ACKNOWLEDGEMENTS

The Working Group was sponsored through the generous support of the International Atomic Energy Agency, the United Nations Industrial Development Organization and the International Maritime Organization. GESAMP wishes to thank the thorough peer review of the manuscript by Scott Fowler, Michael Angelidis and Albert Palanques and the following GESAMP Members Robert Duce, Manmohan Sarin, Jan Linders, Ahmad Hamed Abu-Hilal, Peter Kershaw and Alex Baker. Any omissions or errors remain the responsibility of the editors. Special recognition is given to Sandor Mulsow, for his collaboration in the inception phase of the Working Group; León Felipe Álvarez León, head of the Unidad de Informática Marina (UNINMAR, UNAM) for the development of the Working Group 39 web platform; to Brenda Berenice Martínez Villa for data collection and production os several figures; Dunia Ríos Yunes and Marisol Meneses Fernández for their assistance in the collection of information; and NOAA-NMFS (National Marine Fisheries Service) Large Marine Ecosystems Program for assistance with maps of the new LMEs 18 and 66 (accompanying the LME report titles). Thanks are due to Syvlia Sanders, Iolanda Osvath, Michael Angelidis and Jae Oh for their dedicated encouragement and administrative support during the development of this work.

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GLOSSARY

Acronyms

AMAP	Arctic Monitoring and Assessment Programme
ATSDR	Agency for Toxic Substances and Disease Registry
DOI	Digital Object Identifier
EMODnet	European Marine Observation and Data Network
EPA	Environmental Protection Agency
FAO	Food and Agriculture Organization
GESAMP	Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection
HABs	Harmful Algal Blooms
HELCOM	Baltic Marine Environment Protection Commission - Helsinki Commission
IAEA	International Atomic Energy Agency
IMO	International Maritime Organization
IOC	Intergovernmental Oceanographic Commission
ISO	International Organization for Standardization
LME	Large Marine Ecosystem
MED POL	Programme for the assessment and control of marine pollution in the Mediterranean
NMFS	National Marine Fisheries Service (NOAA)
NOAA	National Oceanic and Atmospheric Administration
NORM	Naturally occurring radioactive material
PAME	Protection of the Arctic Marine Environment
SI	International System of units
TWAP	Transboundary Waters Assessment Programme
UN	United Nations
UNAM	Universidad Nacional Autónoma de México (National Autonomous University of Mexico)
UNDP	United Nations Development Programme
UNEP	United Nations Environment Programme
UNESCO	United Nations Educational, Scientific and Cultural Organization
UNIDO	United Nations Industrial Development Organization
UNINMAR	Unidad de Informática Marina (Marine Informatics Unit, UNAM; www.icmyl.unam.mx/uninmar)
WG39	Working Group 39 (GESAMP)
WHO	World Health Organization
WMO	World Meteorological Organization
WWII	World War II

Contaminants

Metals and metalloids (for simplicity, referred as Metals in this work)

Ag	silver	Hg	mercury
As	arsenic	Ni	nickel
Ba	barium	Pb	lead
Cd	cadmium	Sn	tin
Cr	chromium	V	vanadium
Cu	copper	Zn	zinc

Radionuclides

Am	americium (^{241}Am)	Ra	radium (^{226}Ra)
Cs	caesium (^{137}Cs)	Th	thorium (^{232}Th)
Pb	lead (^{210}Pb)	U	uranium (^{238}U)
Pu	plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu)		

Pesticides

CHL	chlordane	HCB	hexachlorobenzene
DDT	dichlorodiphenyltrichloroethane		

Industrial organic chemicals and byproducts

PCB	polychlorinated biphenyl	PCDD	polychlorinated dibenzo-p-dioxins (dioxins)
PBDE	polybrominated diphenyl ether	PCDF	polychlorinated dibenzofurans (furans)

Polynuclear Aromatic Hydrocarbons

PAHs	polynuclear aromatic hydrocarbons		
Ace	acenaphthene	Ch	chrysene
Acy	acenaphthylene	D[a,h]An	dibenzo(a,h)anthracene
An	anthracene	Flt	fluoranthene
B[a]An	benzo(a)anthracene	Flu	fluorene
B[a]Py	benzo(a)pyrene	Ipy	indeno(1,2,3-cd)pyrene
B[b]Flt	benzo(b)fluoranthene	Na	naphthalene
B[ghi]Pe	benzo(g,h,i)perylene	Phe	phenanthrene
B[k]Flt	benzo(k)fluroanthene	Py	pyrene

Nutrients

N	nitrogen	Si	silicon
P	phosphorus		

GESAMP Working Group 39 members.

Lead Agency: International Atomic Energy Agency
Iolanda Osvath, IAEA Technical Secretary for GESAMP

Co-sponsor: United Nations Industrial Development
Christian Susan, UNIDO Technical Secretary for GESAMP

WG39 Member	Country	Affiliation	Expertise
Ms. Ana Carolina Ruiz-Fernández (chairperson)	Mexico	Universidad Nacional Autónoma de México	Global change, ^{210}Pb dating, marine pollution by heavy metal and nutrients
Ms. Roberta Delfanti	Italy	Marine Environment Research Centre-ENEA	Radionuclides as ocean tracers of physical processes
Mr. José Marcus Godoy	Brazil	Pontificia Universidade Catolica do Rio de Janeiro	Environmental analytical chemistry
Mr. Elvis Nyarko	Ghana	University of Ghana	Biomonitoring of aquatic contaminants, radionuclide dating
Mr. Joan-Albert Sanchez-Cabeza	Mexico	Universidad Nacional Autónoma de México	Radioactive tracers for marine environmental processes, global change
Mr. José L. Sericano	United States of America	Texas A&M University	Determination, impact and fate of trace organic contaminants in the environment
Ms. Elvira Sombrito	Philippines	Consultant (previously at Philippine Nuclear Research Institute)	Marine environmental pollution, environmental impact assessment
Mr. Norbert Theobald	Germany	Federal, Maritime and Hydrographic Agency	Persistent organic pollutants in the marine environment

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ISSN 1020–4873