



Trace metals and polycyclic aromatic hydrocarbons in the Eastern Mediterranean sediments: Concentration ranges as a tool for quality control of large data collections

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ABSTRACT

Assessing the status of marine pollution at regional and sub-regional scales requires the use of comparable and harmonized data provided by multiple institutions, located in several countries. Standardized data management and quality control are crucial for supporting a coherent evaluation of marine pollution. Taking the Eastern Mediterranean Sea as a case study, we propose an approach to improve the quality control procedures used for sediment pollution data, thus supporting a harmonized environmental assessment. The regional ranges of contaminant concentrations in sediments were identified based on an in-depth literature review, and the lowest measured concentrations were evaluated to determine the “background concentrations” of chemical substances not yet targeted in the Mediterranean Sea. In addition, to verify the suitability of the approach for validating large data collections provided by multiple sources, the determined ranges were used to validate a regional dataset available through EMODnet data infrastructure.

1. Introduction

About one third of the population of the Mediterranean countries is concentrated in coastal zones, particularly in the western Mediterranean, the west coast of the Adriatic Sea, the east coast of the Aegean and the Levantine, and the Nile Delta (UNEP/MAP, 2017). Industry, which is often located along the region's coasts, in areas with high population density, is a major source of pollutants to coastal and marine environments and interacts with other types of anthropogenic pressures (e.g. urban wastes, maritime traffic, offshore oil and gas extraction). The

Mediterranean Sea is one of the busiest seas in the world, harvesting 20 % of seaborne trade, 10 % of world container throughput and over 200 million passengers.

About 2/3 of the oil spilled in the Mediterranean occurs in its eastern part (UNEP/MAP and Plan Bleu, 2020). The Eastern Mediterranean Sea, as a semi-enclosed basin, surrounded by densely populated areas (>315 million people; UNDESA, 2019) is under the influence of a large number of pollutants, including trace metals (TMs) and a large variety of organic compounds which include also polycyclic aromatic hydrocarbons (PAHs). PAHs inputs reach the sea through rivers (Albaigés, 2005),

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atmospheric deposition (Albaigés, 2005; Castro-Jiménez et al., 2012; Tsapakis and Stephanou, 2007), wastewater discharges, maritime traffic, offshore oil and gas extraction (Giani et al., 2020; Hatzianestis et al., 2020; Tornero and Hanke, 2016); furthermore, there are also natural sources due to fires and natural seepage (e.g.: Benlahcen et al., 1997).

Trace metals, on the other hand, may have mineralogical origin due to soil erosion (Roussiez et al., 2011) as well as anthropogenic origin due to mining, urban and industrial waste water discharges (e.g. Elbaz-Poulichet et al., 2011; Oursel et al., 2013), shipping, offshore activities, seabed mining (Tornero and Hanke, 2016), atmospheric deposition from land and from mineral dust from Africa (Bonnet and Guieu, 2006; Güllü et al., 1998; Jordi et al., 2012; Luck and Ben Othman, 2002; Migon, 2005; Ternon et al., 2010). In the Mediterranean countries, according to the National Baseline Budget (NBB) inventory (UNEP/MAP, 2012), atmospheric emissions of metals are mostly related to the cement industry (Hg, Cu), production of energy (As, Cd, Ni) and the metal industry (Pb, Zn). Metal emissions to water seem to be mostly related to the fertilizer industry (Hg, As, Pb), metal industry (Ni, Zn) and wastewater treatment plants (Cd, Cu), with significant contributions also from the energy sector and the chemical industry. Oil refining is the main source of Cr releases, both in air and water.

In coastal areas, contaminants inputs are mainly related to the land watersheds loads and their direct releases, whereas atmospheric fallouts dominate contaminant inputs in more remote offshore areas of the open sea (Berrojalbiz et al., 2014; Castro-Jiménez et al., 2012; Dang Duc et al., 2015; Durrieu de Madron et al., 2011; Guigue et al., 2014 and references therein). Several land and sea-based specific sources are reported e.g. mining activities, metallurgic industries, refineries, municipal- sewage outflows, harbors, offshore platforms and maritime traffic (Cukrov et al., 2011; Di Leo et al., 2013; Horvat et al., 2014; Joksimović et al., 2020) where pollution by metals is caused by inputs derived from mining, metal product fabrication, solid waste disposal, fossil fuel combustion, and municipal/industrial waste effluent (Cossa et al., 2018; Elbaz-Poulichet et al., 2011). In the marine environment, toxic metals readily accumulate in the sediments because of particles scavenging and sedimentation (Qian et al., 2015). Polycyclic aromatic hydrocarbons are semi-volatile organic contaminants (SOCs), ubiquitous in the global environment. In the marine environment, petroleum fuels and crude oil release operational and accidental spills and natural seeps are substantial sources of PAHs. Other natural PAH sources (e.g. forest fires, volcano eruptions, biogenic formation) also have a significant regional contribution on PAH occurrence (Howsam and Jones, 1998; Ravindra et al., 2008; Stout et al., 2004). Due to their low solubility and hydrophobicity, PAHs are also retained by fine suspended particles via sorption processes once they get into the marine environment and therefore also effectively accumulate in the sediments with deposition processes (Albaigés, 2005; Parinos and Gogou, 2016).

The assessment of the status of chemical pollution in the Mediterranean Sea is based on criteria defined by the Mediterranean Action Plan of United Nations Environment Program, by the Integrated Monitoring and Assessment Programme (IMAP) of the Mediterranean Sea and Coast (UNEP/MAP, 2016) proposed by the Barcelona Convention, by several EU Directives (WFD - Water Framework Directive, MSFD – Marine Strategy Framework Directive, EQS – Ecological Quality Standards) and by national level control and prevention plans. Both IMAP and MSFD require comparable data and harmonized and integrated approaches for environmental status assessment at the regional and sub-regional level. Large efforts are being conducted at European as well as at Mediterranean scale (e.g. EMODnet¹, IMAP Info System², INFO-RAC³) to overcome data fragmentation among different laboratories, research

institutions and environmental agencies, and to promote easy and open access to marine chemistry data needed to assess environmental status (Giorgetti et al., 2020; Vinci et al., 2017). Coherent data management and quality control become very relevant when environmental status is assessed at regional or sub-regional scale (e.g., for the Mediterranean or the Adriatic Sea), thus requiring the access to data from different sources (Lipizer et al., 2021b; Molina Jack et al., 2020). Data comparability depends, in fact, not only on sampling and analytical protocols under quality assurance and control, but also on how data and metadata are managed, validated and made accessible. Procedures for quality control on heterogeneous datasets, provided by multiple data sources, are not yet uniform and consolidated, and are needed. In the case of marine contaminants, the large and heterogeneous number of substances determined in the different matrices (seawater, sediment, marine organisms), with different sampling and analytical procedures make data quality control and, lastly, data comparability challenging.

Good Environmental Status (GES) with regard to chemical pollution can be assessed in relation to reference conditions, such as background concentrations (BC) of a compound in the pristine environment (European Commission, 2017). BCs of naturally occurring elements and compounds may show significant geographical variability in sediments, which pose an additional challenge for their assessments (Covelli et al., 2013; Ospar, 2006; Pérez-Fernández et al., 2019). The definition of BC in the Mediterranean is periodically revised, as new data become available (UNEP/MED, 2022).

Over the last decades, large efforts have been dedicated to implement and consolidate European data infrastructures (e.g. SeaDataNet⁴, EMODnet⁵, Giorgetti et al., 2020; Míguez et al., 2019) to overcome the fragmentation in data and information availability. However, quality of available data as well as data comparability, particularly in the case of marine contaminants, are still critical issues and procedures for data quality control need to be consolidated. Within the project HarmonIA⁶ (Harmonization and networking for contaminant assessment in the Ionian and Adriatic Seas), financed by the INTERREG VB-ADRION programme (2018–2020), research institutions and environmental agencies from countries bordering the Adriatic and Ionian Seas (Italy, Slovenia, Croatia, Montenegro, Albania and Greece) contributed to the collection, harmonization and creation of accessible datasets for marine contaminants through a common and already consolidated infrastructure, and shared knowledge and information to improve a coherent and harmonized assessment of marine pollution.

The aim of this work is to propose a methodology that improves data quality control procedures for regional or sub-regional (e.g. Eastern Mediterranean) datasets on marine pollutants determined in sediments. This was done by identifying regional ranges of contaminant concentrations, with a focus on the Eastern Mediterranean Sea. The proposed method targets selected trace metals and PAHs assessed in marine sediments in the frame of previously mentioned environmental directives and monitoring programs for this region (e.g. MSFD, IMAP). Contaminant selection was done because of the great heterogeneity of monitored pollutants by the different countries (Lipizer et al., 2020). The approach was based on an *in-depth* literature review and methods comparison of regional concentration ranges of a group of TMs and PAHs for a variety of environments, to be used as references for data quality control, as common practice in data management activities (Cabanes et al., 2013; Lipizer et al., 2021a). The proposed regional ranges of concentrations are, in the end, tested on a sub-regional dataset covering the Adriatic - Ionian Seas, in order to evaluate the adequacy of these concentration limits to improve the internal coherence and quality of the dataset. Lastly, re-analysis of literature would support the evaluation of background concentrations of a set of substances for which information is

¹ <https://www.emodnet-chemistry.eu/>

² <http://imappilot.info-rac.org/app/>

³ <http://www.info-rac.org/en>

⁴ <https://www.seadatanet.org/>

⁵ <https://emodnet.ec.europa.eu/en>

⁶ <https://harmonia.adrioninterreg.eu>

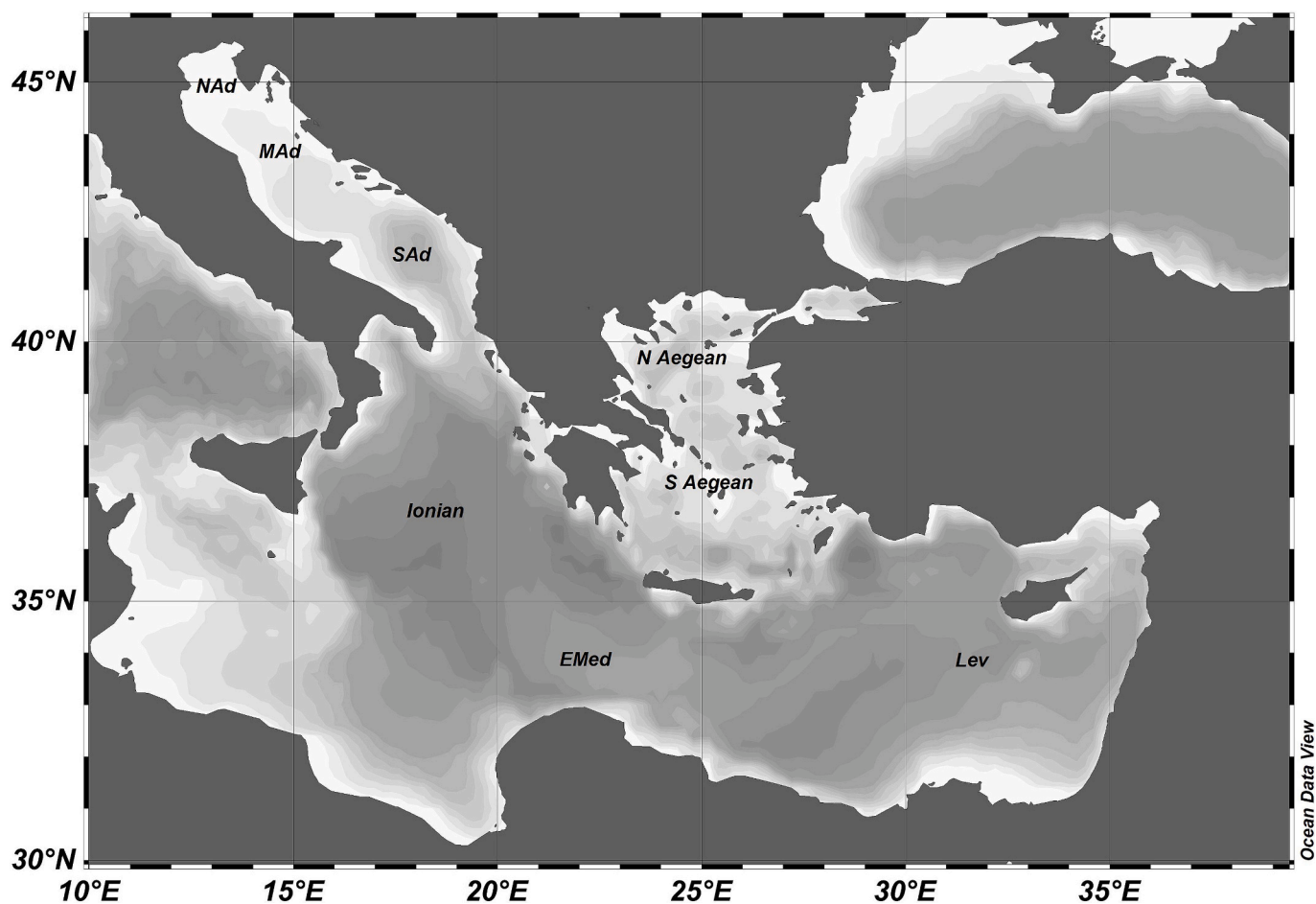


Fig. 1. Area considered in this study: EMed: Eastern Mediterranean Sea; NAd: North Adriatic Sea; MAd: Middle Adriatic Sea; SAd: South Adriatic Sea; Ionian: Ionian Sea; N Aegean: North Aegean Sea; S Aegean: South Aegean Sea; Lev: Levantine Sea.

still limited, for different Mediterranean areas.

2. Study area

The Mediterranean Sea is the largest and deepest semi-enclosed sea on Earth with an area of about $2.5 \times 10^6 \text{ km}^2$ and an average depth of 1.5 km. The greatest depth measured in the Mediterranean Sea is approx. 5.3 km in the Ionian Sea (Calypso Deep, offshore West Peloponnese, Greece). Rivers drain an area of $1.5 \times 10^6 \text{ km}^2$ (Ludwig et al., 2009) and the densely populated coastal zone extends for about $46 \times 10^3 \text{ km}^2$. The Sicilian-Tunisian sill, that is 400 m deep, acts as a geographical and hydrological frontier dividing the Mediterranean basin into two large distinct sub-basins: the western and the eastern basins. The Eastern Mediterranean Sea is known to be the largest sub-region of the Mediterranean and is subdivided geographically as several distinct basins: (a) the Adriatic Sea, (b) the Ionian Sea, (c) the Aegean Sea and (d) the Levantine Sea (Fig. 1).

3. Methods

The first step was an extensive scientific literature review carried out to identify ranges of TMs and PAHs concentrations in marine sediments reported for the Eastern Mediterranean Sea. The review also included analysis of sampling, sample processing and analytical procedures adopted, thus allowing better data comparability among considered scientific references. Upon this examination, a list of key articles was selected, according to the most suitable methodological approaches. Particular focus was dedicated to evaluate the lowest concentrations for

selected TMs and PAHs that are included in national and international monitoring programmes (e.g. MSFD, WFD, UNEP-MAP-MEDPOL), in order to propose “background concentrations (BC)” for substances where BCs have not been proposed for the Mediterranean yet (UNEP/MED, 2021). These values should represent the concentration that would be expected in a “pristine” or “remote” site based on contemporary and/or historical data (Bignert et al., 2004; UNEP (DEPI)/MED, 2011). To define a background concentration, the “geochemical method” was used (Dung et al., 2013). It is based on mean or median values of trace metals in core sediment samples at depth layers that are assumed to have a pre-industrial origin. A similar approach was adopted also for PAHs, as sediment cores were used for the estimation of pre-industrial fluxes of these compounds towards the seafloor (Azoury et al., 2013; Pérez-Fernández et al., 2019). In the present work, background concentrations are derived from subsurface sediment concentrations, which, according to sedimentation rates or to the dating of sediment cores, can be considered as representative of the pre-industrialization period. Lagoon and river delta sediments were not considered in this review, as they represent highly human perturbed environments also during the pre-industrial period. Data collected in different environments, at variable distances from pollution sources (e.g. deep offshore stations, shallow coastal stations close to ports), were used to evaluate ranges of contaminant concentrations, from “hot spots” to sites far from direct pollution impacts.

Because of the large number and the heterogeneity of substances measured in marine sediments by different research institutions and environmental agencies, this study addresses a selection of TMs and PAHs which are targets of several environmental directives

(Supplementary material S1 derived from [Tornero and Hanke, 2017](#)).

As a second step, to validate data collections based on data provided by multiple laboratories and to detect possible outliers, ranges established from the literature were compared with data gathered within the framework of the HarmonIA project and open access data available through EMODnet Chemistry data infrastructure (EMODnet [Chemistry, 2018](#)).

3.1. Analytical methods

To guarantee data comparability from different sediment cores, offshore and coastal sediments, the evaluation of the concentration ranges was based on a list of studies selected according to comparability of analytical methods used for contaminants determination. Due to the heterogeneity in sediment samples treatment (e.g. unsieved, sieved through different mesh sizes), only concentrations determined on unsieved sediments or sieved through 2 mm, were considered, because the fine-grained sediment (<0.063 mm) contains mainly the pelitic fraction which is characterized by higher metal and PAHs content ([Salomons and Förstner, 1984](#)). However, due to incomplete or not comparable information on sediment grain size or on cofactors commonly used for normalization purposes (e.g. Al, Li or organic carbon content), concentration variability between samples arising from differences in bulk sediment properties (e.g. grain size, organic carbon content) could not be ruled out.

Regarding TMs and PAHs, BCs were obtained from sediment cores available for the Eastern Mediterranean region, as outlined below. However, due to the scarcity of sampling sites, only a limited variety of sedimentological environments is represented by these cores. In order to derive ranges of TMs and PAHs concentrations and to take into account the natural variability of sedimentological environments, data collected from deep offshore surface sediments were also considered ([Fig. 2a, b](#)). In the above-mentioned cores, the deepest sediment layer corresponds to 1910 or earlier, depending on the length of the core and the sedimentation rate. The direct methods ([Gałuszka and Migaszewski, 2011](#); [Shahabi-Ghahfarokhi et al., 2021a, 2021b](#)) were used to estimate the BC if not reported in the selected works.

3.1.1. Trace metals

A summary of analytical methods used in the considered literature is presented in Supplementary material S2 Parts A and B. Only data measured according to analytical methods able to determine the total concentration in the sediment of the elements targeted in this study (As, Cd, Hg, Pb, Cu, Cr, Ni, Zn, Co, V, Supplementary material S1) were used to evaluate ranges of concentrations. Specifically, X Ray Fluorescence Spectroscopy (XRF) ([Barra et al., 2020](#); [Karageorgis et al., 2005a, 2005b](#); [Lucchini et al., 2003](#)) applied to the bulk sediment, and Flame, Graphite Furnace or Cold Vapor Atomic Absorption Spectroscopy (FAAS; GFAAS; CVASS) and Inductively Coupled Plasma Atomic Emission Spectroscopy and Inductively Coupled Plasma Mass Spectrometry (ICP-AES, ICP-MS) applied to the solution obtained after the total dissolution of the bulk sediment by strong acid mixtures containing HF in teflon bombs (after heating or digestion in microwave oven), guarantee a correct determination of the total concentrations of trace metals ([Acquavita et al., 2010](#); [Aloupi and Angelidis, 2001](#); [Angelidis et al., 2011](#); [Angelidis and Aloupi, 1995](#); [Buccolieri et al., 2006](#); [Covelli and Fontolan, 1997](#); [Giani et al., 1994](#); [Ilijanić et al., 2014](#); [Lopes-Rocha et al., 2017a, 2017b](#)). Exception was Hg which was determined by direct thermal treatment of the bulk sediment, preconcentration on an amalgamator, desorption and determination by AAS ([Azoury et al., 2013](#); [Buccolieri et al., 2006](#); [Joksimović et al., 2020](#)), and CVAAS ([Acquavita et al., 2010](#); [Covelli et al., 2006](#); [Ergin et al., 1996](#); [Giani et al., 1994](#); [Ilijanić et al., 2014](#); [Soliman, 2015](#)) (Supplementary material S2 Parts A and B).

All the reported accuracies for the targeted TMs were within $\pm 10\%$ from the certified values ([Acquavita et al., 2010](#); [Aloupi and Angelidis, 2001](#); [Buccolieri et al., 2006](#); [Dolenec et al., 1998](#); [Ergin et al., 1996](#);

[Giani et al., 1994](#); [Ilijanić et al., 2014](#); [Karageorgis et al., 2005a, 2005b](#); [Lopes-Rocha et al., 2017b](#); [Lucchini et al., 2003](#); [Ujević et al., 1998](#)) with the exception of Cd ([Acquavita et al., 2010](#)), Pb ([Lopes-Rocha et al., 2017b](#)) and Co ([Ergin et al., 1996](#)) for which the accuracy was <15%. The reproducibility reported in the same studies was <5% for Cd, As, Cu, and V, <6% for Cr, <8% for Zn, <10% for Pb and Hg, and <14% for Co. The cumulative error in the determination of TMs in sediments was assumed to be $\pm 10\%$ ([Birch et al., 2001](#)).

3.1.2. PAHs

Background concentrations of PAHs can derive from natural sources such as petrogenic or pyrogenic (biomass burning) sources which determine a non-zero level also in pre-industrial time. The background levels were derived from concentrations earlier reported in sediment cores ([Fig. 2b](#)). As the number of studied cores is limited, they may represent a narrow range of a variety of sedimentological environments. Therefore, the surface sediments of offshore stations were also considered to define the regional ranges of concentrations. Among available literature, a set of publications were chosen according to the analytical methods adopted, the sediment pre-treatment protocols and regional availability of data for each PAH compound.

PAHs can be determined in marine sediments with a variable percent of error, depending on the targeted compounds. Unfortunately, the recoveries, accuracies, precisions, and Limits of Detection (LOD) are usually not reported for the single compounds but as a range for the whole class of determined PAHs (Supplementary material S3) or for subgroups depending on the number of aromatic rings.

Procedures used for PAHs extraction to derive background levels and ranges in offshore and coastal sediments were based on pure solvent or a mixture of solvents (*n*-hexane, hexane/acetone, or dichloromethane/methanol) usually with a Soxhlet apparatus ([Bihari et al., 2006](#); [Botsou and Hatzianestis, 2012](#); [Cardellicchio et al., 2007](#); [Caricchia et al., 1993](#); [Gogou et al., 2000](#); [Hatzianestis et al., 2004, 2020](#); [Tsapakis et al., 2010](#)). In a few selected cases the extraction was carried out with dichloromethane by Accelerated Solvent Extraction (ASE) ([Azoury et al., 2013](#); [Combi et al., 2020](#); [Mandić et al., 2018](#); [Mandić and Vrancic, 2017](#)) or by ASE and Supercritical Fluid Extraction (SFE) ([Heath et al., 2006](#); [Notar et al., 2001](#)). After purification of the extracts, PAHs were determined in most of the cases by Gas Chromatography - Mass Spectrometry (GC-MS) ([Azoury et al., 2013](#); [Botsou and Hatzianestis, 2012](#); [Cardellicchio et al., 2007](#); [Gogou et al., 2000](#); [Hatzianestis et al., 2004, 2020](#); [Mandić and Vrancic, 2017](#); [Notar et al., 2001](#); [Parinos et al., 2013](#); [Tsapakis et al., 2010](#)), or by Gas Chromatography coupled to a Flame Ionization Detector (GC-FID) and GC-MS ([Zaghden et al., 2017](#)) or by HPLC with a fluorescence detector ([Caricchia et al., 1993](#)) or by a fluorescence and UV detector ([Bihari et al., 2006](#)).

The Limits of detection (LOD) or quantification (LOQ) were not usually reported for single compounds, but rather as ranges for all of the determined PAHs varying from 0.01 to 9 $\mu\text{g kg}^{-1}$ (Supplementary material S3). Reported recoveries ranged from 60 to 107%, the precision was generally between 5 and 19% (Supplementary material S3).

A cumulative error (analytical + sampling) in the determination of a PAH in a sediment was assumed to be $\pm 15\%$ ([Jiang et al., 2009](#)).

3.2. Identification of regional ranges of contaminant concentrations for data Quality Control

Based on the literature review, regional concentration ranges (RMin-RMax) for the EMed were identified for selected TMs and PAHs (Supplementary material S1) on the basis of the lowest and the highest concentrations, respectively, reported for BCs, for offshore and coastal sediments. These concentration ranges were used to calculate concentration limits (R'Min - R'Max), which take into account also possible sampling and analytical errors, to be proposed for data Quality Control (QC). A maximum error (analytical + sampling) in the determination of TMs concentrations in a sediment was assumed to be $\pm 10\%$ ([Birch](#)

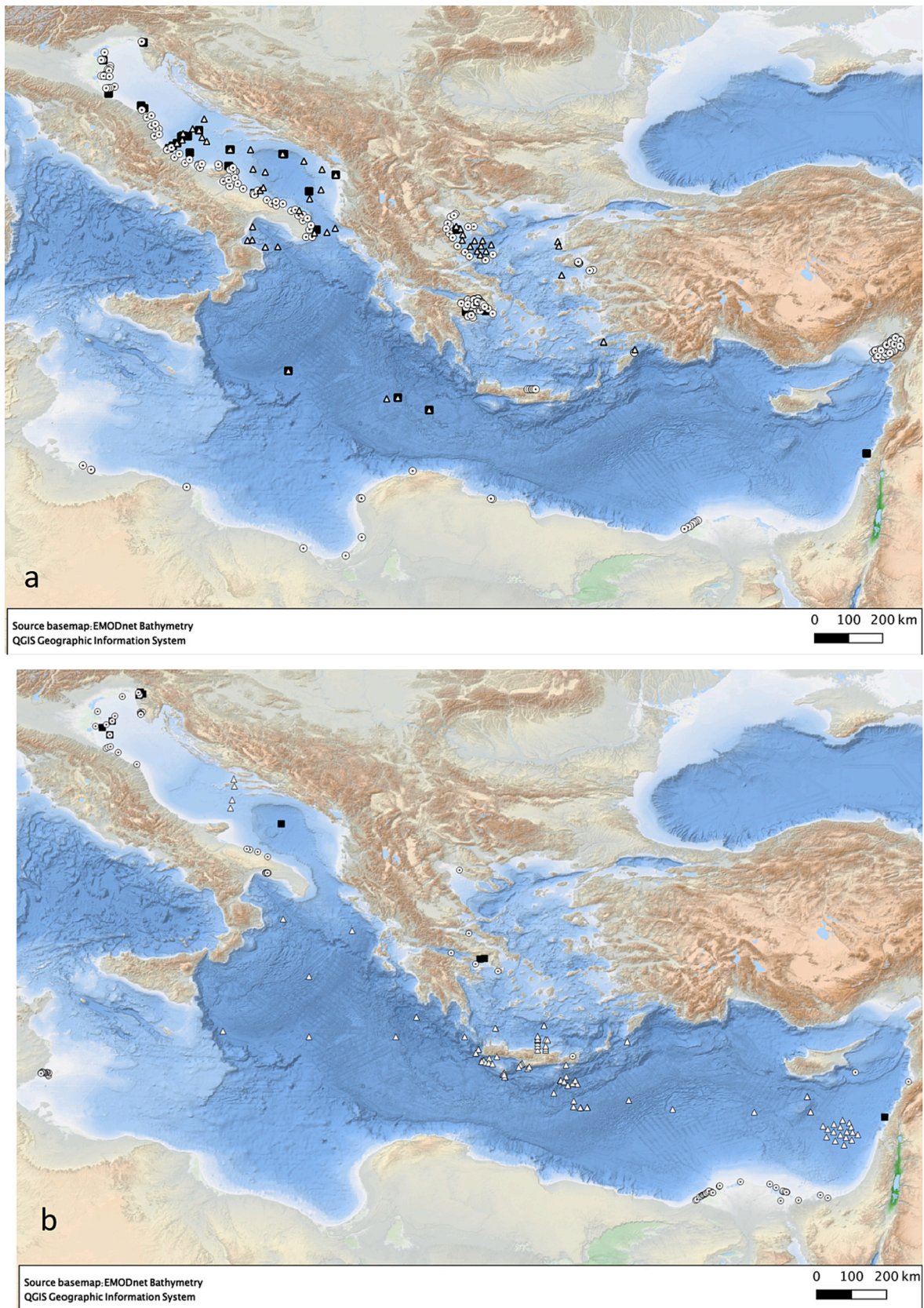


Fig. 2. Spatial distribution of stations considered to derive concentration ranges in sediment cores (black squares), surface offshore (white triangles) and coastal sediments (white dots) for trace metals (a) and polycyclic aromatic hydrocarbons (b).

et al., 2001), which is in accordance with the precision of the methods reported in the scientific articles used to define the ranges (Supplementary material S2 - Part A). Therefore, TMs concentrations higher than $R_{Max} + 10\%$ (R'_{Max}) or lower than $R_{Min} - 10\%$ (R'_{Min}) were considered “to be cross-validated”, meaning that they should be verified with the data originators. In the case of single PAH determination, an error of $\pm 15\%$ was assumed, in accordance with the precision reported in the papers used for the definition of the ranges (Supplementary material S2 - Part B). Therefore, PAHs concentrations below the lowest limit, $R_{Min} - 15\%$, or higher than $R_{Max} + 15\%$, were considered “to be re-checked”.

3.3. Data used for the Quality Control

For evaluation of the proposed concentration ranges as suitable reference to control the quality of large datasets deriving from different laboratories and to propose main targets of data QC procedures, unrestricted data available for the Adriatic and Ionian Seas through the European Marine and Data Network (EMODnet Chemistry), together with data made accessible within the ADRION HarmonIA project, were used (Fig. 3). These data are part of the Mediterranean Sea - Contaminants aggregated datasets 1974/2017 v2018 (EMODnet Chemistry, 2018), spanning the period from July 1981 – July 2017, and contain the following categories of contaminants: antifoulants, heavy metals, hydrocarbons, pesticides and biocides, pharmaceuticals and PCBs, which include >190 different substances.

Data available through EMODnet Chemistry were managed according to standard protocols developed within the framework of the European SeaDataNet (SDN) marine data infrastructure, that has implemented consolidated communication standards and tools, common data and metadata models and common file formats (Giorgetti et al., 2020). Consequently, the whole dataset is homogeneous as measured parameters are coded with controlled vocabularies (BODC Parameter Usage Vocabulary terms, P01 vocabulary) and described with standard metadata (with information on cruises, spatial and temporal coordinates, data originators, observing systems, sampling protocols, data access conditions) required for describing geographic information and services (ISO 19115 metadata content standard). Furthermore, data have undergone a first validation according to common procedures (Lipizer et al., 2021a), have been flagged according to a common scheme (SDN L20 vocabulary) and are available through standard Ocean Data

View (ODV) data format (Schlitzer, 2021). Yet, high heterogeneity in analyzed substances by the different laboratories, in measurement units, in matrix characteristics (i.e. un-sieved, sieved, different size classes), in sampling, analytical and normalization protocols, all contribute to make data management, validation and comparability challenging (Molina Jack et al., 2020).

3.4. Data Quality Control

The proposed data validation method followed standard QC procedures proposed in the framework of EMODnet Chemistry (Lipizer et al., 2021a), adopting the Quality Flag (QF) scheme used within SeaDataNet pan-European data infrastructure (SDN L20). The comparison of experimental data with regional ranges of concentration proposed in this study (R'_{Min} and R'_{Max}) was then carried out as an innovative QC phase. The following QC steps were carried out on the original EMODnet dataset, reporting errors and inconsistencies to data originators to correct erroneous data:

- check of inconsistent measurement units
- inspection of anomalous data distribution
- identification of data with “zero” value of concentration and flagging as “value below detection limit (QF = 6)”
- identification of negative values of concentration (not related to variables that admit negative values) and flagging as “bad values (QF=4)”
- identification of data below R'_{Min} and flagging as QF = 3 - probably bad data
- identification of data within the range R'_{Min} - R'_{Max} and flagging as “good (QF = 1)”
- identification of data above R'_{Max} : further analysis of data spatial and temporal distribution to distinguish between possible erroneous data (flagged as QF = 4 - bad data or QF = 3 - probably bad data) and actual pollution “hot spots”.

Data passing these checks maintain QF assigned by data originators or are flagged as “good (QF = 1)”. Lastly, dataset formal check included inspection of availability of relevant metadata and of supporting information such as: station bottom depth, thickness of sampled sediment layer, depth of the sampled sediment layer, spatial and temporal coordinates, sediment grain size, organic carbon, and aluminum content,

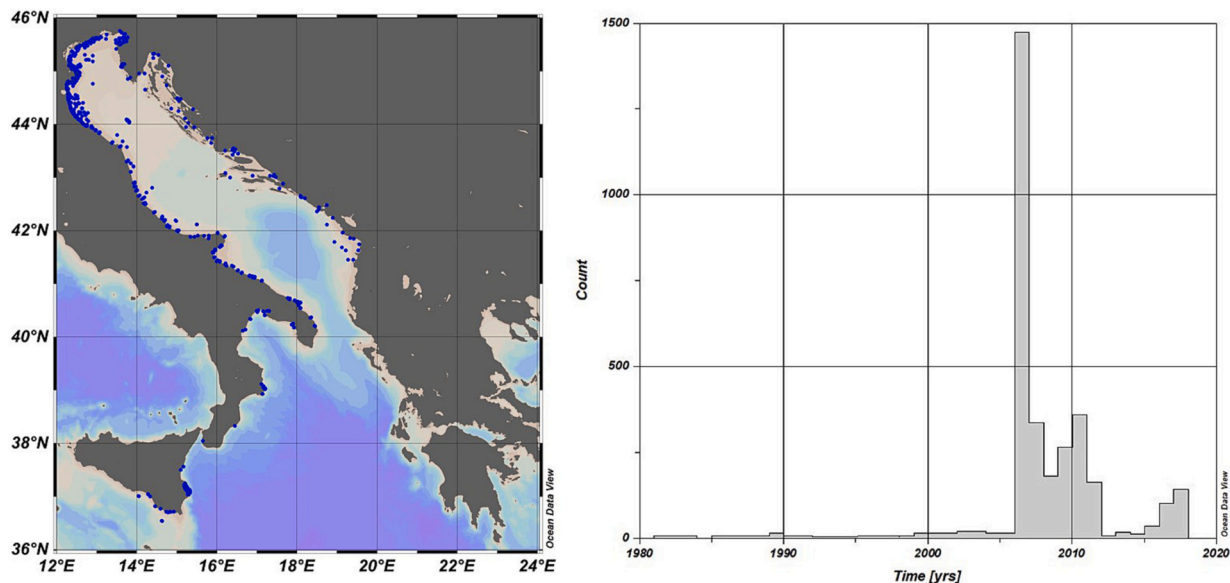


Fig. 3. Study area with spatial (left) and temporal (right) distribution of sediment sampling stations in the Adriatic and Ionian Seas (EMODnet Chemistry 1974/2017 v2018 and HarmonIA data).

which are needed for normalization procedures. The availability of detailed and accurate metadata is of fundamental relevance for data QC and for its improvement.

3.5. Data elaboration

Basic statistics (minimum and maximum value) were extracted or calculated from published data, to assess the ranges of contaminant concentrations of the substances listed in Supplementary material S1. As a second step, data available for the Adriatic - Ionian Seas (EMODnet Chemistry, 2018) already flagged as “good” and “probably good” (Sea-DataNet QF 1 and 2, respectively) were selected and min-max ranges of concentrations of selected substances (Supplementary material S1) within the whole dataset were calculated in order to identify possible inconsistencies in the data.

Figures presented in this work were produced by open source Ocean Data View Software, ODV (Schlitzer, 2021).

4. Results and discussion

4.1. Trace metals concentrations

4.1.1. Trace metal background concentrations derived from sediment cores data

There is a shortage of background concentrations derived from sediment cores for the Eastern Mediterranean (Fig. 2a). Therefore, besides BCs directly published in several articles (Barra et al., 2020; Karageorgis et al., 2020; Lopes-Rocha et al., 2017a), other BCs were estimated from data available in the literature (Angelidis et al., 2011; Azoury et al., 2013), or provided directly by the authors (Ilijanić et al., 2014; Ogrinc et al., 2007) (Table 1 and Supplementary material S4). All data are expressed as $\mu\text{g kg}^{-1}$ dry weight as requested by the Integrated Monitoring and Assessment Protocol (IMAP) of the Barcelona Convention (UNEP/MED, 2019), and as proposed in the framework of EMODnet Chemistry data management to facilitate data interoperability (Lipizer et al., 2022).

Table 1 and Supplementary material S4 present the review of BCs derived from sediment cores in several areas of the Mediterranean and the Adriatic Seas (Fig. 2a). The ranges of BCs reported in the literature may span up to 1 order of magnitude as in the case of Hg and Cu. The variability in BC ranges may be ascribable to different sediment size as well as to differences in geochemical provinces. Although detailed information on sediment grain size was not always available in the considered articles (Supplementary materials S4, S6, S7), positive

Table 1

Background Concentrations for trace metals in sediments of the Eastern Mediterranean Sea ($\mu\text{g kg}^{-1}$ dry weight), based on the following references: Angelidis et al., 2011; Azoury et al., 2013; Barra et al., 2020; Covelli and Fontolan, 1997; Frignani et al., 2005; Giani et al., 1994; Heath et al., 2006; Ilijanić et al., 2014; Karageorgis et al., 2005a, 2020; Lopes-Rocha et al., 2017a; Lucchini et al., 2003; Ogrinc et al., 2007; Oliveri et al., 2013; Van Santvoort et al., 1996. UNEP/MED, 2021 (used as comparison, not to propose BC ranges). For full reference and value overview see Supplementary material S4.

Element	Units	BC ranges	
		BCMin	BCMax
Cd	$\mu\text{g kg}^{-1}$ dw	118	600
Hg	$\mu\text{g kg}^{-1}$ dw	12	130
Pb	$\mu\text{g kg}^{-1}$ dw	10,000	46,000
As	$\mu\text{g kg}^{-1}$ dw	4,000	19,100
Cr	$\mu\text{g kg}^{-1}$ dw	35,990	194,200
Cu	$\mu\text{g kg}^{-1}$ dw	4,100	49,150
Ni	$\mu\text{g kg}^{-1}$ dw	23,000	195,000
Zn	$\mu\text{g kg}^{-1}$ dw	34,600	103,000
Co	$\mu\text{g kg}^{-1}$ dw	11,800	29,100
V	$\mu\text{g kg}^{-1}$ dw	34,465	109,000

relationships between some TMs (Cu, Co, Zn, Supplementary material S5) and the percentage of silt and clay suggest that part of the variability may be due to sediment granulometry. On the other hand, differences in geochemical provinces of origin and the presence of ancient anthropogenic activities, which have impacted trace metal concentrations for a long time, may also contribute to the observed BC variability. The Northern Adriatic (NAd) is, in fact, influenced by the Hg mine of Idrija (Covelli et al., 2001, 2006) which represents a geochemical anomaly in European soils (Ottesen et al., 2013). The historical development of Hg contamination is well recorded in core sediments, and it varies according to Hg data production since the beginning of the 1800s when extraction activity began to grow exponentially (Covelli et al., 2006). The Hg baseline ($130 \mu\text{g kg}^{-1}$) determined by Covelli et al., 2006 in the Gulf of Trieste was therefore considered as the average of concentrations in the basal levels of the cores. This geographical difference is also evident in background concentrations for Hg reported by UNEP/MED, 2021 for the Eastern Mediterranean, which range from 31 in the Levantine Sea to $107 \mu\text{g kg}^{-1}$ in the Adriatic, and which are in line with BCs calculated in this review. In addition, Pb could have an anthropogenic origin according to the high enrichment factors estimated in this area by (Cibic et al., 2008). Besides, a clear increasing gradient of Pb, Zn and PAHs concentrations was recognized after 1900 and could be related to the increase of harbor and industrial activities and oil and coal burning (Vidovic et al., 2016). Considering Cr range, high concentrations found in the Adriatic and in the Thermaikos Gulf are attributed to weathering of ophiolite complexes from catchments Mirdita in Albania (Ilijanić et al., 2014) or from ultrabasic rocks in the Po River catchment (Amorosi and Sammartino, 2007), and possibly from ophiolite complexes in the Axios and Aliakmon River catchments in northern Greece (Karageorgis et al., 2005a).

Regarding the higher Pb background (2–4 fold higher than the BCs found in other EMed areas) in the outer Saronikos Gulf, this can be attributed to the mineral geology of the area, with deposits of Pb-Zn-Ag sulfides and/or oxides, where the mining activities (Lavrion, South Attica) date back to the third millennium BC (Voudouris et al., 2021). Another high background ($34,000 \mu\text{g kg}^{-1}$) value was found in the Holocene sediments in SAd (Lucchini et al., 2003), which could have been influenced by the presence of the organic-rich sediment layers (sapropel) (Van Santvoort et al., 1996), as the BC of Pb in the Adriatic generally ranges from 10,000 to $23,000 \mu\text{g kg}^{-1}$ (Table 1 and Supplementary material S4) and in the Eastern Adriatic coastal sediments from 15,000 to $22,000 \mu\text{g kg}^{-1}$ (Ujević et al., 1998). BCs reported by UNEP/MED, 2021 for Cd and Pb are lower than those calculated from this review. Our review proposes BCs for additional TMs for which BCs were not yet consolidated in the Mediterranean. Assessment criteria and BCs for additional TMs have been proposed by OSPAR convention (Ospar Commission, 2009) for the Northeast Atlantic with which BCs proposed for the Eastern Mediterranean in this review are in line, always with lower BCMin. Lastly, this review also proposes BCs for Co, V and Cu, which are included in the list of marine specific contaminants identified as potentially entering the marine environment from sea-based sources (Tornero and Hanke, 2016).

4.1.2. Trace metal concentrations in offshore sediments

The ranges of metal concentrations reported for the surface sediments of the Eastern Mediterranean were derived from the critical review and are summarized in Table 2 and Supplementary material S6. The sediment samples considered were collected at depths ranging from 100 to 3375 m, mostly at least 5 nautical miles from the coast.

The comparison between the lowest concentrations determined in offshore sediments (OffshoreMin, Table 2 and Supplementary material S6) and the BCMin derived from the cores (Table 1 and Supplementary material S4) showed that concentrations were very similar in the case of Cd, Ni, Zn and V. Regarding Pb, As, Cr and Co the OffshoreMin were 0.3–0.7 folds lower than the BCMin. Conversely, Hg and Cu OffshoreMin were 1.5 and 2.6 times higher than the BCMin. These discrepancies

Table 2

Ranges of TM concentrations in offshore sediments of the Eastern Mediterranean Sea ($\mu\text{g kg}^{-1}$ dry weight). For full reference and value overview see Supplementary material S6. References: Angelidis et al., 2011; Buccolieri et al., 2006; Dolenc et al., 1998; Ergin et al., 1996; Ilijanić et al., 2014; Karageorgis et al., 2005a; Lopes-Rocha et al., 2017b; Ogrinc et al., 2007; Oliveri et al., 2013. Stations with bottom depth < 150 m were excluded.

Element	Units	Offshore sediments concentration ranges	
		OffshoreMin	OffshoreMax
Cd	$\mu\text{g kg}^{-1}$ dw	120	400
Hg	$\mu\text{g kg}^{-1}$ dw	32	419
Pb	$\mu\text{g kg}^{-1}$ dw	5,000	72,500
As	$\mu\text{g kg}^{-1}$ dw	1,000	36,500
Cr	$\mu\text{g kg}^{-1}$ dw	24,000	312,000
Cu	$\mu\text{g kg}^{-1}$ dw	6,000	60,000
Ni	$\mu\text{g kg}^{-1}$ dw	23,000	407,000
Zn	$\mu\text{g kg}^{-1}$ dw	29,880	124,000
Co	$\mu\text{g kg}^{-1}$ dw	5,000	31,000
V	$\mu\text{g kg}^{-1}$ dw	36,630	142,000

could arise from physico-chemical characteristics of the sediments (e.g. grain size), however, the considered literature did not provide sufficient information on sample granulometry and on ancillary parameters normally used in the normalization procedures to ascertain this aspect. Higher Hg concentrations have been reported in offshore sediments of the central Adriatic, an area affected by anthropogenic as well as mineralogical inputs of this element (Živković et al., 2017).

4.1.3. Trace metals concentrations in coastal sediments

The ranges of metal concentrations reported since 1995 for contaminated surface coastal sediments in the Eastern Mediterranean

Table 3

Ranges of TM concentrations in coastal sediments of the Eastern Mediterranean Sea ($\mu\text{g kg}^{-1}$ dry weight). For full reference and value overview see Supplementary material S7. References: Acquavita et al., 2010; Aloupi and Angelidis, 2001; Angelidis and Aloupi, 1995; Droghini et al., 2019; Ergin et al., 1996; Karageorgis et al., 2005b, 2020; Khaled et al., 2021; Koukounari et al., 2020; Kucuksezgin et al., 2006; Lopes-Rocha et al., 2017b; Soliman, 2015.

Element	Unit	Coastal sediments concentration ranges	
		CoastMin	CoastMax
Cd	$\mu\text{g kg}^{-1}$ dw	20	10,500
Hg	$\mu\text{g kg}^{-1}$ dw	10	14,920
Pb	$\mu\text{g kg}^{-1}$ dw	5,000	373,900
As	$\mu\text{g kg}^{-1}$ dw	3,100	178,500
Cr	$\mu\text{g kg}^{-1}$ dw	14,800	694,000
Cu	$\mu\text{g kg}^{-1}$ dw	2,900	365,300
Ni	$\mu\text{g kg}^{-1}$ dw	9,100	808,000
Zn	$\mu\text{g kg}^{-1}$ dw	11,600	982,200
Co	$\mu\text{g kg}^{-1}$ dw	1,100	99,000
V	$\mu\text{g kg}^{-1}$ dw	4,600	157,300

Table 4

Ranges of TMs concentrations in sediments in the Eastern Mediterranean (RMin - RMax, EMed) derived from this study; ranges for coastal sediments in the Western Mediterranean (Min - Max, WMed) derived from [Ministere de l'Ecologie et du developpement durable, 1998](#); proposed limits (R' Min - R' Max) for data QC ($\mu\text{g kg}^{-1}$ dry weight).

Element	Units	EMed		WMed		Limits for the QC			
		RMin	RMax	Min	Max	R' Min	R' Max		
Cd	$\mu\text{g kg}^{-1}$ dw	CoastMin	20	10,500	CoastMax	<100	3,600	18	11,550
Hg	$\mu\text{g kg}^{-1}$ dw	CoastMin	10	14,920	CoastMax	10	4,900	9	16,412
Pb	$\mu\text{g kg}^{-1}$ dw	CoastMin	5,000	373,900	CoastMax	9,500	321,000	4,500	411,290
As	$\mu\text{g kg}^{-1}$ dw	OffshoreMin	1,000	178,500	CoastMax			900	196,350
Cr	$\mu\text{g kg}^{-1}$ dw	CoastMin	14,800	694,000	CoastMax			13,320	763,400
Cu	$\mu\text{g kg}^{-1}$ dw	OffshoreMin	2,900	365,300	CoastMax	2,500	208,000	2,610	401,830
Ni	$\mu\text{g kg}^{-1}$ dw	CoastMin	9,100	808,000	CoastMax			8,190	888,800
Zn	$\mu\text{g kg}^{-1}$ dw	CoastMin	11,600	982,200	CoastMax	21,000	741,000	10,440	1,080,420
Co	$\mu\text{g kg}^{-1}$ dw	CoastMin	1,100	99,000	CoastMax			990	108,900
V	$\mu\text{g kg}^{-1}$ dw	CoastMin	4,600	157,300	CoastMax			4,140	173,030

are summarized in [Table 3](#) and Supplementary material S6. In order to improve the data comparability across sites, the literature considered here was selected according to the analytical methods applied to sediment pre-treatment procedures and to the availability of data for the target TMs. Coastal sediments are characterized by a wide range of variability, being closer to possible land-based pollution sources and characterized by more heterogeneous sediment-size composition (silt+clay ranging between 0.4 and 99.9 %, [Table 3](#) and Supplementary material S6). The lowest concentration in coastal sediments (CoastMin) was, in most cases, lower than the BCMin and OffshoreMin, as coastal sediments are characterized by prevailing sandy fraction (silt+clay <5 %, [Table 3](#) and Supplementary material S6). The highest concentrations in coastal sediments (CoastMax) exceeded the highest ranges identified in offshore sediments (OffshoreMax) and were, thus, selected as the upper range to be used for data QC ([Table 3](#) and Supplementary material S7).

4.1.4. Selection of the regional ranges of TM concentrations for data QC

The minimum among BCMin ([Table 1](#) and Supplementary material S4), OffshoreMin ([Table 2](#) and Supplementary material S6) and CoastMin ([Table 3](#) and Supplementary material S7) was selected as the lowest limit of the range proposed for data QC procedures ([Table 4](#)). Accordingly, the maximum among BCMax ([Table 1](#) and Supplementary material S4), OffshoreMax ([Table 2](#) and Supplementary material S6) and CoastMax ([Table 3](#) and Supplementary material S7) was taken as the upper limit for data QC ([Table 4](#)). In the proposed limits possible sampling and analytical errors (10 %; [Birch et al., 2001](#)) as detailed in [Section 4.3](#), have been taken into account. [Table 4](#) summarizes the regional ranges of TMs concentrations in sediments in the Eastern Mediterranean (EMed RMin-RMax) derived from this study, together with the ranges determined in the framework of an extensive monitoring of sediments in the NW Mediterranean French coast ([Ministere de l'Ecologie et du developpement durable, 1998](#)) (WMed Min-Max) and the proposed limits to be used for the data QC in this study (R' Min-R' Max). The report of the NW French coast was selected as it provides an extensive and harmonized data collection for a large number of pollutants.

The comparison of the ranges in the Eastern and Western Mediterranean basins showed that the minimum concentrations of Cd, Hg, Pb, Cu and Zn were quite comparable between the two basins; conversely, the upper values of all elements were higher in the EMed, with RMax of Cd and Hg three times higher than the Max in the WMed ([Ministere de l'Ecologie et du developpement durable, 1998](#)).

The limits proposed here for data QC were not derived from purely sandy sediments, as most research and monitoring were carried out in sediments containing pelitic fractions, where contaminants accumulate the most. A few works which considered sandy sediments were carried out along the Egypt coast ([Okbah et al., 2014](#)), where, with respect to the proposed ranges, lower concentrations were measured for Cu, Ni, Zn,

Co, Cr (460, 1,650, 2,050, 430, 4,080 $\mu\text{g kg}^{-1}$, respectively).

On the other hand, higher concentrations were reached in pollution hot spots, heavily impacted by human activities. As an example, in heavily contaminated harbor areas, such as the Alexandria harbor, Egypt, concentrations of 1,070,000 $\mu\text{g kg}^{-1}$ of Pb, 378,000 $\mu\text{g kg}^{-1}$ of Cu, and 1,221,000 $\mu\text{g kg}^{-1}$ of Zn were found (Mostafa et al., 2004), while even higher concentrations (As: 165,000 $\mu\text{g kg}^{-1}$; Cd: 82,000 $\mu\text{g kg}^{-1}$; Cu: 18,388,000 $\mu\text{g kg}^{-1}$; Hg: 788,300 $\mu\text{g kg}^{-1}$; Pb: 5,393,000 $\mu\text{g kg}^{-1}$; Zn: 22,027,000 $\mu\text{g kg}^{-1}$) were reported in the Italian SIN - Sito di Interesse Nazionale - Site of National Concern (Priolo, Gela, Taranto and Crotona; Ausili et al., 2020). In specific hot spots contaminant concentrations may be >50 times higher than the proposed upper limit; thus, it is useful to recognize high values that should be examined in depth in order to verify potential presence of pollution sources or, conversely, of data outliers.

4.2. PAHs concentration

4.2.1. Background PAHs concentration

The range of BC spans from below limit of detection (LOD) to concentrations up to 30 $\mu\text{g kg}^{-1}$ in the case of phenanthrene reported in the Northern Adriatic Sea (Combi et al., 2020) (Table 5 and Supplementary material S7). Lower BCs are usually found in the South Adriatic and in the Levantine basin, farther from the coasts, densely inhabited for a long time, in contrast to Northern Adriatic characterized by higher values. It is remarkable that there are measurable concentrations for all PAHs compounds (i.e. values above detection limit) also in deep basins of the open Levantine Sea. This is likely due to the natural, petrogenic origins of PAHs and to forest fire combustion (Parinos et al., 2013).

With regards to the lowest limit, BC ranges derived from cores collected in the EMed are quite comparable with those reported by UNEP/MED, 2021 for most compounds. However, BC minima identified in this review for fluoranthene, pyrene, benzo[a]anthracene, benzo[a]pyrene and phenanthrene are from 4 to 10 times lower than those reported by UNEP/MED, 2021. BC indicated by UNEP/MED, 2021 were here presented as a comparison, however, they are not used to propose regional BC ranges as precise information on analytical protocols used in

Table 5

Background concentrations (BC, $\mu\text{g kg}^{-1}$ dry weight) of PAHs in sediments of the Eastern Mediterranean Sea. For full reference and value overview see Supplementary material S8. References: Azoury et al., 2013; Caricchia et al., 1993; Combi et al., 2020; Hatzianestis et al., 2004; Heath et al., 2006; Notar et al., 2001. UNEP/MED, 2021 (used as comparison, not to propose BC ranges). LOD: limit of detection.

Compound	Unit	BC ranges	
		BCMin	BCMax
Naphthalene	$\mu\text{g kg}^{-1}$ dw	4.17	17.88
Acenaphthene	$\mu\text{g kg}^{-1}$ dw	0.20	1.00
Acenaphthylene	$\mu\text{g kg}^{-1}$ dw	1.10	1.63
Fluorene	$\mu\text{g kg}^{-1}$ dw	LOD (0.6)	4.33
Phenanthrene	$\mu\text{g kg}^{-1}$ dw	1.04	30.13
Anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.24)	2.40
Fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.39	23.16
Pyrene	$\mu\text{g kg}^{-1}$ dw	0.56	18.76
Benzo[a]anthracene	$\mu\text{g kg}^{-1}$ dw	0.21	10.63
Chrysene	$\mu\text{g kg}^{-1}$ dw	0.75	19.04
Benzo[a]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.3)	10.10
Benzo[ghi]perylene	$\mu\text{g kg}^{-1}$ dw	LOD (0.5)	19.04
Indeno[1,2,3-cd]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.2)	12.70
Dibenzo[ah]anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.1)	3.60
Perylene	$\mu\text{g kg}^{-1}$ dw	0.60	7.69
Benzo[b]fluoranthene	$\mu\text{g kg}^{-1}$ dw	1.10	25.21
Dibenzothiophene	$\mu\text{g kg}^{-1}$ dw	0.04	
Benzo[k]fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.36	10.24
TPAH	$\mu\text{g kg}^{-1}$ dw	14.92	193.19

the several references is not available in the documents. Finally, this review permitted also to propose BC for dibenzothiophene and perylene which are not yet targeted in the Mediterranean Sea by the Barcelona Convention but are pollutants from sea-based sources, included in the List of Substances of Possible Concern by the OSPAR Convention (Tornerio and Hanke, 2016).

4.2.2. PAHs concentration in offshore sediments

Data from offshore stations were selected in order to evaluate contaminant levels found in surface sediments far from direct land-based pollution sources. However, with the increasing number of offshore gas and oil extraction platforms also into the deep sea, the direct impact of pollution in deep offshore sediments is becoming more relevant. As an example, the area shallower than 1490 m in the Levantine basin was not considered, as data showed indications of contamination by oil/gas extraction or dumping (Astrahan et al., 2017). The maxima of total PAHs concentrations in that area can reach 159–190 $\mu\text{g kg}^{-1}$ at depths between 452 and 698 m, i.e. almost 5 fold higher than in the deepest sediments.

The maximum concentrations found in offshore sediments were close to BCMax for indeno[1,2,3-cd]pyrene while they are 1.2 to 6.2 fold lower than BCMax for most of the other compounds. Only in the case of benzo[a]anthracene and perylene, OffshoreMax concentrations were 2.3–4.5 fold higher than BCMax.

4.2.3. PAHs concentrations in coastal sediments

Close to the coast, the PAHs concentration range was wider than in surface offshore sediments and in sediment cores. The lowest limits presumably correspond with detection limits which were, however, not always specified for the single compound, in the references used. The highest concentrations corresponded to heavily polluted areas such as the Taranto harbor (Cardellicchio et al., 2007), Sibenik Bay (Mandić et al., 2018), the Saronikos Gulf, the Corinth Gulf (Botsou and Hatzianestis, 2012) and the Egyptian coast (Nemr et al., 2007). However, beside industrial areas and big harbors, touristic ports can also represent hot spots for PAHs pollution in sediments (Mali et al., 2017) (Supplementary material S10).

As in the case of trace metals, higher values of PAHs, with respect to the proposed regional range, can be found in particularly polluted areas such as the Italian SIN of Priolo, Gela, Taranto and Crotona where PAHs concentrations can reach 2,394 mg kg^{-1} (Ausili et al., 2020), that is three orders of magnitude higher than the upper limit of the proposed range.

4.2.4. Selection of regional PAHs ranges for data QC

According to literature review and analysis of data available for different environments, the lowest concentration range was selected as the minimum value among the BCMin, OffshoreMin and CoastMin (Tables 5, 6 and 7). Conversely, the maximum concentrations were based on the highest concentrations reported for BC, coastal and offshore sediments of the Eastern Mediterranean (Table 8).

The proposed range limits were also compared with the ranges derived from data of the extensive monitoring carried out along the French Mediterranean coast (Ministere de l'Ecologie et du developpement durable, 1998). Information was available only for a limited number of PAHs substances in the two basins. PAHs concentration ranges were quite comparable between the two basins for e.g. benzo[a]pyrene, indeno[1,2,3-cd]pyrene while differences were evident in the case of other hydrocarbons (fluoranthene, benzo[ghi]perylene, benzo[b]fluoranthene).

Table 6

Range concentrations of PAHs in offshore sediments of the Eastern Mediterranean Sea ($\mu\text{g kg}^{-1}$ dry weight). For full reference and value overview see Supplementary material S9. References: Astrahan et al., 2017; Gogou et al., 2000; Mandalakis et al., 2014; Mandić et al., 2018; Parinos et al., 2013. UNEP/MED, 2021 not used to propose BC ranges, only for reference. Excluded shallower stations near dumping and gas drilling sites. LOD: limit of detection.

Compound	Unit	Concentration range offshore sediments	
		OffshoreMin	OffshoreMax
Naphthalene	$\mu\text{g kg}^{-1}$ dw	0.22	2.80
Acenaphthene	$\mu\text{g kg}^{-1}$ dw	LOD (0.005)	0.25
Acenaphthylene	$\mu\text{g kg}^{-1}$ dw	LOD (0.03)	0.38
Fluorene	$\mu\text{g kg}^{-1}$ dw	LOD (0.06)	0.70
Phenanthrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.62)	10.72
Anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.04)	0.90
Fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.45	17.83
Pyrene	$\mu\text{g kg}^{-1}$ dw	0.30	9.71
Benzo[a]anthracene	$\mu\text{g kg}^{-1}$ dw	0.08	47.39
Chrysene	$\mu\text{g kg}^{-1}$ dw	LOD (0.25)	11.37
Benzo[a]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.10)	8.76
Benzo[ghi]perylene	$\mu\text{g kg}^{-1}$ dw	LOD (0.51)	10.79
Indeno[1,2,3-cd]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.75)	12.82
Dibenzo[ah]anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.10)	2.88
Perylene	$\mu\text{g kg}^{-1}$ dw	0.19	18.35
Benzo[b]fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.61	14.46
Dibenzothiophene	$\mu\text{g kg}^{-1}$ dw	0.01	0.71
Benzo[k]fluoranthene	$\mu\text{g kg}^{-1}$ dw	LOD (0.20)	12.62
TPAH	$\mu\text{g kg}^{-1}$ dw	5.59	164.91

Table 7

Range of concentrations of PAHs in coastal sediments of the eastern Mediterranean Sea ($\mu\text{g kg}^{-1}$ dry weight). For full reference and value overview see Supplementary material S10. References: Bihari et al., 2006; Botsou and Hatzianestis, 2012; Cardellicchio et al., 2007; Caricchia et al., 1993; Magi et al., 2002; Mali et al., 2017; Merhaby et al., 2015; Nemr et al., 2007; Notar et al., 2001; Tzapakis et al., 2010; Zaghden et al., 2017. LOD: limit of detection.

Compound	Unit	Range of concentrations	
		CoastMin	CoastMax
Naphthalene	$\mu\text{g kg}^{-1}$ dw	LOD (0.04)	1,170
Acenaphthene	$\mu\text{g kg}^{-1}$ dw	LOD (0.01)	462
Acenaphthylene	$\mu\text{g kg}^{-1}$ dw	LOD (0.01)	285
Fluorene	$\mu\text{g kg}^{-1}$ dw	LOD (0.01)	531
Phenanthrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.02)	2,350
Anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.01)	765
Fluoranthene	$\mu\text{g kg}^{-1}$ dw	LOD (0.03)	3,551
Pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.03)	2,976
Benzo[a]anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.04)	1,888
Chrysene	$\mu\text{g kg}^{-1}$ dw	LOD (0.04)	2,327
Benzo[a]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (0.05)	2,756
Benzo[ghi]perylene	$\mu\text{g kg}^{-1}$ dw	LOD (0.08)	1,924
Indeno[1,2,3-cd]pyrene	$\mu\text{g kg}^{-1}$ dw	LOD (<0.1)	1,944
Dibenzo[ah]anthracene	$\mu\text{g kg}^{-1}$ dw	LOD (0.06)	1,379
Perylene	$\mu\text{g kg}^{-1}$ dw	1.70	678
Benzo[b]fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.05	3,807
Dibenzothiophene	$\mu\text{g kg}^{-1}$ dw	0.70	93.40
Benzo[k]fluoranthene	$\mu\text{g kg}^{-1}$ dw	0.05	1,707
TPAH	$\mu\text{g kg}^{-1}$ dw	2.10	26,633

4.3. Data QC on “real data”

The proposed limits for data QC (R'Min – R'Max) of selected chemical contaminants (Table 4 and Table 8) were used to compare monitoring and research data collected in the Adriatic – Ionian region (extracted from Mediterranean Sea - Contaminants aggregated datasets (EMODnet Chemistry, 2018) in order to identify potential inconsistencies and to obtain a validated and harmonized regional dataset.

With regard to the wide diversity in monitored substances, variety of sampling and analytical protocols, the procedure for data QC was tested

on selected trace metals (Pb, Cd, Cu and Hg) and PAHs (anthracene and benzo[a]pyrene) which are monitored in sediments by most countries in the Adriatic and Ionian Seas (Berto et al., 2020; Molina Jack et al., 2020). According to SeaDataNet standards adopted by EMODnet Chemistry for marine data management, measured parameters were coded with standard controlled vocabularies (P01 vocabulary) which describe the chemical substance analyzed (e.g. Pb) but also include information on matrix characteristics and methodological aspects (Supplementary material S4). Consequently, the same substance can be coded by different P01 terms due to heterogeneous sampling and analytical protocols adopted by the different laboratories (Giorgetti et al., 2020). In this study, all data related to Pb, Cd, Cu, Hg, anthracene and benzo[a]pyrene were compared with the proposed concentration ranges, regardless of differences in sediment grain-size class or measurement protocols. The list of P01 terms related to Pb, Cd, Cu, Hg, anthracene and benzo[a]pyrene available in the Adriatic – Ionian dataset is provided in Table S11 (Supplementary material S10).

4.3.1. Trace metals: examples of Pb, Hg, Cu and Cd

All P01 related to the same TM were analyzed together and descriptive statistics along with the number of data lower than R'Min or higher than R'Max are presented in Table 9. In the case of Pb, the comparison showed that, in some cases, in situ values exceeded the ranges proposed in this study (Table 4), with the lowest concentration ($232 \mu\text{g kg}^{-1}$) being one order of magnitude lower than the R'Min ($4,500 \mu\text{g kg}^{-1}$) and the highest ($646,000 \mu\text{g kg}^{-1}$) 1.6 fold the R'Max ($411,290 \mu\text{g kg}^{-1}$). To support data QC, distribution of stations and temporal and spatial variability of Pb concentration are displayed in Fig. 4. Concentrations lower than R'Min (Fig. 4c) are flagged as “below detection limit (QF=6)”, according to standard SeaDataNet qualify flag scheme⁷, and this outcome is subsequently reported to data originators who are ultimately responsible for data revision and validation. Conversely, higher concentrations need further inspection in order to correctly assign quality flags. Comparison with literature (Cibic et al., 2017; Cukrov et al., 2011; Robinson et al., 2017; Sakellariadou, 1987; Stamatis et al., 2002; Zonta et al., 2007) indicated that such high concentrations are compatible with pollution hot spots such as harbors and industrial areas, and analysis of spatial and temporal data distribution (Fig. 4a, b, d) allowed to ascribe the “outliers” to pollution hot spots. The proposed limits can, thus, assist to identify erroneous data, in case of samples collected far from direct pollution sources.

The same approach was used for mercury. Table 9 presents the descriptive statistics for the data displayed in Fig. 5.

In the case of Hg, values below the lowest limit (R'Min) were measured in several coastal areas (Fig. 5c), conversely values higher than R'Max (up to $63,000 \mu\text{g kg}^{-1}$) were measured only on a few occasions (in 2006 in Priolo Site of National Concern, Ionian Sea, and in 2009 in Monfalcone harbor, North Adriatic) (Fig. 5d). These high concentrations may be attributed, respectively, to the oil refining activities in Sicily (Musmeci et al., 2014) and to Hg-reach sediments from the Isonzo river drainage basin, an area of long history of Hg extraction (Covelli, 2001).

As in the case of Pb, the spatial analysis allowed to ascribe apparent data outliers to pollution hot spot areas. This data QC approach enables users to easily identify possible data inconsistencies to be revised and, eventually, corrected by data originators.

4.3.2. PAH: examples of anthracene and benzo[a]pyrene

P01 codes used for anthracene and benzo[a]pyrene concentration in the sediment, available in the dataset are listed in table S11 (Supplementary material S11) and Table 9 presents the descriptive statistics for the data displayed in Figs. 6 and 7.

With regard to anthracene (Fig. 6), only one value was lower than

⁷ <https://vocab.nerc.ac.uk/collection/L20/current/>

Table 8

Ranges of PAHs concentrations in sediments in the Eastern Mediterranean (RMin - RMax, EMed) derived in this study; ranges for coastal sediments of the Western Mediterranean (Min - Max, WMed) derived from [Ministere de l'Ecologie et du developpement durable, 1998](#); proposed limits (R'Min - R'Max) for data Quality Check, which take into account possible sampling + analytical errors (See [Section 4.3](#)).

Compound	Unit		EMed			WMed		Limits for QC	
			RMin	RMax		Min	Max	R'Min	R'Max
Naphthalene	µg kg ⁻¹ dw	CoastMin	0.04	1,170	CoastMax			0.03	1,346
Acenaphthene	µg kg ⁻¹ dw	CoastMin	0.005	462	CoastMax			0.004	532
Acenaphthylene	µg kg ⁻¹ dw	CoastMin	0.01	285	CoastMax			0.01	328
Fluorene	µg kg ⁻¹ dw	CoastMin	0.01	531	CoastMax			0.01	611
Phenanthrene	µg kg ⁻¹ dw	CoastMin	0.02	2,350	CoastMax			0.02	2,702
Anthracene	µg kg ⁻¹ dw	CoastMin	0.01	765	CoastMax			0.01	879
Fluoranthene	µg kg ⁻¹ dw	CoastMin	0.03	3,551	CoastMax	4.5	8,200	0.03	4,083
Pyrene	µg kg ⁻¹ dw	CoastMin	0.03	2,976	CoastMax			0.03	3,422
Benzo[a]anthracene	µg kg ⁻¹ dw	CoastMin	0.04	1,888	CoastMax			0.03	2,171
Chrysene	µg kg ⁻¹ dw	CoastMin	0.04	2,327	CoastMax			0.03	2,676
Benzo[a]pyrene	µg kg ⁻¹ dw	CoastMin	0.05	2,756	CoastMax	<1	2,340	0.04	3,169
Benzo[ghi]perylene	µg kg ⁻¹ dw	CoastMin	0.08	1,924	CoastMax	<1	2,525	0.07	2,212
Indeno[1,2,3-cd]pyrene	µg kg ⁻¹ dw	CoastMin	<0.1	1,944	CoastMax	<1	1,900	0.09	2,235
Dibenzo[ah]anthracene	µg kg ⁻¹ dw	CoastMin	0.06	1,379	CoastMax			0.05	1,585
Perylene	µg kg ⁻¹ dw	OffshoreMin	0.19	678	CoastMax			0.16	779
Benzo[b]fluoranthene	µg kg ⁻¹ dw	CoastMin	0.05	3,807	CoastMax	<1	2,790	0.04	4,378
Dibenzothiophene	µg kg ⁻¹ dw	OffshoreMin	0.01	93.4	CoastMax			0.01	107
Benzo[k]fluoranthene	µg kg ⁻¹ dw	CoastMin	0.05	1,707	CoastMax	<1	1,320	0.09	1,963
TPAH	µg kg ⁻¹ dw	CoastMin	2.1	26,633	CoastMax			1.79	30,628

Table 9

Descriptive statistics for the time-series data set of lead (Pb), mercury (Hg), copper (Cu), cadmium (Cd), anthracene, benzo[a]pyrene (BaP) in sediments and comparison with the limits for the QC. Valid N: number of valid data; Min: lowest concentration in the dataset; Max: highest concentration in the dataset; R'Min and R'Max: ranges for data QC; N < R'Min and N > R'Max: number of data below R'Min and above R'Max, respectively.

Element	Valid N	Mean	Min	Max	R'Min	R'Max	N < R'Min	N > R'Max
Pb [µg kg ⁻¹]	2,755	22,916	232	646,000	4,500	411,290	85	28
Hg [µg kg ⁻¹]	2,562	1,290	1	63,000	9	16,412	28	56
Cu [µg kg ⁻¹]	2,595	26,856	43	2,610,000	2,610	401,830	186	14
Cd [µg kg ⁻¹]	2,789	213	1	29,410	18	11,550	5	8
Anthracene [µg kg ⁻¹]	2,249	15.1	0	1,700	0.01	879	1	2
BaP [µg kg ⁻¹]	2,329	29	0	2,900	0.04	3,169	151	0

R'Min and was flagged as below the limit of detection (QF = 6) ([Fig. 6c](#)), and 2 values were higher than R'Max (880 µg kg⁻¹), the highest being 2 fold higher than the limit ([Fig. 6a](#)) ([Table 9](#)). As in the case of lead ([Fig. 4d](#)), and mercury ([Fig. 5d](#)), concentrations exceeding R'Max were measured in long-recognized pollution “hot spots” such as Priolo (eastern Sicily, Ionian Sea), where very high PAHs concentrations have been reported ([Ausili et al., 2020](#); [Di Leonardo et al., 2014](#); [Romano et al., 2013](#)).

With regard to benzo[a]pyrene concentration, regardless of size fraction, no data exceeded R'Max, conversely around 6 % of all data, collected also in previously documented pollution “hot spots” ([Fig. 4d](#), [5d](#), [6d](#)) fell below R'Min ([Fig. 7c](#)) and were flagged as below limit of detection (QF = 6), also considering that the proposed R'Min for benzo [a]pyrene is lower than LOD and LOQ indicated by all institutions providing data. The presence of values below data QC limits (R'Min) proposed in this study, also in areas of high concentrations of other contaminants, indicates the need to evaluate the quality of data available as well as the suitability of the monitoring (sampling and analytical) protocols adopted. In the case of benzo[a]pyrene data, the comparison with regional ranges (R'Min – R'Max) and with concentrations of other parameters measured in the same stations helped in identifying possibly erroneous data which require a closer evaluation of information on monitoring protocols, laboratory performance and data management procedures. These metadata, which need to be available with the data, are crucial to evaluate data quality and fitness for use.

[Figs. 4, 5, 6 and 7](#) display all data related to Pb, Hg, anthracene and benzo[a]pyrene measured in marine sediments, regardless of sediment grain-size and analytical protocols. Despite this heterogeneity, patterns in data concentrations are generally coherent and the proposed concentration ranges effectively help to apply an overall data QC and to

identify possible inconsistencies. Nevertheless, in order to support harmonized assessment of ecosystem status in cross-border areas and to enable comparability among data collected by different laboratories, detailed metadata such as information on station coordinates and depth, sampled depth layer, matrix characteristics (i.e. whether the sample was sieved), on sampling, sample pre-treatment and analytical protocols are required. The list of terms used (P01 vocabulary, Supplementary material S11) indicates that, in many cases, methodological information is extremely limited, even though the adopted vocabulary is specifically structured to handle detailed information (BODC Parameter Usage Vocabulary terms, P01 vocabulary⁸), and is continuously upgraded, upon request, by data originators in order to include additional information. Additional efforts are, thus, required by organizations managing data to use the most complete P01 vocabulary terms and to provide detailed metadata and, possibly, useful ancillary data (e.g. organic carbon content, water content, sediment granulometry, etc.). Shortage in information may be, however, overcome by direct contact with data originators which can be reached, thanks to the detailed mandatory metadata associated with the datasets managed by EMODnet Chemistry data infrastructure.

5. Conclusions

Data validation according to a common approach is useful to improve access to harmonized and comparable data which are prerequisite for environmental status evaluation and, consequently, for the adoption of suitable measures for environmental management and

⁸ https://www.bodc.ac.uk/resources/vocabularies/parameter_codes/

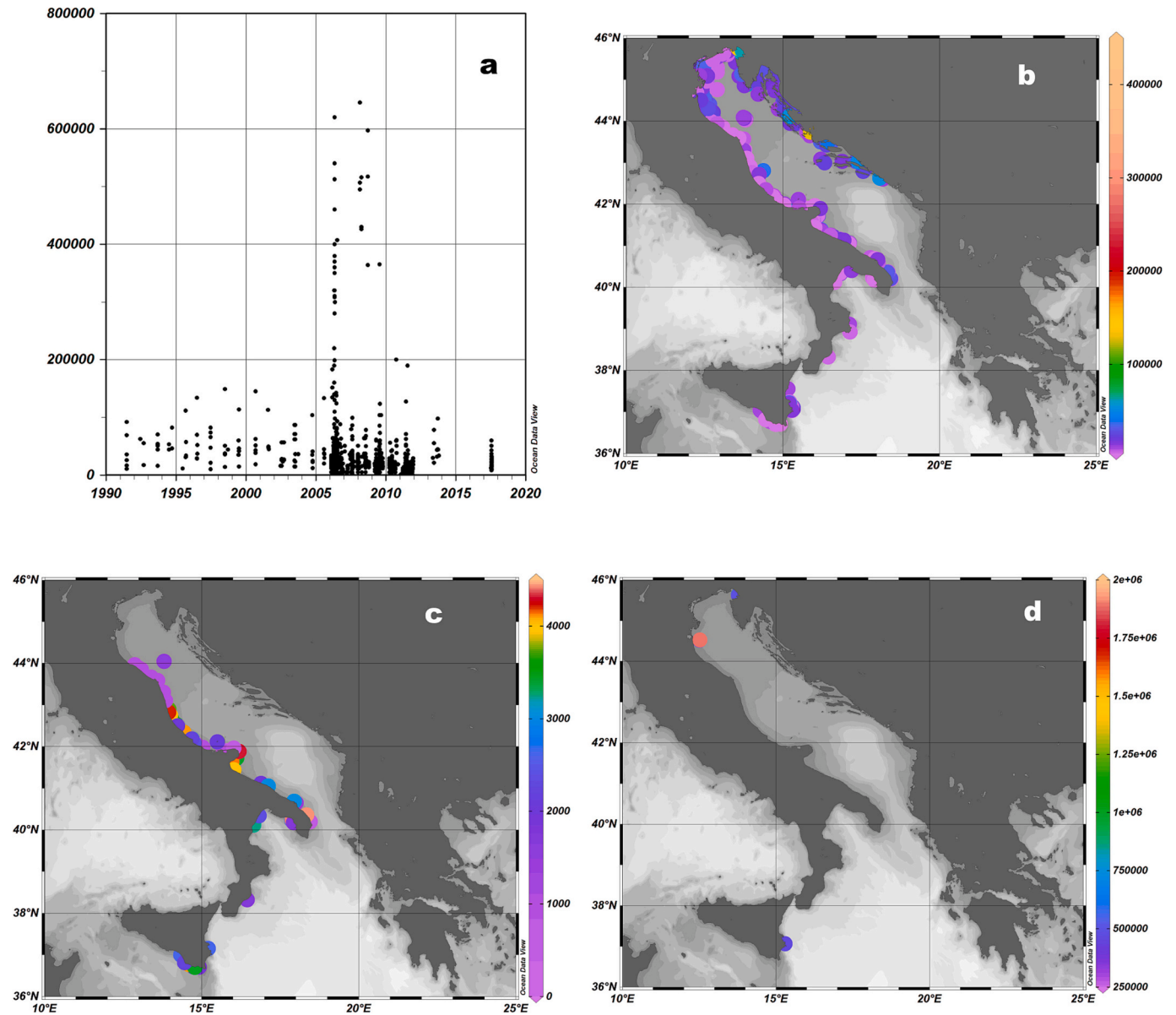


Fig. 4. Lead (Pb) concentrations ($\mu\text{g kg}^{-1}$): data distribution as a function of time (a) spatial distribution in surface sediments (b), data below R'Min (c) and above R'Max (d).

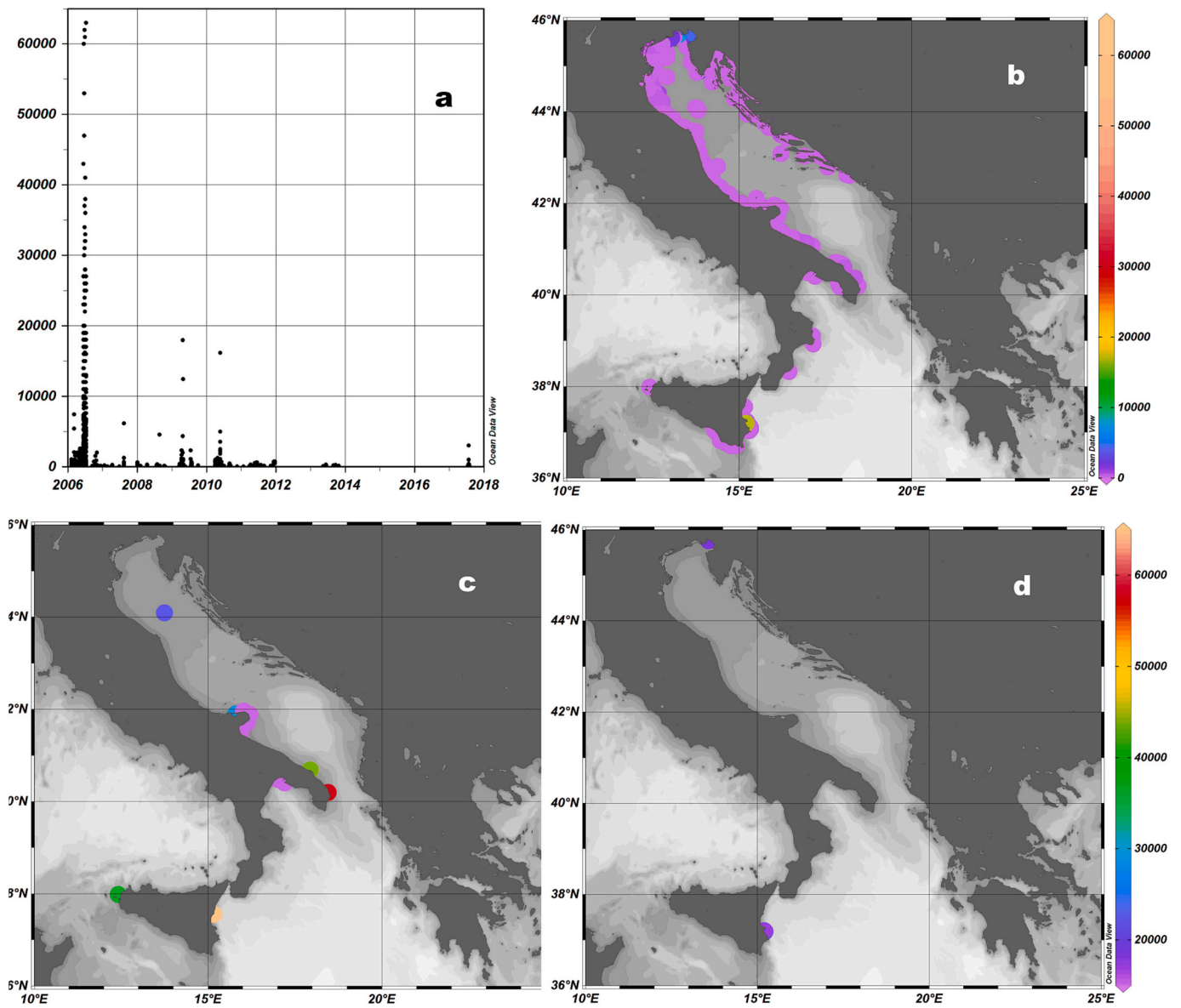


Fig. 5. Mercury (Hg) concentrations ($\mu\text{g kg}^{-1}$): data distribution as a function of time (a), spatial distribution in surface sediments (b), data below R'Min (c) and above R'Max (d).

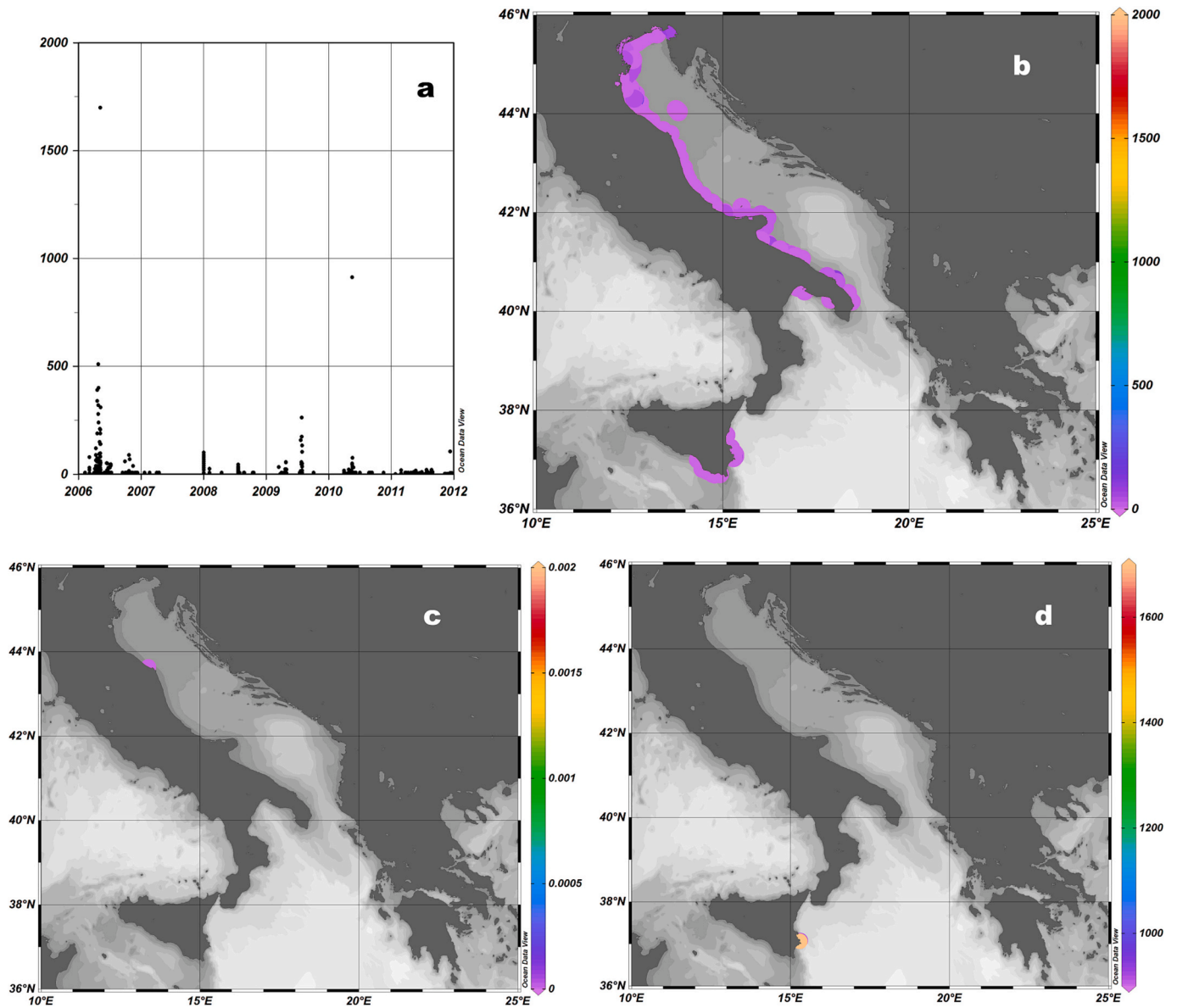


Fig. 6. Anthracene concentrations ($\mu\text{g kg}^{-1}$): data distribution as a function of time (a), spatial distribution in surface sediments (b), data below $R'Min$ (c) and above $R'Max$ (d).

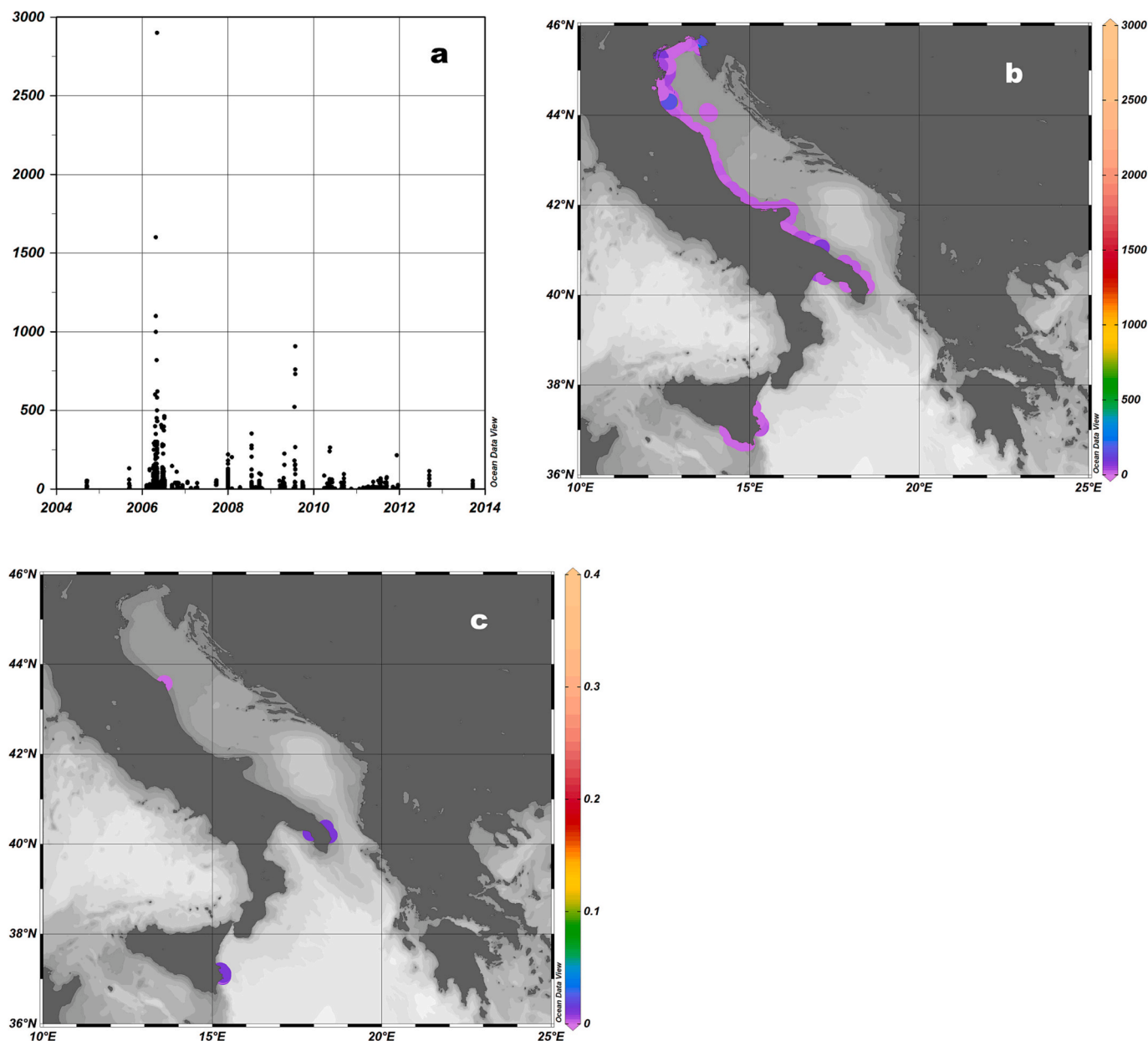


Fig. 7. Benzo[a]pyrene (BaP, $\mu\text{g kg}^{-1}$): data distribution as a function of time (a), spatial distribution in surface sediments (b), data below R*Min (c).

pollution reduction, required for GES achievement. The aim of this study was to propose a methodology that improves the data QC procedures regarding selected TMs and PAHs in marine sediments by identifying regional concentration ranges encountered in the Eastern Mediterranean Sea. An extensive review of the existing literature led to the definition of minimum and maximum contaminant concentrations in the study area. The scientific literature review showed a significant variability in natural background contaminant concentrations. Even though complete information was not always available in the selected references, this study showed that natural background concentrations of TMs are, to some extent, related to granulometry, with finer size fractions related to higher concentrations, and also significantly vary among sea areas.

Testing the proposed regional concentration ranges against EMODnet datasets collected during monitoring programs in the Adriatic and the Ionian Seas proved to be a valid tool for checking the regional data collections. Spatial analysis of contaminant concentrations allowed to

evaluate whether outliers could be ascribed to pollution hot spots or to data that needed further quality check. The analysis of the EMODnet datasets also revealed that, in several cases, relevant information such as matrix characteristics, sample pre-treatment and analytical methods is poorly documented, thus limiting data comparability. Furthermore, the presence of additional information (e.g. sediment grain size, organic carbon and aluminum content, indication of LOD and LOQ for each element or compound analyzed, participation in intercalibration rounds, etc.) can be of great value in the assessment of data quality.

Thus, this study has shown that, above all, data and information management need to be improved to support a harmonized assessment of the state of the environment at regional and subregional levels, e.g., for the entire Mediterranean region. Limited data availability and low information on data quality at the basin level still prevent a coherent and large-scale marine pollution assessment and thus an efficient transnational approach to marine pollution control.

Finally, this review proposes BC for additional substances of

potential concern, not yet defined specifically for the Eastern Mediterranean, which may support a harmonized assessment of pollution status in this region.

Acronyms

AAS	Atomic Absorption Spectroscopy
ASE	Accelerated Solvent Extraction
BC	Background Concentration – the concentration of a compound in the pristine environment
BCMax	highest concentrations in deep core layers
BCMin	lowest concentrations in deep core layers
bdl	below detection limit
CoastMax	highest concentrations in coastal sediments
CoastMin	lowest concentrations in coastal sediments
CVAAS	Cold Vapor Atomic Absorption Spectrometry
FAAS	Flame atomic absorption spectrometry
GC-FID	Gas Chromatography coupled to a Flame Ionization Detector
GC-MS	Gas Chromatography - Mass Spectrometry
GES	Good Environmental Status
GFAAS	Graphite Furnace Atomic Absorption Spectrometry
ICP-AES	Inductively Coupled Plasma Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IMAP	Integrated Monitoring and Assessment Programme of the Mediterranean Sea and Coast
LOD	Limit of Detection
LOQ	Limit of Quantification
MSFD	Marine Strategy Framework Directive
OffshoreMax	highest concentrations in offshore sediments
OffshoreMin	lowest concentrations in offshore sediments
PAH	polycyclic aromatic hydrocarbon
QC	Quality Control
QF	Quality Flag
SFE	Supercritical Fluid Extraction
WFD	Water Framework Directive
XRF	X Ray Fluorescence Spectroscopy

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Declaration of competing interest

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Data availability

Part of the data have been cited in the text with the related doi.

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