



**UNITED  
NATIONS**

**EP**

UNEP (DEPI)/MED WG.444/Inf.10

---



**UNEP**



**UNITED NATIONS  
ENVIRONMENT PROGRAMME  
MEDITERRANEAN ACTION PLAN**

3 August 2017  
English only

---

6<sup>th</sup> Meeting of the Ecosystem Approach Coordination Group

Athens, Greece, 11 September 2017

**Agenda item 4: Review of Quality Status Report (QSR)**

**Temporal Trends and Analysis for Chemical Contaminants from the MED POL Database**

For environmental and economic reasons, this document is printed in a limited number. Delegates are kindly requested to bring their copies to meetings and not to request additional copies.

UNEP/MAP  
Athens, 2017



UNITED  
NATIONS

EP

UNEP(DEPI)/MED WG.439/Inf.10



UNITED NATIONS  
ENVIRONMENT PROGRAMME  
MEDITERRANEAN ACTION PLAN

12 May 2017  
Original: English

Meeting of the MED POL Focal Points

Rome, Italy, 29-31 May 2017

**Agenda item 4: Progress Achieved regarding the Implementation of the Programme of Work 2016-2017 related to Land Based Pollution and Governance Themes**

**Temporal Trends and Analysis for Chemical Contaminants from the MED POL Database**

For environmental and economic reasons, this document is printed in a limited number. Delegates are kindly requested to bring their copies to meetings and not to request additional copies.



UNITED  
NATIONS

EP

UNEP(DEPI)/MED WG.427/Inf.4



UNITED NATIONS  
ENVIRONMENT PROGRAMME  
MEDITERRANEAN ACTION PLAN

5 October 2016  
Original: English

---

Meeting of the Ecosystem Approach Correspondence Group on Pollution Monitoring

Marseille, France, 19-21 October 2016

**Agenda item 4. Marine Pollution Monitoring Regional Data Base and Reporting, MED POL Database Gaps and Related Quality Assurance Issues**

**Temporal Trends and Levels Analysis for Chemical Contaminants from the MED POL Database**

For environmental and economy reasons, this document is printed in a limited number and will not be distributed at the meeting. Delegates are kindly requested to bring their copies to meetings and not to request additional copies.

## **PREAMBLE**

This document presents an update of temporal trends and analysis for hazardous chemical contaminants in the Mediterranean Sea for the most recent data received (up to the end of 2015) and uploaded in the MEDPOL Database. The data analysis and statistical methods in this report build on previous UNEP/MAP work undertaken in 2011<sup>1</sup>. This report has been prepared by Dr. Carlos Guitart (Marine Environment Consultant, Spain) in collaboration with Dr. Juan Miguel Marín (Department of Statistics, University Carlos III of Madrid, Spain) and Dimitris Poursanidis (University of the Aegean, Marine Sciences Department), under the supervision and edition of UNEP/MAP-MEDPOL.

---

<sup>1</sup> Analysis of the trend monitoring activities and data for the MED POL Phase III and IV (1999-2010) UNEP(DEPI)/MED WG.365/Inf.5 and Hazardous substances in the Mediterranean: A spatial and temporal assessment UNEP(DEPI)/MED WG.365/Inf.4.

## INDEX

PREAMBLE .....	
INDEX .....	
1. BACKGROUND.....	1
2. OBJECTIVES AND SCOPE .....	1
3. CONCEPTUAL FRAMEWORK .....	2
3.1. Assessment of levels and temporal trends of chemical contaminants.....	2
3.2. Statistical methods for levels and trends analysis .....	3
3.3. Uncertainty analysis .....	4
4. UPDATE ON CONTAMINANT LEVELS AND TEMPORAL TRENDS .....	5
4.1. Croatia.....	5
4.2. Cyprus .....	9
4.3. Israel.....	13
4.4. Morocco .....	36
4.5. Slovenia.....	38
4.6. Tunisia.....	45
4.7. Turkey .....	53
5. CONCLUSIONS AND RECOMMENDATIONS.....	62
6. REFERENCES.....	62
ANNEX I. Data trend analysis comparison with the PIA statistical software .....	

## **1. BACKGROUND**

The Programme for the Assessment and Control of Marine Pollution in the Mediterranean (MEDPOL) of UNEP/MAP has supported the implementation of a coordinated monitoring programme in the Mediterranean Sea since 1975 (MEDPOL Phases I-IV). The MEDPOL Programme was designed to support Contracting Parties to the Barcelona Convention to qualify and quantify the marine pollution levels, sources and impacts on their marine and coastal environments, and to undertake coordinated measures and implement national and regional actions plans for the control, phase out and elimination of pollution from land based sources and activities.

One of the major components of the pollution monitoring and assessment activities (MEDPOL Phase III and IV) is the temporal trend assessment of contaminants in selected coastal hotspots and environments under specific monitoring requirements and assessment criteria. Thus far, several assessment studies and background documents have evaluated the monitoring datasets of hazardous chemical substances in marine sediments and biota in order to determine the levels and temporal trends that can be considered of concern and identified coastal hotspots for priority action. The evaluations of the MEDPOL database were made in 2003 (UNEP(DEC)/MED WG.243/3), 2005 (UNEP(DEC)/MED WG.273/2), 2009 (UNEP(DEPI)/MED WG.343/3) and 2011 (UNEP(DEPI)/MED WG.365/Inf.5) in view to be presented to MEDPOL review meetings of monitoring activities. In parallel, the spatial and temporal assessments on hazardous chemical substances were also reported (UNEP(DEPI)/MED WG.365/Inf.4).

In this sense, this document reports on temporal trends and levels analysis using the latest datasets up to the end of 2015, submitted by Contracting Parties to the MAP/MEDPOL Database. The assessment in this report is performed ensuring continuity to the statistical framework designed for the MED POL monitoring programme and their implementation phases in the Mediterranean Sea (MEDPOL Phase III and IV). However, in line with the Integrated Monitoring and Assessment Program of the Mediterranean Sea and Coast and Related Assessment Criteria (IMAP) (UNEP(DEPI)/MED WG.421/Inf.9) adopted in the COP19 (UNEP(DEPI)/MED IG.22/7 Decision), we have merged the temporal trend and data analysis evaluations in this report with the assessment criteria developed for the Mediterranean Sea (UNEP(DEPI)/MED WG.427/Inf.3).

## **2. OBJECTIVES AND SCOPE**

The objective of this report is to update on hazardous chemical substances temporal trends in sediment and biota in the Mediterranean Sea using the MEDPOL datasets up to the end of 2015, with the aim to provide information by means of the monitoring and assessment activities on the general progresses made towards the fulfilment of the Barcelona Convention Protocols in the Mediterranean Sea.

Therefore, the scope of this report is to serve as an information document for further revision and preparation of national strategies and monitoring programmes by Contracting Parties in the Mediterranean in line with the Ecological Objective 9 of the current IMAP Programme under the EcAp framework.

### 3. CONCEPTUAL FRAMEWORK

#### 3.1. Assessment of levels and temporal trends of chemical contaminants

The first evaluation of the data collected in the MEDPOL Database was made in 2003 to identify the sampling and analytical variances underlying each national monitoring strategy, as the set up towards a common monitoring program implementation initiated during the period 1998-2001. In 2005 a second evaluation was made mainly to identify the weakest parts of the adopted both monitoring and sampling strategies. In 2009 when the 10 years benchmark was reached a detailed analysis of variances and trends – where possible - for each monitoring site was performed. In 2011, the latest detailed report (UNEP(DEPI)/MED WG.365/Inf. 5), included an innovative statistical software suite (PIA© software) based on the methodology followed by the Arctic Monitoring and Assessment Programme (AMAP) for its use in temporal trend assessment activities. The PIA© software is a computer program for analysing trends in time series datasets by applying a sum of standard statistical methods (Bignert et al., 2006).

Within the MEDPOL Monitoring Program implementation activities, the statistical objective for the programme was agreed by Contracting Parties to detect a minimum linear trend of 10 % per year in 10 years with a statistical power of 90% with a standard confidence level of  $\alpha=0.05$  (Type I error ( $\alpha$ ), set to assume a 5% of probability of committing a false positive, thus to reject the null hypothesis being true,  $H_0 = \text{no trend}$ ). The concept of statistical power was included within the assessment of the monitoring datasets (Power =  $1-\beta$ ; Type II error ( $\beta$ ), the probability to commit a false negative, thus accept the null hypothesis when is false in statistical hypothesis testing), for statistical optimization, as well as calculated with the implemented algorithm in the PIA© software (Nicholson et al., 1995 and 1997). Both statistical “type” errors are desirable to be minimised, but whilst  $\alpha$  is arbitrarily fixed,  $\beta$  cannot be controlled and depends on a number of parameters, such as the datasets variances and the number of data (years) in the time series.

On the whole, the idea behind was to continuously evaluate the correct implementation of the monitoring programs and their optimization by using the statistical power concept ( $P=1-\beta$ ), which directly depends on a strong sampling strategy within the long-term monitoring programs. Nicholson et al. (1997), reported a calculated table for  $|b|/\psi$  (thus, the data variability within the time series ( $\psi$ ) and the expected trend detection ( $|b|$ ) relationship, the signal-to-noise ratio  $|b|/\psi$ ), corresponding to different powers as T varies from 5 to 25 (number of years). With a 90% statistical power and 10 years, the signal-to-noise ratio was calculated to be  $|b|/\psi=0.409$  to assess at least a 10% trend change. Thus, if it is supposed that  $b=0.1$  (10% linear trend), then the acceptable within year monitoring standard deviation to fulfil the statistical objectives would be  $\psi = (0.1/0.409)$ , and the acceptable programme variance  $\psi^2=0.060$ . Even if the underlying trend for a dataset is not always linear, the programme optimization objective is said to be fulfilled if the within year variance is below the threshold of 0.060. Further, this limit is correct if we assume that in general the between year variance is always significantly lower than the within year variance (Nicholson et al., 1997).

However, after almost 20 years of ongoing monitoring activities, the statistical power of the existing long-term monitoring datasets is far to reach the 90% statistical power target agreed for the statistical temporal trend assessments and ranges between 10% and 30%, with a large number of years (15 to 40 years) of monitoring predicted (*ca.* required), as reported in the summary table in the latest trends report (UNEP/DEPI/MED WG.365/Inf.5). A part from the practicalities of the implementation of cost-effective long-term monitoring programs, surely, the complex and fast-changing environment (e.g. abrupt changes) hinders to reach the statistically monitoring proposed criteria (Guitart et al., 2012; Solaun et al., 2013), although statistically significant upward and downward trends ( $\alpha=0.05$ ) with an associated power value ( $P=1-\beta$ ) have been detected so far.

In this report, we have continued to evaluate the time series following this conceptual approach, both for the ongoing time series with added datasets (from 1 to 3 years) and the new datasets and time series for some biota species which now spans for at least five years of data. The within year monitoring variances have been calculated to evaluate the robustness, systematic and reliable monitoring activities. In this report, the trend analysis have been undertaken using the standard statistical package software SSPS 17.0 (SPSS Inc., Chicago, USA), including the statistics described in the following sections. Nevertheless, a statistical assessment with the PIA© software is presented in the Annex in order to compare the outputs of both approximations.

### 3.2. Statistical methods for levels and trends analysis

The work within OSPAR Convention has proposed three trend detection methods all at once with the idea to take the benefits of all methods, because there is not only one statistical method which always offers the best analysis of the environmental monitoring datasets. Each one has its own capabilities and underlying assumptions. The three methods (Mann-Kendall, linear regression and LOWESS smoother) are amongst the most commonly used in this field. The priority is given from the simplest to the more complex method. The non parametric Mann-Kendall is the most robust to outliers, but in case of a linear trend, the linear regression has more statistical power. The smoother algorithms are used to detect non-linear trends. However, in the MEDPOL Database the contaminant probability distributions shown different data distributions and in fact exceptional cases (*ca.* monitored stations) show a normal probability distribution (such as theoretically expected for example in reference areas). Therefore, in this report, we have given more weight to a non-parametric statistical approach (without any log-data transformations) both for basic statistics and temporal trends analysis.

For the level assessments, medians, interquartile ranges (IQR), concentration ranges and lower and upper quartiles (Box-and-Whisker plots) for each chemical have been calculated in order to characterise the populations and to identify the extreme values and outliers (including also the arithmetic mean for reference). The median values are generally considered as a better central tendency statistic estimators in large asymmetric distributions, such as those found at regional/national level in the MEDPOL Database. Nevertheless, the normality of the datasets for the different populations has been evaluated by means of the Kolmogorov-Smirnov test. Since datasets were not normally distributed, the use of non-parametric statistics was confirmed. The potential data outliers, located above the upper limit at a distance higher than 1.5 times the interquartile range (IQR, grouping the 50% of data, between the 25<sup>th</sup> and 75<sup>th</sup> quartile) were not automatically discarded, with the exception of some data judged as a gross errors and clearly inconsistent. Therefore, these high values might influence the overall levels, the variances of the time series and the trends analysis, thus should be taken into account if not confirmed wrong. The Mann-Whitney test was used for comparing two data sets and the Kruskal-Wallis test when more than two groups were considered. In these two non-parametric tests, the differences were considered statistically significant at a standard confidence level of  $\alpha=0.05$ .

The non-parametric alternative test to the linear regression analysis, the Mann-Kendall trend test (Gilbert, 1987; Helsel and Hirsch, 1995) was chosen for temporal trend analysis. This test generally has lower power than the regression analysis. It counts the number of consecutive years where the concentration increases or decreases compared with the year before, but does not take into account the magnitude of the concentrations. If the regression analysis yields a significant result but the Mann-Kendall test does not, one explanation could be the lower power of the Mann-Kendall test. An alternative explanation might be the undue influence of endpoints in parametric regression analysis. Hence, the robust Kendall's 'tau' value ( $\tau$ -Kendall B), 'overlook' these values within a linear time series and the corresponding correlation coefficients with a confidence level of  $\alpha=0.05$  or higher are reported. The Kendall's 'tau' ranges from 0 to 1 (see tables through the sections), like the traditional Pearson's correlation coefficient 'r' (shown also in the plots in this report), but will generally be lower. In general, linear correlations of 0.9



or above correspond to  $\tau$ -values of about 0.7 or greater (Helsland Hirsch, 1995, p. 212). In an evaluation comparing several other trend tests, the Mann-Kendall test was recommended by the USEPA for use in water quality monitoring programmes with annual samples (Loftis et al., 1989). In order to evaluate non-linear trend components the time series have been fitted to diverse parameterisations (namely, linear, quadratic and cubic), according the best correlation coefficient. The latest two, provide some kind of a ‘smoothed’ trend line to identify potential non-linear trends. More than one parameterisation plot has been included to improve interpretations in some cases.

### 3.3. Uncertainty analysis

In this report we have included uncertainty bands in time trend plots. This new feature, a continuous 95% confidence interval for the mean within the parameterisation selected, allows for an additional interpretation of the between and within year variability. The uncertainty bands are fitted either to the individual or the mean value of the regression line (linear, quadratic or cubic), depending on the parameter and the monitoring variability observed. In the plots we have also indicated any significant trend, otherwise the contrary. As an example, the figure below shows time trend plots used in this report. Further, the proposed BACs and EACs have also been included in these plots to further assess the levels and temporal trends.

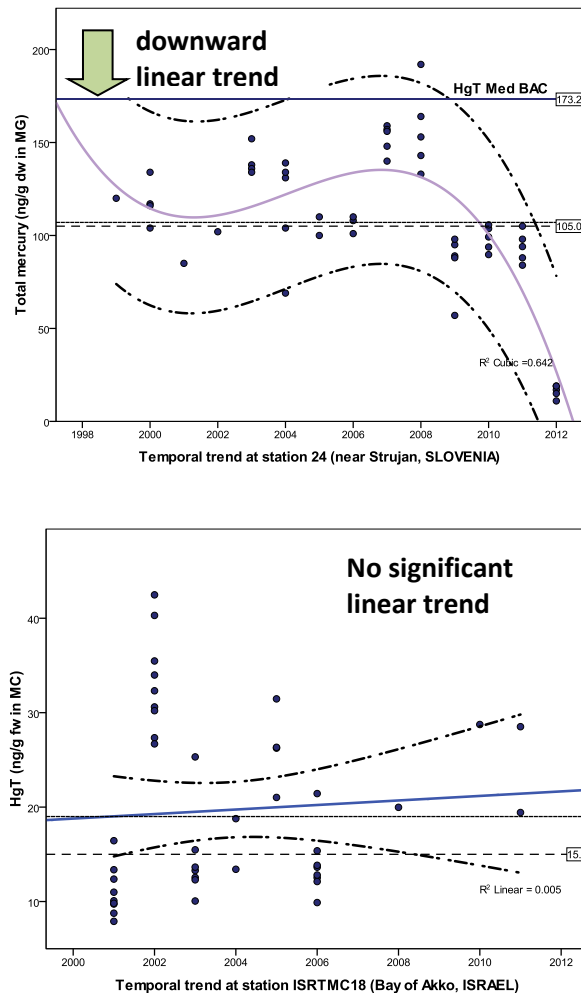


Figure 1. Example of time trend plots (this report) fitted to the best parameterisation (linear, quadratic or cubic), including the uncertainty bands and a written indication of the  $\tau$ -Kendall B test result, as well as the assessment criteria when appropriate.

## 4. UPDATE ON CONTAMINANT LEVELS AND TEMPORAL TRENDS

### 4.1. Croatia

Datasets from 2011 to 2014 were available in the MEDPOL Database for biota and sediment). However, monitoring datasets were only provided for the historical stations for 2011, and thus no temporal trends could be further investigated since the last assessment report (UNEP(DEPI)/MED WG.365/Inf.5).

#### Trace metals in *Mytilus galloprovincialis* (MG)

The biota datasets were for metals and organic compounds in common mussel *Mytilus galloprovincialis* (MG). Data for Cd, HgT and Pb, as well as organic compounds (eg. PCBs, DDT, DDD, DDE, Dieldrin, HCB) were reported although some values as BDL for the latter groups. Determinations were performed in pooled samples with 25-40 individuals. Table Cro.1 presents a summary of the concentrations in historical stations for 2011 for the stations in the areas of Dubrovnik, Martin and Split.

Table Cro.1. Summary for trace metals in MG at the historical stations in Croatia ( $\mu\text{g}/\text{Kg dw}$ ) for 2011.

Station	Cd	Cr	Cu	HgT	Pb	Zn
GR	652	2491	11470	205	3258	156136
SI	494	2331	7425	168	1197	74900
MA	927	2051	927	121	1580	97000
IN	749	1971	8560	2378*	3452	171900
VR	734	2204	20667	319	9655	416300

\*see text

The high value for HgT in the table above is lower than the previous value in 2009 (3879  $\mu\text{g}/\text{Kg dw}$ ) and the the maximum measured level in 2004 (8578  $\mu\text{g}/\text{Kg dw}$ ) at this station. During the period 2012-2014, MG specimens were collected in new coastal stations, including other species (*ca.* AN, CC, OE and VV). The whole soft tissue pooled samples (15 individuals of 6-7 cm each) were used for the determination of Cd, Cr, Cu, HgT, Pb and Zn and organic chemicals. For the MG biota samples, the plots below (Figures Cro.1 - Cro.3) show the levels determined in the new stations reported to the MEDPOL Database. In this period 1 o 2 samples were determined at each location per year.

#### Trace metals in sediments

For the years 2011 and 2013, Croatia submitted datasets for Cd, Cu, HgT, Pb and Zn in sediment samples collected at different coastal stations. However, the references to the sampled layer and sediment fraction information are both missing (and thus these data is considered as unsieved samples). More, as it was reported in the previous assessment (UNEP(DEPI)/MED WG.365/Inf.5), a single sediment sample per location was determined for hazardous chemicals making difficult to study of temporal trends. These sediment samples were collected nearby the MG stations. The following plots (Figures Cro.4 – Cro.6) presents the data for Cd, HgT and Pb determined in these samples. It should be noticed that these sediment stations were classified as hotspot stations, and therefore, the levels are above the Med BAC assessment criteria.

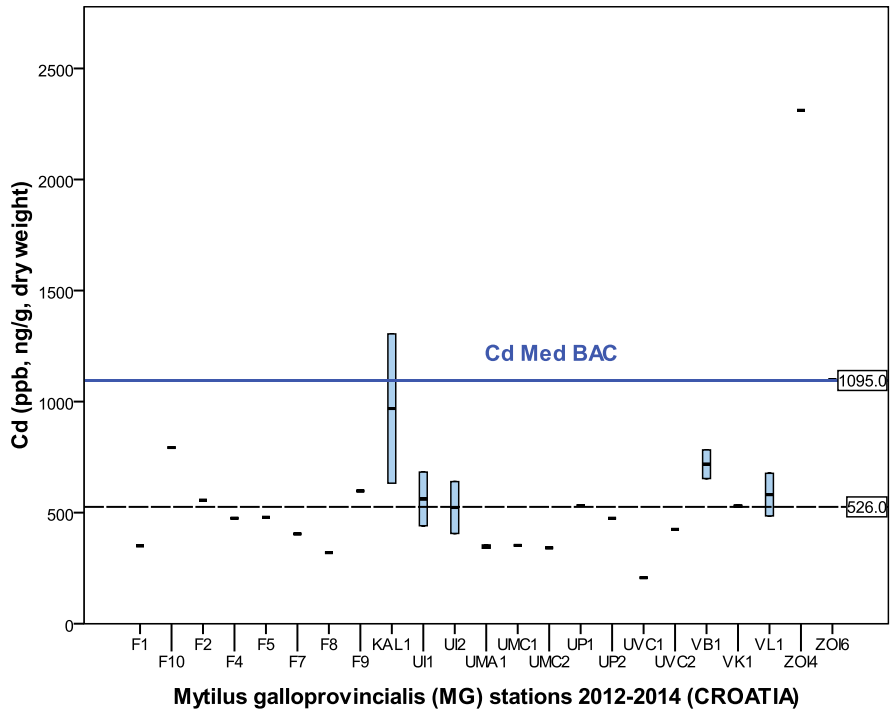


Figure Cro.1. Cadmium box-plots for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the Cd Med BAC is also presented for reference.

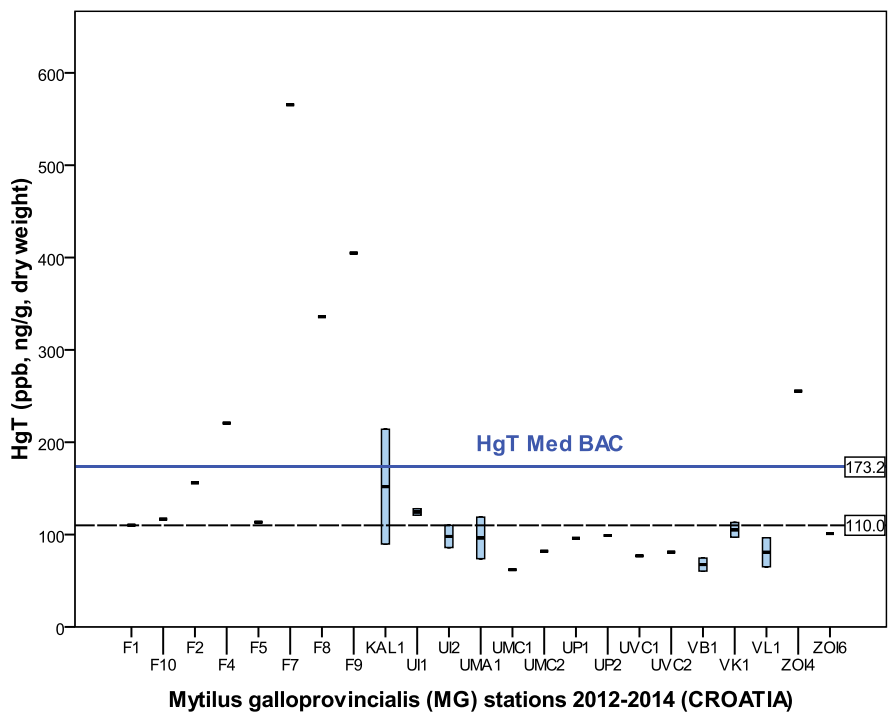


Figure Cro.2. Total mercury box-plots for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the HgT Med BAC is also presented for reference.

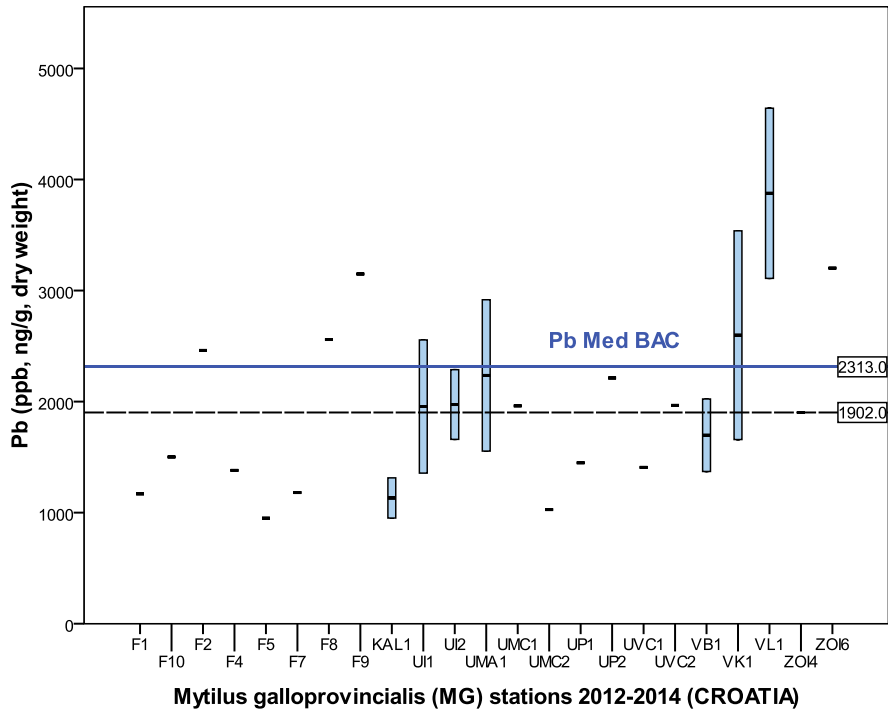


Figure Cro.3. Lead box-plots for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the Pb Med BAC is also presented for reference.

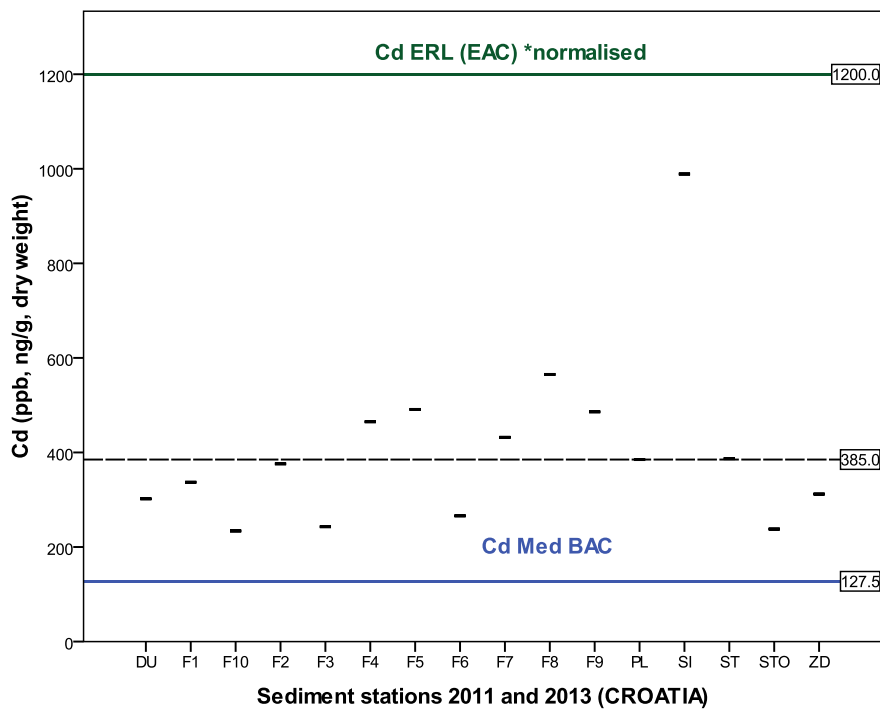


Figure Cro.4. Cadmium data for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the Med BAC and EAC are also presented for reference.

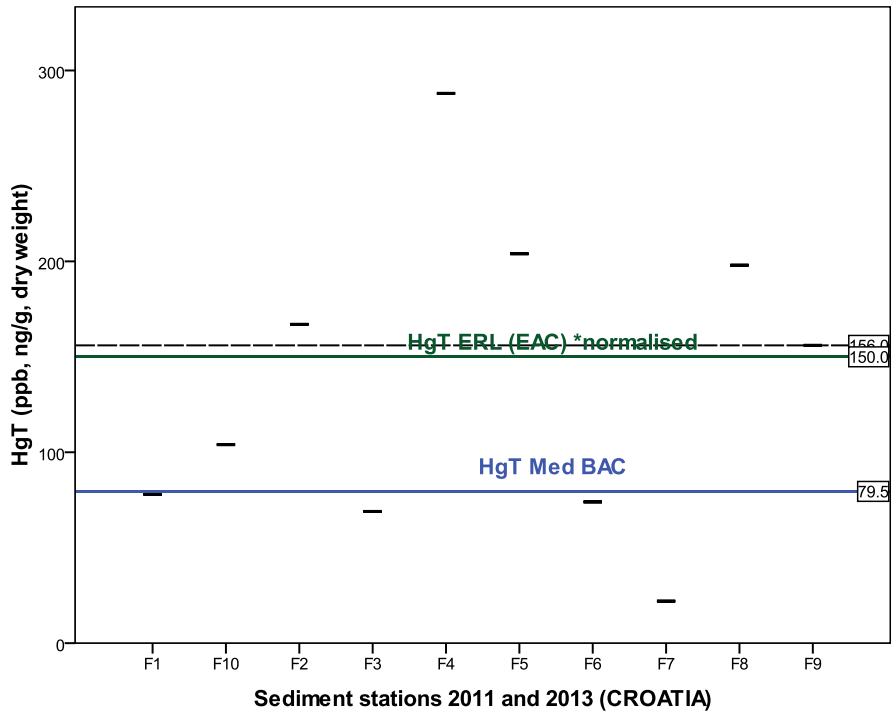


Figure Cro.5. Total mercury data for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the Med BAC and EAC are also presented for reference.

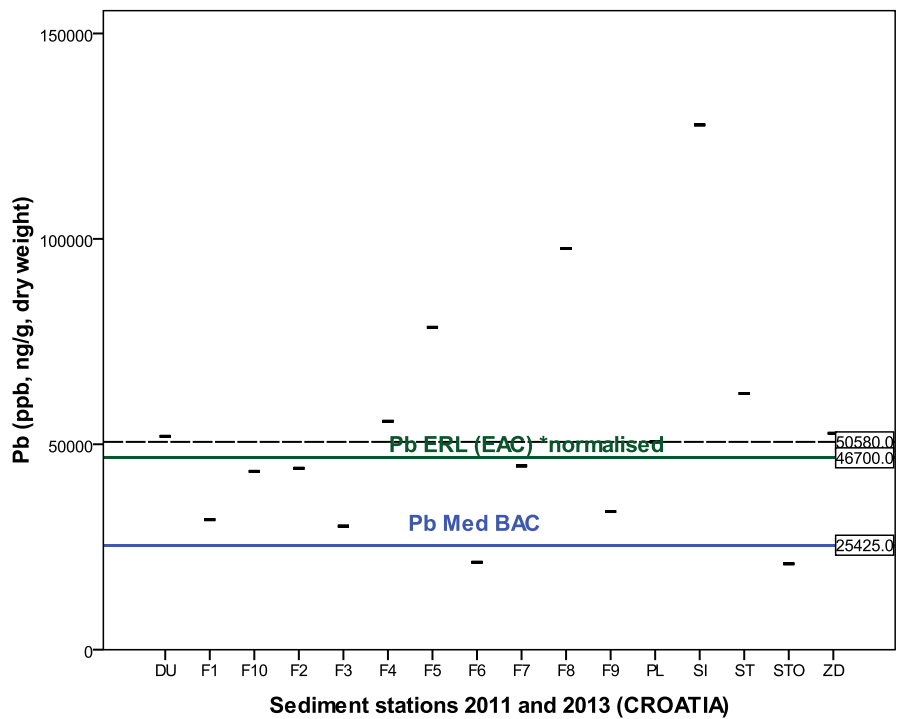


Figure Cro.6. Lead data for 2012-2014 (new stations in the MEDPOL database). The dashed line represents the median and the Med BAC and EAC are also presented for reference.

## 4.2. Cyprus

New datasets were available in the MEDPOL database for the periods 2008-2010 and 2012-2015. The previous submitted and evaluated datasets, contained information for trace metals and organic contaminants in biota (*Mullus barbatus*, MB) (UNEP/(DEPI)/MED WG.365/Inf.5). These new submitted datasets were for samples collected at 3 out of the 5 stations monitored in the south coast of Cyprus, namely Larnaca, Limassol and Paphos.

However, expectations to produce further statistical and temporal trend analyses with an increased number of environmental information were frustrated as the chemical contaminants were only above the analytical detection limits for total mercury (HgT) determinations, thus only this parameter is valuable to perform statistical analysis. However, only the year 2014 reports HgT above the DL for the period 2012-2015. Cadmium (Cd), in the first place, followed by lead (Pb) concentrations are almost all, reported as undetectable concentrations in specimens of MB species for the period 2008-2010 (93% of the values reported as BDLs for Cd and 73% for Pb) and below BDLs for this period, whilst for 2012-2015 are reported as BDLs, except Pb for Limassol and Paphos in 2015 (ranging from 56-180 and 78-120 µg/Kg fw, respectively). For the case of OCs compounds datasets, such as polychlorinated biphenyls (PCBs), pesticides (DDTs, DDDs and DDEs), aldrin, endrin, hexachlorobenzene, these were almost 100% reported as BDLs for all these continuing years, and similarly, no updated statistical analyses could be provided in the present report.

### Total mercury in *Mullus barbatus* (MB)

The fillet tissue was selected as sample tissue. Six samples (each pooled in general using 6 or 8 individuals) were analyzed yearly. The datasets units were converted from dry weight to fresh weight using DW/FW ratio provided in the database (or a 0.2519 averaged for the whole DW/FW ratios reported). Thus, data was transformed to fresh weight for further comparison with other countries, if necessary.

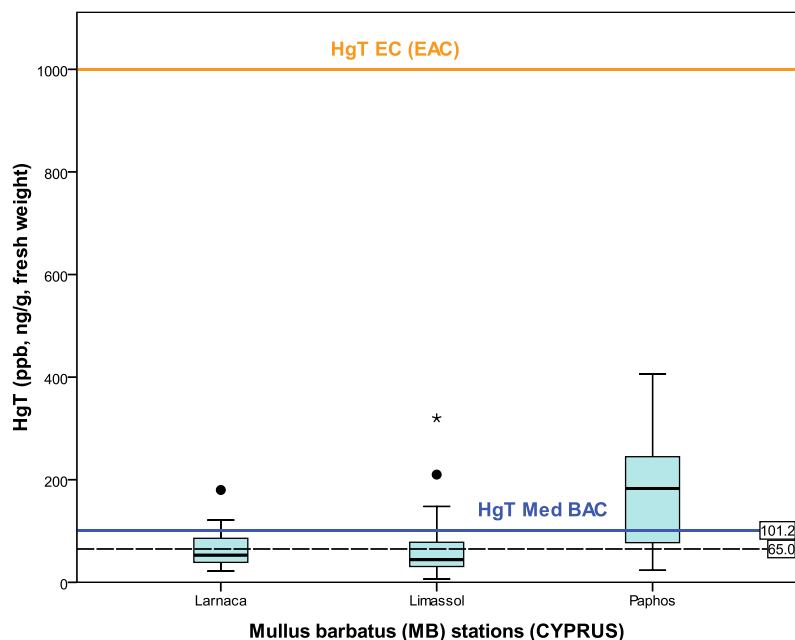


Figure Cyp.1. Total mercury box-plots in Cyprus (all years in the MEDPOL database) at the 3 selected stations with updated datasets for 2008-2015. The dashed line represents the median and the Med BAC and EAC are presented for reference.

As mentioned, HgT was the solely trace metal available to compare levels between stations. The non-parametric statistical Kruskal-Wallis test (independent samples, Chi-square statistic,  $\alpha=0.05$ ) was performed to establish significant differences between levels and stations. Further comparisons were done in pairs using the non-parametric Mann-Whitney test (U statistics,  $\alpha=0.05$ , 2-tailed). The results confirm statistically significant differences between the levels of total mercury between Paphos and the other two stations, Larnaca and Limassol. The Table Cyp.1. below, summarizes the main statistics of the datasets and Figure Cyp.1. includes the threshold Background Assessment Criteria (BAC) and the recommended Environmental Assessment Criteria (EAC) for HgT (UNEP/DEPI WG. 427/Inf.3).

Table Cyp.1. Summary table statistics for HgT (all years database,  $\mu\text{g}/\text{Kg}$  fw).

Station	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Larnaca	27	65	53	51	38	89	22	180
Limassol	27	69	44	50	31	81	6	320
Paphos	22	186	185	167	86	253	24	406

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

When looking closely at the individual stations over the years (Figures Cyp.2 - Cyp.4.) also some features become apparent. The levels of mercury determined at the stations of Larnaca and Limassol indicates these two areas are the lowest impacted by mercury pollution, thus the median values are 53 and 44  $\mu\text{g}/\text{Kg}$  fw, respectively, despite in 2008 and 2014 at both stations the box-plots show a median value around the 101.2  $\mu\text{g}/\text{Kg}$  fw, the Med BAC.

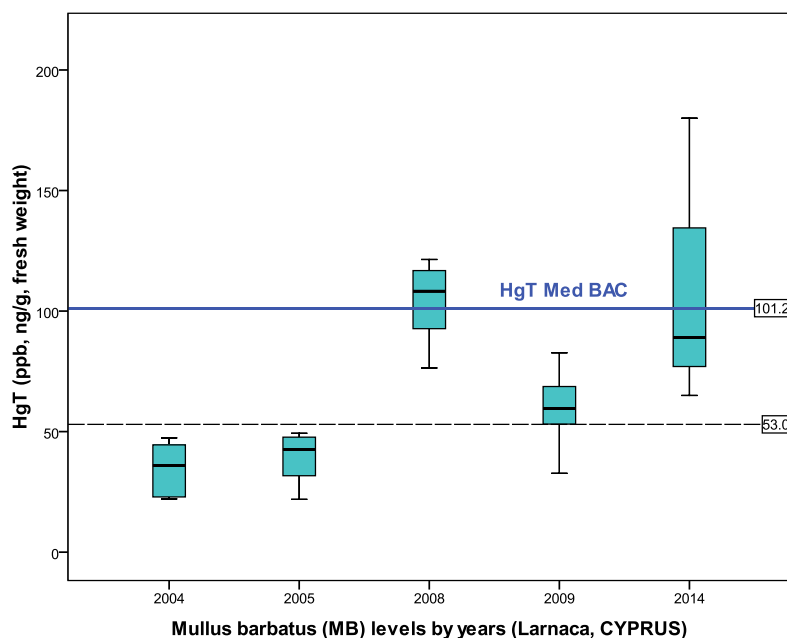


Figure Cyp.2. Total mercury box-plots at the Larnaca station. The dashed line represents the median.

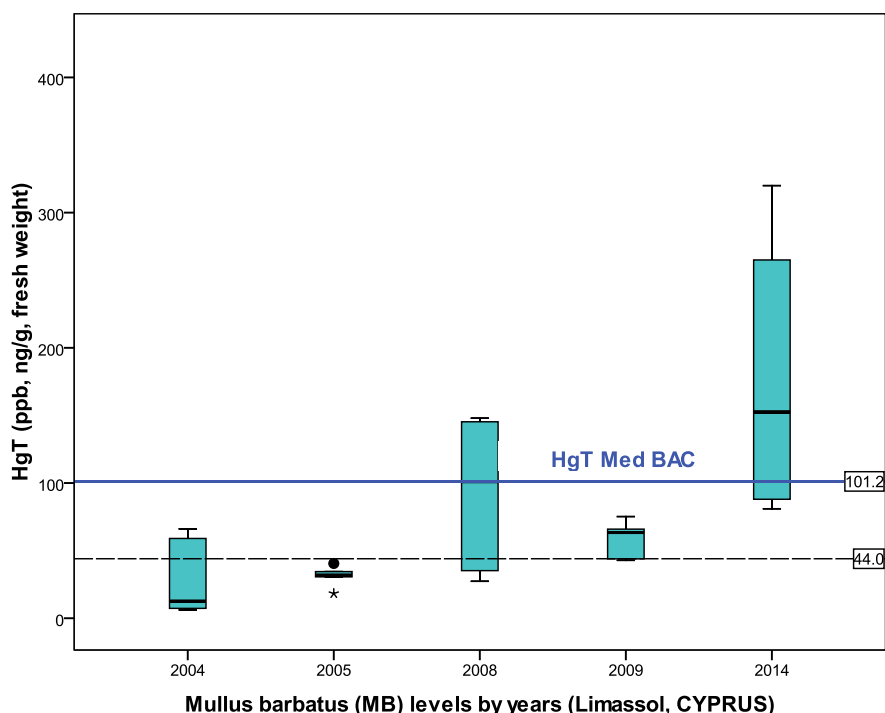


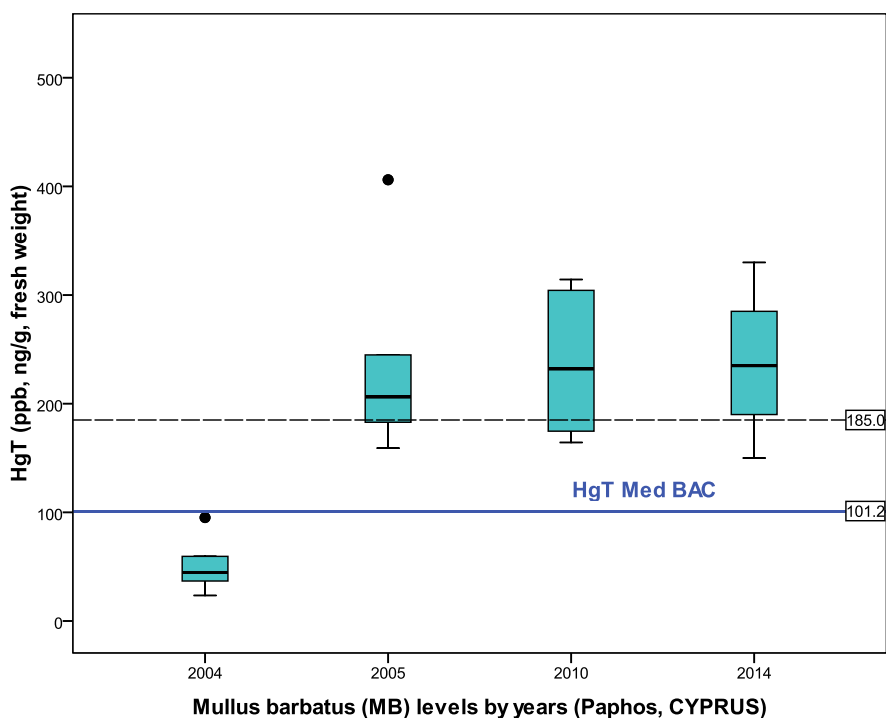
Figure Cyp.3. Total mercury box-plots at the Limassol station. The dashed line represents the median.

It is not possible to evaluate with the current data gaps if the variability and tendency between the years 2008 and 2014 at the Larnaca and Limassol stations are related to minor inputs of mercury in the area, due to slightly changes in the sampling months and geographical coordinates or simply responds to random between year variability. The year data gaps for HgT should be taken into account to this regard. At Larnaca station Cd was reported in two samples, with 83 and 153  $\mu\text{g}/\text{Kg}$  fw for 2008. For Pb, values of 175 and 200  $\mu\text{g}/\text{Kg}$  fw for 2008 and 2009 were reported, respectively. In both cases, these data was reported amongst a majority of BDLs for the same year and sampling campaign. In 2015, lead was reported as mentioned earlier.

On the other hand, in the Paphos area (Figure Cyp.4.) significant differences were found with respect the two other stations in Cyprus, the non-parametric Kruskal-Wallis (independent samples, Chi-square statistic,  $\alpha=0.05$ ) and Mann-Whitney (U statistics,  $\alpha=0.05$ , 2-tailed) tests were also applied to detect significant differences between years. Accordingly, the total mercury measured in 2004 is significantly lower at Paphos compared to 2005, 2010 and 2014. The overall median value of HgT at this station is 185  $\mu\text{g}/\text{kg}$  fw. above the Med BAC.

It should be mentioned here, that the monitoring strategy to investigate temporal trends in benthic fish (MB) in Cyprus could not be assessed due to the non-detected values for the majority of contaminants, although levels can be evaluated. The time-trend series at these 3 stations are broken due to missing years, but fundamentally because the lack of reported quality controlled values above the detection limits.





Figures Cyp.4. Total mercury box-plots at the Paphos station. The dashed line represents the median.

Therefore, in Cyprus, the sampling strategy need to be revised thus it is based solely in this species (as reported in the MEDPOL database) with no cost-efficient results, either to properly understand chemical contamination (levels and trends) or their biological effects in the marine environment. At present, this would suggest a new start of a long-term monitoring program for biota samples in Cyprus (or another alternative), but what is clear is that the ratio between monitoring and analytical efforts versus the environmental information obtained is not cost-effective. Cyprus have not yet submitted to the MEDPOL database any datasets for trace metals and organic contaminants in sediments, and therefore a shift to a long-term sediment monitoring program would be desirable and complementary.

The reason why many of the MEDPOL chemical contaminant parameters, in *Mullus barbatus* (MB) species, are reported as BDLs should be further discussed in the context of a long-term monitoring program (a similar pattern will be discussed in the present report for other countries). However, the main reason might be simply that this species (and the selected fillet tissue sample) is not a good proxy to monitor levels of metals and organic contaminants at the low end of trace concentrations in the marine environment. To overcome this problem to some extent, the analytical methodologies could be refined (eg. increase the amount of sample, tissue selection, use of advanced analytical instrumentation, etc.) and the natural variability of these compounds in this species controlled through an improved sample collection (e.g. number of individuals per sample, pooled samples, best length/weight ratio determined, etc.). However, as seen for the datasets from Cyprus, even if the sample collection was performed adequately (with a 6 pooled samples of fillet tissue collected regularly) the data show a large variability and a majority of BDLs. More, because the latest datasets submitted to MEDPOL do not report analytical results for the majority of chemical compounds any evaluation of the biological effects could not be correlated with the presence of these hazardous substances.

### 4.3. Israel

Further datasets for 2010, 2011, 2012 and 2013 were available in the MEDPOL database from Israel for biota and sediments. In the present report we have updated the 3 species monitored most recently, *Mullus barbatus* (MB), *Donax trunculus* (DT) and *Macra corralina* (MC) (fish and two bivalves, respectively). The temporal trends in the two bivalve species (DT and MC) have been evaluated, thus datasets spans now from 5 to 7 years in some locations (Haifa and Akko Bays). Sediments samples were continued to be collected at the majority of historical sites, although the sampling strategy still limited to a single sample per site (without replicates), thus temporal trends analysis supported by yearly replicate samples (within year variability) could not be performed. Instead an update and level comparisons plots for sediment stations are presented.

A part from the study of Cd and HgT in biota, we have also included in the present report the datasets for Cu, Fe and Zn up to 2011 (with HgT and Zn up to 2013). For sediments we have included HgT, Cu, Fe, Zn, Mn, Ni and Pb up to 2013. These datasets are available in the MEDPOL database.

#### Trace metals in *Mullus barbatus* (MB)

The target fish species was chosen to be MB. The fillet tissue was selected as sample tissue (fewer determinations in liver tissue were omitted in this report). These samples were not pooled and individuals were collected and processed individually to perform chemical analyses. There are 3 main trawling areas along the Israel coast for the study of levels and trends, TRAWL N, TRAWL C and TRAWL S. These stations were renamed after the year 2000 (previously, HMF8, HFM9 and HMF9, respectively), and therefore these datasets have been combined. The TRAWL C area is the most complete time series for the study of temporal trends (1999-2013), despite all the areas have data gaps for this period (depending on the parameter). From the whole datasets, 4 gross outliers were deleted for TRAWL\_S samples in 2004 (exceptionally high for Fe, Cu and Zn) within the 21 individuals, and 2 gross outliers were also deleted for Cu and HgT for the same reason (plots were scaled accordingly). For TRAWL C data for HgT and Zn is available only for 2012, and two individual gross outliers in TRAWL N for mercury (1182 and 1686 µg/kg fw) were also omitted from the calculations and plots.

The levels of Cd were reported below detection limits (BDLs) in MB for the updated datasets in the MEDPOL database. The Cd, HgT, Cu, Fe and Zn levels were evaluated in MB samples including the most recent datasets available. Both historical level comparisons between areas and temporal trends (except for Cd) are presented in the figures below (Figures Isr.1.-Isr.9.). From the box-plots, it can be observed that the median at the TRAWL N (located offshore Haifa Bay) is always above the median concentrations for the whole period and stations datasets, although the overall median calculated for each element fit within the 25<sup>th</sup> and the 75<sup>th</sup> quartiles in all the stations (Figures Isr.1-Isr.5). The robust non-parametric Kruskal-Wallis test (independent samples, Chi-square statistic,  $\alpha=0.05$ ) was applied to elucidate if there were significant differences between the sampling areas. Significant differences were found for all the metals and trace elements, although the groups were somewhat unequally weight in terms of data measurements (N). Further, Mann-Whitney tests (U statistics,  $\alpha=0.05$ , 2-tailed) were performed and significant differences were found mainly between the TRAWL N and TRAWL S for HgT and Cu, whilst the significant differences between TRAWL N and TRAWL C were found for Fe and Zn. No attempts were made for Cd (Figure Isr.1), as datasets for this element were reported >90% to be BDLs (see table below). It is worth to mention that when standard analytical methodologies are applied to samples with low concentration levels, the total uncertainty in analytical measurements increases, and therefore, the 10% of the values reported for Cd in the datasets need to be interpreted also with caution. The Mediterranean BACs and recommended EACs are included in the Figures for Cd and HgT.

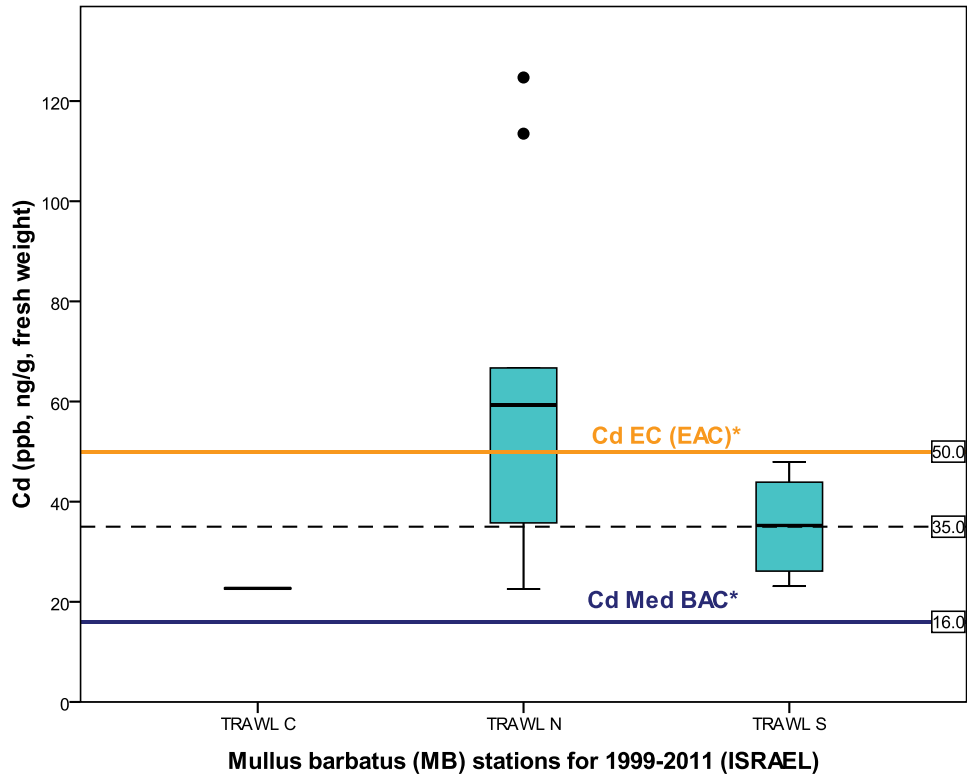


Figure Isr.1. Cadmium box-plots in MB offshore Israel coast for all the years (MEDPOL database) at the 3 selected stations with updated datasets for 2010-2011. The dashed line represents the median.

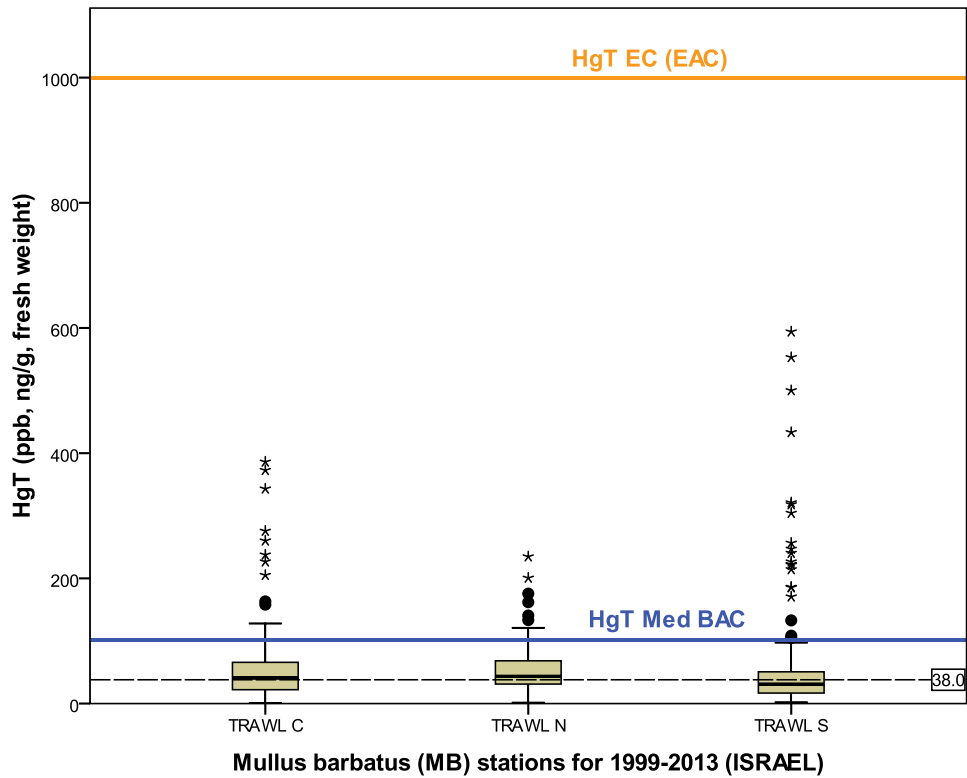


Figure Isr. 2. Total mercury box-plots in MB offshore Israel coast for all the years (MEDPOL database) at the 3 selected stations with updated datasets for 2010-2013. The dashed line represents the median.

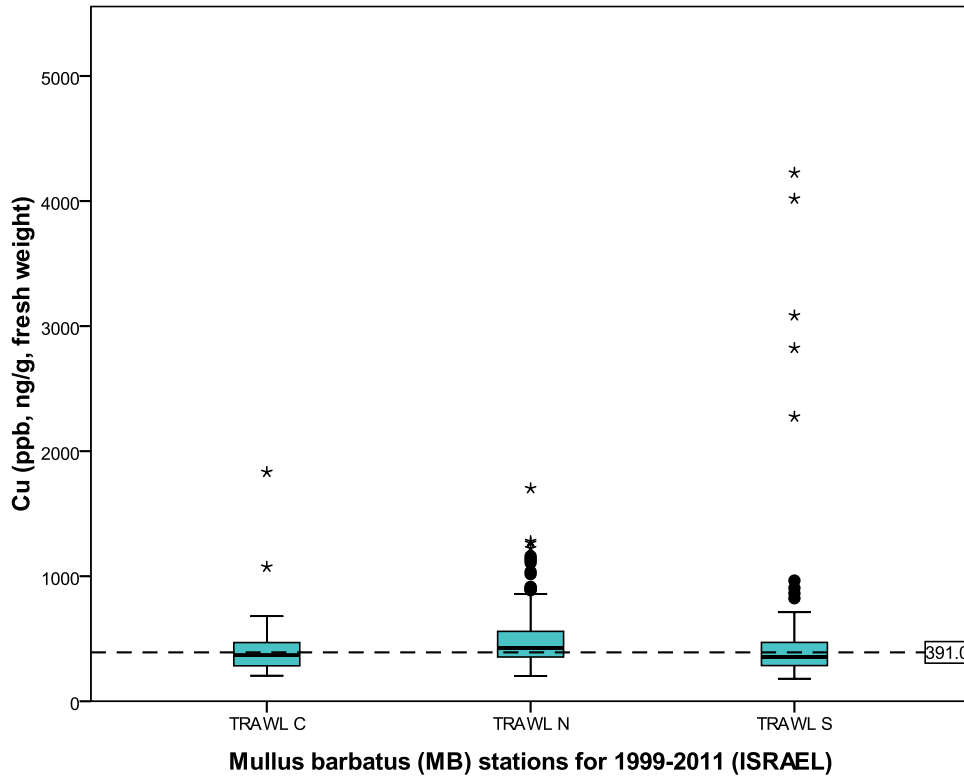


Figure Isr.3. Copper box-plots in MB offshore Israel coast for all the years (MEDPOL database) at the 3 selected stations with updated datasets for 2010-2011. The dashed line represents the median.

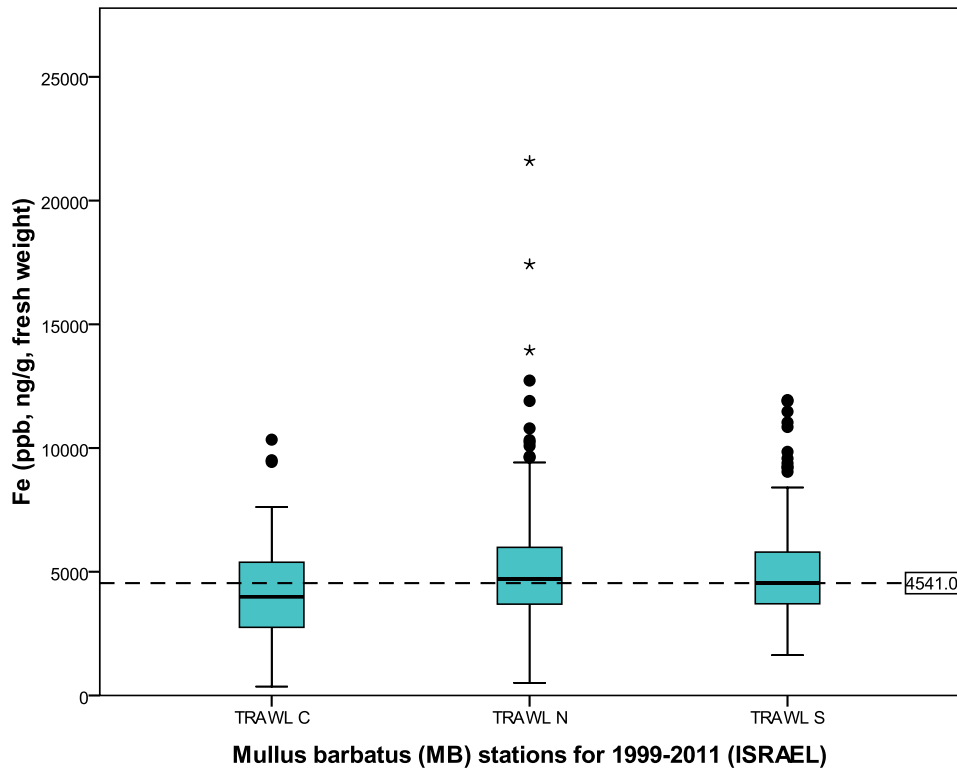


Figure Isr.4. Iron box-plots in MB offshore Israel coast for all the years (MEDPOL database) at the 3 selected stations with updated datasets for 2010-2011. The dashed line represents the median.

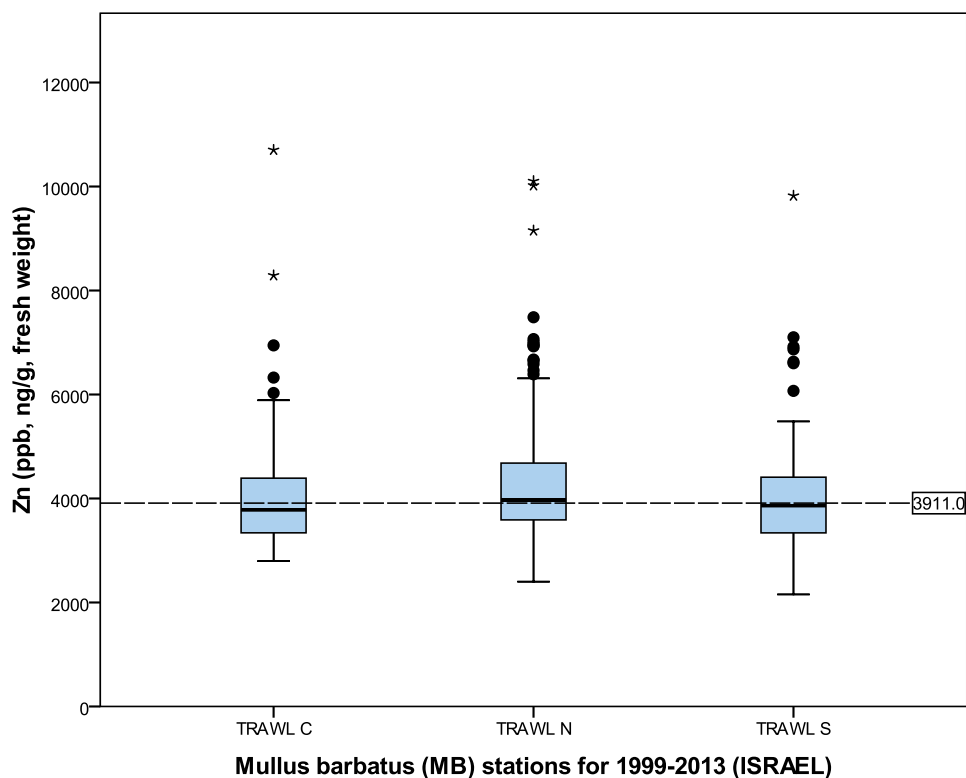


Figure Isr.5. Zinc box-plots in MB offshore Israel coast for all the years (MEDPOL database) at the 3 selected stations with updated datasets for 2010-2013. The dashed line represents the median.

The Table Isr.1 below summarizes the statistics for the 3 stations located offshore Israel taking into consideration the new datasets (2010 - 2013).

Table Isr.1. Summary table of statistics for trace metals in MB (all years and stations database,  $\mu\text{g}/\text{Kg}$  fw).

Trace element	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cd	23	44	35	24	24	47	23	125
HgT	492*	65	39	42	23	65	1	1686
Cu	461	461	391	179	306	485	181	4226
Fe	454	4900	4540	2294	3489	5783	360	21598
Zn	503*	4080	3872	1105	3382	4487	2155	10703

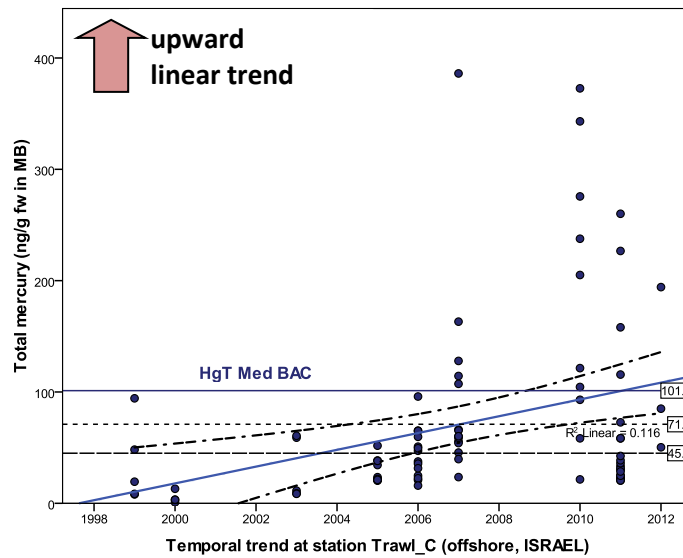
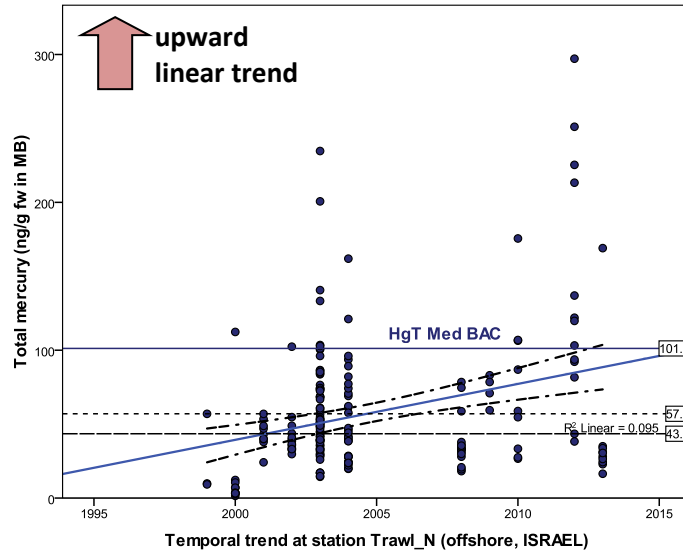
\*includes data up to 2013, otherwise 2011

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

### Temporal trends

The time series for each trace element were investigated for linear trends by means of the non-parametric  $\tau$ -Kendall B test (at the significant levels of  $\alpha=0.05$  and  $\alpha=0.01$ , 2-tailed). A two-tailed test has been chosen to allow similar weights at both sides of the data distribution.

As mentioned in the methodology section, plots are presented for trends with the best fitted line (linear, quadratic or cubic) according to the correlation coefficient, although the statistical analysis is performed for linear trends exclusively (see Table Isr.2). Further, all the plots show their inherent uncertainty bands, either fitted to individual measurements as a whole or to the linear regression mean, which gives a real view of the between year and within year variability, and therefore the potential for future changes (Figures Isr.6.-Isr.9.).



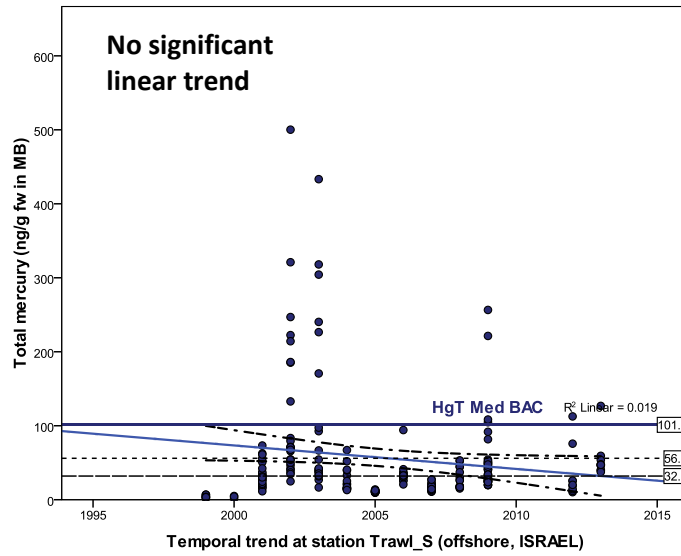
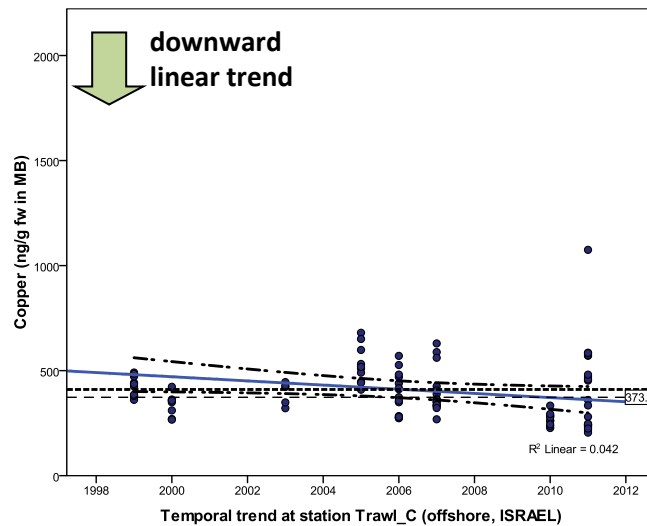
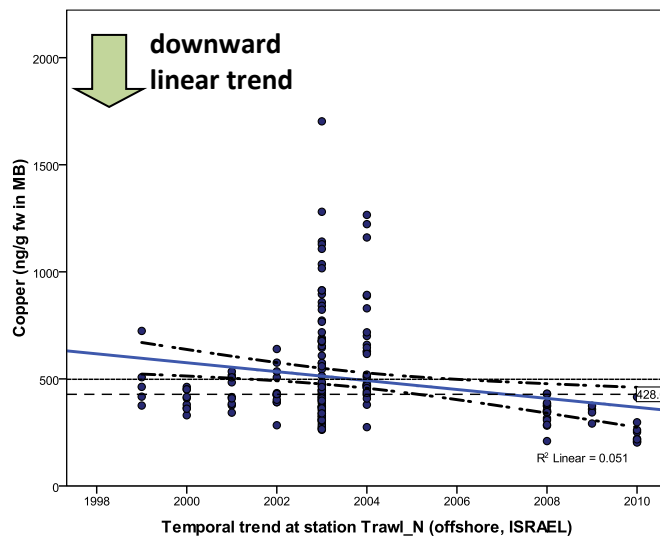


Figure Isr.6. Total mercury temporal trends (1999-2013) offshore Israel coast. (top: Trawl N sampling area, middle: Trawl C sampling area, bottom: Trawl S sampling area; uncertainty bands are fitted to the mean, dotted line=mean, dashed line=median).



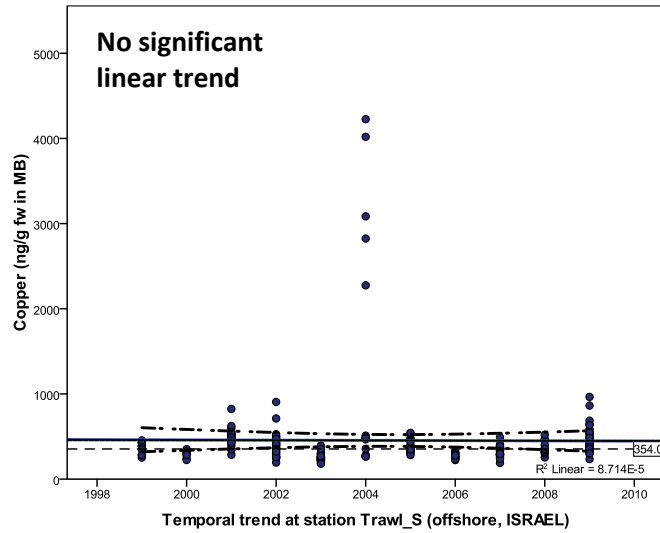
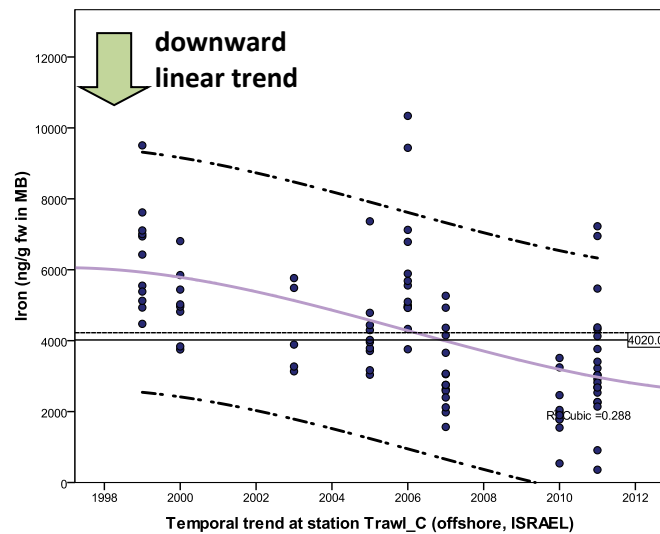
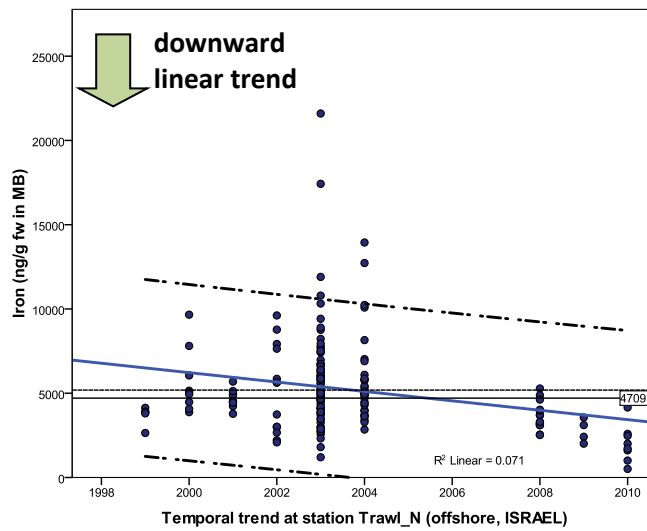


Figure Isr.7. Copper temporal trends (1999-2011) offshore Israel coast (top: Trawl N sampling area, middle: Trawl C sampling area, bottom: Trawl S sampling area; uncertainty band are fitted to the mean, dotted line=mean, dashed line=median).





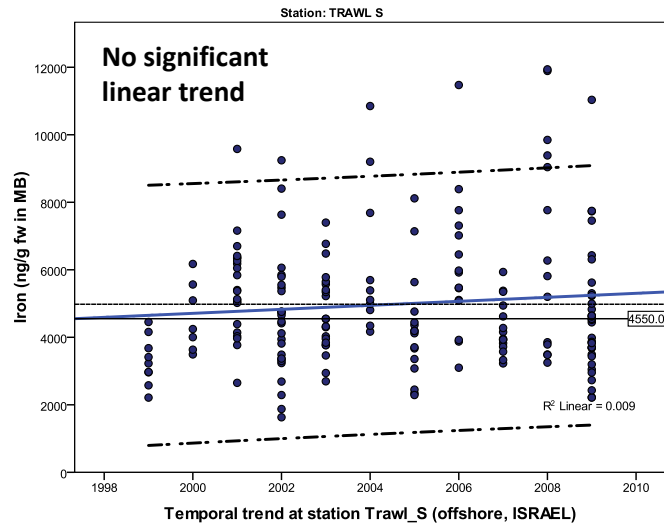
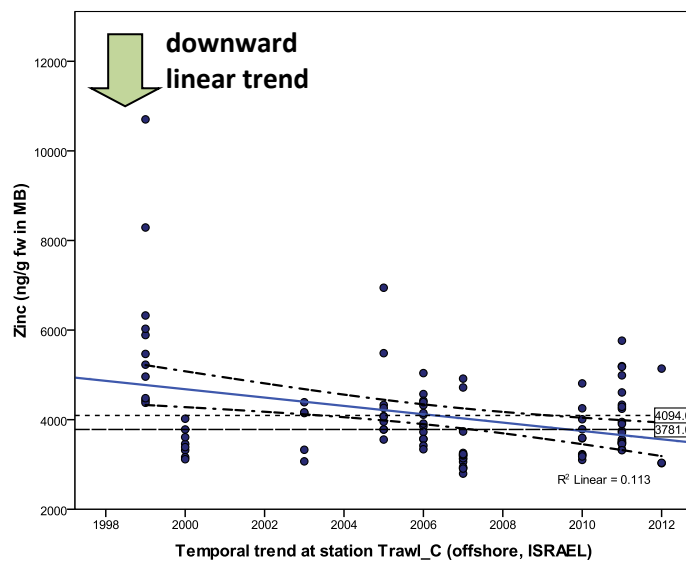
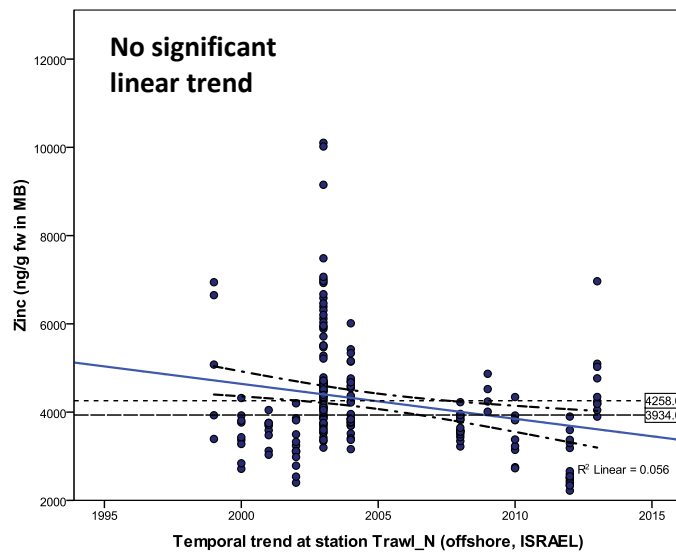


Figure Isr.8. Iron temporal trends (1999-2011) offshore Israel coast. (top: Trawl N sampling area, middle: Trawl C sampling area, bottom: Trawl S sampling area; uncertainty band are fitted to individual measurements as a whole, dotted line=mean, dashed line=median).



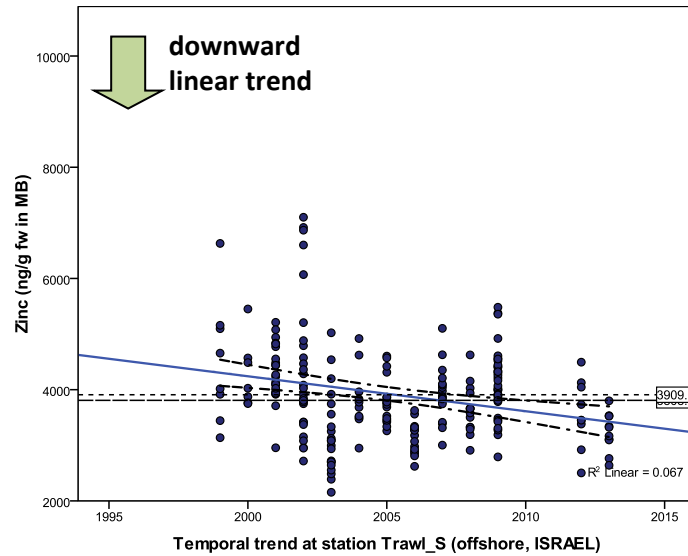


Figure Isr.9. Zinc temporal trends (1999-2013) offshore Israel coast. (top: Trawl N sampling area, middle: Trawl C sampling area, bottom: Trawl S sampling area; uncertainty band are fitted to individual measurements as a whole, dotted line=mean, dashed line=median).

With three more years' datasets (2010, 2012 and 2013) at the station TRAWL N, some changes can be observed from the previous temporal trends analysis (UNEP(DEPI)/MED WG.365/Inf.5). The HgT tendency to increase, observed previously, has become a statistically significant upward trend ( $\tau$ -Kendall B test,  $\alpha=0.05$ , 2-tailed), although the large within and between year variability in MB samples need to be taken into account (Figure Isr.6). As mentioned, for Cd no further trends could be evaluated as were reported as BDL for all the recent datasets. Statistical significant downward trends were found for Cu and Fe also at this station.

At the station TRAWL C, including 2010, 2011 and 2012 datasets, there is a significant upward trend for HgT, whilst Cu, Fe and Zn show significant downward trends. The Table Isr.2. below summarizes the  $\tau$ -Kendall B correlation coefficients for each trace element and station. It is not clear yet, what are the reasons of this large natural sampling within year variability (*c.a.* variance) in *Mullus barbatus* (MB) for some parameters, as mentioned for datasets from Cyprus. This occasional and random variability has been constantly observed for Cd, HgT, Cu and Fe without regard to the sampling area (Figure Isr.6. and Isr.7.), whilst for Zn (the occurrence of this element is not necessarily linked to anthropogenic activities) the within year data variability shows a more predictable and regular pattern ( $\Psi^2 < 0.060$ ), despite the higher ranges (see Table Isr.3). The latter, can be observed in the TRAWL S station (Figure Isr.9.) where little anthropogenic impacts should be expected at this station.

Table Isr.2. Summary of  $\tau$ -Kendall B correlation coefficients for each trace element and station.

Station	HgT	Cu	Fe	Zn
Trawl N	0.148**	-0.135*	-0.185**	-0.091
Trawl C	0.322**	-0.240**	-0.431**	-0.192*
Trawl S	0.033	0.041	0.014	-0.142**

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed)

Table Isr.3. Calculated within year variances and summary of statistics for the latest years (2009 to 2013) at each station in MB (unit in  $\mu\text{g}/\text{kg}$  fw).

Station	Year	Parameter	N	Min	Max	Mean	Median	Std.Dev.	Variance $\Psi^2(\text{LogC})$
TRAWL_N	2010	HgT	9	27	176	75	58	49	0.084
		Cu	9	203	416	253	220	68	0.010
		Fe	9	510	4661	2300	2005	1365	0.089
		Zn	9	2728	4341	3460	3379	553	0.005
TRAWL_N	2012	HgT	13	38	297	140	120	81	0.075
		Cu	-	-	-	-	-	-	-
		Fe	-	-	-	-	-	-	-
		Zn	15	2221	3899	2794	2543	529	0.006
TRAWL_N	2013	HgT	10	16	169	42	29	45	0.074
		Cu	-	-	-	-	-	-	-
		Fe	-	-	-	-	-	-	-
		Zn	10	3898	6965	4676	4292	902	0.006
TRAWL_C	2010	HgT	10	21	373	183	163	122	0.151
		Cu	10	227	333	270	270	31	0.002
		Fe	10	539	3512	2080	1938	845	0.050
		Zn	10	3102	4809	3675	3587	552	0.004
TRAWL_C	2011	HgT	21	20	260	64	33	69	0.118
		Cu	21	205	1075	395	334	209	0.040
		Fe	21	360	7226	3357	3026	1676	0.080
		Zn	21	3318	5763	4050	3750	717	0.005
TRAWL_C	2012	HgT	3	50	194	110	85	75	0.092
		Cu	-	-	-	-	-	-	-
		Fe	-	-	-	-	-	-	-
		Zn	3	3028	5140	3735	3036	1217	0.018
TRAWL_S	2009	HgT	34	20	257	54	40	52	0.076
		Cu	34	235	965	463	420	158	0.018
		Fe	34	2214	11034	4637	4480	1840	0.025
		Zn	34	2791	5482	4128	4118	618	0.004
TRAWL_S	2012	HgT	9	10	113	35	20	35	0.126
		Cu	-	-	-	-	-	-	-
		Fe	-	-	-	-	-	-	-
		Zn	9	2503	4494	3567	3451	617	0.006
TRAWL_S	2013	HgT	10	38	127	55	47	26	0.023
		Cu	-	-	-	-	-	-	-
		Fe	-	-	-	-	-	-	-
		Zn	10	2640	3799	3229	3245	353	0.002

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev (standard deviation statistic), Variance (variance statistic).

#### Trace elements in *Donax trunculus* (DT)

The selected stations for DT evaluations were ISRTMH8, ISRTMC18, ISRTMC23 (all located in Haifa and Akko Bays) and ISRTMC39 (located southwards Israel shoreline in front the TRAWL C sampling area). *Donax trunculus* is a bivalve species and has been monitored

historically by Israel within the monitoring program. The individuals were pooled in subsamples with a dissimilar number of individuals each year. There are sufficient data years to evaluate temporal trends at the selected stations for this report, except for station ISRTMC23 where there are too many years of data gaps.

Datasets for As, Cd, Cu, Fe, HgT, Mn and Zn for 2010 and 2011 were available, despite the datasets for DT at the different stations along the Israel coast present some gaps in parameters and years. The DT datasets reported also a number of Cd concentrations as BDLs. For the years 2012 and 2013, datasets for HgT were available for ISRTMC23, although only for 2012 for the rest of stations, and have been considered in this report.

The levels and temporal trends are studied for HgT (Figure Isr.10.) The data was reported on a fresh weight basis and show a median value of 18 µg/Kg fw (a ratio of 0.1 for this species could be used to transform the data to dry weight if necessary). Few mercury values (169, 570 and 1530 µg/Kg fw) were removed as gross outliers from station ISRTMH8 datasets.

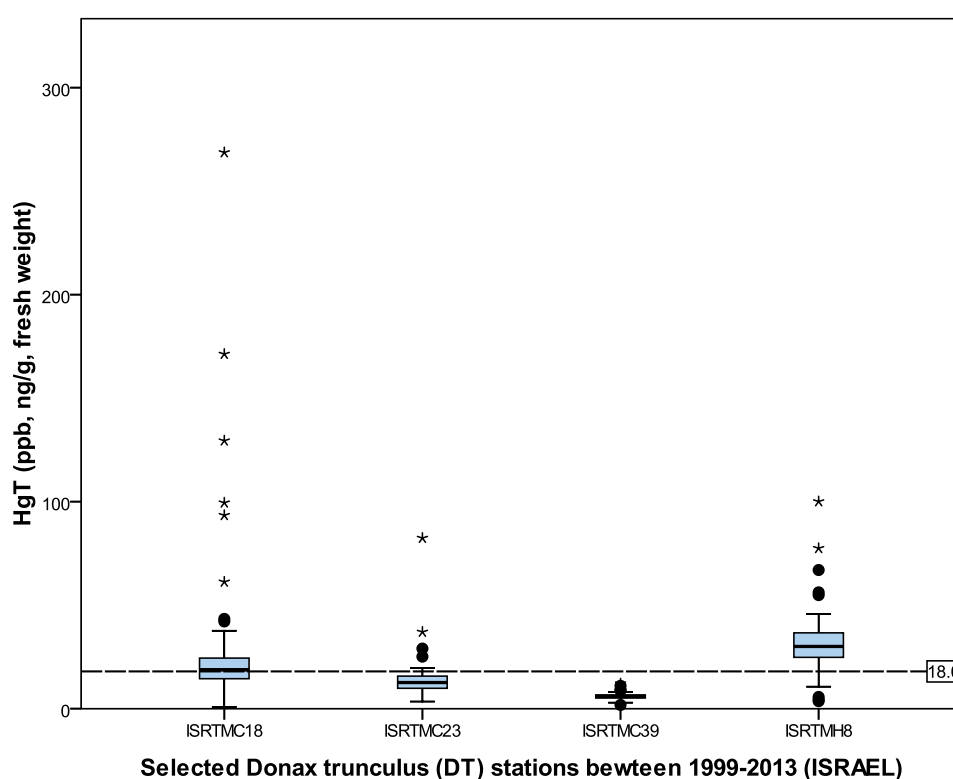


Figure Isr.10. Total mercury box-plots for all the years in the MEDPOL database at the 3 selected stations with updated datasets for 2010-2011. The dashed line represents the median.

There are significant differences between locations in Figure Isr.10. based on the robust non-parametric Kruskal-Wallis test (independent samples, Chi-square statistic,  $\alpha=0.05$ ). Further, the HgT levels at the 4 stations are all different from each other (Mann-Whitney tests, U statistics,  $\alpha=0.05$ , 2-tailed). As can be observed in Table Isr.4., the most regular trace metals determined in the database are Cu, Fe, HgT and Zn, according the number of data (N). On the contrary, as seen also for MB species, the number of samples for Cd with a valid analytical result is very low due to reported BDLs. In the case of arsenic, this element has started to be monitored later on, since 2006.

Table Isr.4. Summary table of statistics for trace metals in DT (all years and stations, µg/Kg fw).

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
As	181	999	970	324	798	1121	453	2393
Cd	27	132	101	113	65	179	36	333
Cu	327	1730	1284	895	992	1887	538	13013
Fe	327	29724	29020	15926	21448	37374	533	101348
HgT	324*	22	18	16	11	26	2	269
Zn	329	10221	9307	4231	7645	11877	2838	29096

\*includes datas up to 2013, otherwise 2011

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

#### Temporal trends

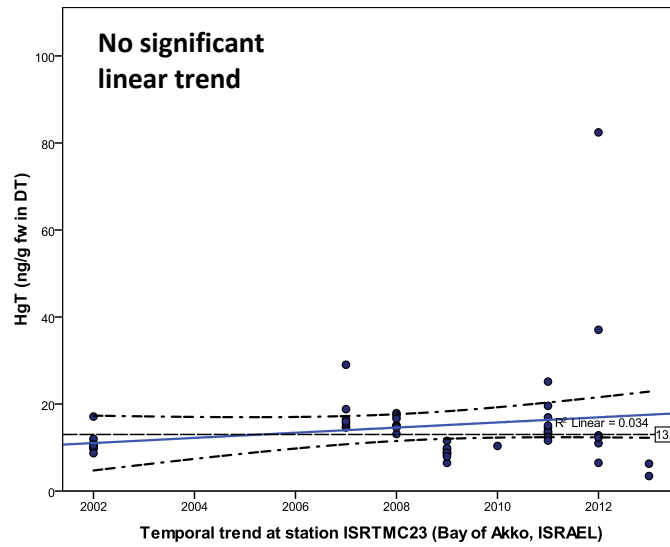
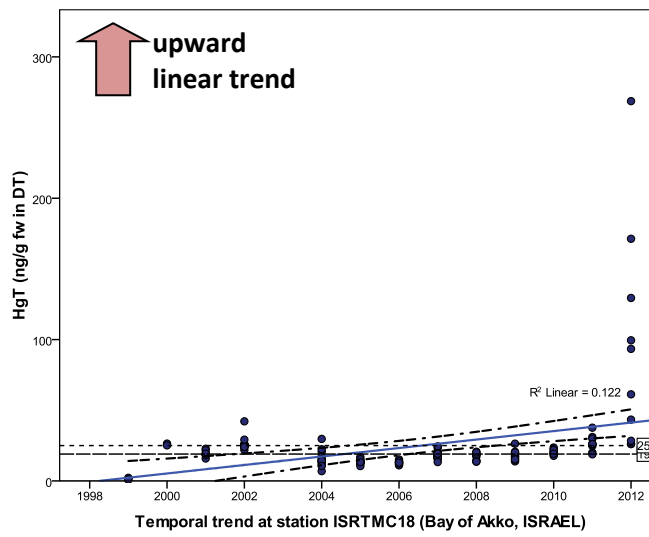
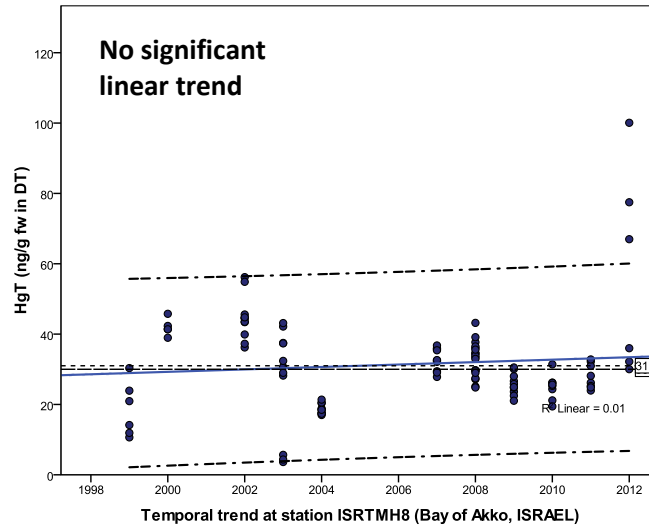
First of all it should be observed the concentration ranges for the HgT determinations at each station (Figure Isr.11.). The station ISRTMC39 (located southwards Israel coast) presents the lowest range with reported HgT levels near the detection limits of the analytical methodologies for biota (2.5 µg/kg fw), whilst the values of ISRTMH8 are the highest and are explained by its location (Akko Bay), although not relevant. Linear and cubic fitting allow for the interpretation of the HgT trends at this stations. The Table Isr.5. below, summarizes the correlation coefficients for linear regressions.

Table Isr.5. Summary of  $\tau$ -Kendall B correlation coefficients for HgT at each station.

Station	HgT
ISRTMH8	0.094
ISRTMC18	0.287**
ISRTMC23	-0.018
ISRTMC39	0.086

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed). *Note: plots show the Pearson coefficient which might differ.*

At the station ISRTMH8 (located in front of a chloralkaly plant in Haifa Bay (a hot spot), there is a slightly upward trend but not statistically significant, although the time series presents some within year variability, especially before 2007 and for the last available year 2012. The overall uncertainty bands are fitted for the individual measurements, the low correlation coefficients and the earlier higher values with high within year variance, however, point to a rather stable HgT trend at this station (Figure Isr. 11). Further datasets will confirm upward or downward linear trends at this station. On the contrary, the coastal station located south in Haifa Bay, ISRTMC18, presents a significant upward linear trend with a high within year variance for the latest year 2012. The station located in the southern coast, ISRTMC39, do not present significant a linear trend, with an acceptable within year variability (see Table Isr.6.), in accordance with its location (not impacted area). In Figures Isr.10 and Isr.11, it can be also observed the scale (Y axis) of the mean and median values for each station, confirming a gradient from Haifa Bay to the southern coast of Israel.



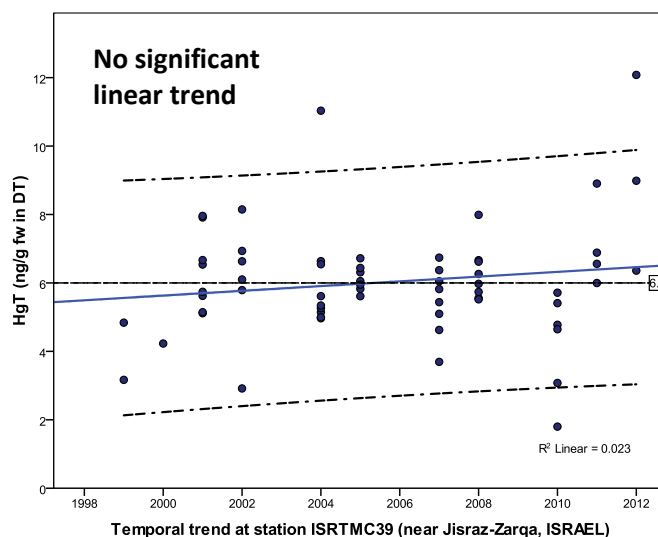


Figure Isr.11. Total mercury temporal trends (1999-2013) offshore Israel coast. (from top: station ISRTMH8, ISRTMC18, ISRTMC23 and ISRTMC39; uncertainty bands are fitted to individual or mean measurements as a whole, dotted line=mean, dashed line=median).

Table Isr.6. Calculated within year variances for the time series and summary of statistics for *Donax trunculus* (DT) datasets at each station (unit in µg/Kg fw).

Station	Year	Parameter	N	Min	Max	Mean	Median	Std.Dev.	Variance $\Psi^2(\text{LogC})$
ISRTMH8	1999	HgT	6	11	30	18	17	7	0.033
	2000		5	39	46	41	41	2	0.001
	2001		-	-	-	-	-	-	-
	2002		11	36	56	44	44	6	0.004
	2003		11	4	43	26	30	15	0.178
	2004		10	17	21	18	18	2	0.001
	2005		-	-	-	-	-	-	-
	2006		-	-	-	-	-	-	-
	2007		10	28	37	32	10	3	0.002
	2008		19	25	43	32	33	5	0.004
	2009		10	21	31	25	25	10	0.003
	2010		8	19	31	24	25	4	0.004
	2011		8	24	33	28	27	4	0.003
2012	6	30	100	57	51	29	0.389		
ISRTMC18	1999	HgT	3	1	2	1	1	1	0.045
	2000		2	25	26	25	25	1	0.000
	2001		7	16	23	19	19	2	0.003
	2002		13	22	42	26	24	5	0.005
	2003		-	-	-	-	-	-	-
	2004		12	7	30	15	14	6	0.027
	2005		11	10	17	13	13	2	0.003
	2006		10	11	15	12	13	1	0.002
	2007		18	13	24	18	17	3	0.004
	2008		10	14	21	17	18	3	0.005
	2009		10	14	26	18	17	4	0.007
	2010		9	18	24	20	20	2	0.002
	2011		10	19	38	26	26	5	0.008
2012	10	26	269	95	77	78	0.130		

ISRTMC39	1999	HgT	2	3	5	4	4	1	0.017
	2000		1	-	-	4	4	-	-
	2001		8	5	8	6	6	1	0.006
	2002		6	3	8	6	6	2	0.024
	2003		-	-	-	-	-	-	-
	2004		9	5	11	6	5	2	0.012
	2005		7	6	7	6	6	.3	0.001
	2006		-	-	-	-	-	-	-
	2007		8	4	7	5	5	.9	0.007
	2008		8	6	8	6	6	.8	0.003
	2009		-	-	-	-	-	-	-
	2010		6	2	6	4	4	2	0.037
	2011		4	6	9	7	6	1	0.005
2012	3	6	12	9	9	3	0.019		

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev (standard deviation statistic), Variance (variance statistic).

#### Trace metals in *Mactra corralina* (MC)

This bivalve species was also monitored by Israel along DT in the same stations. Further datasets for Cd, Cu, Fe, HgT, Mn and Zn from 2010 to 2013 were provided for different stations and years along the Israel coast. Similarly to the other two species reported in this report, the levels for Cd were reported as BDLs in the majority of the cases. Levels were reported on a fresh weight basis and a ratio of 0.1 for this species could be used to transform the data to dry weight if necessary. We have selected the stations ISRTMH8 and ISRTMC18 (to perform the levels and trend analysis on HgT. The overall data levels in this species for the Israel coast are summarized in the Table Isr.7. below and plots below up to 2011. For 2012, HgT for stations ISRTMC18 and ISRTMH8 were 45 and 29 µg/Kg fw, respectively.

Table Isr.7. Summary table of statistics for trace metals in MC (all years up to 2011 and stations, µg/Kg fw).

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cd	33	113	99	83	64	147	25	319
Cu	122	801	739	465	541	1006	296	1722
Fe	125	142801	83040	152232	35841	188073	6496	794938
HgT	111	30	28	26	16	42	2	82
Zn	125	10470	8784	4237	7177	11414	2748	35122

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).



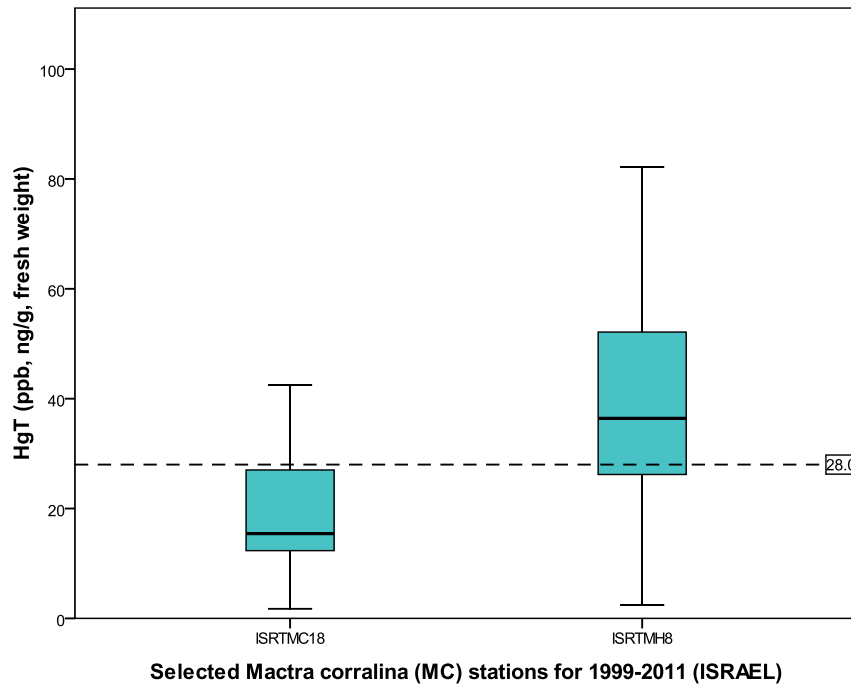


Figure Isr.12. Total mercury box-plots for all the years in the MEDPOL database at the two selected stations with updated datasets for 2010-2011. The dashed line represents the median.

Significant differences were confirmed based on the robust non-parametric Mann-Whitney tests (U statistics,  $\alpha=0.05$ , 2-tailed) between the two stations selected in Haifa and Akko Bays, as can be observed in Figure Isr.12. The station ISRTMH8 (hot spot) present the highest values for HgT as also observed for DT in the same station, although these are in the low range of the mercury values for bivalves (36  $\mu\text{g}/\text{kg}$  fw for MC and 29  $\mu\text{g}/\text{kg}$  fw for DT, respectively). It can be observed that the median for ISRTMH8 is above the overall median, whilst the southern station ISRTMC18 median (15  $\mu\text{g}/\text{kg}$  fw) is placed below.

#### *Temporal trends*

The uncertainty bands fitted to individual measurements and to the mean value of the linear trend for the stations ISRTMH8 and ISRTMC18, respectively, indicates large between year variability (Figure Isr.13.). Further, few data values for some monitoring years increase the within year uncertainty (or inexistent, for single points within the time series). Therefore, the non-parametric  $\tau$ -Kendall B correlation coefficients are low and the test for linear trends was not statistically significant in any case (Tables Isr.8. and Isr.9.).

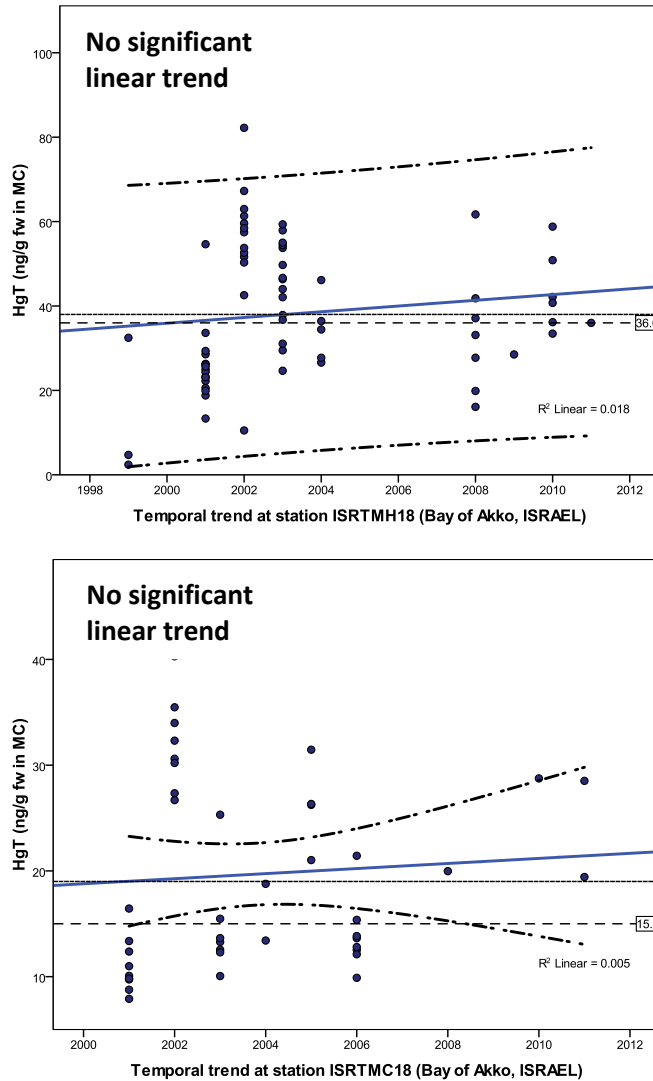


Figure Isr.13. Total mercury temporal trends (1999-2011) offshore Israel coast. (top: station ISRTMH8, bottom: ISRTMC39; uncertainty bands are fitted to individual measurements and to the mean, top and bottom respectively; dotted line=mean, dashed line=median).

It should be noticed that for the station ISRTMC18 the latest datasets contain a low number of samples (2008-2010) in comparison with previously reported datasets for this species. Obviously, this has an effect when calculating the uncertainty bands fitted to the mean, as can be observed in Figure Isr.13., where the uncertainty bands wider its range after 2006. Therefore, statistical significance, despite using a robust non-parametric  $\tau$ -Kendall B test, is difficult to achieve. Accordingly, no significant trends were observed for this species (Table Isr.8.). The monitoring datasets should be improved in the future to be able to statistically perform a more detailed trend analysis.

Table Isr.8. Summary of  $\tau$ -Kendall B correlation coefficients for HgT at each station.

Station	HgT
ISRTMH8	0.166
ISRTMC18	0.110

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed)

Table Isr.9. Calculated within-year variances for the time-series and summary of statistics for *Mactra corralina* (MC) datasets at each station (unit in µg/Kg fw).

Station	Year	Parameter	N	Min	Max	Mean	Median	Std.Dev.	Variance $\Psi^2(\text{LogC})$
ISRTMH8	1999	HgT	3	2	32	13	4	16	0.344
	2001		16	13	55	25	24	9	0.017
	2002		13	11	82	54	57	16	0.047
	2003		15	25	59	44	46	11	0.115
	2004		5	27	46	34	34	8	0.009
	2008		7	16	62	33	33	15	0.039
	2009		1	-	-	-	-	-	-
	2010		6	33	59	43	41	10	0.008
	2011		1	-	-	-	-	-	-
	ISRTMC18		1999	HgT	-	-	-	-	-
2000		-	-		-	-	-	-	-
2001		9	8		16	11	10	3	0.010
2002		9	27		42	33	32	5	0.005
2003		7	10		25	14	13	5	0.016
2004		2	13		19	16	16	4	0.011
2005		4	21		31	26	26	4	0.005
2006		8	10		21	14	13	3	0.009
2007		-	-		-	-	-	-	-
2008		1	-		-	-	-	-	-
2009		-	-		-	-	-	-	-
2010		1	-		-	-	-	-	-
2011		2	19		29	23	23	6	0.014

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev (standard deviation statistic), Variance (variance statistic).

#### Trace metals in sediments

Israel provided new datasets for sediment samples from 2010 and 2013. These correspond to the historical sampling sites (namely, ISRTMH0, ISRTMH3, ISRTMC14, ISRTMC18, ISRTMC22, ISRTMC23, ISRTMC26, ISRTMC39, ISRTMC43, ISRTMC49, ISRTMC55a, ISRTMH1, ISRTMH2, ISRTMH10, ISRTMH11, ISRTMH12, ISRTMH8, ISRTMH9 and ISRTMH27), being reference, coastal and hotspot stations within the monitoring program along the Israel coast since 1999.

The analytical results for Cu, Fe, HgT, Mn, Ni, Pb and Zn were reported in µg/kg dry weight for sediments sieved through the fraction <250 µm in the majority of the cases with the exception of the samples for the stations ISRTMH0 and ISRTMH3 (<1000 µm). Thus, these 2 stations were overlooked for further analyses, as initial exploratory analysis identified the highest concentrations of trace metals in these stations and would need a particular study. Further, gross outliers (out of scale data) for the year 2007 were removed for 4 stations (ISRTMC39, ISRTMC43, ISRTMC49 and ISRTMC55a). No replicates were provided (with few exceptions for some years and stations), and therefore the within year variability and the

study of temporal trends has not been attempted, as the uncertainty could not be computed. However, the large geographical distribution of the datasets provide a good spatial assessment of the levels along the Israel coast, which allow the differentiation of impacted from coastal and reference areas.

Although different analytical methodologies were employed over the years, data was considered coherent and of sufficient quality to perform a combination under the premise that quality controlled data is submitted to the MEDPOL database. Tables Isr.10 (1999-2011) and Isr.11 (2012-2013) below summarizes the main statistics for all the years and sampling stations with the exception of the stations ISRTMH0 and ISRTMH3, as mentioned. We have included in this report the datasets for manganese (Mn) and niquel (Ni). The median values obtained are comparable to those calculated previously for Israel, for example for Zn (10070 µg/kg dw for 1999-2009, 9892 µg/kg dw for 1999-2011 and 8175 µg/kg dw for 2012-2013 periods). These concentrations in sediments relates to natural baseline levels (UNEP/DEPI/MED WG.365/Inf.4).

Table Isr.10. Summary table of statistics for trace metals in sediment (all years and stations, µg/Kg dw).

Trace element	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cu	169	3380	2225	1537	1544	3081	745	38404
Fe	171	4060348	3105496	2412213	2255389	4667603	1972	25125940
HgT	161	113	63	196	21	216	1	302
Mn	171	233584	191499	158179	129783	287962	70328	135934
Ni	169	4145	2951	2588	2122	4709	816	19781
Pb	149	6170	5513	4429	3374	7803	1567	21786
Zn	192	14202	9892	9715	6289	16005	1198	119187

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

Table Isr.11. Summary table of statistics for trace metals in sediment (all years and stations, µg/Kg dw).

Trace element	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cu	37	3220	2182	1198	1648	2847	1049	27114
Fe	38	6988199	2986928	5299053	2387931	7687025	1512313	49664435
HgT	38	101	41	190	16	207	1	260
Mn	38	218844	153326	143355	107994	251349	68440	638080
Ni	35	4072	2975	1928	2062	3990	1482	22573
Pb	38	5758	5306	1695	4546	6241	3398	16154
Zn	38	11304	8175	4205	6155	10359	2545	70523

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

The following box-plots (Figures Isr.14.-Isr.20) are useful to explore the levels between the selected stations for the whole period 1999-2013, as well as for the elucidation of potential reference stations.

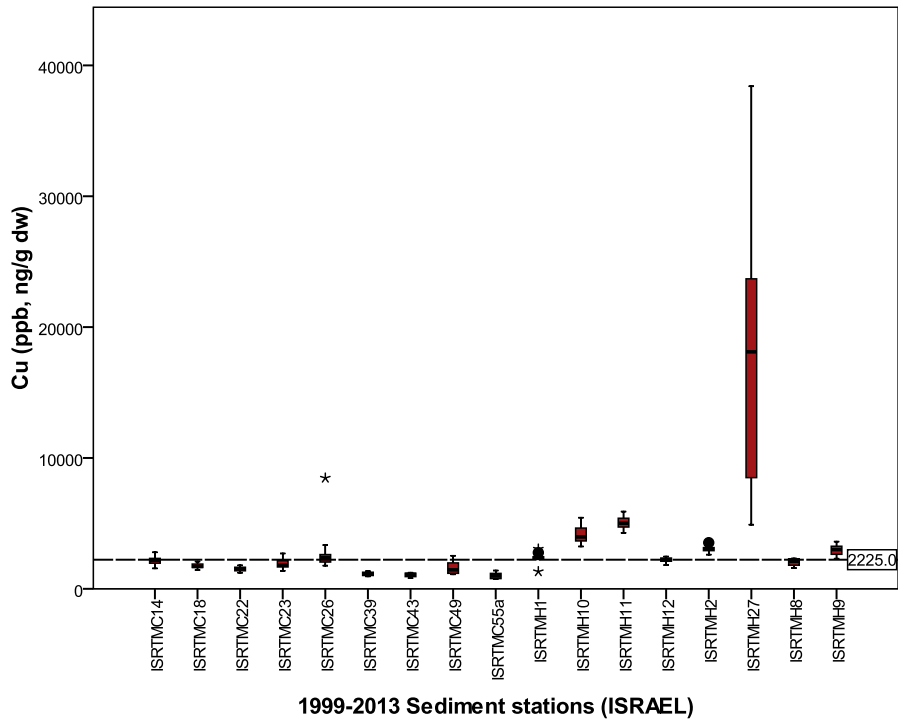


Figure Isr.14. Copper box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

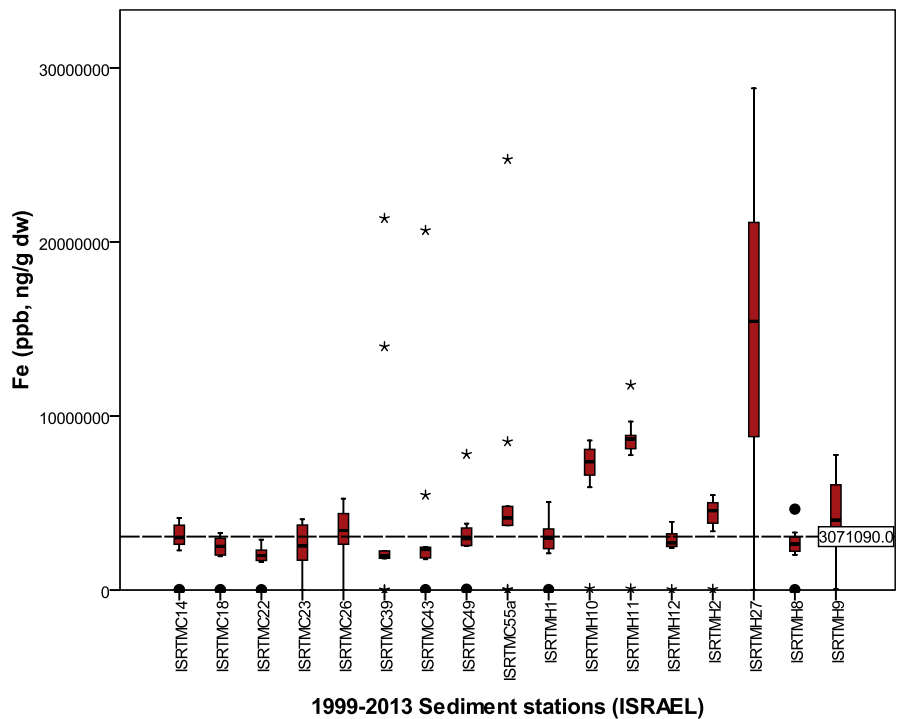


Figure Isr.15. Iron box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

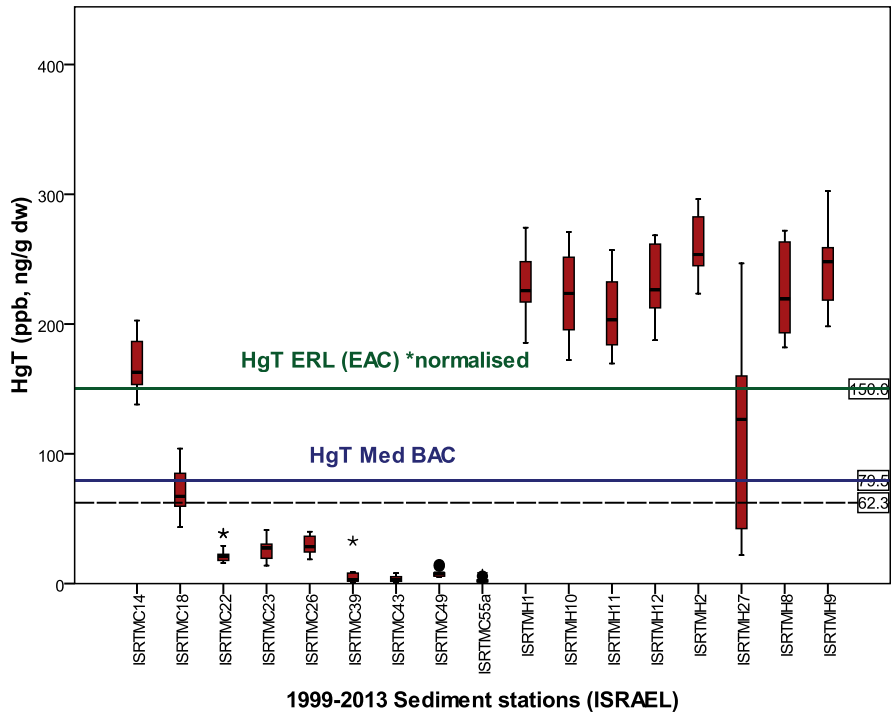


Figure Isr.16. Total mercury box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

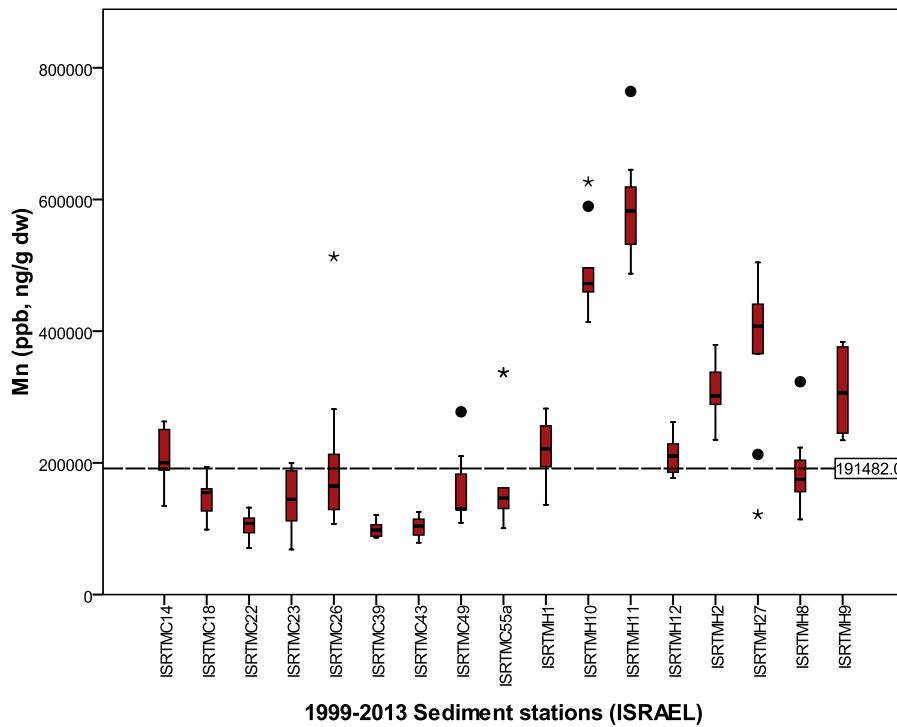


Figure Isr.17. Manganese box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

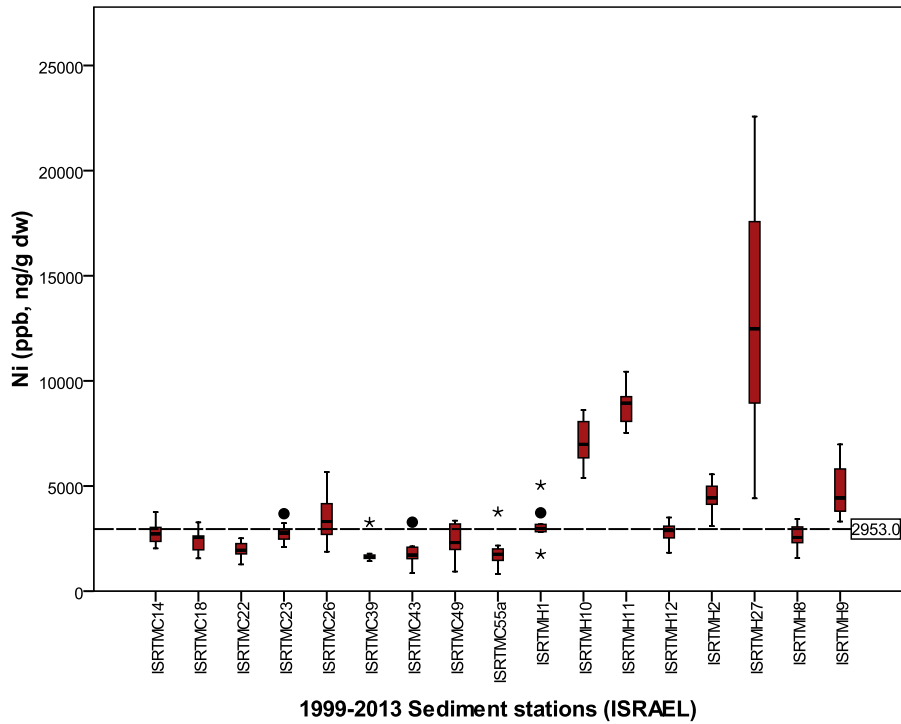


Figure Isr.18. Nickel box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

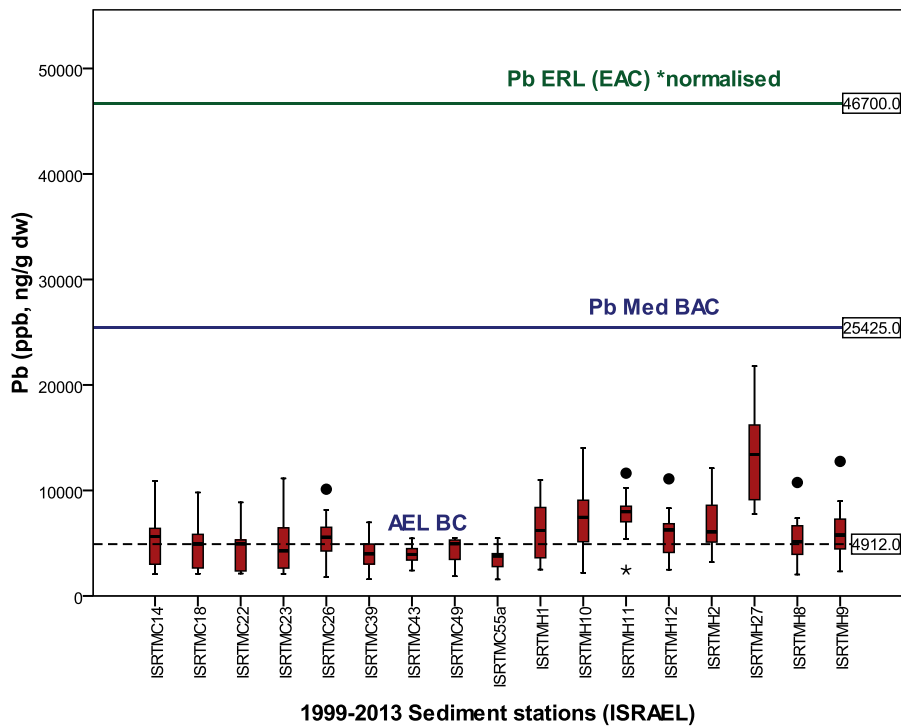


Figure Isr.19. Lead box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

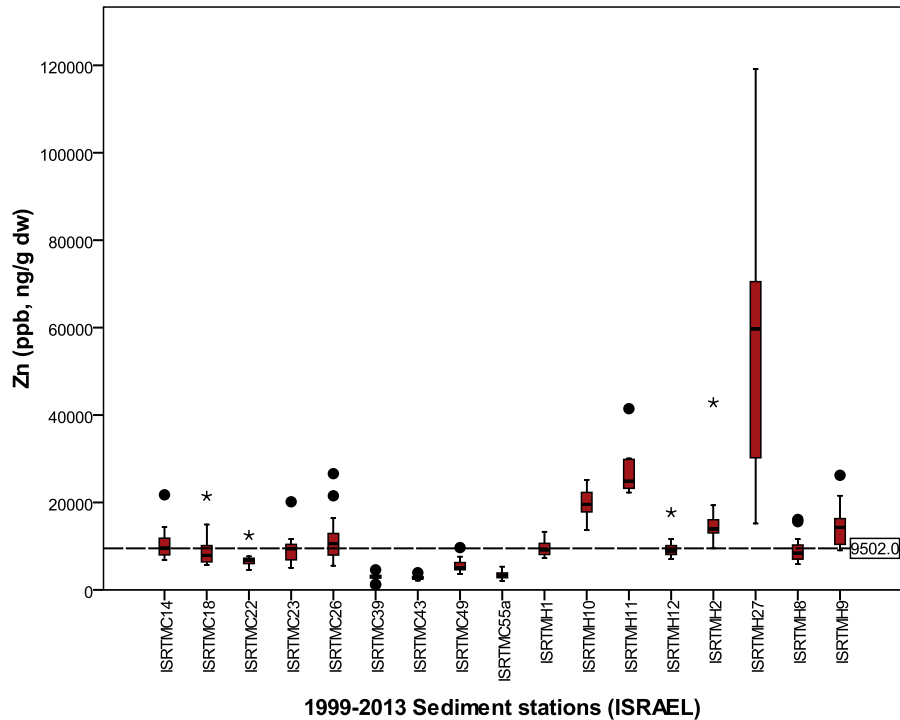


Figure Isr.20. Zinc box-plots for all the years in the MEDPOL database at the stations with updated datasets for 2010-2013. The dashed line represents the median.

A first look shows clear differences between the left and the right hand side of the plots where the coastal samples and the hotspots are placed, respectively. The station ISRTM27 (located in Haifa Bay) stands out as being the highest for Cu, Fe, Ni, Pb and Zn, despite the high data dispersion for the whole period. Surprisingly, the lowest value for HgT within the stations considered hotspots (those on the right) was found at this station; whilst the highest HgT levels were found in sediments about 10 Km north from Haifa Bay, in the surroundings of a chloralkaly plant (note: stations ISRTMH8, ISRTMH9, ISRTMH10, ISRTMH11 and ISRTMH12 conform a offshore transect from the plant, whilst ISRTMH1 and ISRTMH2 are located in front of the city of Acre).

It can also be observed that the stations ISRTMC39, ISRTMC43, ISRTMC49 and ISRTMC55a, represent a group of stations in the southern coast of Israel and far from main pollution inputs (central stations in plots), as the median values are well below the overall median for trace metals. Particularly, Figure Isr.16, shows clearly this pattern for HgT where hotspot levels are above Med BAC and EAC (normalized EAC values need to be used as reference with caution). There is a gradient between the northern hot spot stations, followed by the southern coastal stations in the same area (within the 10 km shoreline between Acre and Haifa), which are well differentiated from the third group of stations (central) along the southern coast outside Haifa and Akko Bays, as observed in biota samples (Figures above). Further, there are clear differences for Mn and Ni between the coastal stations from Haifa Bay and those for the southern coast of Israel (left and central stations, respectively). The Cu, Fe and Pb spatial distributions show a similar pattern to some extent, despite lead interpretation should take into account the AEL BC/BAC rather than the Med BAC (Figure Isr.19).



#### 4.4. Morocco

One more year was available for 2007 in the MEDPOL database for the monitored stations in the coasts of Morocco. Unfortunately, the *Mytilus galloprovincialis* (MG) species selected for the level and trends monitoring program at these stations, seems to have been interrupted and substituted with another species, namely VA. Despite an effort was done to enlarge the trace metals determinations for new targets, such as chromium (Cr), iron (Fe) and zinc (Zn), no reporting information about dry/fresh weight units was provided. Further, only 2 samples were included for MG at the stations of Kabila and Mdiq. However, the latest, have only two years of data (2006 and 2007) and no trends could be performed.

On the other side, the datasets for OC compounds in MG submitted along the mentioned samples in 2007 for Aldrin, Chlordane, DDT, DDD, DDE, Endrin, Endosulfan, Hexachlorobenzene and Lindane, does not seem correct despite reported in  $\mu\text{g/Kg}$  in dry weight as required (parts per billion), thus the concentrations ranges between 50000 and 3000000  $\mu\text{g/Kg}$  in dry weight pointing to a gross error in data units and/or analytical methods. Typical concentrations in marine biota, as previously reported for the Mediterranean basins range many orders of magnitude below (typically between 0.01 and 1000  $\mu\text{g/Kg}$  in dry weight). Therefore, this new dataset submitted for 2007 need to be revised for OCs in order to perform an assessment of levels, and therefore no further evaluation is provided in the present report.

##### Trace metals in *Mytilus galloprovincialis* (MG)

There is no new available information to study levels or temporal trend in the coasts of Morocco (as available in the MEDPOL database). Further, the historical time series have been interrupted. The solely station to perform trend analysis for trace metals is Oued Laou (MOR 1), however, there is no Cd, HgT and Pb data for 2007; and thus, the temporal trend analysis have been plotted again in this report for these trace metals to include the uncertainty bands, mean and median values, as well as assessment criteria; but no new information is provided since the last report (UNEP(DEPI)MED WG.365/Inf.5) .

Table Mor.1. Summary table of statistics for trace metals (all years,  $\mu\text{g/Kg}$  dw) at the Oued Laou station between (1999-2006).

Metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cd	36	291	220	250	150	400	90	970
HgT	32	234	145	190	67	257	13	1600
Pb	36	1114	735	900	462	1362	50	3870

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

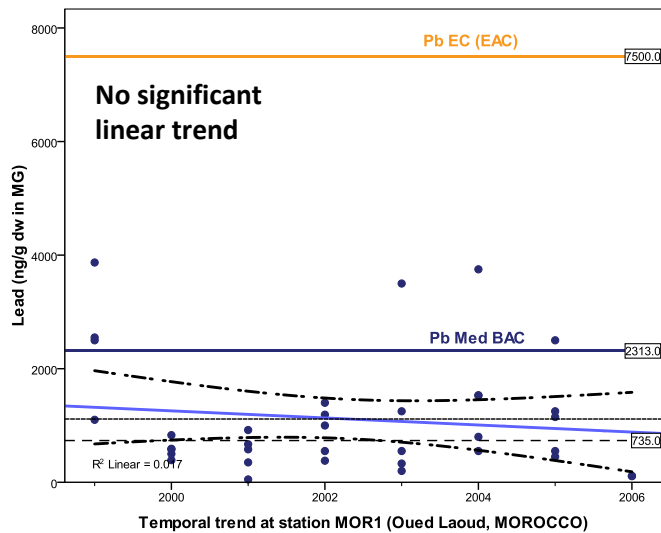
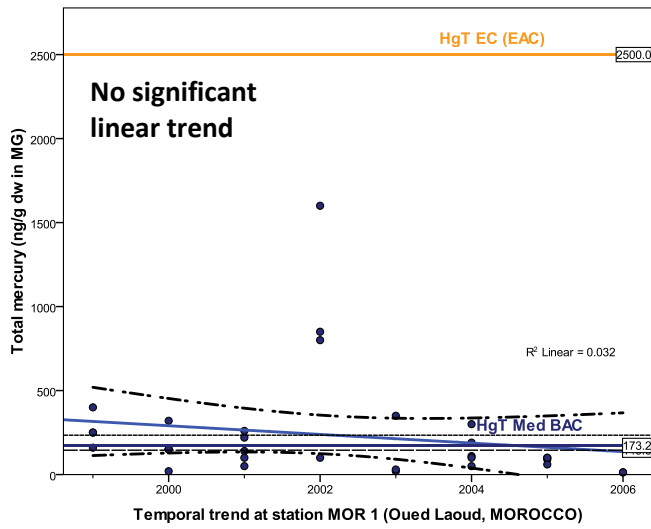
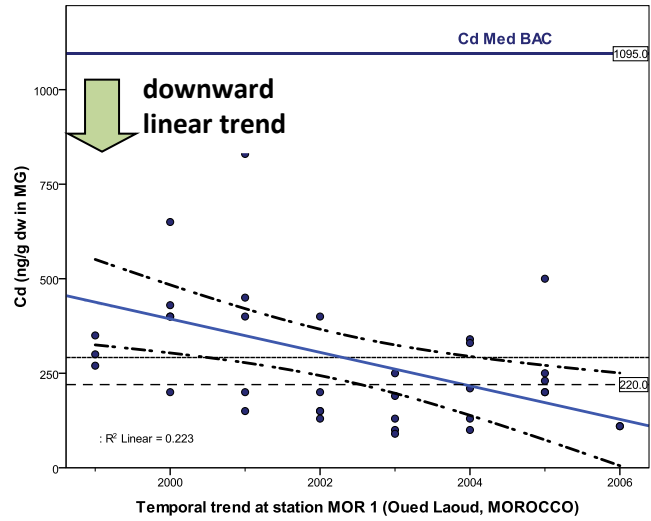


Figure Mor.1. Cadmium and total mercury temporal trends (1999-2006) at the Oued Laoud offshore Israel coast. (top: cadmium, middle: total mercury, bottom: lead; uncertainty bands are fitted to the mean,; dotted line=mean, dashed line=median).

It is not possible to perform an assessment of the Moroccan coast as datasets are too scattered between years (including different sampling months), stations and parameters determined. As previously reported, the within year variance does not fulfill the criteria established to detect a 10% change in 10 years with a statistical power of 90%, specially for total mercury and lead (see within year variances table in UNEP/DEPI/MED WG.365/Inf.5 report). Nevertheless, the location off the coast of the Oued Laou station, might represent the reference levels for trace metals in Morocco. The median values of the datasets are shown in plots for Cd, HgT and Pb (Figure Mor.1). The significant correlation was obtained for Cd (Table Mor.1.) as it was also previously reported.

Table Mor.1. Summary of  $\tau$ -Kendall B correlation coefficients at Oued Laou 1999-2006.

Station	Cd	HgT	Pb
Oued Laou	-0.472**	-0.179	-0.131

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed)

#### Trace metals in sediment

Morocco submitted single datasets for 2007 to the MEDPOL database. However, the data reported is scattered and no replicate samples for the sediments were collected. At present, no levels and trend assessments can be reported.

#### **4.5. Slovenia**

Slovenia continued the marine monitoring program for *Mytilus galloprovincialis* (MG) and sediments and provided datasets for 2010, 2011 and 2012, at the stations monitored regularly in the Gulf of Trieste. For the period 2013-2015, only few data for Cd was provided for these stations. In 2011 and 2013-2015, few data for aliphatic and aromatic petroleum hydrocarbons (the latest below detection limits, BDLs) was also reported for MG. However, the petroleum hydrocarbons results in MG need a revision before an assessment can be performed for these compounds, as will be also suggested for sediment samples later.

#### Trace metals in *Mytilus galloprovincialis* (MG)

The data provided for arsenic in 2005, and not yet evaluated, was compared with the data provided in 2011. Overall, the yearly observations under the monitoring strategy were carefully prepared with 5 replicate samples with 15 individuals each, collected after the spawning period (September-November). Different parts of the organisms have been also dissected and analysed for trace metals in occasions, although we have combined the whole soft tissue (WST) and the soft parts (SO) data from mussels to perform the assessment of levels and trends in this report. Datasets were also available for Cd and HgT and are evaluated in this report consistently up to 2012. For the period 2013-2015, only Cd data for 2013 was available and the ranges were 720-968  $\mu\text{g/Kg dw}$  for TM and 530-1022  $\mu\text{g/Kg dw}$  for station 24. These latest 2013 datasets have not been included in the present assessment.

The Kolmogorov-Smirnov and Shapiro-Wilk tests (to contrast the null hypothesis of the datasets to come from a normal distributed population,  $H_0$ ), indicates that these datasets for trace metals cannot be discarded as being normally distributed taken as a whole ( $H_0$  was not rejected, significance levels  $\alpha=0.05$ ). The Table Slo.1. summarizes the main statistics in MG samples for all the years and stations.

Table Slo.1. Summary table of statistics for trace metals in MG at the two station in Slovenia ( $\mu\text{g}/\text{Kg dw}$ ) from 1999-2012.

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
As	15	813	830	220	700	920	590	970
HgT	104	110	111	45	90	135	11	235
Cd	100	867	885	216	763	978	470	1300

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

The As comparison with the available datasets indicates the order of magnitude of the concentrations found in MG at both stations (Figure Slo.1.). The time window gap between the measurements (6 years) might have introduced some elements of analytical variability in the data (eg. analytical methods, laboratory operators, use of different CRMs, etc) a part from the natural environmental variability. However, it provides a reference value for As concentrations in biota for the northern Adriatic Sea area.

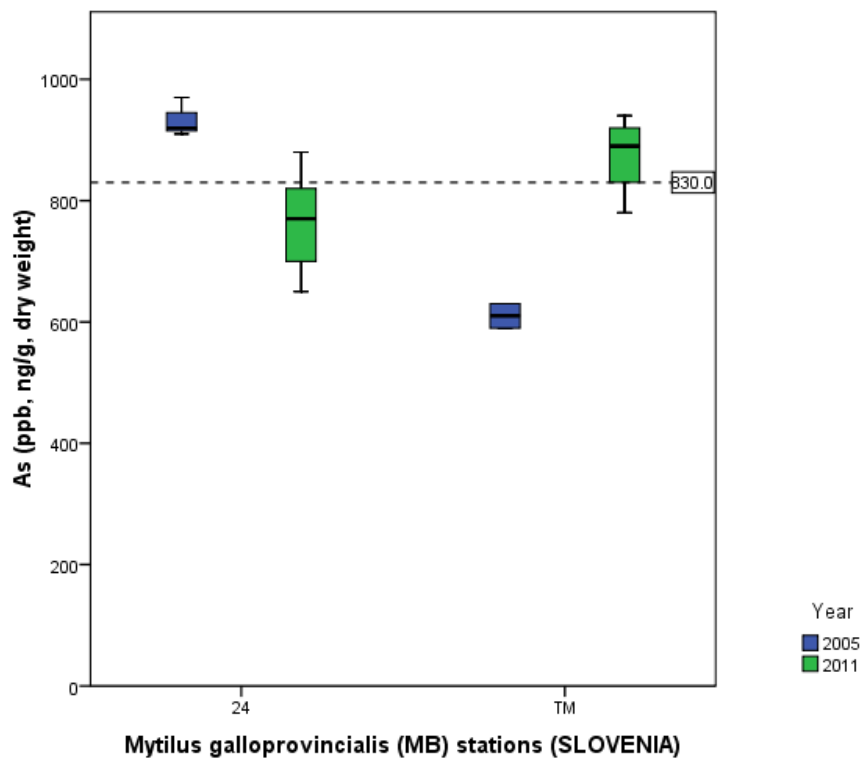


Figure Slo.1. Box-plots for comparison of arsenic datasets in 2005 and 2011 at the 2 selected stations. The dashed line represents the median.

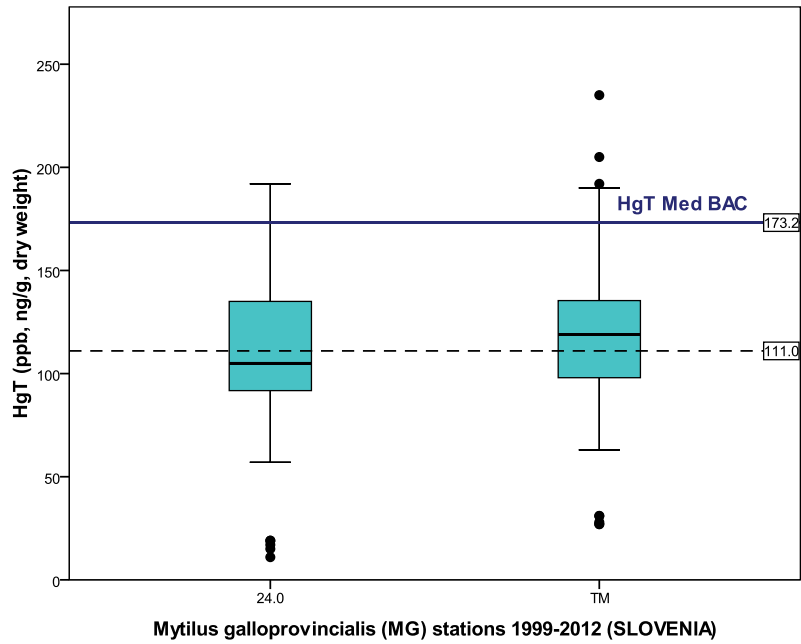


Figure Slo.2. Total mercury box-plots for all the years in the MEDPOL database at the 2 selected stations with updated datasets for 2010-2012. The dashed line represents the median.

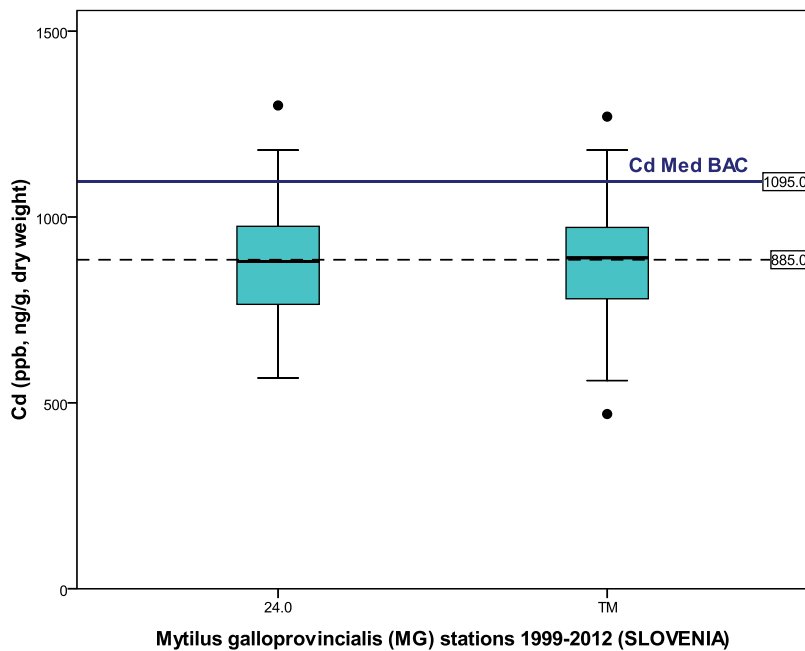


Figure Slo.3. Total mercury box-plots for all the years in the MEDPOL database at the 2 selected stations with updated datasets for 2010-2012. The dashed line represents the median.

In the Figure Slo.2. and Slo.3 the median levels of HgT and Cd at both stations nearly match the median value for the whole dataset and can be contrasted against the Med BACs. The Mann-Whitney test for two independent samples was applied to contrast the differences between the two stations (U statistics,  $\alpha=0.05$ , 2-tailed) and was not significant, thus there are no differences between both stations.

*Temporal trends*

The additional datasets submitted for HgT have significantly changed the previous trends assessment for this time series (UNEP(DEPI)/MED WG.365/Inf5).

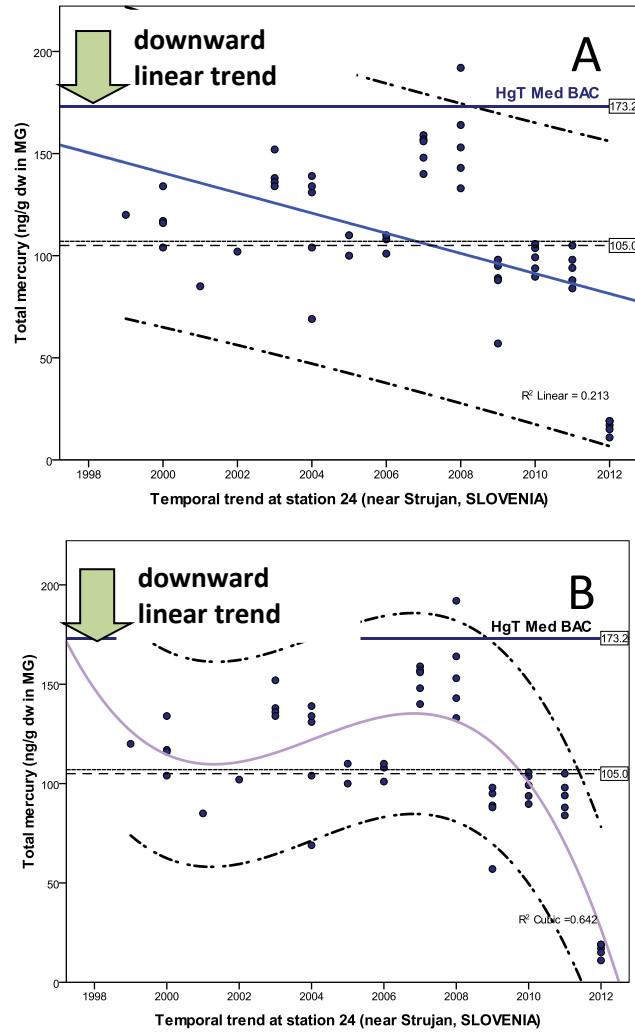


Figure Slo.3. Total mercury temporal trends (1999-2012) at the station 24. (A) total mercury with a linear fit, B) total mercury with a cubic fit; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).

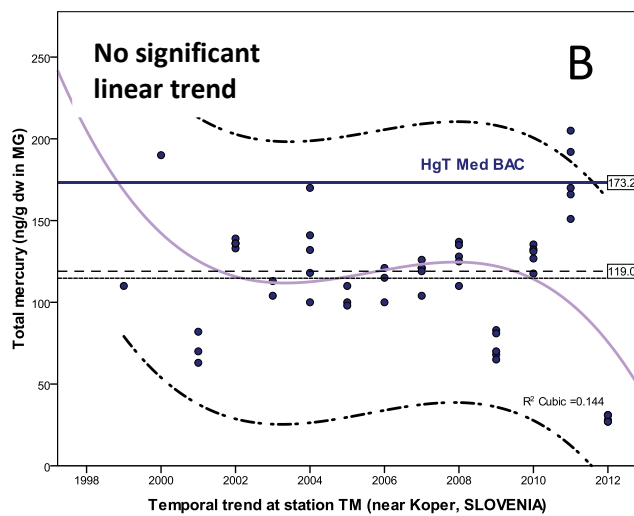
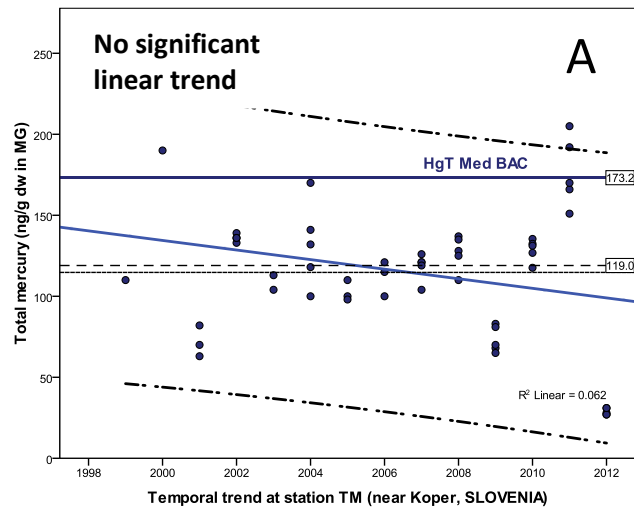


Figure Slo.4. Total mercury temporal trends (1999-2012) at the station TM. (A) total mercury with a linear fit B) total mercury with a cubic fit; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).

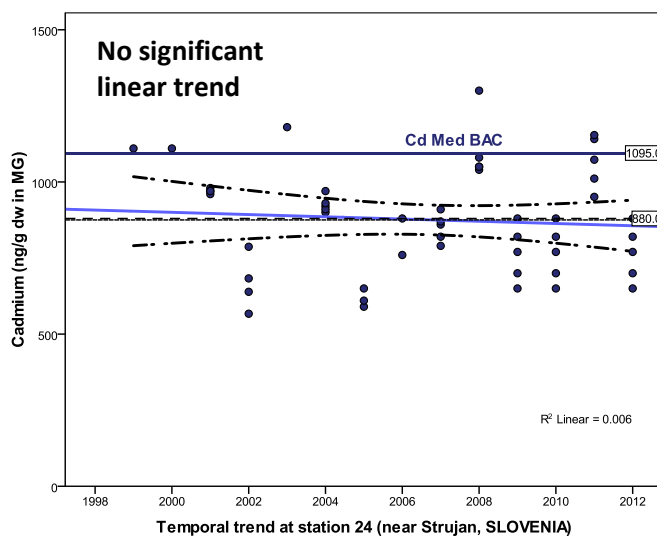


Figure Slo.5. Cadmium temporal trends (1999-2012) at the station 24. (Cadmium with a linear fit; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).

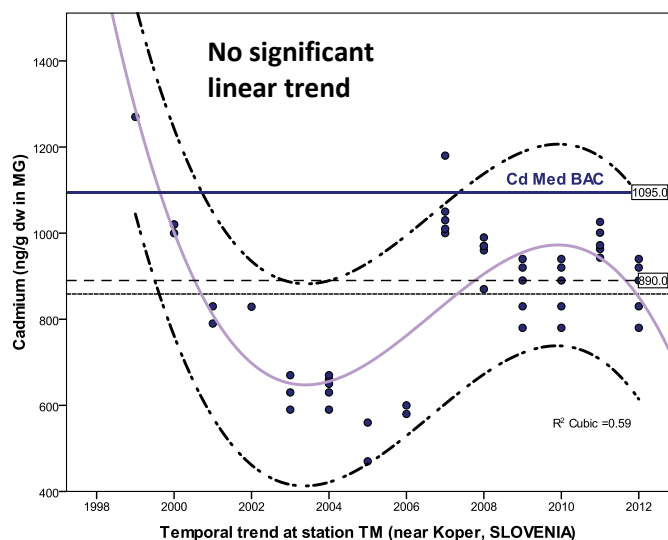


Figure Slo.6. Cadmium temporal trends (1999-2012) at the station TM. (Cadmium with a cubic fit; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).

Therefore, a HgT downward trend was statistically significant at the station 24 when 3 more years of data were added based on  $\tau$ -Kendall B correlation coefficients (monotonic, linear, confidence level of  $\alpha=0.01$ ). The dataset was also fitted to a cubic parameterization to refine the interpretations, thus it can be observed that consecutive lower values were determined since 2007 (Figure Slo.3. A and B). At the station TM the trend was not significant (Figure Slo.4. A and B) in line with the previous assessed temporal trends (UNEP/DEPI/MAP WG.365/Inf.5). For Cadmium, at the station 24, despite the number of monitoring years, no trends were found and the levels over the whole monitoring period fit well below the Med BAC threshold value calculated for the Mediterranean mussel (*Mytilus galloprovincialis*) in the present report (Figure Slo.5.). For station TM a similar conclusion can be assessed, although differences in between year variances are much larger (Figure Slo.6.). The average concentrations for 2103 were 779 and 864  $\mu\text{g/Kg dw}$  for 24 and TM, respectively (not plotted), both below the median within the time series.

Table Slo.2. Summary of  $\tau$ -Kendall B correlation coefficients at the stations 24 and TM for the period 1999-2012.

Station	HgT	Cd
24	-0.358**	-0.088
TM	-0.078	0.067

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed)

The Table Slo.3. shows the variances calculated for the whole HgT times series where it can be observed that the monitoring strategy (5 pooled samples with 15 individuals each) fulfill the criteria established to monitor the long-term monitoring program, although a high between year variability impedes to conclude significant trends. A comparative statistics between the statistical approach and PIA software statistical analysis can be found in the Annex VII of this report.



Table Slo.3. Calculated within year variances for the time series and summary of statistics for *Mytilus galloprovincialis* (MG) datasets for HgT at each station (unit in µg/Kg dw).

Parameter	Station	Year	N	Min	Max	Avg	Median	Std.Dev	Variance Ψ <sup>2</sup> (LogC)
Hg	TM	1999	1	-	-	110.0	-	-	-
		2000	2	190	235	212.5	212.5	31.8	0.004
		2001	3	63	82	71.6	70.0	9.6	0.003
		2002	4	133	139	136.0	136.0	2.4	0.000
		2003	2	104	113	108.5	108.5	6.4	0.001
		2004	5	100	170	132.2	132.0	26.2	0.007
		2005	3	98	110	102.6	100.0	6.4	0.001
		2006	3	100	121	112.0	115.0	10.8	0.002
		2007	5	104	126	118.2	121.0	8.3	0.001
		2008	5	110	137	127.0	128.0	10.7	0.001
		2009	5	65	83	73.4	70.0	8.1	0.002
		2010	5	118	135	128.6	131.2	6.9	0.001
		2011	5	151	205	176.8	170.0	21.5	0.003
2012	5	27	31	28.8	28.0	2.0	0.001		
Hg	24	1999	1	-	-	120.0	-	-	-
		2000	4	104	134	117.7	116.5	12.3	0.002
		2001	1	-	-	85.0	-	-	-
		2002	1	-	-	102.0	-	-	-
		2003	4	134	152	140.0	137.0	8.2	0.001
		2004	5	69	139	115.4	131.0	29.3	0.016
		2005	2	100	110	105.0	105.0	7.1	0.001
		2006	3	101	110	106.3	108.0	4.7	0.000
		2007	5	140	159	152.0	156.0	7.9	0.001
		2008	5	133	192	157.0	153.0	22.7	0.004
		2009	5	57	98	85.4	89.0	16.4	0.009
		2010	5	90	106	98.4	99.2	6.7	0.001
		2011	5	84	105	93.8	94.0	8.3	0.001
2012	5	11	19	16.2	17	3.3	0.010		

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev (standard deviation statistic), Variance (variance statistic).

#### Organic compounds in sediments

Slovenia provided data for polycyclic aromatic hydrocarbons (PAH) in sediments for 3 more years from 2010 to 2015, although the majority of data was for 2010, 2011 and 2012. However, data levels were reported as below detection limits (BDLs) at all the stations (000F, 000K, 0014, 00CZ, 00KK, 00MA and 00MP). The reported detection limit for petroleum hydrocarbons is 1 ppm (probably determined by spectrometric analytical methods). To note, however, that is also reported 1ppb for earlier data generated for PAH, despite the analytical results being consistent with the latest datasets. Therefore, it was expected to evaluate trends and uncertainty, although no replicate samples are collected, but at present it is not possible. The fact that the latest datasets do not show detectable concentrations of PAH (<BDL) should be confirmed by using other analytical methods, as well as determining individual congeners (by spectrofluorimetry or mass spectrometry, if possible). Therefore, it is not possible to evaluate the environmental impact of PAH contamination in this report.

#### 4.6. Tunisia

Datasets for 2010 and 2011 were available in the MEDOPL database for biota (few species, MG, ML and RD) and sediments. However, these datasets were not entirely useful, as the sampling strategy seems to have been directed towards the RD species and just few stations were available for MG. Thus, no further continued time series analysis can be done for this species. On the other hand, the sediment analytical results presents a gross units mismatch between 2010 and 2011, and no further attempts for data assessment can be made until revised.

##### Trace metals in *Mytilus galloprovincialis* (MG)

Only two new samples were available for MG in 2010. Further, these samples report the same value, although different coordinates are described (only one coincides with station B3 – Lagoon of Bizerte) and the analytical result is reported without replicates (from a pooled sample with 20 individuals). The levels of HgT and Pb were determined 48 and 515 µg/Kg dry weight, respectively, in these samples. Both values are lower than the latest in 2008. Cadmium values were reported as zero value. No further data analysis or trends can be studied. It should be confirmed if the temporal series have been stopped for MG within the marine monitoring strategy in Tunisia.

##### Trace metals in *Ruditapes decussatus* (RD)

New datasets up to 2011 for the bivalve species *Ruditapes decussatus* (RD) were submitted for 4 of the 5 regularly sampled stations in Tunisia (B3, G1, S2 and T2, with the exception of M1). These new datasets for Cd, HgT and Pb, provides further information to perform evaluations for both for levels and temporal trends. The Pb levels and trends in RD are presented in this report for first time. Additionally, at the station B3, analytical results were reported for another species coded ML, but which no corresponds with the MEDPOL codes (the samples were collected at two different geographical locations in the Lagoon of Bizerte). The Table Tun.1. summarizes the major statistics for trace metals in RD along the Tunisian coast .

Table Tun.1. Summary of statistics for trace metals in RD (all years and stations, µg/Kg dw) in the Tunisian coast (2001-2011).

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
Cd	123	313	289	178	218	396	22	984
HgT	125	132	86	170	44	213	9	492
Pb	121	445	372	302	263	565	86	1087

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).

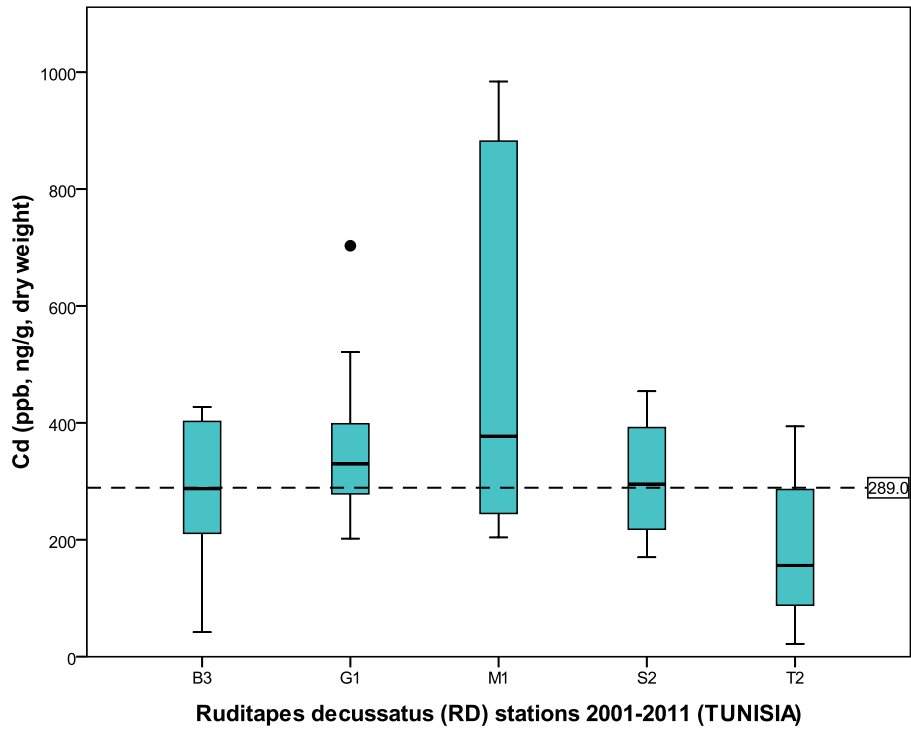


Figure Tun.1. Cadmium box-plots for all the years in the MEDPOL database at the Tunisian stations with updated datasets for 2010 and 2011. Station M1 is not updated. The dashed line represents the median.

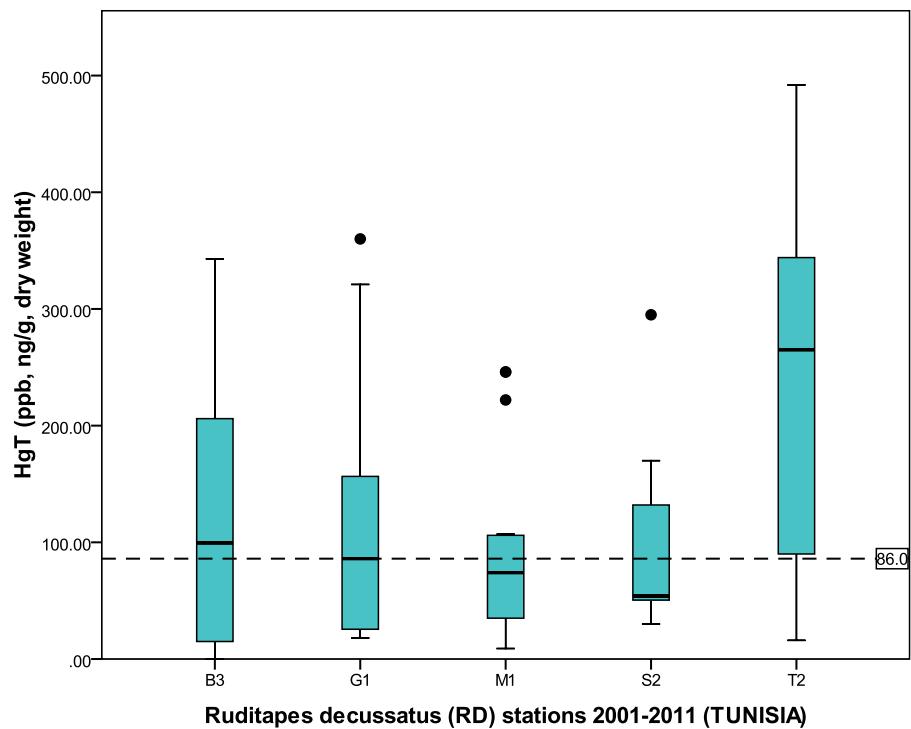


Figure Tun.2. Total mercury box-plots for all the years in the MEDPOL database at the Tunisian stations with updated datasets for 2010 and 2011. Station M1 is not updated. The dashed line represents the median.

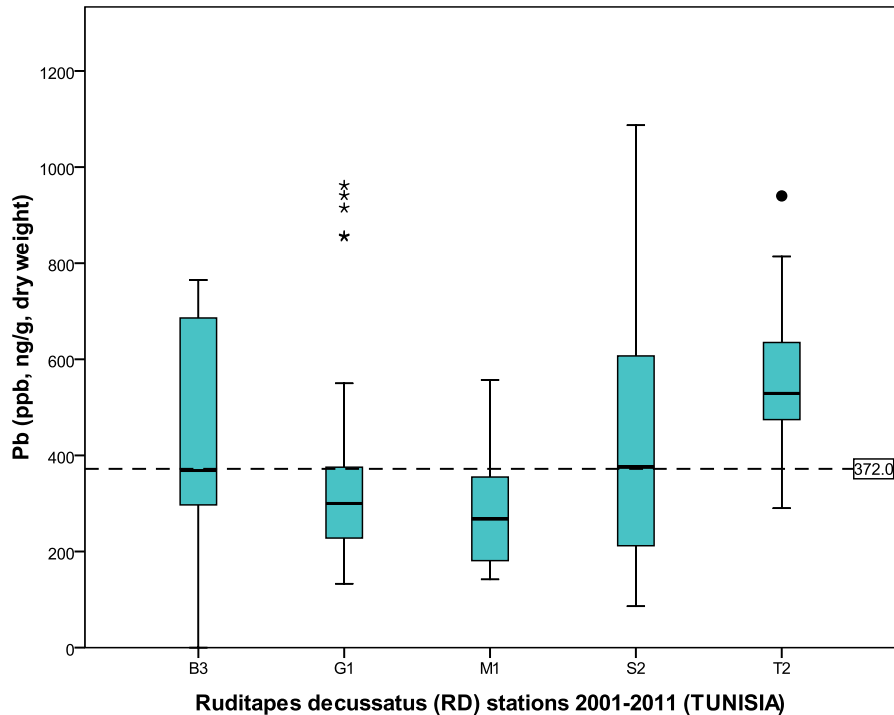
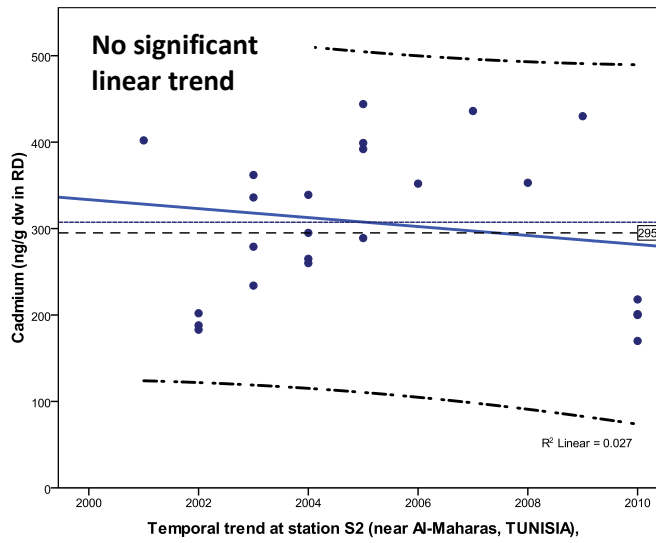
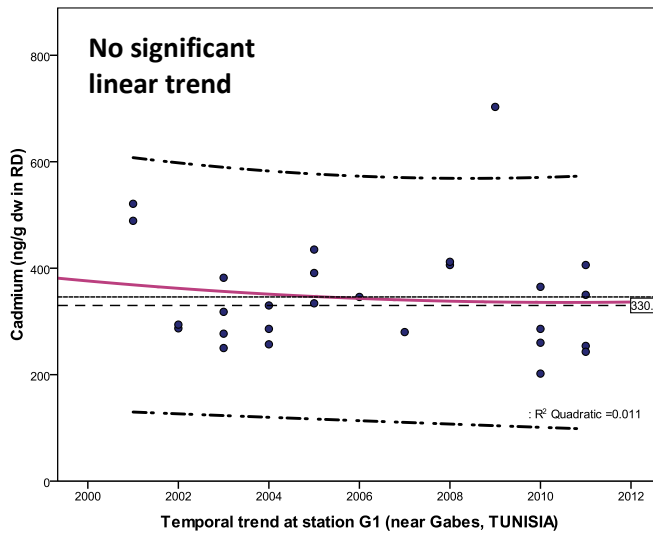
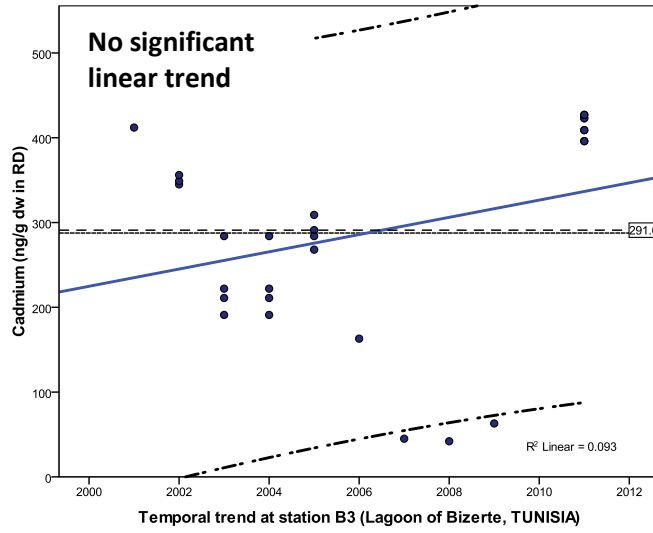


Figure Tun.3. Lead box-plots for all the years in the MEDPOL database at the Tunisian stations with updated datasets for 2010 and 2011. Station M1 is not updated. The dashed line represents the median.

The robust non-parametric Kruskal-Wallis test (independent samples, Chi-square statistic,  $\alpha=0.05$ ) was applied to elucidate if there were significant differences between the stations and it was significant for all the trace elements (Cd, HgT and Pb), although these areas were slightly unequally weight in terms of data measurements (N). Further, Mann-Whitney tests (U statistics,  $\alpha=0.05$ , 2-tailed) were performed and significant differences were found mainly for Cd, HgT and Pb at the station T2. The Cd median at T2 is almost 3-fold lower compared to the overall datasets medians (Figure Tun.1.), which might respond to some depletion process at this station (Lagoon of Tunisia, near La Goleta), rather than a real difference in inputs. On the contrary, HgT and Pb at T2 exceed the overall median and the differences with the levels observed with the rest of stations are statistically significant (Figures Tun .2. and Tun.3.). In the case of Pb, the stations G1 and M1, both located in the Gulf of Gabes, are also differentiated from the rest of stations exhibiting lower levels in this area.

#### *Temporal trends*

The extended time series including the new datasets available in the MEDPOL database show the same trends as previously assessed (Figures Tun.4.-Tun.6.), except at the Lagoon of Bizerte (station B3), where higher values of Cd were observed in 2011 making the whole trend interpretation at this station to shift from a downward to an upward trend for this element, although not statistically significant (Table Tun.2.). However, some caution is necessary to interpret high and low end points in the time series, and further datasets will confirm the sign of the trends. A significant downward trend for Cd was also found at station T2.



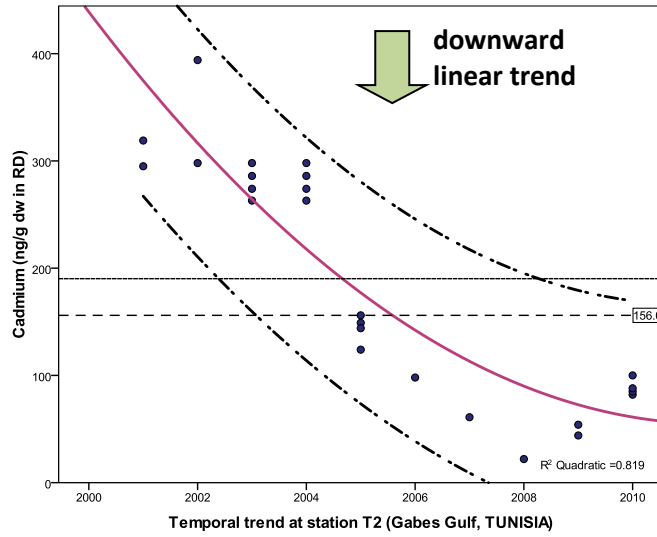
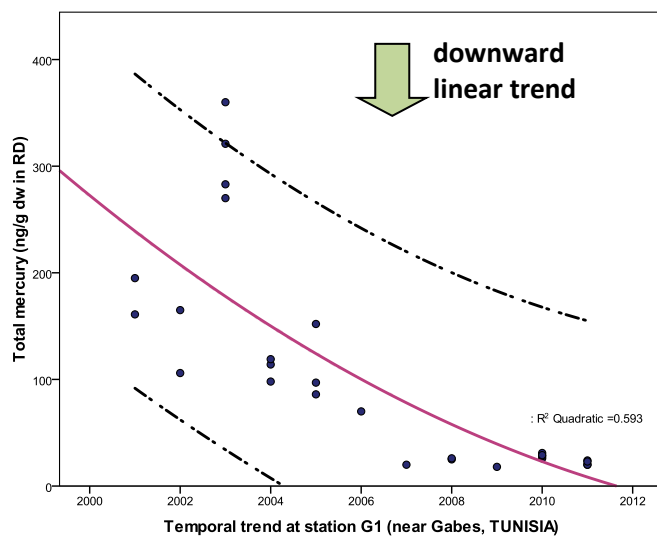
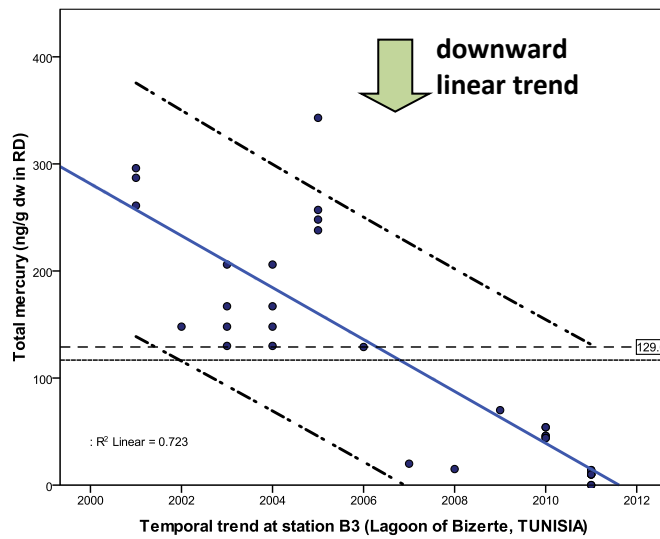


Figure Tun.4. Cadmium temporal trends (2001-2011) at the Tunisian stations. (from top to bottom: linear fit at B3, quadratic fit at G1, linear fit at S2 and quadratic fit at T2; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).



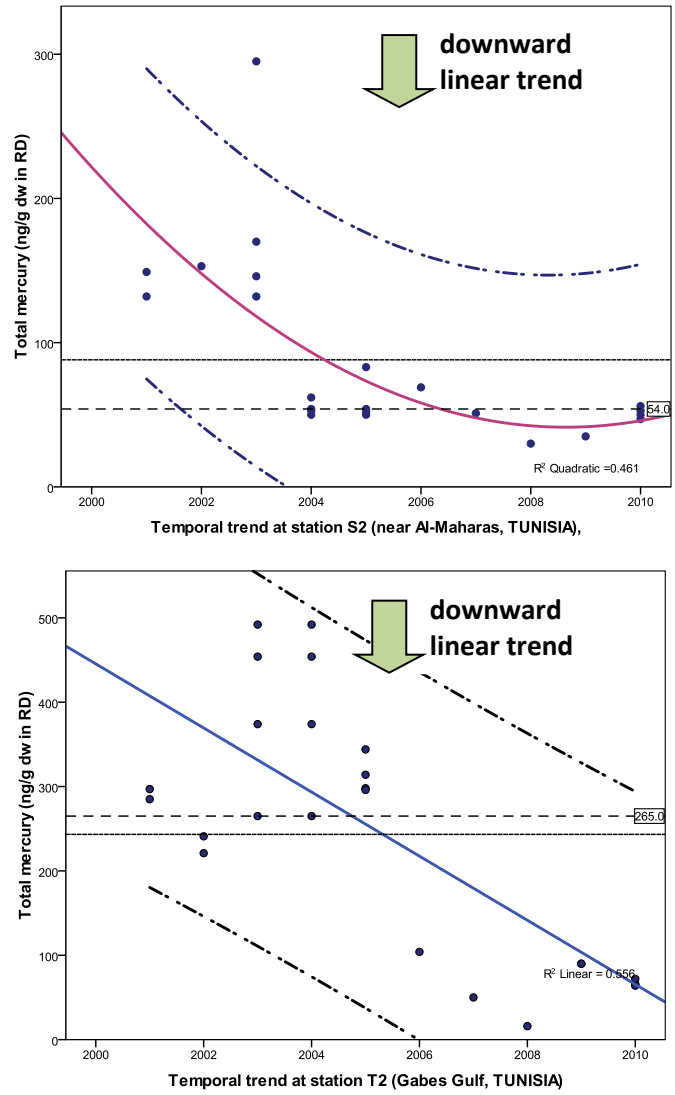
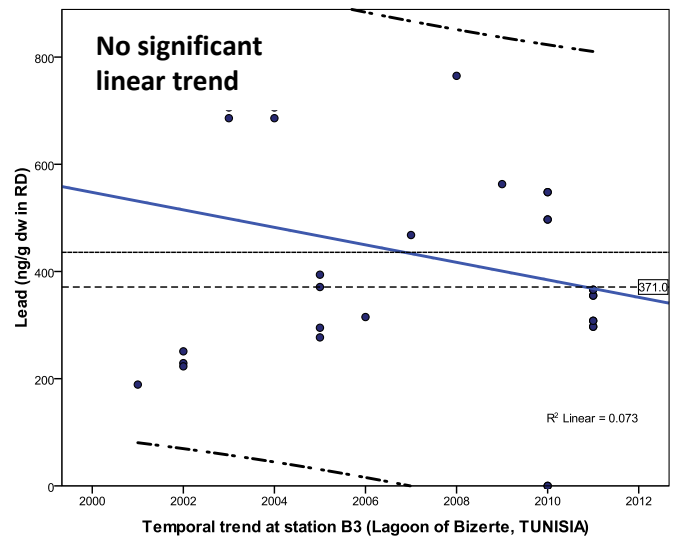


Figure Tun.5. Total mercury temporal trends (2001-2011) at the Tunisian stations. (from top to bottom: linear fit at B3, quadratic fit at G1, quadratic fit at S2 and linear fit at T2; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).



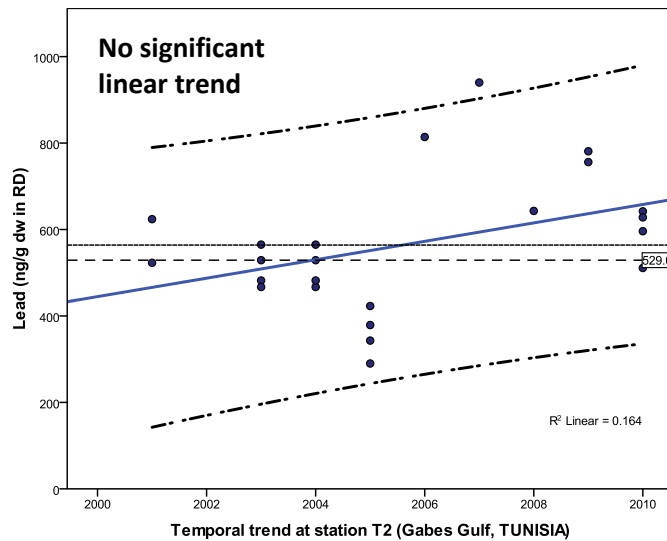
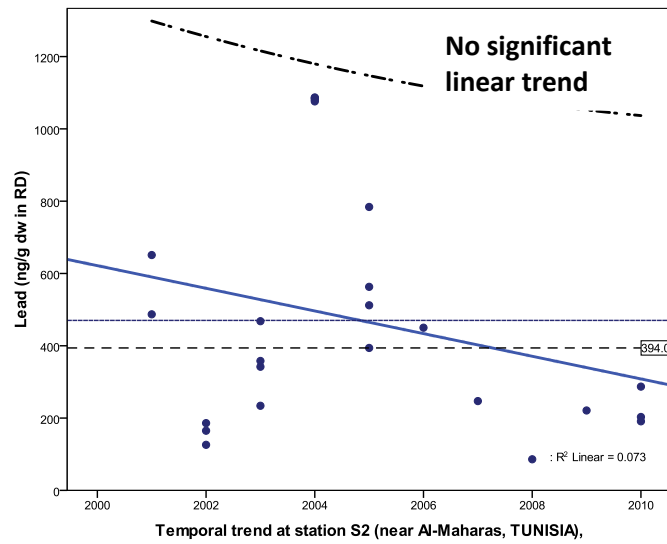
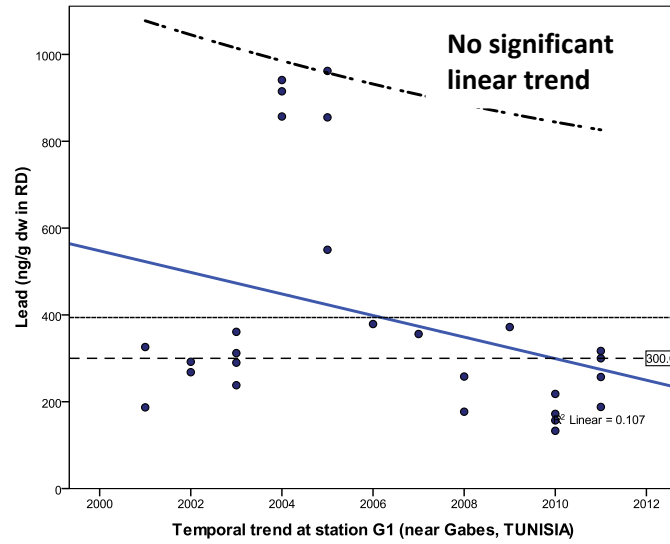


Figure Tun.6. Lead temporal trends (2001-2011) at the Tunisian stations. (from top to bottom: linear fit at B3, G1, S2 and T2 stations; uncertainty bands are fitted to individual measurements; dotted line=mean, dashed line=median).



Further, stations G1 and S2 which have been evaluated in the present report show significant statistical downward trends for HgT, similarly to stations B3 and T2 as previously reported. The Pb temporal trends at all the stations do not show either significant downward or upward trends, despite the most recent datasets fulfill with the within year variance threshold (Table Tun.3.), thus the between year variance might lead to large uncertainty bands which impedes a significant statistical conclusion (Figure Tun.6. and Table Tun.2.).

Table Tun.2. Summary of  $\tau$ -Kendall B correlation coefficients at the 4 stations with data updated for 2010 and 2011.

Station	Cd	HgT	Pb
B3	0.175	-0.702**	-0.151
G1	-0.095	-0.683**	-0.202
S2	-0.007	-0.556**	-0.125
T2	-0.731**	-0.462**	0.172

\*\*Correlation is significant at the 0.01 level (2-tailed); \*Correlation is significant at the 0.05 level (2-tailed)

Table Tun.3. Calculated within-year variances for the timeseries and summary of statistics for *Ruditapes decussatus* (RD) datasets at each station (unit in  $\mu\text{g}/\text{Kg dw}$ ).

Station	Year	Parameter	N	Min	Max	Mean	Median	Std.Dev.	Variance $\Psi^2(\text{LogC})$
B3	2010	Cd	-	-	-	-	-	-	-
		HgT	6	44	54	48	46	5	0.002
		Pb	4	497	548	522	522	29	0.001
B3	2011	Cd	8	396	427	413	416	13	0.000
		HgT	6	10	14	11	10	2	0.006
		Pb	8	297	366	331	331	31	0.002
G1	2010	Cd	4	202	365	278	273	67	0.011
		HgT	4	26	31	28	28	2	0.001
		Pb	4	133	218	170	164	35	0.008
	2011	Cd	4	243	406	313	302	78	0.012
		HgT	4	20	24	21	21	2	0.002
		Pb	4	188	317	265	278	57	0.010
S2	2010	Cd	4	170	218	197	200	19	0.002
		HgT	4	47	56	51	51	4	0.001
		Pb	3	191	287	227	203	52	0.009
T2	2010	Cd	4	82	100	88	86	8	0.001
		HgT	4	64	72	66	65	4	0.001
		Pb	4	511	642	594	612	58	0.002

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev. (standard deviation statistic), Variance (variance statistic).

### Trace metals in sediment

Tunisia updated the datasets for the trace metals in sediment for the years 2011 and 2012 at the stations B3, CHOU and S3A (the latest considered hotspot stations). However, there might be a gross mismatch between the Cd units for 2011 and 2012. The reason is that the levels reported for the same station (with exact geographical coordinates) varies over 4 orders of magnitude, which is almost environmentally impossible. Further, HgT and Pb levels seem out of range as well, and were only reported for 2012 and for a single sample with no replicates. These data should be carefully revised as well as the submissions to the MEDPOL database. No attempts for data correction or evaluation could be made in the present report.

### **4.7. Turkey**

New datasets for 2011 and 2013 were available in the MEDPOL database. In 2011, 27 new stations classified as coastal stations were sampled for biota. The samples were collected for 4 biota species, 3 fish species *Mullus barbatus* (MB), *Mullus surmuletus* (MS) and *Upeneus moluccensis* (UM), as well as the bivalve *Mytilus galloprovincialis* (MG). The data for chemical contaminants was available to statistically evaluate metal contamination (HgT, Pb and Zn), although organochlorine compounds and petroleum hydrocarbons datasets in this species were almost all, reported as BDLs. Turkey submitted also new sediment datasets for 2011 and 2013, within a monitoring strategy which included from 1 to 3 stations per area (in total 89 stations in 2011 and 60 stations in 2013) and tentative reference sites (with single, duplicate or triplicate samples at each station). In total 48 areas were available for trace metal evaluation in 2011 in sediments for HgT, Pb and Zn, increasing substantially the datasets submitted earlier to the MEDPOL database (from 25 to 48 areas since 1999).

Nevertheless, it should be noticed that the historical time series for *Mullus barbatus* (MB) at the stations of Goksu, Tirtar and Mersin seems to have been stopped, as well as the time series for *Mytilus galloprovincialis* (MG) at Izmir Bay, started since 2000. Further, only few sediment stations have been continued to be sampled within new the large spatial sampling undertaken in 2011 (and these have been renamed accordingly).

However, because no further datasets for the historical locations have been submitted for either 2010, 2011 or 2013, the temporal trends in biota cannot be performed. A spatial exploratory statistical analysis for these largest 2011 datasets, both for biota and sediments, is considered in the present report and the statistical information provided up to 2013.

### Trace metals in *Mullus barbatus* (MB)

A part from the new 18 station datasets submitted for MB, also new samples for *Mytilus galloprovincialis* (MG) from 2 new locations at Izmir Bay were submitted (one for metals in 2011). Further, at 7 locations, the samples collected were from other species, such as *Upeneus moluccensis* (UM), and other non-identified species by MEDPOL codes (UP, UB), which would need to be confirmed.

These new dataset for MB covered the entire coast and gives a first spatial view in both the Aegean Sea coast (12 stations: ALTMB, BMRMB, BODMB, CABMB, DATMB, GEDMB, GOBMB, GULMB, ILBMB, KMRMB, MESMB and SABMB) and the Levantine Sea coast (6 stations: ALBMB, ANBMB, FETMB, GREMB, KARMB and TIRMB) in the Mediterranean Sea. The HgT, Pb and Zn were determined in fillet tissue (for pooled samples between 15 and 30 individuals in general. The Table Tur.1. summarizes the main statistics for the 2011 datasets in MB. In 2013, 6 more stations were sampled for MB and not other species (ANAMB, DALMB, DIBMB, FIBMB, MARMB, SAMMB, YUMMB) and a statistical summary for HgT,

Pb and Zinc is presented in Table Tur.1a. ). It should be noticed that the Pb concentrations for MB in 2011 seems too elevated and should be confirmed (Table Tur.1 and Figure Tur.2).

Table Tur.1. Summary of statistics for trace metals ( $\mu\text{g}/\text{Kg}$  fw) in MB from Turkey in 2011.

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
HgT	44	163	148	215	51	266	24	654
*Pb	37	4012	3954	4142	1634	5776	279	9100
Zn	44	4510	4256	1238	3717	4955	2340	11760

\*concentrations to be revised

Table Tur.1a. Summary of statistics for trace metals ( $\mu\text{g}/\text{Kg}$  fw) in MB from Turkish sampled stations in 2013.

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
HgT	75	56	59	27	43	70	12	94
Pb	73	65	50	60	30	90	10	220
Zn	75	6364	6100	2005	5330	7335	4160	9210

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset. \*to be revised)

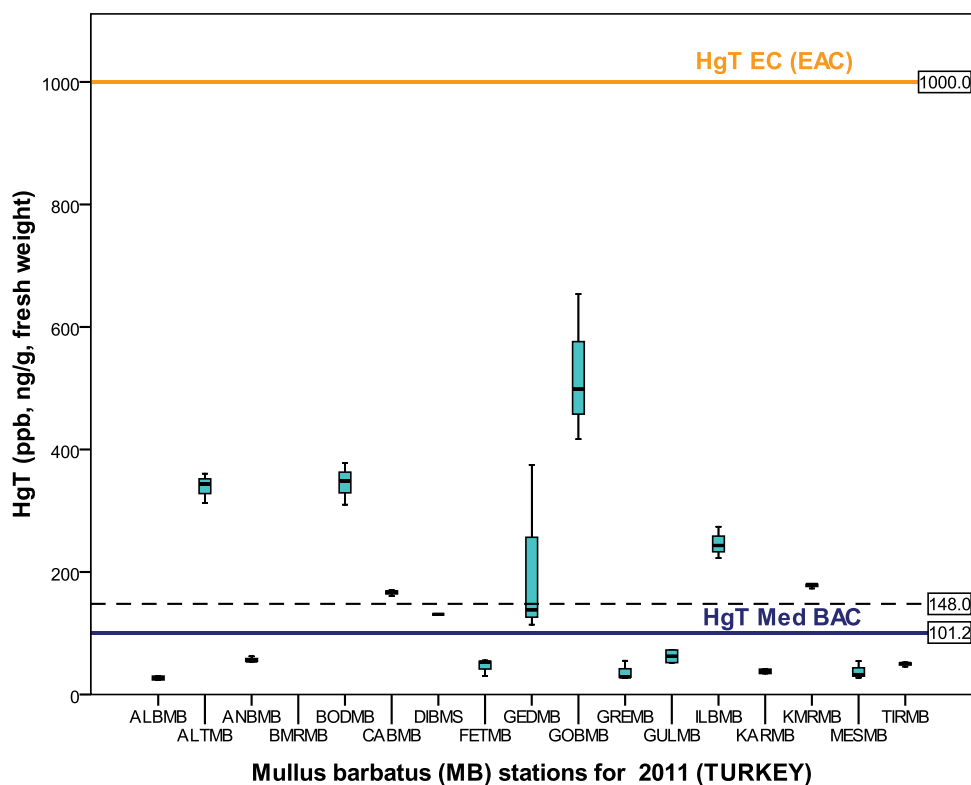


Figure Tur.1. Total mercury box-plot for 2011 datasets in the MEDPOL database in the Turkish coast. The dashed line represents the median.

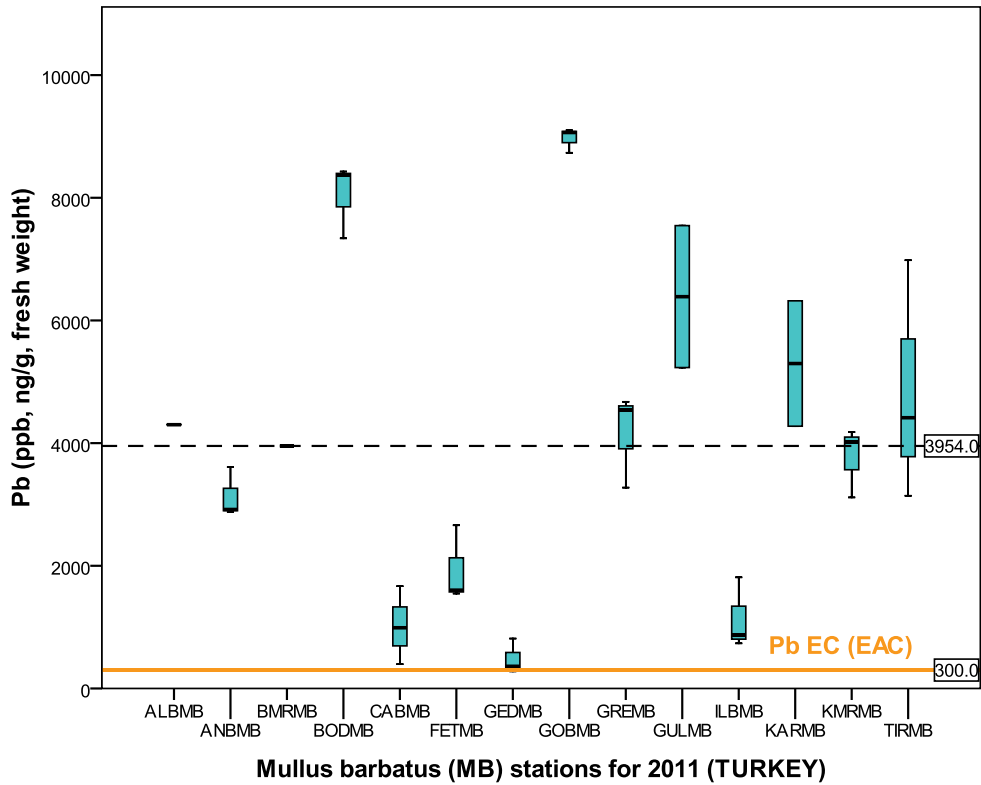


Figure Tur.2. Lead box-plot for 2011 datasets in the MEDPOL database in the Turkish coast. The dashed line represents the median. (Levels to be revised; see text)

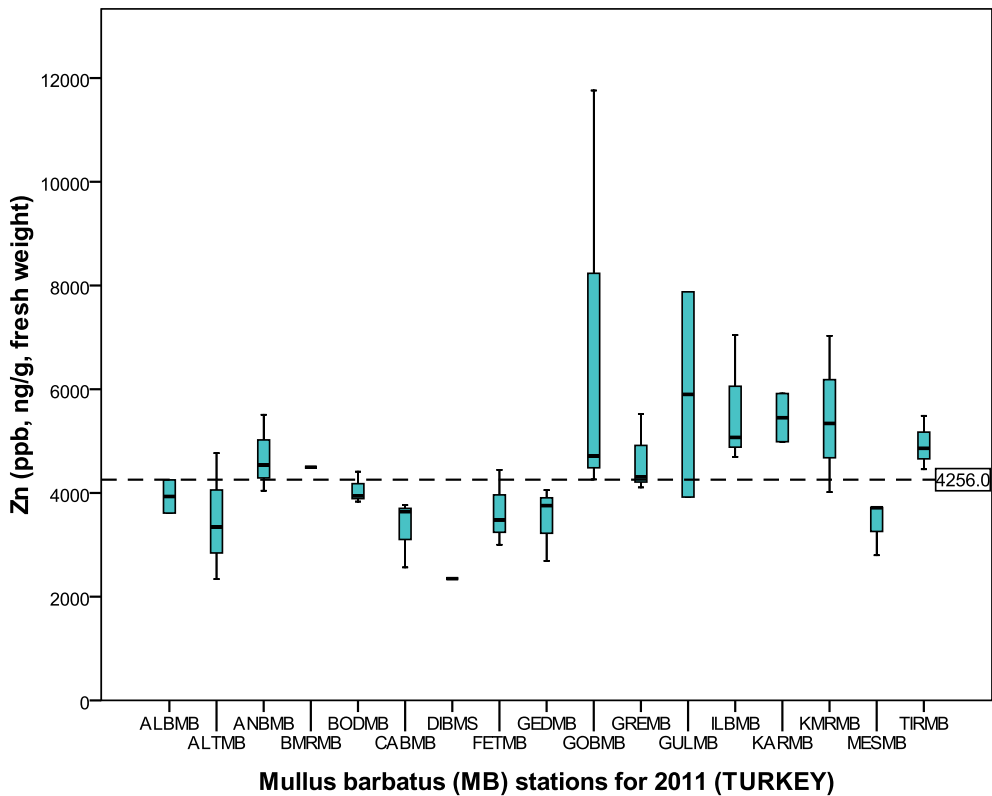


Figure Tur.3. Zinc box-plot for 2011 datasets in the MEDPOL database in the Turkish coast. The dashed line represents the median.

The robust non-parametric Kruskal-Wallis test (independent samples, Chi-square statistic,  $\alpha=0.05$ ) was applied to elucidate if there were significant differences between the stations for each metal and it was significant for all (HgT, Pb and Zn). It can be further observed that the stations with the lowest values for HgT are located in the Levantine Sea (open Mediterranean Sea) as clearly shown when grouping all the stations (Figure Tur.4.). The Mann-Whitney test (U statistics,  $\alpha=0.05$ , 2-tailed), further confirmed the statistical differences between the HgT levels determined in MB sampled in both seas. As mentioned, Pb levels for the sampled stations in 2011 might have some analytical troubleshooting and should be revised, thus no attempt to contrast both sub-regions has been made.

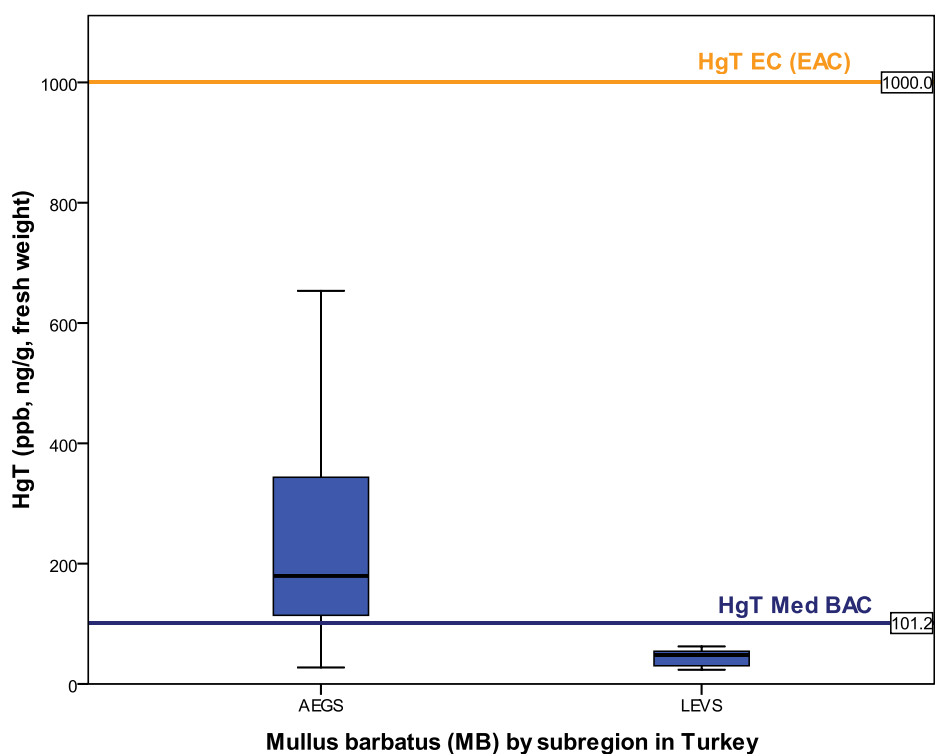


Figure Tur.4. Total mercury box-plots for 2011 datasets in the MEDPOL database in the Turkish coast grouped by sub-region (Aegean Sea and Levantine Sea coasts).

#### Trace metals in sediment

Turkey included also new sediment areas in 2011 and submitted datasets correspondingly, for 89 stations in 48 areas. In 2013, the datasets are reported for 60 stations within the ones sampled in 2011. The present report evaluates HgT, Pb and Zn. Sampling was performed by taking from 1 to 3 replicate samples per station, including tentative reference sites. These samples were reported to be analyzed as a whole sample without sieving, although in earlier datasets the 63  $\mu\text{m}$  fractions were considered. An exploratory analysis of trace metal levels by grouping stations (by main areas) are show in the figures below (Figures Tur.5.-Tur.7.) for the largest 2011 datasets and information provided in tables.

Tables Tur.2 and Tur2a below, summarizes the main statistics for the 2011 and 2013 sediment datasets for HgT, Pb and Zn (in  $\mu\text{g}/\text{Kg}$  dw) in the Turkish coasts. For other chemical contaminants, such as organochlorine compounds and petroleum hydrocarbons, the majority of

the analytical results were not reported or reported below detection limits (BDLs). Further, the sediment datasets reported THC (Total Hydrocarbons Content), but both the units and the analytical methodology were not clear, as was the case also for measurements of TOC, which will need to be revised. These datasets constitute the first step towards the establishment of a long-term monitoring program for sediment samples, although a refinement for cost-effective monitoring might be needed.

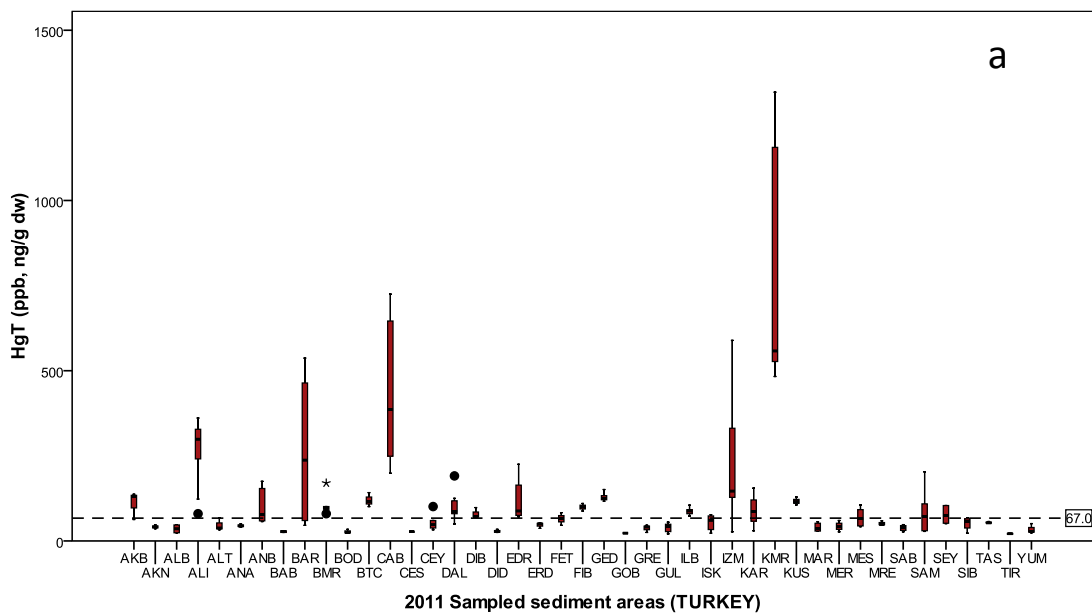
Table Tur.1. Summary of statistics for trace metals in sediments by areas ( $\mu\text{g}/\text{Kg dw}$ ) in Turkish coast in 2011.

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
HgT	217	119	67	75	42	116	20	1318
Pb	240	11488	8456	7891	5532	13422	189	78480
Zn	258	32062	27145	24678	16167	40845	4547	147700

Table Tur.1a. Summary of statistics for trace metals in sediments by areas ( $\mu\text{g}/\text{Kg dw}$ ) in Turkish coast in 2013.

Trace metal	N	Mean	Median	IQR	25 <sup>th</sup>	75 <sup>th</sup>	Min	Max
HgT	118	360	310	220	200	420	28	1335
Pb	118	13538	8621	8124	6440	14565	1694	96229
Zn	118	68169	52725	51023	31835	82858	7339	323158

N (number of data), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), IQR (Interquartile range, robust measure of data dispersion assuming a non-Normal distributed dataset), 25<sup>th</sup> percentile and 75<sup>th</sup> percentile (values of the top and bottom of the box equivalent to Q1 and Q3 quartiles, respectively), Min. (minimum value in the dataset) and Max. (maximum value in the dataset).



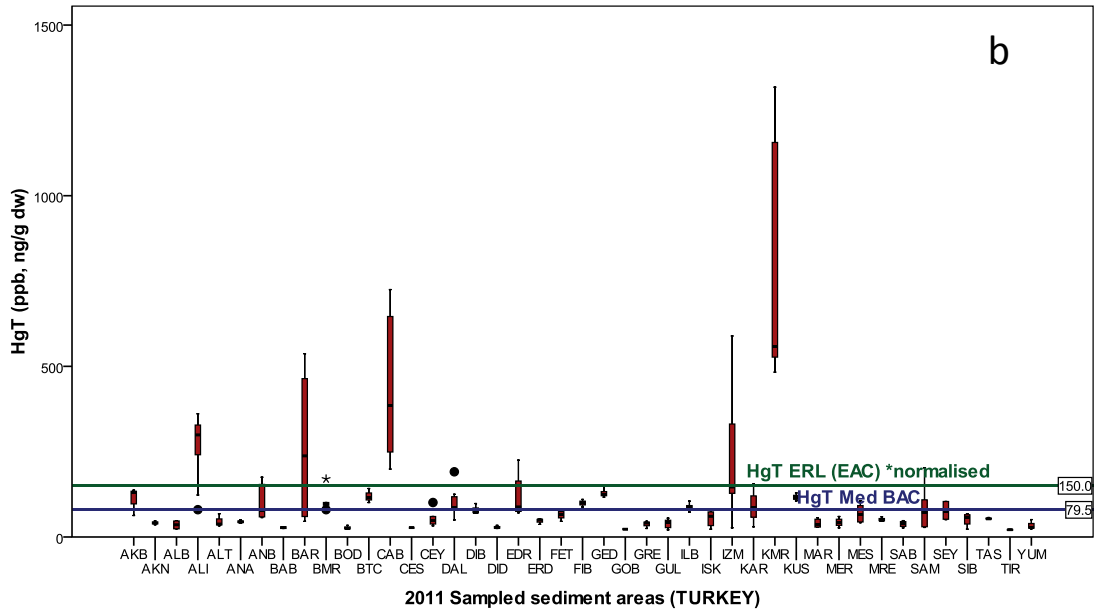
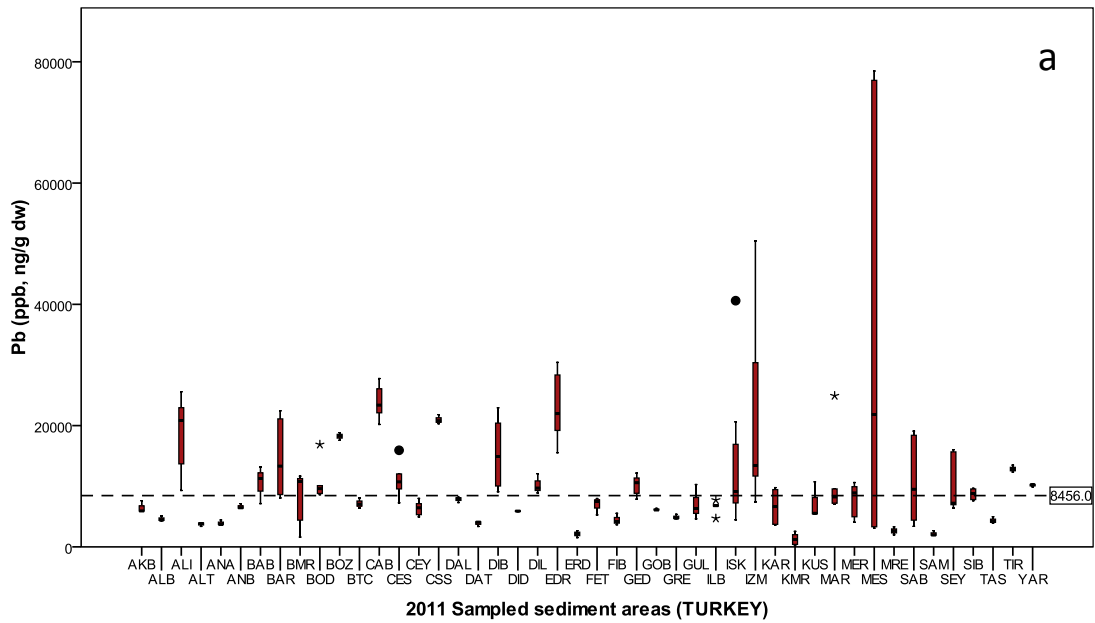


Figure Tur.5a. and 5b. Total mercury box-plots for 2011 sediment datasets in the MEDPOL database in the Turkish coast grouped by area. The dashed line represents the median (a) and (b) shows the Med BAC and ERL thresholds.



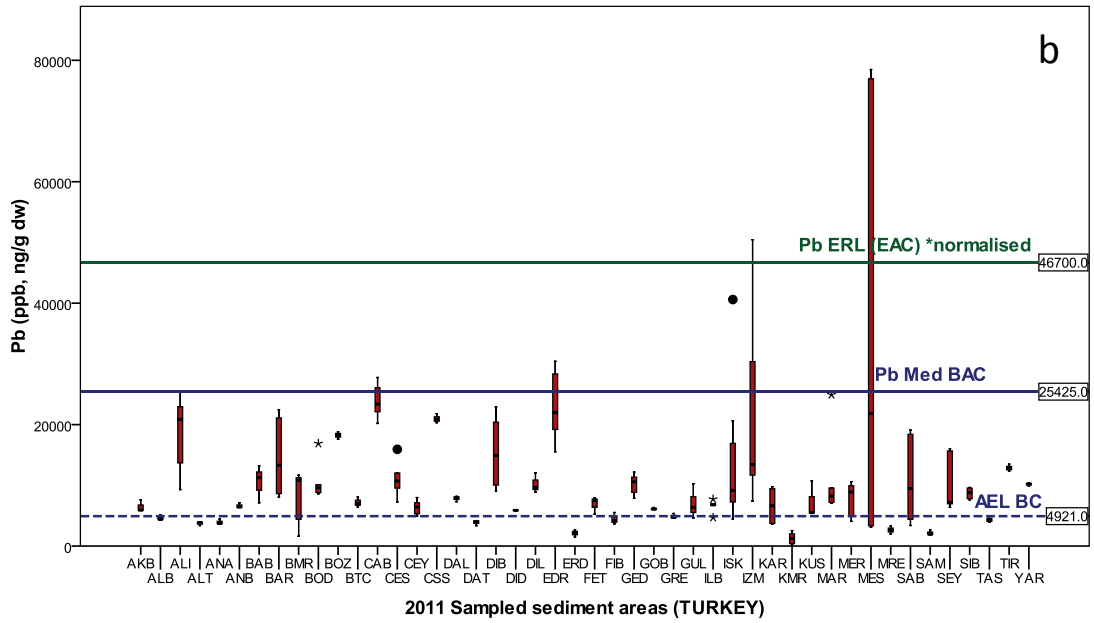


Figure Tur.6a and 6b. Lead box-plot for 2011 sediment datasets in the MEDPOL database in the Turkish coast grouped by area. The dashed line represents the median (a) and (b) shows the Med BAC and ERL thresholds.

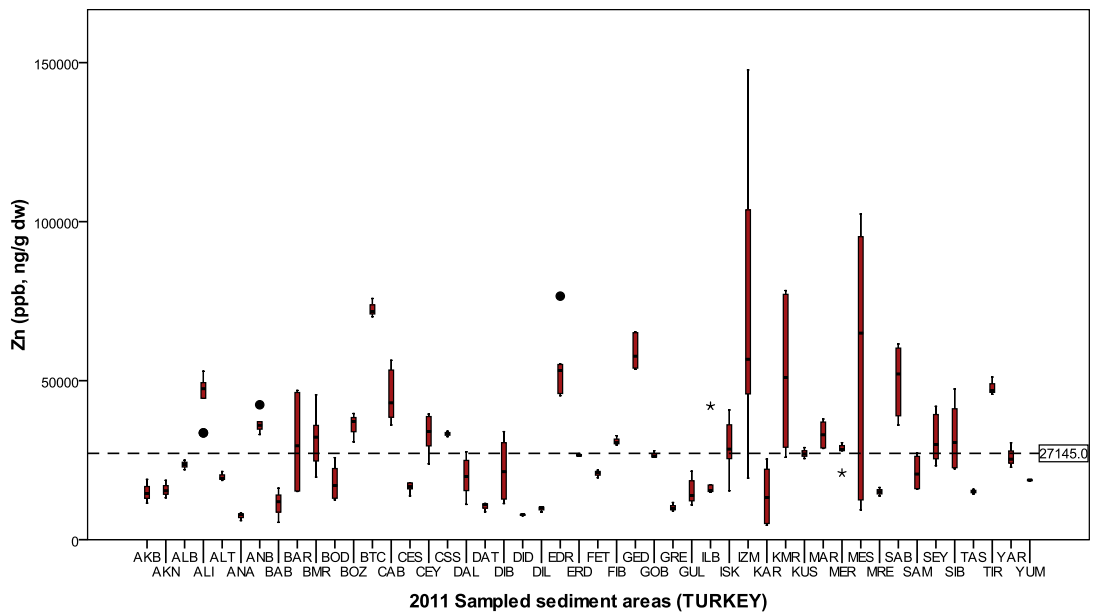


Figure Tur.7. Zinc box-plot for 2011 sediment datasets in the MEDPOL database in the Turkish coast grouped by area. The dashed line represents the median.

A close look, indicates some higher levels for HgT and Pb for some stations, at the same time as the majority of them has low levels (under the overall median), with some stations, clearly, being potential reference stations (the lowest box-plots). For Zn, the data show a large variability for the entire dataset, but similarly, stations could be classified as potentially hotspots, coastal and reference areas.



The Figures Tur.8 – Tur.10 below show the HgT, Pb and Zn datasets grouped by sub-regions in the Eastern Mediterranean Sea. The Mann-Whitney test (U statistics,  $\alpha=0.05$ , 2-tailed) for two independent samples was not applied in this case as the difference in the number of samples (N) within groups was too large (almost 2-fold), despite the robustness of the non-parametric test in this sense. Nevertheless, it can be observed a majority of extreme values and outliers measured for HgT in the Aegean Sea pointing to potential sources of contamination in the marine environment. Further, the Aegean Sea HgT datasets are above the median and the Med BAC. For Pb and Zn levels in the Aegean Sea, slightly above the median, a lower number of extreme concentrations for these two elements can be observed. With respect the Pb concentration, the median value in sediments is 8490 ( $\mu\text{g}/\text{kg dw}$ ), whilst the previously reported median for the Turkish sediments datasets was 26160 ( $\mu\text{g}/\text{kg dw}$ ) (UNEP(DEPI)MED wg.365/Inf.4). This new value is closer to the medians determined for Israel sediment samples (a closer area in the Eastern Mediterranean), which was 5260 ( $\mu\text{g}/\text{kg dw}$ ) previously, and 5513 ( $\mu\text{g}/\text{kg dw}$ ) in this report. Clearly, an increased and spatially representative number of data (N) in the dataset refines earlier assessments. The samples grouped from the Levantine Sea with a Pb median of 6858 ( $\mu\text{g}/\text{kg dw}$ ) are much closer to the Israel value. To this regard, Pb assessments should take into account the AEL BC (and AEL BAC) in the Eastern Mediterranean Sea basin (Figure Tur.9). Similarly, the median value for HgT (67  $\mu\text{g}/\text{kg dw}$ ) is almost halved taking into account the new sediment stations sampled by Turkey in comparison with the previous value of 140 ( $\mu\text{g}/\text{kg dw}$ ). More, Zn exhibit a median value of 27200 ( $\mu\text{g}/\text{kg dw}$ ), 3-fold lower than previously assessed (90810  $\mu\text{g}/\text{kg dw}$ ) with the datasets before 2010, although the average level is doubled from 2011 to 2013 and the reasons need to be investigated (probably not linked to environmental contamination). As mentioned, these datasets constitute a excellent step towards the establishment of a stable long-term monitoring program for sediment, although a refinement of the sampling stations would be needed after an in-depth statistically assessment.

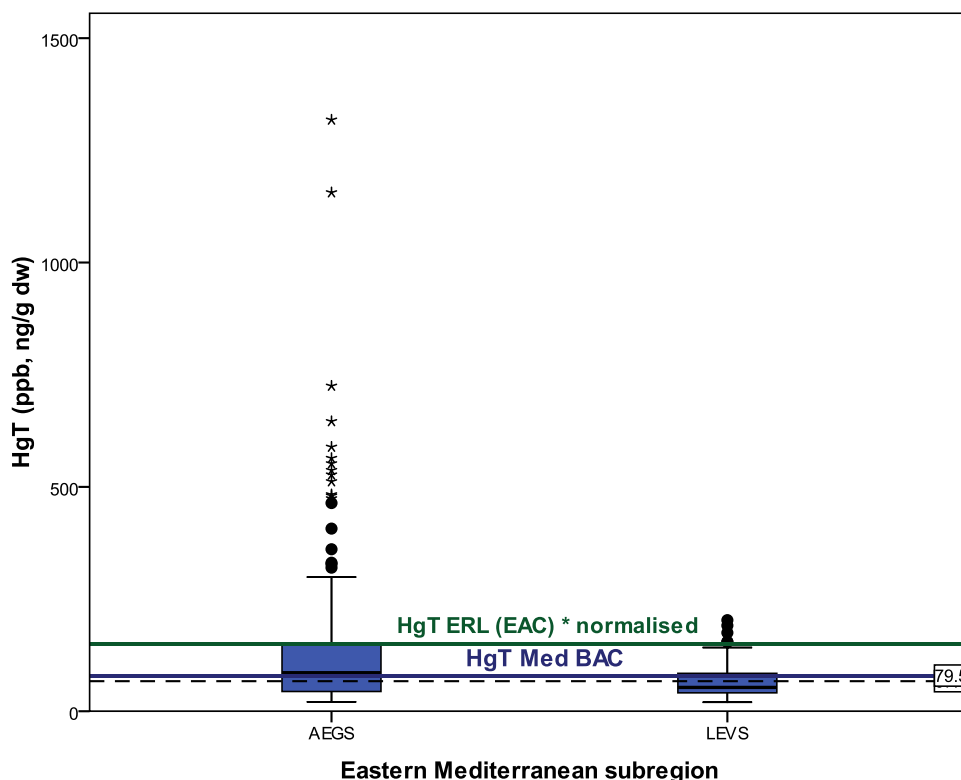


Figure Tur.8. Total mercury box-plots for 2011 datasets in the MEDPOL database in the Turkish coast grouped by subregion (Aegean Sea and Lenvantine Sea coasts).

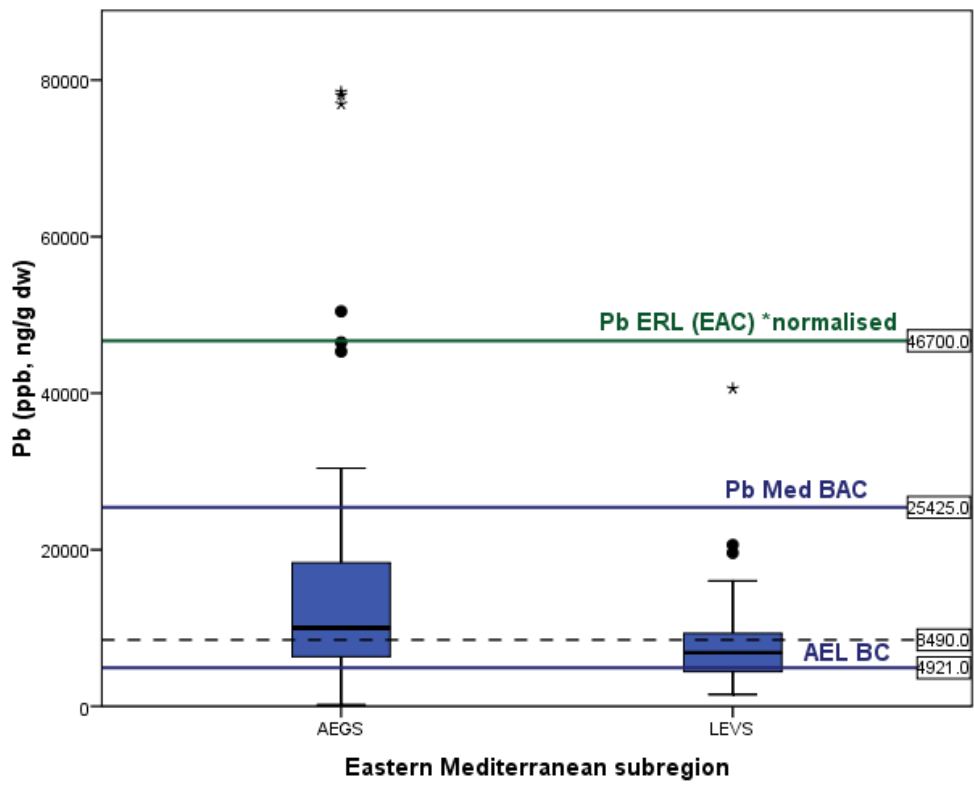


Figure Tur.9. Lead box-plots for 2011 datasets in the MEDPOL database in the Turkish coast grouped by subregion (Aegean Sea and Lenvantine Sea coasts).

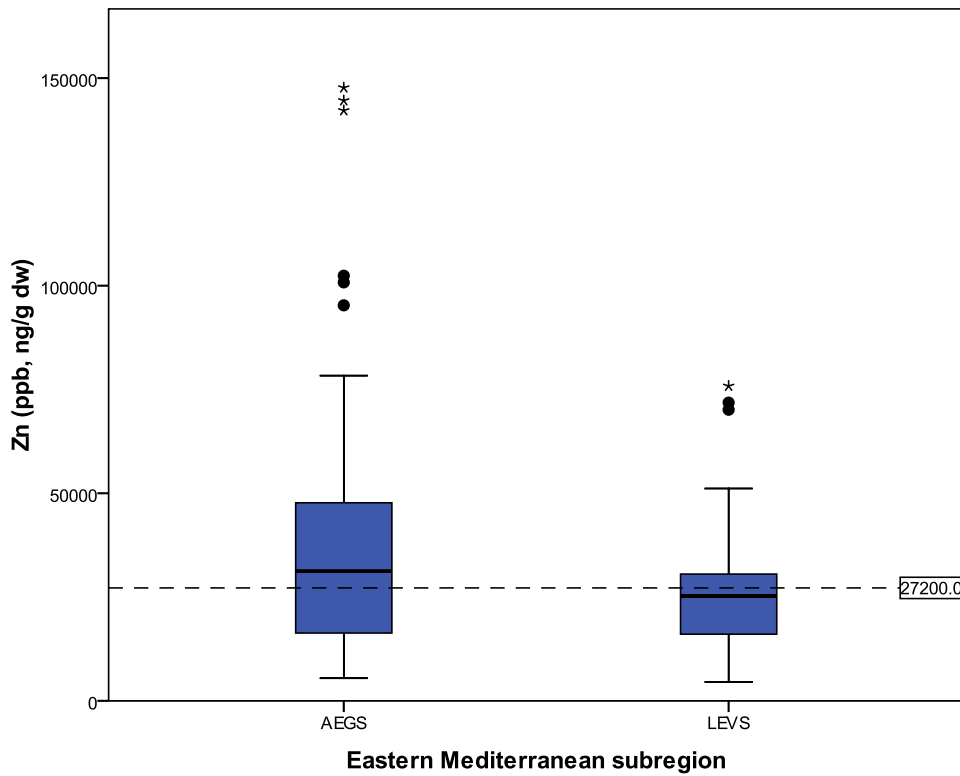


Figure Tur.10. Zinc box-plots for 2011 datasets in the MEDPOL database in the Turkish coast grouped by subregion (Aegean Sea and Lenvantine Sea coasts).

## 5. CONCLUSIONS AND RECOMMENDATIONS

### Monitoring and trend assessments:

1. In general, the monitoring variances comply with the threshold set for the within year variability ( $\sigma^2 < 0.060$ ) for linear trend detection in the MEDPOL monitoring activities. Few exceptions, seems randomly to occur during sampling campaigns for *Mullus barbatus* (MB) samples.
2. Temporal trends have been statistically assessed for different MEDPOL countries taking into consideration the updated datasets up to the end of 2015 and confirmed upward and downward significant trends.
3. A majority of non-reported or below detection limit (<BDL) data for chemical contaminants, within the MEDPOL monitoring database, indicate a contaminant monitoring strategy within MEDPOL monitoring activities far to be optimal.

### Sediments:

4. Sieving methodologies are not yet standardized within the MEDPOL monitoring activities and reported to be from 63 $\mu$ m to 2mm. For hazardous chemical substances (such as trace metals) the sediment fraction size need to be considered to be able to compare datasets at a Mediterranean scales.
5. The sediment monitoring strategies should select sampling stations to study temporal trends (which should include replicate samples in order to calculate the overall within year variability at the selected geographical scale).
6. With respect to hazardous substances and assessment criteria in sediments, at present, it was not possible to perform a detailed study considering data normalization (e.g. Al, Li, TOC), due to insufficient datasets for these parameters in the MEDPOL Database.

### Biota:

7. With respect to hazardous substances in mussel (*Mytilus galloprovincialis*, MG) datasets, some countries seem to have been stopped their long-term monitoring strategies and the reasons should be investigated and discussed.
8. Other species of bivalves are monitored in the Mediterranean and this report presents the first assessments (*Donax trunculus*, DT, *Mactra corralina*, MC) including their temporal trends when possible.
9. With respect to hazardous substances in fish fillet tissue (*Mullus barbatus*, MB) datasets, the environmental concentrations of cadmium and lead are at the low end of the current analytical capabilities and the MEDPOL database show up to 90% of non-detected values. For organic compounds almost the 100% of data are reported as below detections limits. The utility of this target species in both chemical and biomonitoring activities should be revised and alternatives considered.

### Analytical quality assurance:

10. The statistical assessments and calculations have been performed for chemical contaminants, however, the majority of these datasets were not complete and some important information (detection limits, methodologies, etc.) was missing.
11. There is a majority of hazardous chemical substances reported below detection limits (<BDLs). This fact should be further investigated in terms of analytical methodologies by Contracting Parties laboratories.
12. The MEDPOL countries should submit the QA templates on quality assurance for the certified reference materials (sediment and biota) used by MEDPOL and participate in the MEDPOL-IAEA Proficiency testing exercises, along with the monitoring datasets.

## 6. REFERENCES

- Angelidis, M.O., Radakovitch, O., Veron, A., Aloupi, M., Heussner, S., Price, B., 2011. Anthropogenic metal contamination and sapropel imprints in deep Mediterranean sediments. *Marine Pollution Bulletin* 62, 1041–1052.
- Bignert, A., Nyberg E., Asplund L., Eriksson U., Willander A. 2006. Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota. Report to the Swedish Environmental Protection Agency, 2006, 115 pp.
- Gilbert O.R. 1987. *Statistical Methods for Environmental Pollution Monitoring*. Van Nostrand Reinhold Ed., New York.
- Guitart, C., Hernández-del-Valle, A., Marín, J.M., Benedicto, J., 2012. Tracking temporal trend breaks of anthropogenic change in Mussel Watch (MW) databases. *Environmental Science and Technology*, 46, 11515-11523.
- Helsel, D.R., Hirsch., R.M., 1995. *Statistical Methods in Water Resources*, Studies in Environmental Sciences 49. Elsevier, Amsterdam.
- Loftis, J.C., Ward, R.C., Phillips, R.D., 1989. An Evaluation of Trend Detection Techniques for Use in Water Quality Monitoring Programs. EPA/600/3-89/037, 139 pp.
- Long, E.R, McDonald, D.D., Smith, S.L., Calder, F.D., 1995. Incidence of adverse biological effects with ranges of chemical concentrations in marine and estuarine sediments. *Environmental Management*, 19, 81-97.
- Nicholson, M.D., Fryer, R., Larsen, J.R. 1995. A Robust Method for Analysing Contaminant Trend Monitoring Data. *Techniques in Marine Environmental Sciences*. ICES.
- Nicholson M., R. Fryer, C. A. Ross. 1997. Designing Monitoring Programmes for Detecting Temporal Trends in Contaminants in Fish and Shellfish. *Marine Pollution Bulletin*, 34, 821-826.
- Solaun, O., Rodríguez, J.G., Borja, A., González, M., Saiz-Salinas. J., 2013. Biomonitoring of metals under the water framework directive: detecting temporal trends and abrupt changes, in relation to the removal of pollution sources. *Marine Pollution Bulletin*, 67, 26-35.
- UNEP(DEC)/MED WG.243/3. REVIEW OF IMPLEMENTATION OF MED POL PHASE III MONITORING ACTIVITIES. Review Meeting of MED POL - Phase III Monitoring Activities. Saronida (Athens), Greece, 9-11 December 2003.
- UNEP(DEC)/MED WG.273/2. Report of the expert meeting to revise the strategy for trend monitoring of pollutants in coastal water sediments. Expert meeting to revise the strategy for trend monitoring of pollutants in coastal water sediments. Anavissos, Greece, 14-15 April 2005.
- UNEP(DEPI)/MED WG.343/3. REVIEW OF MED POL MARINE MONITORING ACTIVITIES ASSESSMENT OF THE STATE OF ENVIRONMENT AND POLLUTION TRENDS. Consultation Meeting to Review MED POL Monitoring Activities. Rome, 1-2 December 2009.
- UNEP(DEPI)/MED WG.365/Inf.4. HAZARDOUS SUBSTANCES IN THE MEDITERRANEAN:A SPATIAL AND TEMPORAL ASSESSMENT. Consultation Meeting to Review MED POL Monitoring Activities Athens, 22-23 November 2011.

UNEP(DEPI)/MED WG.365/Inf.5. ANALYSIS OF THE TREND MONITORING ACTIVITIES AND DATA FOR THE MED POL PHASE III AND IV (1999-2010). Consultation Meeting to Review MED POL Monitoring Activities. Athens, 22-23 November 2011.

UNEP(DEPI)/MED WG. 365/Inf.8. DEVELOPMENT OF ASSESSMENT CRITERIA FOR HAZARDOUS SUBSTANCES IN THE MEDITERRANEAN. Consultation Meeting to Review MED POL Monitoring Activities. Athens, 22-23 November 2011.

UNEP(DEPI)/MED WG.421/Inf.9. Integrated Monitoring and Assessment Guidance. Agenda item 5.7: Draft Decision on Integrated Monitoring and Assessment Programme (IMAP) of the Mediterranean Sea and Coast and Related Assessment Criteria. Meeting of the MAP Focal Points. Athens, Greece, 13-16 October 2015.

UNEP(DEPI)/MED WG.417/Inf.15. Report of the online groups on eutrophication, contaminants and marine litter. Joint Session MED POL and REMPEC Focal Points Meetings Malta, 17 June 2015.

UNEP(DEPI)/ MED WG.417/Inf.4. Agenda item 7: Report on specific issues (points a and j). Joint Session MED POL and REMPEC Focal Points Meetings. Malta, 17 June 2015.

## **Annex I**

### **Data trend analysis comparison with the PIA statistical software**

## ANNEX I. Data trend analysis comparison with the PIA statistical software

Explanation of trend statistics using PIA©software:

This is a typical PIA statistical suite trend statistics output:

### **Trend statistics:**

Slope = -2.6% (-4.9, -.38)

SD(lr)= .30, 7.0%, 16 yr

Power = .52/.25/11%

y(06) = .27(.18, .41)

r2 = .38, p < .025 \*

tau = -.51, p < .015 \*

SD(sm)= .18, p < .008 \*

Slope = reports the slope, expressed as the yearly percentage change, together with its 95% confidence interval.

SD(lr) = reports the square root of the residual variance around the regression line, as a measure of between-year variation, together with the lowest detectable change in the current time series with a power of 80%, one-sided test, alpha=0.05. The last figure on this line is the estimated number of years required to detect an annual change of 5% with a power of 80%, one-sided test, alpha=0.05.

Power = reports the power to detect a log-linear trend in the time series (Nicholson and Fryer, 1991). The first figure represents the power to detect an annual change of 5% with the number of years in the current time series. The second figure is the power estimated as if the slope were 5% a year and the number of years were ten. The third figure is the lowest detectable change for a ten-year period with the current between-year variation at a power of 80%. This test assumes the time series is based on annual sampling.

y(xx) = reports the concentration estimated from the regression line for the last year in the time series (as indicated by the value of xx), together with a 95% confidence interval. Provided that the regression line is relevant for describing the trend, the residual variance may be more appropriate than the within-year variance in this respect.

r2 = reports the coefficient of determination (r2) together with a p-value for a two-sided test (H0: slope = 0), i.e., a significant value is interpreted as a true change, provided that the assumptions of the regression analysis are fulfilled.

tau = reports Kendall's 'tau', and the corresponding p-value for this non-parametric trend test.

SD(sm) = reports the square root of the residual variance around the smoothed line. The significance of this line could be tested by means of an Analysis of Variance. The p-value is reported for this test. A significant result will indicate a (significant) nonlinear trend component.

**Mercury in mussel *Mytilus galloprovincialis* ( $\mu\text{g}/\text{kg}$  dry weight) at Station 24 - Slovenia**

Data set summary:

Number of values = 51  
Range of years= 1999 - 2012  
Range of data (individuals) = 11.0 - 192  
Number of years = 14  
Range of annual median values= 17.0 - 153  
Overall median value = 106 (89.0, 137 )

Trend statistics:

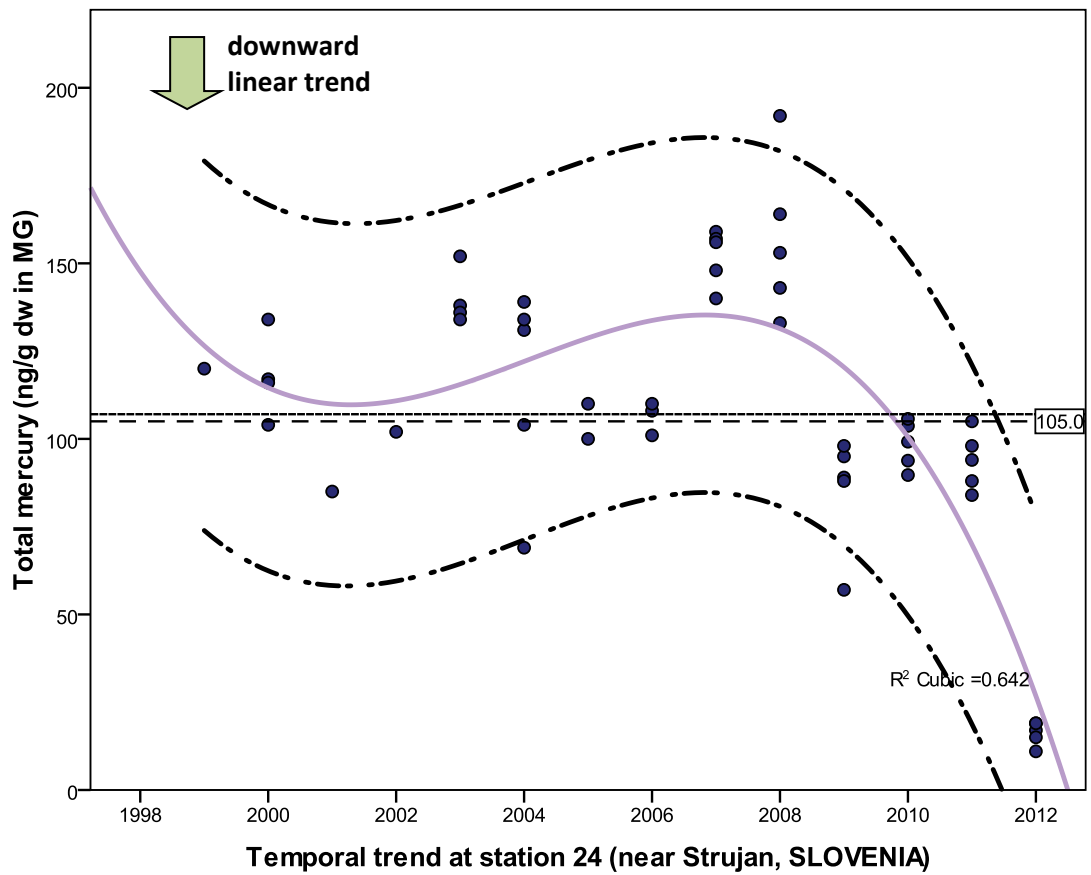
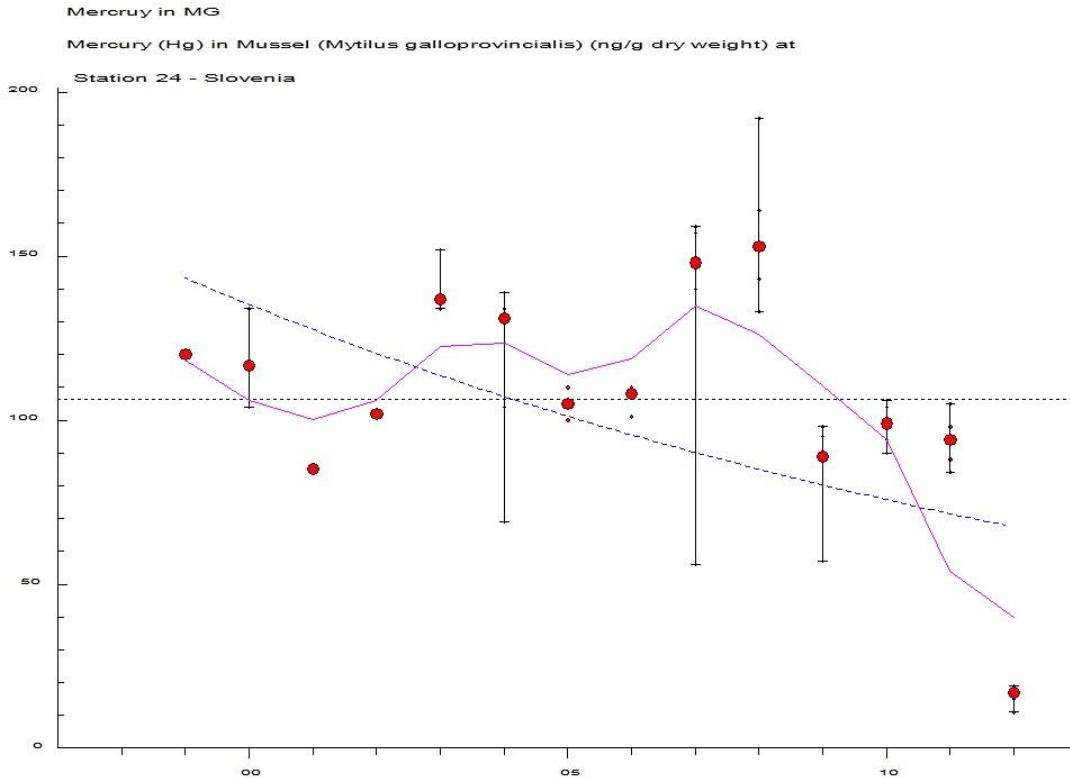
Slope = -5.6% ( -12, 1.4)  
SD(lr)= .50, 13%, 16 yr  
Power = .76/.32/25%  
y(12) = 67 (39, 117)  
r<sup>2</sup> = 0.20, p < 0.102  
tau = -0.21, p < .298  
SD(sm)= .366, p < .035 \*

Interpretation of trend statistics:

The data series show no significant log-linear trend (log-linear regression). The lowest detectable change in the current time series (with a power of 90% and one-sided test, alpha=5 %) is 13 %. 16 years of data are required to detect an annual change of 10% (with a power of 90% and one-sided test, alpha=5 %). The power of the time series to detect a log-linear trend of 10% with the number of years in the current time series is 76%. For a period of 10.0 years, a power of 32% would be expected. The lowest detectable change for a period of 10.0 years with the current between-year variation, at a power of 90% is 25%. The smoother explains significantly more than the log-linear regression line, hence the time series contains non-linear trend components.

The same data evaluated in the present report (see Figures below). The data is fitted to the best regression line (here, in two different regression lines) and plotted with the uncertainty bands (either to the individual measurements or to the mean regression line over the whole time series using a moving average (equivalent to the confidence interval for the mean slope to some extent). Further mean and median values for the datasets are plotted along the time series for reference. Significant tau-Kendall b results are indicated with an arrow both for upward and downward trends (or no trends) and the correlation coefficients and the variances summarized in tables in the text.





**Mercury in mussel *Mytilus galloprovincialis* ( $\mu\text{g}/\text{kg}$  dry weight) at Station TM - Slovenia**

Data set summary:

Number of values = 53  
Range of years= 1999 - 2012  
Range of data (individuals) = 27.0 - 235  
Number of years = 14  
Range of annual median values= 28.0 - 211  
Overall median value = 118 (70.0, 136 )

Trend statistics:

Slope = -4.0% ( -10, 2.8)  
SD(lr)= .47, 12%, 16 yr  
Power = .81/.36/23%  
y(12) = 82 (49, 138)  
r<sup>2</sup> = 0.12, p < 0.216  
tau = -0.07, p < .741  
SD(sm)= .524, p < .871

Interpretation of trend statistics:

The data series show no significant log-linear trend (log-linear regression). The lowest detectable change in the current time series (with a power of 90% and one-sided test, alpha=5 %) is 12 %. 16 years of data are required to detect an annual change of 10% (with a power of 90% and one-sided test, alpha=5 %). The power of the time series to detect a log-linear trend of 10% with the number of years in the current time series is 81%. For a period of 10.0 years, a power of 36% would be expected. The lowest detectable change for a period of 10.0 years with the current between-year variation, at a power of 90% is 23%.

The same data evaluated in the present report (see Figures below). The data is fitted to the best regression line (here, in two different regression lines) and plotted with the uncertainty bands (either to the individual measurements or to the mean regression line over the whole time series using a moving average (equivalent to the confidence interval for the mean slope to some extent). Further mean and median values for the datasets are plotted along the time series for reference. Significant tau-Kendall b results are indicated with an arrow both for upward and downward trends (or no trends) and the correlation coefficients and the variances summarized in tables in the text.

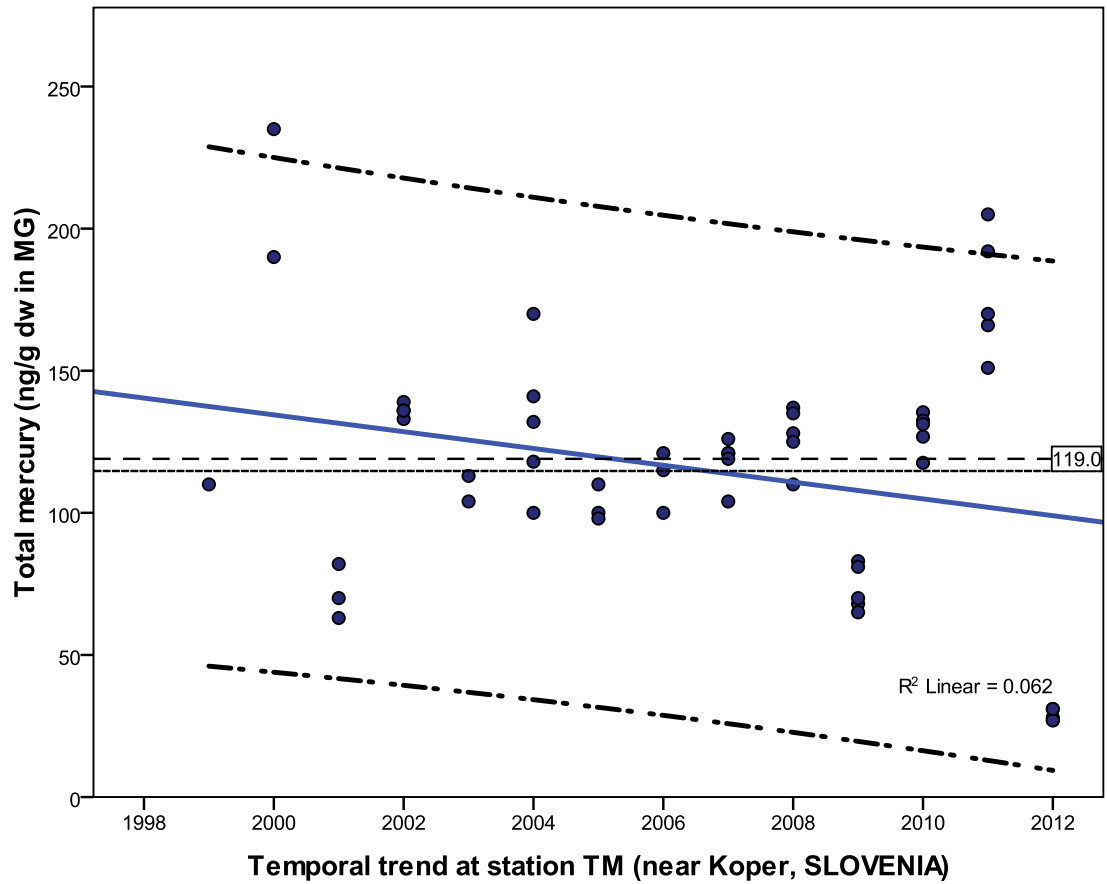
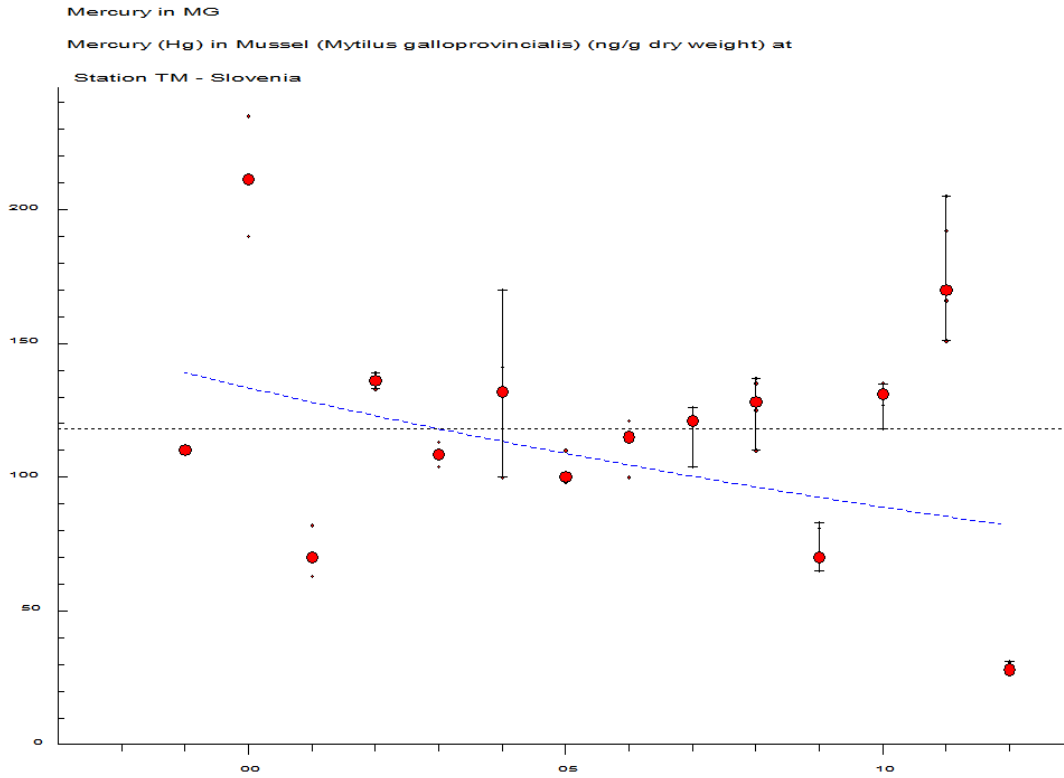


Table A.8.1. Calculated within year variances for the time series and summary of statistics for *Mytilus galloprovincialis* (MG) datasets at both stations (unit in  $\mu\text{g}/\text{Kg dw}$ ).

Parameter	Station	Year	N	Min	Max	Avg	Median	Std.Dev	Variance $\Psi^2(\text{LogC})$
Hg	TM	1999	1	-	-	110.0	-	-	-
		2000	2	190	235	212.5	212.5	31.8	0.004
		2001	3	63	82	71.6	70.0	9.6	0.003
		2002	4	133	139	136.0	136.0	2.4	0.000
		2003	2	104	113	108.5	108.5	6.4	0.001
		2004	5	100	170	132.2	132.0	26.2	0.007
		2005	3	98	110	102.6	100.0	6.4	0.001
		2006	3	100	121	112.0	115.0	10.8	0.002
		2007	5	104	126	118.2	121.0	8.3	0.001
		2008	5	110	137	127.0	128.0	10.7	0.001
		2009	5	65	83	73.4	70.0	8.1	0.002
		2010	5	118	135	128.6	131.2	6.9	0.001
		2011	5	151	205	176.8	170.0	21.5	0.003
2012	5	27	31	28.8	28.0	2.0	0.001		
Hg	24	1999	1	-	-	120.0	-	-	-
		2000	4	104	134	117.7	116.5	12.3	0.002
		2001	1	-	-	85.0	-	-	-
		2002	1	-	-	102.0	-	-	-
		2003	4	134	152	140.0	137.0	8.2	0.001
		2004	5	69	139	115.4	131.0	29.3	0.016
		2005	2	100	110	105.0	105.0	7.1	0.001
		2006	3	101	110	106.3	108.0	4.7	0.000
		2007	5	140	159	152.0	156.0	7.9	0.001
		2008	5	133	192	157.0	153.0	22.7	0.004
		2009	5	57	98	85.4	89.0	16.4	0.009
		2010	5	90	106	98.4	99.2	6.7	0.001
		2011	5	84	105	93.8	94.0	8.3	0.001
2012	5	11	19	16.2	17	3.3	0.010		

Station (name), Year (sampling year), Parameter (chemical), N (number of data), Min. (minimum value in the dataset) and Max. (maximum value in the dataset), Mean (central tendency statistic estimator assuming a Normal distributed dataset), Median (central tendency statistic estimator assuming a non-Normal distributed dataset), Std.Dev (standard deviation statistic), Variance (variance statistic).