OSPAR COMMISSION

Levels and trends in marine contaminants and their biological effects – CEMP Assessment Report 2013

Monitoring and Assessment Series

2014

Levels and trends in marine contaminants and their biological effects – CEMP Assessment report 2013

OSPAR Commission 2014

OSPAR Convention

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the “OSPAR Convention”) was opened for signature at the Ministerial Meeting of the former Oslo and Paris Commissions in Paris on 22 September 1992. The Convention entered into force on 25 March 1998. The Contracting Parties are: Belgium, Denmark, the European Union, Finland, France, Germany, Iceland, Ireland, Luxembourg, Netherlands, Norway, Portugal, Spain, Sweden, Switzerland and the United Kingdom.

Convention OSPAR

La Convention pour la protection du milieu marin de l'Atlantique du Nord-Est, dite Convention OSPAR, a été ouverte à la signature à la réunion ministérielle des anciennes Commissions d'Oslo et de Paris, à Paris le 22 septembre 1992. La Convention est entrée en vigueur le 25 mars 1998. Les Parties Contractantes sont : l'Allemagne, la Belgique, le Danemark, l’Espagne, la Finlande, la France, l’Irlande, l’Islande, le Luxembourg, la Norvège, les Pays-Bas, le Portugal, le Royaume-Uni de Grande Bretagne et d’Irlande du Nord, la Suède, la Suisse et l’Union européenne.

Acknowledgement

This report has been prepared by the Working Group on Monitoring and on Trends and Effects of Substances in the Marine Environment (MIME). Special thanks to Dr Martin M. Larsen, Denmark, and Dr Rob Fryer, UK, for their hard work in producing this report.

Contents

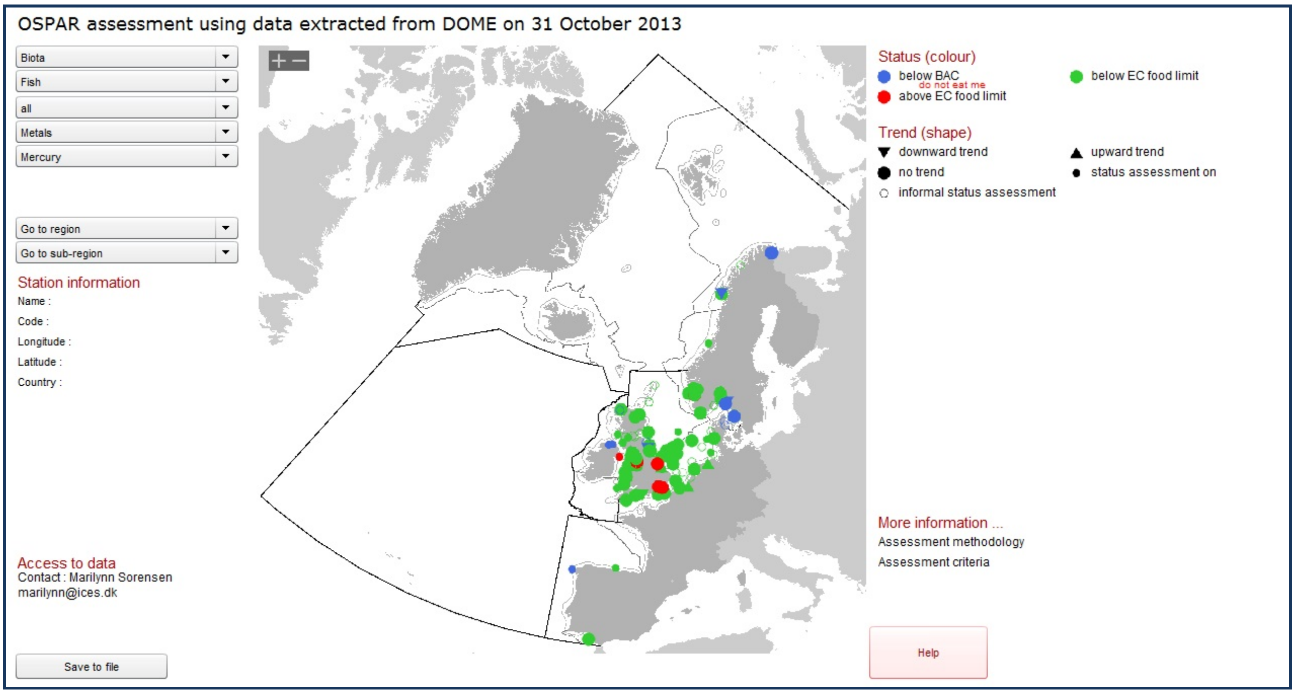
Executive Summary

The CEMP assessment measured progress towards the OSPAR objective of having concentrations of hazardous substances at background levels, or close to zero, by 2020. Concentrations of metals, PCBs, and PAHs were generally stable or decreasing. In biota, 73, 87 and 96% of significant time trends were downwards for metals, PAHs and PCBs respectively. In sediment, the corresponding values were 72, 93 and 91%. For both biota and sediment, only about half of the parameters assessed showed any upward trends and only metal concentrations increased at more than four stations. Cadmium, mercury, lead, and copper concentrations increased at 20, 16, 12, and 8 biota stations respectively; arsenic, copper, chromium, nickel and lead concentrations increased at 14, 14, 7, 5 and 5 sediment stations respectively.

This report focuses in particular on the assessment of CEMP data on TBT and its biological effects, as well as on contamination by copper:

- Concentrations of the antifouling agent TBT and biological effects in marine gastropods resulting from its use have also decreased following the ban on the use of TBT on pleasure boats in 1987 and on all ships in 2001. TBT concentrations decreased significantly in 55% of sediment time series and 81% of biota time series, and the biological effect vas deference index decreased significantly in 61% of time series. Copper is also used as an antifouling agent, and the assessment results for TBT and Cu are addressed in this report. For the information on TBT/Imposex, a separate Assessment Sheet is published by OSPAR alongside this report;
- 7% of the 202 copper time series in sediment were increasing and 11% were decreasing. Most of the upward trends are seen in the vicinity of harbours, marinas and major shipping lanes where concentrations are already at levels possibly causing effects to the ecosystem. The increasing concentrations in sediments should be investigated further and the sources and pathways into the sediment clarified.

These CEMP assessment results can be consulted in full detail on-line at http://dome.ices.dk/osparmime/main.html



Récapitulatif

L'évaluation du CEMP a mesuré les progrès réalisés vers l'objectif d'OSPAR, nommément des concentrations de substances dangereuses aux niveaux de fond, ou proches de zéro, d'ici à 2020. Les concentrations de métaux, de PCB et de HAP ont généralement été stables ou en diminution. Dans le biote, 73 %, 87 % et 96 % des tendances temporelles significatives étaient à la baisse pour les métaux, les HAP et les PCB, respectivement. Dans le sédiment, les valeurs correspondantes étaient de 72 %, 93 % et 91 %. Pour le biote comme pour le sédiment, la moitié seulement des paramètres évalués ont présenté des tendances à la hausse, et seules les concentrations de métaux ont augmenté dans plus de quatre stations. Les concentrations de cadmium, de mercure, de plomb et de cuivre ont augmenté dans 20, 16, 12 et 8 stations de contrôle du biote, respectivement ; les concentrations d'arsenic, de cuivre, de chrome, de nickel et de plomb ont augmenté dans 14, 14, 7, 5 et 5 stations de contrôle du sédiment, respectivement.

Ce rapport concerne en particulier l'évaluation des données du CEMP sur le tributylétain (TBT) et sur ses effets biologiques, ainsi que la contamination par le cuivre :

- Les concentrations de l'agent antifouling TBT et les effets biologiques que son utilisation produit sur les gastéropodes marins ont aussi diminué, suite à l'interdiction de l'utilisation du TBT sur les bateaux de plaisance en 1987 et sur tous les navires en 2001. Les concentrations de TBT ont diminué significativement dans 55 % des séries chronologiques de sédiment et dans 81 % des séries chronologiques de biote, et l'Indice vas deferens (Vas Deferens Index, VDSI) utilisé pour évaluer l'effet biologique a diminué significativement dans 61 % des séries chronologiques. Le cuivre est aussi utilisé comme agent antifouling, et les résultats de l'évaluation pour le TBT et le cuivre sont traités dans ce rapport. En ce qui concerne les informations sur le TBT/Imposex, une Fiche d'évaluation séparée est publiée par OSPAR parallèlement à ce rapport ;
- 7 % des séries chronologiques du cuivre 202 dans le sédiment étaient en augmentation et 11 % étaient en diminution. La plupart des tendances à la hausse sont observées à proximité des ports, des marinas et des grandes voies de navigation, où les concentrations sont déjà à des niveaux qui pourraient avoir des effets sur l'écosystème. Il convient d'étudier davantage les concentrations croissantes dans les sédiments, et de clarifier les sources et les voies qui aboutissent au sédiment.

La version intégrale de ces résultats de l'évaluation du CEMP peut être consultée en ligne à http ://dome.ices.dk/osparmime/main.html

Introduction

The 2013 assessment of data collected under the OSPAR Coordinated Environmental Monitoring Programme (CEMP) was prepared by the OSPAR Working Group on Monitoring and on Trends and Effects of Substances in the Marine Environment (MIME) at, and following, its meeting in December 2013 and is based upon data reported by Contracting Parties to ICES and held in the ICES Environmental databases. This report focuses on case study assessments of TBT, Imposex and Copper. The assessment results on TBT and Imposex are also published as a separate Assessment Sheet.

The CEMP is the monitoring under the OSPAR Joint Assessment and Monitoring Programme where the national contributions overlap and are coordinated through adherence to commonly agreed monitoring guidelines, quality assurance tools and assessment tools. It covers temporal trend and spatial monitoring programmes for concentrations of selected chemicals and nutrients, and for biological effects. Monitoring under the CEMP aims to indicate the extent of contamination of fish, shellfish and sediments with hazardous substances and the intensity of their biological effects. The purpose is to support OSPAR assessments of the effectiveness of measures to reduce releases of hazardous substances to the environment.

The assessment evaluates the status and trend of concentrations of hazardous substances in the marine environment for selected hazardous substances which have been prioritised for action by OSPAR due to their risk for the marine environment and which are being monitored under the CEMP. It builds on experience gained in the first comprehensive trend assessment of CEMP data in 2005 (OSPAR Commission, 2005), and the annual CEMP assessments undertaken in the period 2006 – 2009 (OSPAR Commission, 2006; 2007; 2008; 2009a), which contributed to the OSPAR Quality Status Report 2010 (OSPAR Commission, 2010) accessible at http://qsr2010.ospar.org.

Web-based presentation of CEMP assessment results

The OSPAR assessment for 2013 covers over 400 000 measurements in 73 parameters at 1223 stations. There were 8765 time series (station / parameter combinations with more than three years of data) that were assessed for trends in sediment, biota and for biological effects (Annexes 1 and 2). Overall, there were 1506 significant trends of which 88% were downwards indicating a general decline in pollutant concentrations. Assessment products are presented in the Web assessment tool at http://dome.ices.dk/osparmime/main.html. Assessment data are also available at this location together with supporting tables, plots, help files and methodological information.

Contaminants and matrices covered

The contaminants covered by the CEMP are the metals cadmium (Cd), mercury (Hg), lead (Pb), nickel (Ni), copper (Cu), zinc (Zn), chromium (Cr), arsenic (As) and the organic contaminants PCBs (congeners 28, 52, 101, 105, 118, 126, 138, 153, 156, 169, 180) and PAHs (naphthalene, phenanthrene, anthracene, dibenzothiophene, fluoranthene, pyrene, chrysene/triphenylene, benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*ghi*]perylene, and indeno[12,3-*cd*]pyrene). This report focuses on TBT, the imposex biological effect (from TBT) and copper. The webtool assessment results are available for cadmium, mercury and lead, which are the metals selected by the OSPAR Commission for priority action. The other metals covered are either micronutrients (zinc and copper) or have other biological functions (arsenosugars, chromium, nickel).

Further to the PAHs and CBs mentioned above, the webtool assessment also covers the concentrations of selected indicators from other hazardous substances groups: four organochlorine pesticides, eleven selected organobromines, one dioxin and one furan. The available data for each substance in marine sediment, fish tissue (muscle and liver) and/or shellfish tissues are assessed.

Finally nine biological effects are assessed, three imposex (TBT related) and six other, either general biologic effect or specific for PAH (metabolites) related biological effects. For some biological effects higher numbers indicate better status; this is reflected in the assessment criteria and colour scheme.

Methods

Methods for data screening, treatment of quality assurance information, temporal trend assessment and assessment against criteria used previously by CEMP are described in the CEMP Assessment Manual (OSPAR Commission, 2008b) and in the help files at http://dome.ices.dk/osparmime/main.html.

Criteria used to assess environmental concentrations of hazardous substances are set out in the OSPAR agreement on CEMP Assessment Criteria for the QSR 2010 (OSPAR agreement 2009-2). The derivation of these criteria for hazardous substances is discussed in a Background Document on CEMP Assessment Criteria for the QSR 2010 (OSPAR Commission, 2009b). These criteria reflect a two-stage process in which data are compared to concentrations yielding limited risks of biological effects (see Environmental Assessment Criteria, EACs) and then against Background Concentrations (BCs) or zero, expressed as Background Assessment Concentrations (BACs). The latter reflects the OSPAR Hazardous Substances Strategy that concentrations should be at or close to background levels for naturally occurring substances (thus zero for man-made substances).

An overview of the assessment criteria is at **Annex 3**.

This year, two changes were made to the configuration of the rollover assessment:

1. Time series were only considered if they contained data collected within the previous six monitoring years (2007-2012) instead of five; this is in line with the six-year reporting cycle for MSFD.
2. Trends in sediment were considered over the most recent twenty monitoring years (1993-2012) instead of ten. Trends in biota were considered over the most recent ten monitoring years as before. Sediment concentrations are likely to change more slowly than biota concentrations, so a twenty-year period is more relevant. Furthermore, the monitoring frequency at many sediment stations has been reduced (partly to reflect the slower expected rate of change in sediment concentrations) so a longer period is required to maintain statistical power. The number of significant downward trends (over all parameters) increased from 515 (in last year's assessment) to 594, whilst the number of significant upward trends decreased slightly from 89 to 85. However, the number of time series that were assessed had also increased. The assessment maps suggest no major shift in interpretation.

Overview of data submissions

The data submissions by Contracting Parties are assessed from the ICES website (via http://info.ices.dk/datacentre/accessions/CommissionSummary.aspx?commission=OSPAR).
The status of each submission is:

X: the Contracting Party has submitted data and these data are available for use from the ICES database;
P: the Contracting Party has submitted data, and the data are still being processed (quality checks etc., by the Contracting Party or by the data host ICES).

The date stamp on this overview of data submissions is 17 February 2014.

The parameters reported within the 2005-2011 period are listed (only parameter groups assessed in MIME during the last years, others may occur). Data for each parameter are not necessarily reported every year for each station.

Imposex and other Biological Effects in Biota Data

Country	2007	2008	2009	2010	2011	2012	Parameters #
Denmark	**P**		**P**				**IMP**
France	X	X	X	X	X	X	IMP (EROD)
Germany	X		X				GRS, PAH
Ireland				X	X	X	
Netherlands	X	X	X	X	X	X	IMP, GRS
Norway	P	X	P	X	X	X	IMP, ALAD, EROD, PAH
Spain	X	X	X	X	X	X	IMP, EROD, TOX
Sweden	X	X	X	X	X	X	IMP
United Kingdom	X	P X	P P	P	X	X	IMP, GRS, EROD, PAH, TOX
Portugal		X	X				IMP

IMP: Imposex/intersex; GRS: Fish diseases; PAH: PAH metabolites; ALAD: ALA Dehydratase; EROD: Cytochrome P4501A activity; TOX: Sediment bioassays

Contaminants in Biota Data

Country	2007	2008	2009	2010	2011	2012	Parameters
Belgium	X	X	X	X			Metal, TBT, PAH, PCB, PBDE
Denmark	X	X	X	X	X	X	Metal, TBT, PAH, PCB, PBDE, Dioxin
Faroe Islands					X	X	Metal, TBT, PAH, PCB, PBDE
France	X						Metal, PAH, PCB
Germany	X	X	X	X	X	X	Metal, TBT, PAH, PCB, PBDE
Iceland	X	X	X	X	X	X	Metal, PCB
Ireland	X	X	X	X	X	X	Metal, PAH, PCB, PBDE, Dioxin, PFOS
Netherlands	X	X	X	X	X	X	Metal, PTB, PAH, PCB, PBDE
Norway	P	X	P	X	X	X	Metal, PAH, TBT, PCB, PBDE, Dioxin, PFOS
Portugal		P	P	P			Metal, PAH, PCB, Dioxin
Spain	X	X	X	X	X	X	Metal, TBT, PAH, PCB, PBDE
Sweden	X	X	X	X	X	X	Metal, TBT, PAH, PCB, PBDE, PFOS, Dioxin
United Kingdom	X	X	P	P	X	X	Metal, TBT, PAH, PCB, PBDE

Contaminants in Sediment Data

Country	2007	2008	2009	2010	2011	2012	Parameters
Belgium	X	X		X	P	P	Metals, TBT, PAH, PCB, PBDE
Denmark	X	X	X	X	X	X	Metals, TBT, PAH, PCB, PBDE, dioxin
Germany	X	X		P	X	X	Metals, TBT, PAH, PCB, PBDE
Ireland	X						Metal, PCB, TBT, PAH
Netherlands	X	X	X	X	X	X	Metal, PCB, TBT, PAH, PBDE
Norway		X					Metal, PCB, TBT, PAH, BDE, PFOS
Spain	X	X	X	X	X	X	Metal, TBT, PCB, PBDE, PAH
United Kingdom	X	P	P	P	P	P	Metal, PCB, TBT, PAH, PBDE

France has submitted Metals, PAH and PCB in 2003 sediments as the latest.

For information the submission of seawater data is illustrated below. This is not mandatory monitoring and the data are not included in the CEMP assessment. Not all parameters are monitored and reported every year.

Contaminants in Seawater Data

Country	2007	2008	2009	2010	2011	2012	Parameters
Belgium	X	P	X	X	P	P	Metals, TBT, PBDE, PAH
France							
Germany	P X	P X	X	P X	P	P	PCB, PAH, Triazines
Ireland	X						
Netherlands	X	X	X	X	X	X	Metal
Spain				X			
United Kingdom	X	P	P	X	P	P	Metal, TBT, PCB, Triazines

Status and trends

The focus of the 2013 assessment has been on antifouling components (tributyltin (TBT) and copper), and the effects of the ban on TBT with regard to both concentrations in the marine environment and the biological effects on marine snails (imposex). As TBT is phased out, use of copper in antifouling products may increase. Copper concentrations in sediment and biota are increasing in many areas with heavy ship traffic. Two assessment sheets for imposex and TBT and for copper describe the situation in detail.

Overview of results for biota and biological effects

For biota and biological effects there were 4 604 time series involving 46 parameters, with 827 significant trends of which 88% were downwards. ALAD biological effect and TCCD (2,3,7,8-tetrachloro-dibenzo[*b*,*e*][1,4]dioxin) were the only parameters where all the trends were upwards, but both parameters only had three time series. Dioxin-like CB126 and the furan 2,3,7,8-tetrachloro-dibenzofuran had one upward and one downward trend out of seven time series.

Metals are the most commonly investigated parameters with between 250 and 300 time series each. Of these, between 10% (copper) and 20% (cadmium) were significant. Between 64% (mercury) and 88% (zinc) of the significant trends were downwards, so metals are reasonably well regulated.

Although PCBs were monitored at a similar number of stations as metals there were fewer time series, typically between 100 and 250, due to concentrations close to detection limits. Between 12% (congeners 28, 153, 156) and 29% (congener 126) of the time series had significant trends. Of the significant trends, 96% were downwards, so in general PCB concentrations are declining.

PAHs are monitored at around 270 stations, with typically between 80 and 170 time series. Between 8% (fluoranthene, pyrene) and 38% (naphthalene) of the time series had significant trends. Overall 87% of the significant trends were downwards.

There were no upward trends for 22 of the 46 parameters with time series, including most dioxins, organotins and biological effects related to TBT (Vas Deferens Stage Index, imposex and intersex). The reporting and coverage of other biological effects is still far behind that of the chemical parameters, with at most four time series available for any one parameter, and overall only three significant trends, all downwards. Four PAHs (naphthalene, anthracene, dibenzothiophene, pyrene) and four PCBs (congeners 101, 105, 156, 169) only had downward trends. Legacy chlorinated pesticides (HCB and □-HCH) and BDE47 also only had downward trends, whereas DDE (DDT breakdown product) and gamma-HCH both had 2 upward trends.

Overview of results for sediments

Where biota and biological effects usually show the current status of pollution responding to the "average" over the last months or year, sampling surface sediments is giving an "average" of years depending on the sedimentation rate at the sampling site and sampling depths (usually upper 1 cm).

Around 350 stations were sampled for metals, parent PAHs and PCBs, some 290 stations for brominated substances, and some 100 for organotins and methylated PAHs. Overall, there were 4161 time series involving 43 parameters, with 679 significant trends of which 87% were downwards. There were no upward trends for 19 of the 43 parameters with time series.

For metals, between 7% (lead) and 19% (mercury) of the time series had significant trends. Metals had the highest number of upward trends, with 14 for both arsenic and copper. For arsenic, only 18% of trends were downwards. However, for all other metals, between 62% (copper) and 94% (mercury) were downwards.

For the parent PAHs, typically around 16% of the time series were significant and, overall, 93% of trends were downwards. Similarly, for most of the PCBs around 19% of time series were significant and, overall, 91% of trends were downwards.

There were no time series for phenyltins and only a few for the brominated substances, either because measurements were seldom reported or because measurements were close to or below the detection limits. However, there were significant downward trends in tri- and di-butyltins in 49% of time series, with no upward trends for any of the butyltins.

Imposex and TBT: Status, trends and effects in marine molluscs: an improving situation?

OSPAR COMMISSION

Policy Issue: Impacts of tributylin (TBT) on marine molluscs

Policy Objective: Continued reduction of levels of TBT in the marine environment, so that the exposure of marine molluscs and the adverse imposex effect remains below agreed OSPAR environmental assessment criteria (EACs), and ultimately reduction to 'close to zero' levels.

Background

Antifouling paints are widely used on vessels of all sizes to prevent the growth of marine organisms. Historically, antifoulants were primarily based on the use of copper, creating higher, toxic, concentrations close to the hull and so preventing the attachment of organisms. Around the beginning of the 1980s, a more effective component began to be used, TBT. This compound proved extremely effective at preventing the attachment of algal slimes, which are usually the first organisms to attach and which then provide a coating to which other organisms can attach. By the mid-1980s, oyster growers in both France and Great Britain were becoming extremely concerned about poor growth in their stocks. Cultured Pacific oysters, in particular, were misshapen and contained little meat, so were not marketable. Eventually, the cause was traced to the use of TBT in anti-fouling paints applied mainly to pleasure vessels used in estuaries and moored in marinas, close to the commercial shellfish beds.

TBT is now known to be sufficiently toxic to harm many marine organisms at very low concentrations and is unequivocally linked to impairment of reproductive performance in a number of molluscan species, with some female marine snails developing male sex characteristics in response to TBT exposure; this is termed 'imposex' (OSPAR Commission, 2000). TBT ultimately affects many creatures, but the marine mollusc's sensitivity puts it at the front line making it an important indicator species giving an early warning of trouble to the marine ecosystem. Over the past decade, a range of national and international measures have resulted in a continuous phase-out in the OSPAR area of TBT containing paints and of their use on vessels, in aquaculture and on underwater structures. A global ban on TBT in anti-fouling systems on large vessels came into effect in 2008. Together, these measures address the main TBT-related pressures on the marine environment. Assessment criteria in the form of background assessment criteria (BAC) and environmental assessment criteria (EAC) have been derived by OSPAR for imposex measurement in a variety of molluscs, representing the most sensitive species used in the OSPAR monitoring guidelines.

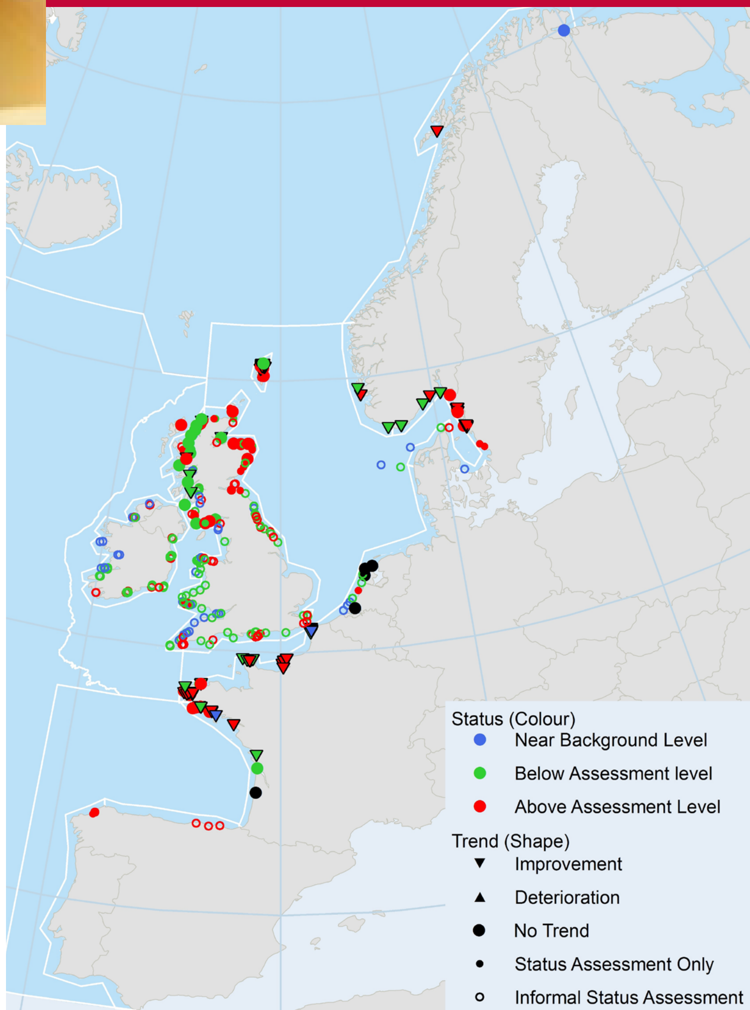
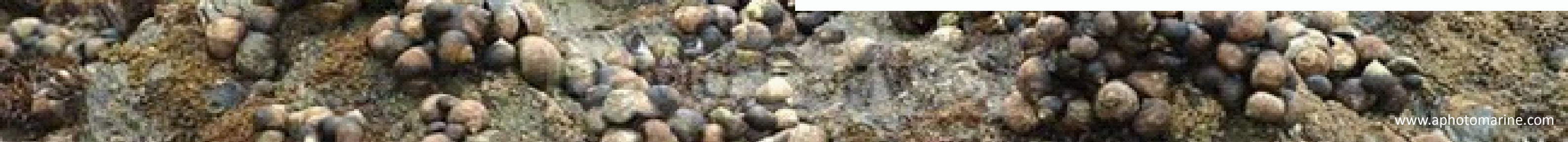


Figure 1: Current monitoring status of imposex in marine molluscs resulting from this assessment

Has regulation proved effective?

Imposex monitoring is currently completed at over 390+ sites on up to 5 marine mollusc species. There is a diversity of approaches to selecting target sampling stations, although there is an emphasis on stations which are in, or proximate to, harbours, ports and marinas where effects are most likely to occur. Currently the OSPAR EAC set for TBT-specific effects is met at most sites (~65%). It is also clear that there is an overall improvement in mollusc imposex status at locations common for both the 2010 QSR assessment (OSPAR Commission, 2009) and for this assessment (n=145 sites).



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Imposex and TBT: Status, trends and effects in marine molluscs: an improving situation?

OSPAR COMMISSION

Has regulation proved effective? (cont.)

Figure 2: Improvement (green) and non-improvement (red) in 5 marine mollusc species sampled at 107 sites in OSPAR region II

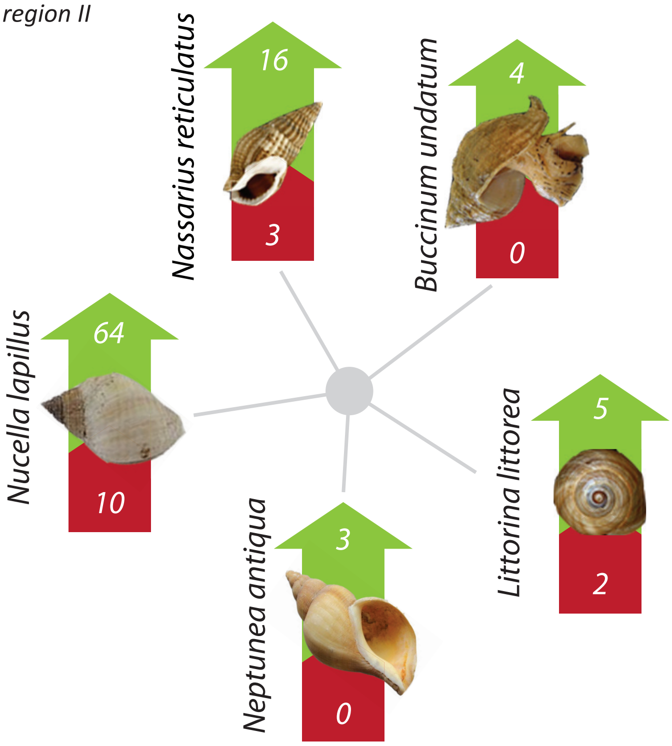
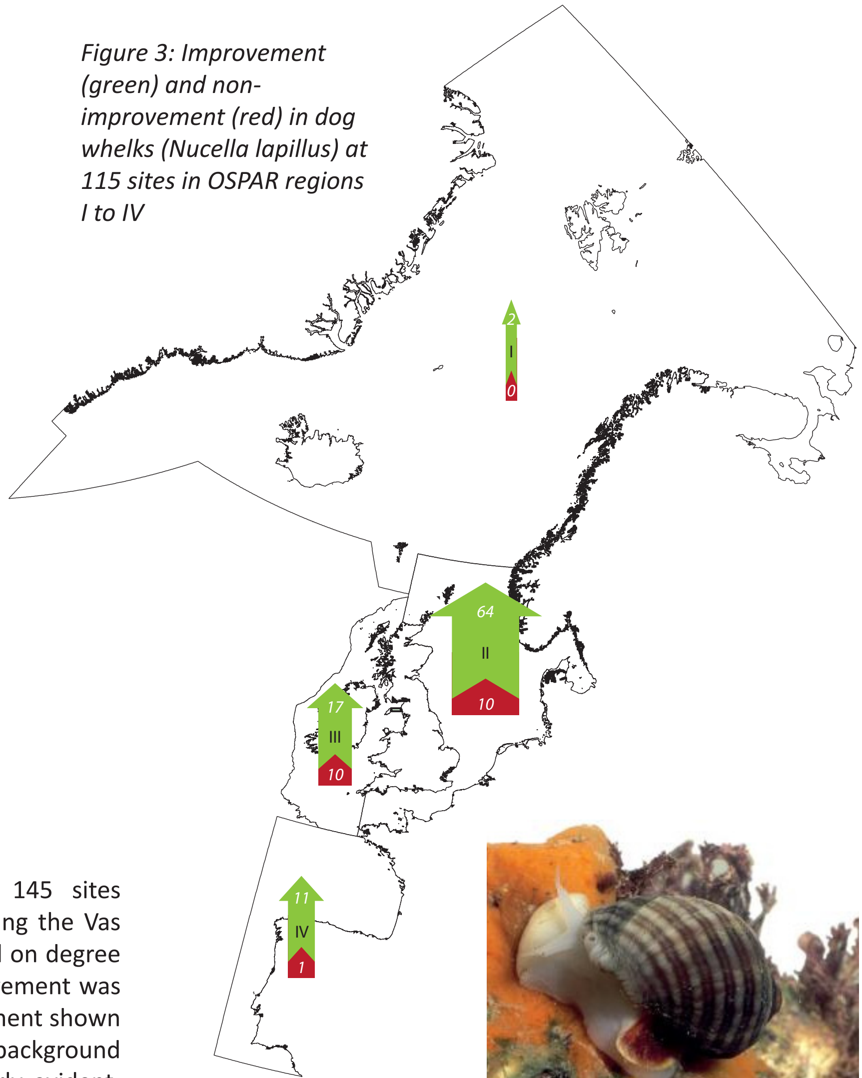


Figure 3: Improvement (green) and non-improvement (red) in dog whelks (Nucella lapillus) at 115 sites in OSPAR regions I to IV



Improvement and non-improvement was measured at 145 sites common to both the QSR and CEMP 2013 assessment, using the Vas Deferens Stage Index (VDSI) – a 7 stage measurement based on degree of penis and Vas Deferens development in females. Improvement was detected at >80% of these common sites with non-improvement shown at 16% of locations, while at 4% of sites the status is at background and continues to be stable so overall improvement is clearly evident.

What happens next?

Ongoing monitoring in marine gastropods will continue to provide a good indicator for TBT pollution and help in identifying illegal use of stocks of TBT-containing anti-foulants or losses of TBT from dockyards, marinas and vessel maintenance activities. One or more further rounds of imposex monitoring will be conducted in order to follow the expected decline in impacts indicated by the reductions in inputs described above.

Most antifouling products have now reverted to the use of copper-based paints, in some cases with the addition of other chemicals ("booster biocides") which enhance their efficacy. These have also been investigated for environmental persistence and effects and have also been banned. OSPAR needs to keep an eye on future developments in this area and avoid further adverse consequences of use of other compounds. In addition, there is a large reservoir of TBT in sediments, particularly in estuaries, which may continue to provide a secondary source to the water column.

Despite the evident significant downward trend in impacts from TBT, some areas are still subject to high imposex levels. Sites which show non-compliance with the EcoQO should be subject to further monitoring. The continued measurement of imposex in marine molluscs offers the most clear-cut tool for monitoring of a contaminant-specific pollution effect under the Marine Strategy Framework Directive.

Sources of data and information:

Law, R.J., Bolam, T., James, D., Barry, J., Deaville, R., Reid, R.J., Penrose, R., Jepson, P.D., 2012. Butyltin compounds in liver of harbour porpoises (Phocoena phocoena) from the UK prior to and following the ban on the use of tributyltin in antifouling paints (1992-2005 & 2009). Marine Pollution Bulletin 64, 2576-2580.

OSPAR Commission, 2000. Quality Status Report 2000. OSPAR Commission, London. 108 + vii pp. http://www.ospar.org/content/content.asp?menu=00650830000000_000000_000000

OSPAR Commission, 2009. CEMP assessment report: 2008/2009 Assessment of trends and concentrations of selected hazardous substances in sediments and biota, [ISBN 978-1-906840-30-3]; Publication Number: 390/2009OSPAR 2010. Quality Status Report 2010. OSPAR Commission. London. 176 pp.

OSPAR Commission, 2011. Background document on organic tin compounds. ISBN 978-1-907390-76-0. Publication Number: 535/2011

ASSESSMENT FOCUS:

Metals in sediment: status and trend of copper burden

Policy issue

Many man-made and naturally occurring chemicals end up in the North-East Atlantic as a result of land-based and sea-based human activities.

Policy objectives

- EU - Marine Strategy Framework Directive:
  - aims to achieve good environmental status of the EU's marine waters by 2020.
  - concentrations of contaminants are at levels not giving rise to pollution effects.
- OSPAR:
  - cessation of discharges, emissions … of hazardous substances by 2020.
  - hazardous substances in the marine environment: (i) achieve concentrations near background values for naturally occurring substances; and (ii) close to zero for man-made substances.

Specific Questions/Background

Copper is an extensively used metal in a variety of applications. Since the early 1980s measures have been taken to reduce riverine inputs, direct discharges and atmospheric inputs. As a result there were important downward trends in concentrations in both sediment and biota. Recently, however, increasing copper levels have been observed in contrast to most other metals and organic substances.

Copper has been used in antifouling treatments since ancient times (Yebra et al (2004)), but especially so from the end of the 18th century when it was used together with other metals (mainly lead, arsenic and mercury) as an antifouling agent. Copper-based paints are usually not used on aluminium hulls as contact between the two metals can form a galvanic element and lead to damage of the hull.

Since a ban on the most efficient antifouling agent TBT (see Assessment Sheet for TBT/Imposex), substitution with copper based paints with Irgarol 1051 (cybutryne) or Diuron booster biocides started on smaller vessels and has now continued for over a decade. These two booster biocides have since been banned in UK, Denmark and Holland (only Diuron), and Danish legislation placed limits on the release rate of copper from antifouling paints on pleasure crafts to 200 µg/cm^2/day in the first 14 days and 350 µg/cm^2/day after 30 days (Danish Ministry of the Environment, 2011).

The new antifouling components, though not causing effects as detrimental as TBT, can have certain adverse effects on biota and be concentrated in sediments, especially in harbour areas (e.g. Briant et al (2013)). The rapid growth in the use of copper-based antifouling systems in aquaculture over the past decade has markedly increased the inputs of copper to the sea in northern Scotland and west and north Norway (OSPAR Commission, 2011). The total use of copper in Norwegian fish farms was estimated to be 261 tons of copper in 2005 (Fitridge et al, 2012), the same range as the 75-311 tons of copper estimated to have been used on UK pleasure boats in 1998 (UK Marine SACs project). The impact on sediments and bottom organisms in the vicinity of fish farms and public awareness of chemical antifouling use opposed to “clean and green” marketing together with EU classification of copper as a R50/R53 dangerous substance (EEC directive 67/548, 1967) may change the focus to other means of protection against fouling. Recent surveys though have not shown increased concentrations outside an impact area of less than 250 m around fish farms.

Findings

Overall, the metal burden in sediments was mostly found to be decreasing in the North Sea, with 9% of 1541 assessed sediment time series showing downward trends compared to 3.5% showing an increasing trend. There were more significant trends for mercury than for any other metal: of the 180 mercury time series, 18% showed a downward trend and just 1% an upward trend.

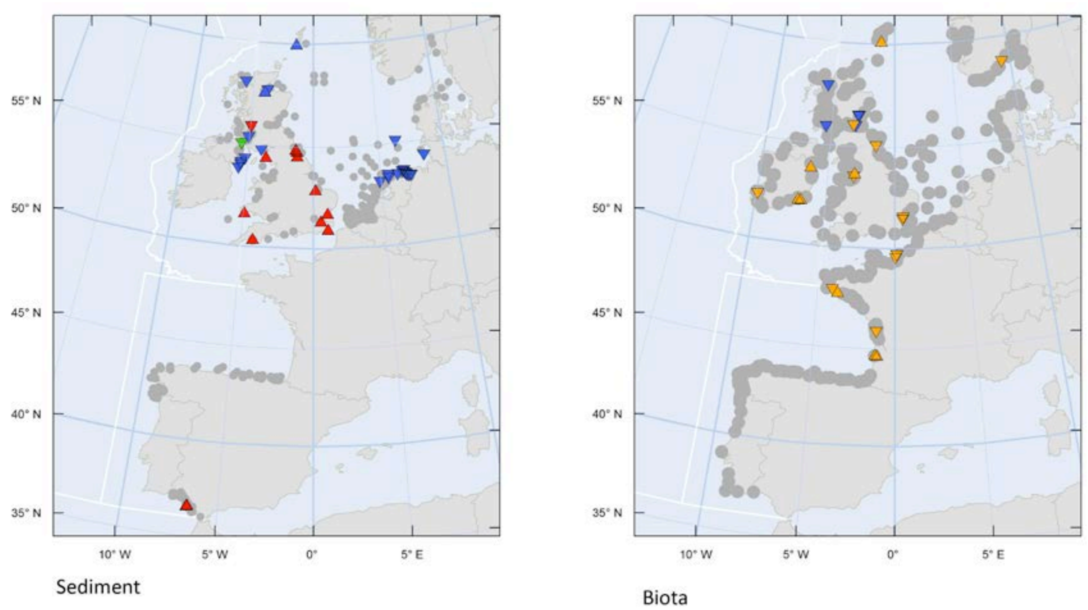
In contrast, 7% of the 202 copper time series in sediment were increasing and 11% were decreasing. The general pattern is (**Figure 3**, left) that sites with high concentrations are further increasing (southern UK, 10 of 11 "red" time series for UK upward, one downward) whereas for the green/blue sites below ERL/background, only two northern UK sites were upward of 22 trends. Also in Biota there have been 3% upward and 7% downward trends (of 267 time series). Most of the increasing trends were observed around the UK, in the Irish Sea, the English Channel and on the English east coast, rising from already elevated concentration levels. No direct geographical link was observed between copper in biota and sediment samples. Upward trends have also been found on the southern Spanish Atlantic coast and, at a lower concentration level, around the Shetland Islands to the north of Scotland.

Most of the upward trends are seen in the vicinity of harbours, marinas and major shipping lanes. There are currently no indications from OSPAR monitoring of aquatic inputs (RID) of increased riverine inputs. In the QSR 2010 (OSPAR Commission, 2010) information about the use of copper, e.g. from Norway, suggested a significant increasing usage of copper in fish farming activities, but the focus on copper as a dangerous substance within the EU may help to reduce inputs and increasing concentration trends. Also, new results have indicated that use of copper and other substances in fish farming have only very local effects on copper concentrations in biota and sediments with elevated levels at 50 m, but not 250 m from the investigated farms in UK and Norway (Russell et al, 2011; Falk, 2014).

Specific Questions/Background

The importance of copper in brakes and tyres, and the losses from railway current supply, was mentioned in the QSR 2000 (OSPAR, 2000). A major source of copper to the environment is brake pad wear, and a recent estimate suggests that this contribution is at the same level as other sources of airborne copper pollution (Hulskotte et al (2006)). California has set up a roadmap with the aim of reducing copper concentrations in brake-pads to 5% in 2021 and to below 0.5% in 2025 (DTSC, 2010). Furthermore, a very important source is the corrosion of copper materials such as drinking water tubing, copper roofs and other copper containing materials (ER, 2014). Although a large part of the corrosion products is removed by adsorption on wastewater treatment sludge the remaining part flows via effluents into surface waters. Other important sources are the runoff and leaching of agricultural soils amended with livestock manure (especially from pigs) and, in some countries, due to application of sewage sludge as fertilizer (Nicholson et al, 2003). In harbours, leaching of copper antifouling from ships' hulls is also of importance. In situ polishing of the brass propellers on ships by diving companies can also contribute to elevated concentrations of copper in harbour water and transport of fine copper particles to the sediment (Berbee, 2014).

The fate of copper entering the marine environment is complex and the freshwater/sewage sources can be removed at the freshwater/seawater interface by flocculation and binding to settling particles or by being bound by natural complexing agents (e.g. humic substances) or substances released by algae before dispersion over larger areas. The increasing concentrations in sediments, but not in mussels, should be investigated further and the sources and pathways into the sediment clarified.



LEGEND	>>>> INCREASING CONCENTRATIONS >>>>		
Sediment	Below background assessment level with increasing trend (Δ) or decreasing trend (∨)	Between background assessment level and the upper assessment criterion (ERL) with increasing trend (Δ) or decreasing trend (∨)	Above the upper assessment criterion (ERL) with increasing trend (Δ) or decreasing trend (∨)
Biota		Above background assessment level and the upper assessment criterion (ERL) with increasing trend (Δ) or decreasing trend (∨)	

Figure 3: trend and status for Copper in sediment (left) and biota (right). Most trends are not significant (grey circles). For significant time trends only, the assessment class is shown according to the table above.

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Conclusion

The general trend for hazardous substances in the environment is downward, in many cases based on actions taken to reduce, minimise or ban use of specific chemicals in general or in specific applications within the individual countries, within the EU or on a worldwide basis. Some naturally occurring substances like metals and PAHs are not following this general trend and knowledge of the sources behind the upward trends are therefore required to make effective legislation or recommendations on future voluntary restrictions on the use or discharge of such substances.

The OSPAR web-based tool for trends in the marine environment is one tool for following the OSPAR aim of reaching levels of contaminants not giving rise to pollution effects and the cessation of discharges, emissions and losses of hazardous substances by 2020. This report's Assessment Focus sections and the associated Assessment Sheet outline specific substances and the problems or solutions related to meeting these goals and will be produced and updated regularly in the yearly CEMP assessment process.

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Annex 1 – Analysed substances and significant trends in sediment

Substance (code)	# stations	# TT	% Up	%Down
Anthracene (ANT)	354	138	1.4%	15.2%
Arsenic (AS)	339	185	7.6%	1.6%
Benzo[a]anthracene (BAA)	354	162	1.9%	12.3%
Benzo[a]pyrene (BAP)	362	165	1.2%	16.4%
BD100 (BD100)	298			
2,2',4,4',5,5'-hexabromodiphenyl ether (PBDE153) (BD153$)	295	4	0.0%	0.0%
BD154 (BD154)	297	0		
BD183 (BD183)	296	0		
BD209 (BD209)	168	0		
BDE28 (BDE28)	285	0		
2,2',4,4'-tetrabromodiphenyl ether (PBDE47) (BDE47$)	298	7	0.0%	14.3%
2,3',4,4'-tetrabromodiphenyl ether (BDE66) (BDE66$)	294	2	0.0%	0.0%
benzo[ghi]perylene (BGHIP)	364	165	0.6%	14.5%
2,2',4,5,5'-pentachlorobiphenyl (CB101)	356	94	1.1%	18.1%
2,3,3',4,4'-pentachlorobiphenyl (CB105)	272	20	%	0.0%
2,3',4,4',5-pentachlorobiphenyl (CB118)	356	96	3.1%	18.8%
2,2',3,4,4',5'-hexachlorobiphenyl (CB138)	355	100	2%	25%
2,2',4,4',5,5'-hexachlorobiphenyl (CB153)	356	112	2.7%	17.0%
2,3,3',4,4',5-hexachlorobiphenyl (CB156)	175	13	0.0%	0.0%
2,2',3,4,4',5,5'-heptachlorobiphenyl (CB180)	356	77	1.3%	15.6%
2,4,4'-trichlorobiphenyl (CB28)	356	74	1.4%	21.6%
2,2',5,5'-tetrachlorobiphenyl (CB52)	356	51	0.0%	9.8%
Cadmium (CD)	363	171	1.8%	13.5%
Chrysene (CHR)	347	148	0	18.9
Chromium (CR)	366	202	3.5%	6.9%
Copper (CU)	368	202	6.9%	11.4%
Dibenzothiophene (DBT)	90	21	0.0%	23.8%
C1 Dibenzothiophene (DBTC1)	83	27	3.7%	14.8%
C2 Dibenzothiophene (DBTC2)	77	28	0.0%	25.0%
C3 Dibenzothiophene (DBTC3)	77	28	0.0%	21.4%
dibutyltin (DBT) - expressed as Sn-atom (DBTIN)	129	42	0	40.5
Diphenyltin (DPTIN)	114	0		
Fluoranthene (FLU)	364	179	1.7%	14.5%
Mercure (HG)	366	180	1.1%	17.8%
indeno[1,2,3-cd]pyrene (ICDP)	354	164	1.8%	19.5%
monobutyltin (MBT) - expressed as Sn-atom (MBTIN)	130	43	0	11.6
Monophenyltin (MPTIN)	114	0		
Naphthalene (NAP)	170	56	1.8%	12.5%
C1 Naphthalene (NAPC1)	19	12	0.0%	0.0%
C2 Naphthalene (NAPC2)	101	36	0.0%	16.7%
C3 Naphthalene (NAPC3)	101	44	0.0%	15.9%
Nickel (NI)	368	202	2.5%	5.4%

Substance (code)	# stations	# TT	% Up	%Down
Phenanthrene (PA)	366	182	0.5%	13.7%
C1 Phenanthrene (PAC1)	86	32	0.0%	15.6%
C2 Phenanthrene (PAC2)	86	32	0.0%	21.9%
C3 Phenanthrene (PAC3)	77	25	0.0%	20.0%
Lead (PB)	367	201	2.5%	4.5%
pyrene (PYR)	364	181	1.7%	13.8%
tributyltin (TBT) - expressed as Sn-atom (TBTIN)	140	60	0	55
triphenyltin (TPTIN)	124	0		
Zinc (ZN)	367	198	2.0%	12.1%

$: BD28, BD85, BD99, BD100, BD154, BD183, BD209: No TT data in 80 – 291 stations monitored

Annex 2 – Analysed substances and significant trends in biota

Substance	# stations	# TT	% Up	%Down
Acetylcholine esterase activity (ACHE)	24	0		
Aminolevulinic acid dehydrase (ALAD)	3	3	33.3	0
Anthracene (ANT)	271	90	0	8.9
Benzo[a]anthracene (BAA)	271	127	3.1%	18.9%
Benzo[a]pyrene (BAP)	268	87	4.6%	9.2%
3-hydroxy benzo(a)pyrene (BAP3OH)	4	1	0	0
2,2',4,4'-tetrabromodiphenyl ether (PBDE47) (BDE47$)	210	42	0.0%	47.6%
benzo[ghi]perylene (BGHIP)	269	115	1.7%	8.7%
2,2',4,5,5'-pentachlorobiphenyl (CB101)	436	178	0.0%	12.9%
2,3,3',4,4'-pentachlorobiphenyl (CB105)	301	117	0	27.4
2,3',4,4',5-pentachlorobiphenyl (CB118)	437	215	0.9	15.3
3,3',4,4',5-pentachlorobiphenyl (CB126)	37	7	14.3	14.3
2,2',3,4,4',5'-hexachlorobiphenyl (CB138	403	209	0.5	20.6
2,2',4,4',5,5'-hexachlorobiphenyl (CB153)	438	248	0.4	12.1
2,3,3',4,4',5-hexachlorobiphenyl (CB156)	301	83	0	12
3,3',4,4',5,5'-hexachlorobiphenyl (CB169)	39	7	0	14.3
2,2',3,4,4',5,5'-heptachlorobiphenyl (CB180)	434	150	2	24
2,4,4'-trichlorobiphenyl (CB28)	435	98	1	11.2
2,2',5,5'-tetrachlorobiphenyl (CB52)	432	130	1.5	13.8
Cadmium (CD)	477	298	6.7	13.8
2,3,7,8-tetrachloro-dibenzofuran (CDF2T)	45	7	14.3	14.3
Chrysene (CHR)	266	135	1.5%	14.8%
Copper (CU)	457	267	3	7.5
Dibenzothiophene (DBT)	145	26	0	19.2
Dibutyltin (DBTIN)	68	19	0	47.4
DDE (p,p') (DDEPP)	232	99	2%	8.1%
Diphenyltin (DPTIN)	53	0		
EROD (EROD)	64	3	0	0

Substance	# stations	# TT	% Up	%Down
Fluoranthene (FLU)	270	167	1.8	6
1,2,5,6,9,10-hexabromocyclododecane (HBCD)	6	2	0	100
Hexachlorobenzene (HCB)	238	66	0	10.6
alpha-HCH (alpha-hexachlorocyclohexane) (HCHA)	248	25	0	20
gamma-HCH (gamma-hexachlorocyclohexane) (HCHG)	244	60	3.3	25
Mercury (HG)	482	286	5.6	9.8
indeno[1,2,3-cd]pyrene (ICDP)	267	105	2.9	5.7
Imposex stage (IMPS)	6	0		
Intersex stage (INTS)	9	7	0	42.9
Monobutyltin (MBTIN)	68	10	0	30
Monophenyltin (MPTIN)	53	0		
(Naphthalene) NAP	213	80	0	37.5
2-hydroxy naphthalene (NAP2OH)	4	0		
Phenanthrene (PA)	271	163	2.5	9.8
1-hydroxy phenanthrene (PA1OH)	4	4	0	25
Lead (PB)	471	269	4.5	15.2
Pyrene (PYR)	252	164	0	7.9
1-hydroxy pyrene (PYR1OH)	25	4	0	50
1-hydroxy pyrene equivalent (PYR1OHEQ)	51	0		
tributyltin (TBT) - expressed as Sn-atom (TBTIN)	78	27	0	81.5
polychlorinated dibenzo-p-dioxins (TCDD)	41	3	66.7	0
Triphenyltin (TPTIN)	62	2	0	0
Vas Deferens Sequence (VDS)	379	133	0	60.9
Zinc (ZN)	457	266	1.5%	10.9%

Annex 3 - Assessment criteria used in the CEMP data assessment[1]

Group	Substance	SEDIMENT (µg/kg dry weight) Background/low concentrations BC	LC Spain	Blue (T_0) < BAC	< BAC Spain	Green (T_1) < EAC	< ERL	MUSSELS (M) AND OYSTERS (O) (µg/kg dry weight except EC for metals: wet weight (ww)) BC/LC	Blue (T_0) < BAC	Green (T_1) < EAC	< EC	FISH (µg/kg wet weight, except: EAC^{passive} for CB: lipid weight (lw)) BC/LC	Blue (T_0) < BAC	Green (T_1) < EAC passive	Amber (T_1) < EC max. food limit
Metals	Cd	200	86	310	129		1200	M-600 O-1800	M-960 O-3000		M-1000 O-1000	a	26		1000 (bivalve. tissue)
	Hg	50	53	70	91		150	M-50 O-100	M-90 O-180		M-500 O-500	a	35		500
	Pb	25000	15500	38000	22400		47000	M-800 O-800	M-1300 O-1300		M-1500 O-1500	a	26		1500 (bivalve. tissue)
	As	15000		25000			---								
	Cr	60000		81000			81000								
	Cu	20000		27000			34000		6000						
	Ni	30000		36000			---								
	Zn	90000		122000			150000		63000						
	TBT	---		---		---		1.0	5.0	12.0					
PAHs	Naphthalene	5	---	8	---		160	---	---	340					
	C1-naphthalene						155[2]								
	C2-naphthalene						150								
	Phenanthrene	17	4.0	32	7.3		240	4.0	11.0	1700					
	C1-phenanthrene						170								
	C2-phenanthrene						200								
	Anthracene	3	1.0	5	1.8		85	---	---	290					
	Dibenzothiophene (DBT)	0.6	---	---	---		190	---	---	---					
	C1-dibenzothiophene						85								
	Fluoranthene	20	7.5	39	14.4		600	5.5	12.2	110					
	Pyrene	13	6.0	24	11.3		665	4.0	9.0	100					
	Benz[a]anthracene	9	3.5	16	7.1		261	1.0	2.5	80					

[1] The assessment criteria are discussed in OSPAR 2009, Background Document on CEMP Assessment Criteria for QSR 2010 (OSPAR Publication 461), which is based *inter alia* on:

- OSPAR 2004, OSPAR/ICES Workshop on the evaluation and update of background reference concentrations (B/RCs) and ecotoxicological assessment criteria (EACs) and how these assessment tools should be used in assessing contaminants in water, sediment and biota (OSPAR Publication 214);
- OSPAR 1996, Report of the Third OSPAR Workshop on Ecotoxicological Assessment Criteria The Hague: 25-29 November 1996 (OSPAR Publication 81).

[2] Sum of 1-methyl naphthalene and 2-methyl naphthalene

Group	Substance	SEDIMENT (µg/kg dry weight)						MUSSELS (M) AND OYSTERS (O) (µg/kg dry weight except EC for metals: wet weight (ww))				FISH (µg/kg wet weight, except: EAC[passive] for CB: lipid weight (lw))			
		Background/low concentrations		Blue (T_0)		Green (T_1)			Blue (T_0)	Green (T_1)			Blue (T_0)	Green (T_1)	Amber (T_1)
		BC	LC Spain	< BAC	< BAC Spain	< EAC	< ERL	BC/LC	< BAC	< EAC	< EC	BC/LC	< BAC	< EAC passive	< EC max. food limit
	Chrysene (Triphenylene)	11	4.0	20	8.0		384	4.0	8.1	---					
	Benzo[a]pyrene	15	4.0	30	8.2		430	0.5	1.4	600					
	Benzo[ghi]perylene	45	3.5	80	6.9		85	1.5	2.5	110					
	Indeno[1,2,3-cd]pyrene	50	4.0	103	8.3		240	1.0	2.4	---					
PCBs	CB28	0.0/0.05		0.22		1.7		0.0/0.25	0.75	3.2		0.0/0.05	0.10	67 lw	
PCBs	CB52	0.0/0.05		0.12		2.7		0.0/0.25	0.75	5.4		0.0/0.05	0.08	108 lw	
PCBs	CB101	0.0/0.05		0.14		3.0		0.0/0.25	0.70	6.0		0.0/0.05	0.08	121 lw	
PCBs	CB105	---		---		---		0.0/0.25	0.75	---		0.0/0.05	0.08	---	
PCBs	CB118	0.0/0.05		0.17		0.6		0.0/0.25	0.60	1.2		0.0/0.05	0.10	25 lw	
PCBs	CB138	0.0/0.05		0.15		7.9		0.0/0.25	0.60	15.8		0.0/0.05	0.09	317 lw	
PCBs	CB153	0.0/0.05		0.19		40		0.0/0.25	0.60	80		0.0/0.05	0.10	1585 lw	
PCBs	CB156	---		---		---		0.0/0.25	0.60	---		0.0/0.05	0.08	---	
PCBs	CB180	0.0/0.05		0.10		12		0.0/0.25	0.60	24		0.0/0.05	0.11	469 lw	
Pesticide	γ-HCH	0.0/0.05	0.13				3.0	0.0/0.25	0.97	1.45	---	---	---	11[b]	
Pesticide	α-HCH	---	---				---	0.0/0.25	0.64	---	---	---	---	---	
Pesticide	DDE (p,p')	0.0/0.05	0.09				2.2	0.0/0.25	0.63	---	---	0.0/0.05	0.10	---	
Pesticide	Hexachlorobenzene	0.0/0.05	0.16				20.0	0.0/0.25	0.63	---	---	0.0/0.05	0.09	---	
Pesticide	Dieldrin	0.0/0.05	0.19				2.0	---	---	---	---	---	---	---	

[a] datasets too limited to allow recommendation for BCs for metals in fish; [b] EAC for fish liver derived by applying a conversion factor of 10 on EAC for whole fish

Notes

- No assessment criteria for PBDE.
- Assessment criteria are used to assess contaminant concentrations
  - **B**ackground **A**ssessment **C**oncentration (BAC)
  - **E**nvironmental **A**ssessment **C**riteria (EAC)
  - **E**ffects **R**ange **L**ow (ERL)
  - **E**uropean **C**ommission food standard (EC)
- BACs were developed by the OSPAR Commission (OSPAR) for testing whether concentrations are near background levels. Mean concentrations significantly below the BAC are said to be near background.
- BACs and EAC[passive]s are available for seven CBs.
- EACs were developed by OSPAR and the International Council for the Exploration of the Sea for assessing the ecological significance of sediment concentrations. Concentrations below the EAC should not cause any chronic effects in marine species.
- BACs and / or EACs are available for ten PAHs.
- EAC[passive]s were developed by OSPAR for assessing the ecological significance of sediment concentrations. Concentrations below the EAC[passive] should cause no chronic effects in marine species.
- ERLs were developed by the United States Environmental Protection Agency for assessing the ecological significance of sediment concentrations. Concentrations below the ERL rarely cause adverse effects in marine organisms. Concentrations *above* the ERM will often cause adverse effects in some marine organisms.
- ECs have been used in the absence of any satisfactory criteria for assessing the ecological significance of biota concentrations. ECs are the maximum acceptable concentrations in food for the protection of public health

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OSPAR's vision is of a clean, healthy and biologically diverse North-East Atlantic used sustainably

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