







25 May 2015 Original: English

MED POL Focal Points Meeting Malta, 16-19 June 2015

Joint Session MED POL and REMPEC Focal Points Meetings Malta, 17 June 2015

An updated list of priority contaminants for the Mediterranean

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#### 1. Introduction

#### 1.1Background

The Contracting Parties to the Barcelona Convention and its Protocols are committed to implement the LBS Protocol through the implementation of a 30 years strategic programme to combat pollution from land based sources (SAP MED) until 2025.

In the framework of the implementation of the Strategic Action Programme (SAP MED), adopted by the 12th Meeting of Contracting Parties to the Barcelona Convention (Monaco, November 2001), the Mediterranean countries prepared Sectoral Plans (SPs) and National Action Plans (NAPs) on the basis of National Diagnostic Analyses (NDAs), and Baseline Budgets (BBs) of emissions/releases.

The preparation of NAPs represents the operational long-term objective of the Strategic Action Programme (SAP), as it is expected to describe the pollution reduction process to be achieved by the countries making use of all the results of the individual activities of the SAP. NAPs were prepared by all countries of the region and the process of preparation was formally endorsed by the 14th Meeting of the Contracting Parties held in Slovenia in November 2005.

The 18<sup>th</sup> Meeting of the Contracting Parties to the Barcelona Convention, 2013, mandated the update of the NAPs by 2015 with the main objective to identify the necessary programmes of measures to meet the obligations under the LBS Protocol/SAP MED which were further specified and strengthened by the Regional Plans for a number of contaminants and sectors adopted by COPs 2009, 2012 and 2014 and meet good Environmental Status on targets related to pollution ecological objectives. The focus of the Programme of Measures of the updated NAPs should be on priority contaminants and relevant sectors affecting the marine environment.

The Med Pol Focal Points at their meeting in 2008 on the Implementation of NAPs and the Preparation of Legally Binding Measures and Timetables agreed to establish an exhaustive list of "substances of concern" for the Mediterranean and a list of "action substances" out of the exhaustive list of "substances of concern" according to the following criteria:

- the substance is covered by regional and/or international instruments regulating its use, release or phasing out, and
- the substance has an ELV or its ELV is under development either at national or at regional levels, and
- the substance and /or its high input could represent a risk to the marine environment or human health

The list of "action substances" was not exhaustive. A Party could propose the inclusion of a substance, falling under the above criteria, during the Contracting Parties meetings.

Due to legal and regulatory development in the MAP system as well as at regional and global level since 2008, there is a need to review the priority contaminant list taking also into account the results of reporting of pollutant loads by the Contracting Parties as well as marine pollution data.

#### 1.2Objectives and scope

The aim of this report is to recommend an updated list of priority contaminants, to enable the Contracting Parties to design realistic NAP programmes of measures for their reduction, elimination and/or phasing out. This would also contribute to the setting or updating the ELV and/or environmental targets for such substances accordingly.

To this end, the following objectives have been established:

- To identify common substances of concern for the marine environment through an international comparison of updated lists of priority substances.
- To review available updated information on the <u>occurrence</u> of substances of concern in the Mediterranean marine environment.

To review available data on the emissions of substances of concern.

To identify updated <u>ELVs</u> already adopted in other international legal frameworks or regional marine conventions, to reduce pollution from substances of concern.

The document is structured in four sections each one addressing the objectives indicated above.

#### 2. Substances of concern in the marine environment – an update

In this section an identification of those substances of concern for the marine environment (as a consequence for example of its toxicological properties) is carried out, as well as an analysis on how these substances have been prioritised in different international and regional conventions.

#### 2.1 Addressing substances of concern in the Mediterranean: the LBS Protocol and the SAP

#### 2.1.1 Background

The Protocol for the Protection of the Mediterranean Sea against Pollution from Land-Based Sources and Activities (**LBS Protocol**) is the main legal reference regarding the reduction of pollutants within the region. According to its general provision (art.1), the Parties "shall take all appropriate measures to prevent, abate, combat and eliminate to the fullest possible extent pollution of the Mediterranean Sea Area caused by discharges from rivers, coastal establishments or outfalls, or emanating from any other land-based sources and activities within their territories, giving priority to the phasing out of inputs of substances that are toxic, persistent and liable to bioaccumulate".

To this end, the Parties shall elaborate and implement national and regional action plans and programmes, containing measures and timetables for their implementation (art. 5). The Protocol includes in its Annex 1 some elements to be taken into account in the preparation of these plans and programmes, like sectors of activity to be primarily considered, characteristics of substances in the environment, and categories of substances to be considered.

The 1996 amendments to the LBS Protocol (initially adopted in 1980) entered into force in 2008. This accelerated the process for Mediterranean countries to develop short term and medium term regional action plans and programmes containing legally binding measures and timetables for their implementation, as stated in article15 of the Barcelona Convention. According to article 15, the plans and the programmes must be approved within one year following the entry into force of the protocol and then become legally binding.

Previously, the MED POL Programme had been preparing the ground for the preparation of the above plans and measures through a process started in 1996 that culminated in the adoption in 1997 of the Strategic Action Programme (SAP) to address pollution from land-based sources and activities and of National Action Plans, recently endorsed by the countries.

The **Strategic Action Programme** (**SAP**) is an action-oriented initiative identifying priority target categories of polluting substances and activities to be eliminated or controlled by the Mediterranean countries through a planned timetable (up to the year 2025) for the implementation of specific pollution reduction measures and interventions.

During 2004–2005, all Mediterranean Countries prepared **National Action Plans** (**NAPs**) which describe what pollution reduction measures they will implement according to the SAP, by when and with what funds, taking into consideration the environmental and socio-economic issues, legislative frameworks, and the available infrastructure.

With regard to chemical contaminants the SAP MED has the following targets:

- 1. Phase out inputs of 9 pesticides and PCBs by 2010 (Aldrin, DDT, Dieldrin, Endrin, Chlordane, Heptachlor, Mirex, Toxaphene, PCB/PCT)
- 2. Reduce to the fullest possible extent hexachloro benzene, dioxins and furans by 2010;
- 4. Phase out inputs of PAHs by 2025;

- 5. Phase out to the fullest possible extent discharges and emissions and losses of heavy metals (mercury, cadmium and lead) by 2025;
- 6. Phase out to the fullest possible extent discharges and emissions and losses of organotin compounds by 2010 (Butyltin compounds);
- 7. Reduce discharges, emissions and losses of zinc, copper and chromium by 2010 and eliminate by 2025;
- 8. Reduce discharges, emissions and losses of organohalogen compounds into the Mediterranean sea by 2010 and eliminate by 2025
- 2.1.2 Regulatory developments with regard to priority contaminants Legally binding measures under the LBS Protocol

Six legally binding measures (Regional Plans) were elaborated and adopted in the framework of the implementation of Articles 5 and 15 of the LBS Protocol. The origin of these measures can be traced back to the year 2008 when MED POL Focal Points agreed to establish a List of 'action' priority substances. The 'action' priority substances as regards chemical contaminants consist of Metals, Organic metallic compounds, Polychlorinated Biphenyls (PCBs), Polychlorinated dibenzodioxins (PCDDs), Polychlorinated dibenzofurans (PCDFs), Volatile Organic Compounds and POPs. In principle, it was agreed that in order to propose measures, the substance:

- a) Is covered by regional and/or international instruments regulating its use, release or phasing out;
- b) Should have an Emission Limit Value (ELV) or its ELV is under development either at national or at regional levels;
- c) The substance and/or its high input could represent a risk to the marine environment or human health; and
- d) The Parties may propose additional substances at the Contracting Parties meetings.

Based on this process, the following Regional Plans, as regards chemical contaminants, were prepared and adopted by the Contracting Parties in 2009 and 2012:

- Elimination of the POPs Aldrin, Chlordane, Dieldrin, Endrin, Heptachlor, Mirex and Toxaphene (Decision IG.19/8, effective 2011 and 2012 for stock piles).
- Phasing out of DDT (Decision IG.19/9, effective 2011 and 2012 for stock piles).
- Reduction of inputs of Mercury (Decision IG.20/8).
- Phasing out of the POPs hexabromodiphenyl ether, heptabromodiphenyl ether, tetrabromodiphenyl ether and pentabromodiphenyl ether (Decision IG20/8, effective 2013 latest)
- Phasing out of the POPs lindane and endosulphan (Decision IG20/8, effective 2013 latest)
- Phasing out of the POPs perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride (Decision IG20/8, effective 2013 latest)
- Elimination of the POPs alpha hexachlorocyclohexane, beta hexachlorocyclohexane, chlordecone, hexabromobiphenyl ether, pentachlorobenzene (Decision IG20/8, effective 2013 latest)

2.1.3 Good environmental status with regard to contaminants in the framework of the implementation of the ecosystem approach for the management of human activities in the Mediterranean Sea.

The Contracting Parties to the Barcelona Convention, decided in their 15<sup>th</sup> meeting held in January 2008 in Almeria, Spain (Decision IG 17/6), that UNEP/MAP should gradually implement the ecosystem approach for the management of human activities in the Mediterranean Sea (hereby mentioned as the ecosystem approach) with the overall aim of achieving a good environmental status (GES) of the Mediterranean sea by 2020. The Contracting Parties further decided in their 15th meeting a roadmap for the implementation of the ecosystem approach consisting of seven steps<sup>1</sup>.

In their 17<sup>th</sup> Meeting (Decision IG 20/4), the Contracting Parties adopted 11 Mediterranean Ecological Objectives associated with 28 Operational Objectives and 61 Indicators. In their 18<sup>th</sup> Meeting (Decision IG 21/3) the Contracting Parties adopted an integrated list of Mediterranean Good Environmental Status and related targets, associated with the Operational Objectives and Indicators.

As regards contaminants the Contracting Parties adopted the Operational Objectives, Indicators, GES and targets as shown in the following table:

ECAP Operational Objectives, Indicators, GES and targets for contaminants

Operational Objective	Indicator	GES	Proposed Targets
9.1 Concentration of priority2 contaminants is kept within acceptable limits and does not increase	9.1.1 Concentration of key harmful contaminants <sup>3</sup> in biota, sediment or water	Level of pollution is below a determined threshold defined for the area and species	State Concentrations of specific contaminants below EACs or below reference concentrations <sup>4</sup> No deterioration trend in contaminants concentrations in sediment and biota from human impacted areas, statistically defined.  Pressure Reduction of contaminants emissions from land based sources <sup>5</sup>

<sup>&</sup>lt;sup>1</sup> The roadmap comprises: 1) establishing the vision for an ecosystem approach throughout the Mediterranean; 2) elaborating three strategic goals to achieve this vision; 3) undertaking an initial assessment to determine priority issues, information availability as well as gaps that need to be filled; 4) deciding on ecological objectives; 5) determining operational objectives and associated indicators and identifying targets or thresholds for those indicators; 6) developing a monitoring strategy; and 7) elaborating specific management plans and actions that will ensure that ecological objectives and strategic goals are met

<sup>&</sup>lt;sup>2</sup> Priority contaminants as listed under the Barcelona Convention and LBS Protocol.

<sup>&</sup>lt;sup>3</sup> Use for further work on reference conditions ERL for sediments taking into account specifics of the Mediterranean.

<sup>&</sup>lt;sup>4</sup> Thresholds to be set by COP19.

<sup>&</sup>lt;sup>5</sup> Reduction programmes are already in place through the Protocols of the Barcelona Convention and the Marine Litter Regional Strategy.

Operational Objective	Indicator	GES	Proposed Targets
9.2 Effects of released contaminants are minimized	9.2.1 Level of pollution effects of key contaminants where a cause and effect relationship has been established	Concentrations of contaminants are not giving rise to acute pollution events	State Contaminants effects below threshold <sup>6</sup> Decreasing trend in the operational releases of oil and other contaminants from coastal, maritime and off-shore activities.
9.3 Acute pollution events are prevented and their impacts are minimized	9.3.1 Occurrence, origin (where possible), extent of significant acute pollution events (e.g. slicks from oil, oil products and hazardous substances) and their impact on biota affected by this pollution	Occurrence of acute pollution events are reduced to the minimum.	Pressure Decreasing trend in the occurrences of acute pollution events
9.4 Levels of known harmful contaminants in major types of seafood do not exceed established standards	9.4.1 Actual levels of contaminants that have been detected and number of contaminants which have exceeded maximum regulatory levels in commonly consumed seafood <sup>7</sup>	Concentrations of contaminants are within the regulatory limits for consumption by humans	State Concentrations of contaminants are within the regulatory limits set by legislation
	9.4.2 Frequency that regulatory levels of contaminants are exceeded	No regulatory levels of contaminants in seafood are exceeded	State Decreasing trend in the frequency of cases of seafood samples above regulatory limits for contaminants
9.5 Water quality in bathing waters and other recreational areas does not undermine human health	9.5.1 Percentage of intestinal enterococci concentration measurements within established standards	Concentrations of intestinal enterococci are within established standards	State Increasing trend in the percentage of intestinal enterococci concentration measurements within established standards

## 2.2 International regulatory framework

To undertake an international comparison, we have selected those regional marine conventions and international legal frameworks where a systematic procedure have been established to identify, prioritise and approve lists of substances of concern for the aquatic or marine environment. A brief description of the most recent lists and procedures is provided below.

<sup>&</sup>lt;sup>6</sup> Thresholds to be set by COP19.

<sup>&</sup>lt;sup>7</sup> Traceability of the origin of seafood sampled should be ensured.

#### 2.2.1 **UN Chemicals and Wastes instruments**

The cluster of chemicals and wastes conventions (Basel, Rotterdam and Stockholm Conventions) are multilateral environmental agreements, which share the common objective of protecting human health and the environment from hazardous chemicals and wastes. Since 2008 a 'synergies' process has been formally adopted aiming at enhancing cooperation and coordination among these three conventions. As part of this process, there has been a convergence of procedures, for example joint Conferences of Parties from 2010, and, since 2013, joint meetings of the scientific committees of the Stockholm and Rotterdam Conventions responsible for recommending the addition of new chemicals to the conventions.

While in practice, the priority chemicals covered by these conventions are automatically included in the EU legislation, they represent an absolute priority list, as even the non-EU member Parties to the Barcelona Convention are legally obliged to address these chemicals and wastes as a consequence of (most of) their ratification of the chemicals and wastes conventions.

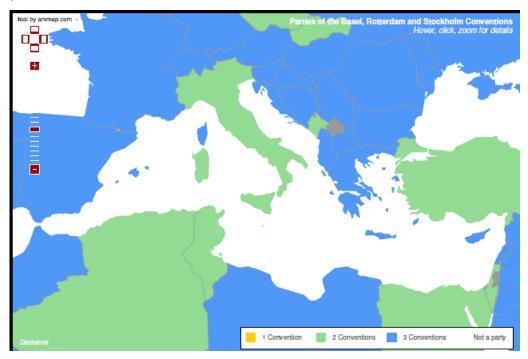


Figure: Status of ratification of Mediterranean countries of the Basel, Rotterdam and Stockholm Conventions (Source: http://synergies.pops.int/)

#### The Stockholm Convention

The Stockholm Convention is a global treaty to protect human health and the environment from persistent organic pollutants that has been signed by 179 governments<sup>8</sup>. In implementing the Convention, which was adopted in 2001 and entered into force 2004, governments will have to take measures to eliminate or reduce the release of POPs into the environment. Under the Convention, any Party can submit proposals for listing new chemicals, which may be added by decisions of the Conference of Parties (CoP) to Annex A for global elimination, Annex B for restriction, and/or Annex C as unintentionally produced products. To date the Stockholm Convention has listed 23 priority **POPs** to its three Annexes: the initial (and largely out of use) 12 POPs, and a futher 11 POPs added in 2009 (9 chemicals), 2011 (1 chemical), 2013 (1 new chemical) and 2015 (3 new chemicals.

The Stockholm Convention on POPs and other international agreements state that monitoring activities should be established to verify the effective implementation of the conventions and the decrease of environmental levels of persistent pollutants. In 2012 the first 'Global Monitoring Plan' report was published, a global assessment of the prevalence of the initial list of 12 POPs in the environment; and in 2015, the second monitoring regional reports<sup>9</sup>, which will include all 23 currently listed POPs. In addition to the Global Monitoring Plan, the convention requires Parties to update their National

<sup>&</sup>lt;sup>8</sup> http://www.pops.int

<sup>&</sup>lt;sup>9</sup> http://chm.pops.int/Programmes/GlobalMonitoringPlan/MonitoringReports/tabid/525/Default.aspx

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Implementation Plans (NIP) to address any newly listed chemicals, including and submit these to the Secretariat within two years. While most of the Mediterranean countries have not submitted their revised NIPs as of early 2015, Croatia, Egypt, Libya, and Montenegro all have active projects developing them. National Reports on the status of implementation of the Stockholm Convention have been submitted<sup>10</sup> under the third round (in 2014) by Albania, Croatia, France, Spain, Tunisia and Turkey.

Listed POPs	Entry into force, Annex
	(es)
Aldrin	2004, A
Alpha hexachlorocyclohexane	2010, A
Beta hexachlorocyclohexane	2010, A
Chlordane	2004, A
Chlordecone	2010, A
DDT	2004, B
Dieldrin	2004, A
Endrin	2004, A
Heptachlor	2004, A
Hexachlorobenzene (HCB)	2004, A and C
Hexabromocyclodeodecane	2014, A
Hexabromobiphenyl	2010, A
Hexabromodiphenyl ether and heptabromodiphenyl ether (commercial	2010, A
octabromodiphenyl ether)	
Lindane	2010, A
Mirex	2004, A
Pentachlorobenzene	2010, A and C
Perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride	2010, B
Polychlorinated biphenyls (PCB)	2004, A and C
Polychlorinated dibenzo-p-dioxins (PCDD)	2004, C
Polychlorinated dibenzofurans (PCDF)	2004, C
Technical endosulfan and its related isomers	2012, A
Toxaphene	2004, A
Tetrabromodiphenyl ether and pentabromodiphenyl ether (commercial	2010, A
pentabromodiphenyl ether)	
Hexachlorobutadiene	2016, A
Polychlorinated napthalenes	2016, A and C
Pentachlorophenol and its salts and esters	2016, A

#### **Rotterdam Convention**

The Rotterdam Convention is a global treaty to promote shared responsibility for chemicals, aiming to promote shared responsibility and cooperative efforts among Parties in the international trade of certain hazardous chemicals in order to protect human health and the environment from potential harm; and to contribute to the environmentally sound use of those hazardous chemicals, by facilitating information exchange about their characteristics, by providing for a national decision-making process on their import and export and by disseminating these decisions to Parties. It has 154 Parties in 2015. While the chemicals listed in the Rotterdam Convention are not banned, they represent priority chemicals that are known to cause unacceptable health and/or environmental effects in more than two regions, and have been banned or severely restricted by national governments for this reason.

The Convention provides for a Prior Informed Consent procedure for imports of chemicals listed in its Annex III, a total of 47 pesticides and industrial chemicals that have been banned or severely restricted for health or environmental reasons by two or more Parties, and which the Conference of the Parties has decided to subject to the PIC procedure (see <u>Appendix 1x</u>). Exports of chemicals listed in Annex

<sup>&</sup>lt;sup>10</sup> http://chm.pops.int/Countries/NationalReports/ThirdRoundPartyReports/tabid/4470/Default.aspx

III must be accompanied by a notificiation by the exporting Party, and comply with labelling and information provision requirements.

#### **Basel Convention**

The Basel Convention was adopted in 1989 with the aims of reducing hazardous waste generation and the promotion of environmentally sound management of hazardous wastes, wherever the place of disposal; restricting transboundary movements of hazardous wastes except where it is perceived to be in accordance with the principles of environmentally sound management; and establishing a regulatory system for cases where transboundary movements are permissible. It has 181 Parties in 2015.

It defines "hazardous wastes" as a function of their source, composition, characteristics, as well as their regulatory status in Party states (e.g. any waste listed as 'hazardous' in national legislation would be covered by the convention even if not explicitly included in its Annexes). Annex I identifies 18 waste streams (e.g. clinical wastes, manufacturing or waste disposal operations), and 27 waste constituents (see Annex 2). Annex III lists 14 Hazard Characteristics; while two additional lists were added in 1998 which identify waste types that are (Annex VIII, List A) and are not (Annex IX, List B) covered by the convention.

Strategic Approach to International Chemicals Management (SAICM)

The Strategic Approach to International Chemicals Management (SAICM) is a policy framework to promote chemical safety around the world. SAICM has as its overall objective the achievement of the sound management of chemicals throughout their life cycle so that, by 2020, chemicals are produced and used in ways that minimize significant adverse impacts on human health and the environment. This "2020 goal" was adopted by the World Summit on Sustainable Development in 2002 as part of the Johannesburg Plan of Implementation.

SAICM comprises the Dubai Declaration on International Chemicals Management, expressing high-level political commitment to SAICM, and an Overarching Policy Strategy which sets out its scope, needs, objectives, financial considerations underlying principles and approaches and implementation and review arrangements. Objectives are grouped under five themes: risk reduction; knowledge and information; governance; capacity-building and technical cooperation; and illegal international traffic. The Declaration and Strategy are accompanied by a Global Plan of Action that serves as a working tool and guidance document to support implementation of SAICM and other relevant international instruments and initiatives. Activities in the plan are to be implemented, as appropriate, by stakeholders, according to their applicability.

One of the functions of SAICM is to call for appropriate action on emerging policy issues as they arise and to forge consensus on priorities for cooperative action. So far resolutions have been adopted on the following issues:

- Lead in Paint
- Chemicals in Products
- Hazardous substance within the life cycle of electrical and electronic products
- Nanotechnolgy and manufactured nanomaterials
- Endocrine-disrupting chemicals
- Perfluorinated chemicals and the transition to safer alternatives

Additional issues have been raised, and it is anticipated that resolutions may be adopted for them in the International Conference on Chemical Management to be held in 2015. These include Highly Hazardous Pesticides and Environmentally Persistent Pharmaceutical Pollutants. Proposed criteria for HHP include:

- Pesticide formulations that meet the criteria of classes Ia or Ib of the WHO Recommended Classification of Pesticides by Hazard;
- Pesticide active ingredients and their formulations that meet the criteria of carcinogenicity Categories 1A and 1B of the Globally Harmonized System on Classification and Labelling of Chemicals (GHS):

- Pesticide active ingredients and their formulations that meet the criteria of mutagenicity Categories 1A and 1B of the Globally Harmonized System on Classification and Labelling of Chemicals (GHS);
- Pesticide active ingredients and their formulations that meet the criteria of reproductive toxicity Categories 1A and 1B of the Globally Harmonized System on Classification and Labelling of Chemicals (GHS);
- Pesticide active ingredients listed by the Stockholm Convention in its Annexes A and B, and those meeting all the criteria in paragraph 1 of annex D of the Convention;
- Pesticide active ingredients and formulations listed by the Rotterdam Convention in its Annex III;
- Pesticides listed under the Montreal Protocol;
- Pesticide active ingredients and formulations that have shown a high incidence of severe or irreversible adverse effects on human health or the environment.

#### 2.2.2 European Union

#### 2.3.2.1 The Water Framework Directive (WFD)

Directive 2000/60/EC, of 23 October 2000, establishing a framework for Community action in the field of water policy (Water Framework Directive WFD)<sup>11</sup>, was set with the intention of providing an adequate global policy framework for the management of the Community water domain in an attempt to reach a final good ecological status for all water bodies. The WFD provides the major common principles that must prevail when adopting new policies to protect and improve the quality of the aquatic environment.

The WFD commits EU Member States to achieve good qualitative and quantitative ecological and chemical status of all water bodies (including marine waters up to one nautical mile from shore) by 2015. In terms of the management of <u>hazardous substances</u>, the WFD requires "the progressive reduction of emissions of dangerous substances to water", the final objective being "the reaching of background values for naturally occurring substances and close to zero for man-made synthetic substances". The term 'hazardous substances' is defined as "substances or groups of substances that are toxic, persistent and liable to bio-accumulate and other substances or groups of substances which give rise to an equivalent level of concern".

Article 16 of the WFD sets out "Strategies against pollution of water" outlining the steps to be taken. The first step was to establish by way of Decision 2455/2001/EC a First list of priority substances to become Annex X of the WFD. These substances were selected from amongst those presenting a significant risk to or via the aquatic environment, using the approaches outlined in Article 16 of the WFD.

This first list was replaced by Annex II of the <u>Directive on Environmental Quality Standards</u> (<u>Directive 2008/105/EC</u>) (EQSD), also known as the Priority Substances Directive, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC. The Priority Substances Directive, published in the Official Journal on 24 December 2008, set environmental quality standards (EQS) for the substances in surface waters (river, lake, transitional and coastal) and confirmed their designation as priority or priority hazardous substances, the latter being a subset of particular concern. The EQSD established:

in Annex I, limits on concentrations of the priority substances in surface waters of <u>33 priority</u> substances and 8 other pollutants;

<sup>&</sup>lt;sup>11</sup>http://eur-lex.europa.eu/resource.html?uri=cellar:5c835afb-2ec6-4577-bdf8-756d3d694eeb.0004.02/DOC 1&format=PDF

the list of 33 priority substances in Annex II as Annex X of the Water Framework Directive (WFD); the possibility of applying EQS for sediment and biota, instead of those for water;

the possibility of designating mixing zones adjacent to discharge points where concentrations of the substances in Annex I might be expected to exceed their EQS;

a requirement for Member States to establish an inventory of emissions, discharges and losses of the substances in Annex I;

an obligation to review the list of priority substances by 13 January 2011.

By replacing <u>five older directives</u>, with effect from 22 December 2012, the EQSD contributed to the Commission's Better Regulation initiative.

As required by the WFD and EQSD, the Commission subsequently reviewed the list of priority substances and in 2012 put forward a proposal (COM(2011)876), for a Directive amending the WFD and the EQSD as regards priority substances.

According to Annex V, point 1.4.3 of the WFD and Article 1 of the EQSD, good chemical status is reached for a water body when it complies with the EQS for all the priority substances and other pollutants listed in Annex I of the EQSD.

<u>COM(2011)876</u>) included a revised (second) list of priority substances, and provisions to improve the functioning of the legislation. The main features of the proposal were:

15 additional priority substances, 6 of them designated as priority hazardous substances;

stricter EQS for four existing priority substances and slightly revised EQS for three others;

the designation of two existing priority substances as priority hazardous substances;

the introduction of biota standards for several substances;

provisions to improve the efficiency of monitoring and the clarity of reporting with regard to certain substances behaving as ubiquitous persistent, bioaccumulative and toxic (PBT) substances;

a provision for a watch-list mechanism designed to allow targeted EU-wide monitoring of substances of possible concern to support the prioritisation process in future reviews of the priority substances list.

Subsequent to the Commission proposal, <u>Directive 2013/39/EU</u> of 12 August 2013 added the following 12 substances to Annex X of the Water Framework Directive:

- Dicofol [CAS# 115-32-2]
- Perfluorooctane sulfonic acid and its derivatives (PFOS) [CAS# 1763-23-1]
- Quinoxyfen [CAS# 124495-18-7]
- Dioxins and dioxin-like compounds
- Aclonifen [CAS# 74070-46-5]
- Bifenox [CAS# 42576-02-3]
- Cybutryne [CAS# 28159-98-0]
- Cypermethrin [CAS# 52315-07-8]
- Dichlorvos [CAS# 62-73-7]
- Hexabromocyclododecanes (HBCDD)
- Heptachlor [CAS# 76-44-8] and Heptachlor epoxide [CAS# 1024-57-3]
- Terbutryn [CAS# 886-50-0]

The full list of the 45 WFD Annex X priority substances is presented in the following table. For newly-identified substances, the maximum permitted concentrations in water set in the Environmental Quality Standards (EQS) will take effect in 2018. The aim is to achieve good chemical status for these substances by 2027. To this end, EU Member States are required to submit supplementary programmes of measures and monitoring programmes to the Commission by 2018. Revised EQS for

existing substances are to be included in River Basin Management Plans in 2015, with the aim of achieving good surface water chemical status for these substances by 2021.

## Next Steps

- 2015: Updated standards for existing substances to be included in River Basin Management Plans
- 2018: New concentration limits start applying for newly-identified substances
- By 2018: EU member states to submit monitoring programmes to the European Commission for newly-identified substances
- 2021: Target date for achieving good chemical status for existing substances (revised standards)
- 2027: Target date for achieving good chemical status for newly-identified

In addition, Article 8b of Directive2013/39/EU states that a new mechanism is needed to provide high-quality monitoring information on the concentrations of potentially polluting substances in the aquatic environment to support future prioritisation exercises in accordance with Article 16(2) of the Water Framework Directive, and thereby to improve the protection of the aquatic environment and of human health via the environment. The mechanism is aimed at emerging pollutants and other substances for which the available monitoring data are either insufficient or of insufficient quality for the purpose of identifying the risk posed across the EU. It involves including a limited number of such substances in a "Watch List", and monitoring them EU-wide at selected representative monitoring stations over at least a 12-month period, and for up to four years.

Frequent reviews of the list will ensure that substances are not monitored for longer than necessary, and that substances posing a significant risk at EU level are identified as candidate priority substances with as little delay as possible. Article 8b sets out the information to be taken into account when identifying substances to include in the Watch List. The first list may contain a maximum of 10 substances or groups of substances and should indicate the monitoring matrices and the possible methods of analysis not entailing excessive costs for each substance. A suspected significant risk at European Union level to, or via, the aquatic environment, and a lack of sufficient monitoring data are both regarded as conditions for a substance selection (Loos, 2015). The Commission, according to Directive 2013/39/EU is to establish the first watch list by 14 September 2014 and shall update it every 24 months thereafter.

The following 3 substances were however immediately included in the first watch list after the adoption of Directive 2013/39/EU for the purpose of facilitating the determination of appropriate measures to address the risk posed by those substances:

- Diclofenac [CAS# 15307-79-6]
- 17-beta-estradiol (E2) [CAS# 50-28-2]
- 17-alpha-ethinylestradiol (EE2) [CAS# 57-63-6]

The Joint Research Centre (JRC) has been tasked with proposing seven substances as candidates for the completion of the first Watch List and identifying analytical methods for their monitoring. The procedure and criteria used to identify the substances for possible inclusion in the Watch List is described in the JRC Science and Policy Report "Development of the 1st Watch List under the Environmental Quality Standards Directive" (Carvalho et al., 2015). The availability of analytical methods was a criterion for the selection of the compounds.

The finally proposed 10 (groups of) substances for inclusion in the 1st Watch List are:

- 1. Diclofenac
- 2. 17-Beta-estradiol (E2), Estrone (E1)
- 3. Alpha-ethinylestradiol (EE2)
- 4. Oxadiazon
- 5. Tri-allate
- 6. Methiocarb
- 7. 2, 6-ditert-butyl-4-methylphenol
- 8. Neonicotinoid insecticides as a group: Imidacloprid, Thiacloprid, Thiamethoxam, Clothianidin, Acetamiprid
- 9. Macrolide antibiotics: Erythromycin Clarithromycin, Azithromycin
- 10. 2-Ethylhexyl 4-methoxycinnamate

Analytical methods for additional substances among those considered for inclusion in the list were searched and investigated but either did not fulfil all selection criteria, in some cases because enough monitoring data were found to exist already, or were not ranked highly enough. These substances were trichlorfon, cyclododecane, aminotriazole (amitrole), dimethenamid-P, diflufenican, dichlofluanid, formaldehyde, triphenyl phosphate, tolylfluanid, ciprofloxacin, and free cyanide. Little or no information on analytical methods was found for trichlorfon, aminotriazole, cyclododecane, and tolylfluanid. For the other compounds analytical methods are available and published. Some of them have already been analysed in the aquatic environment. The analysis of free cyanide in water is difficult. The available analytical methods do not reach the proposed PNEC value of 0.26  $\mu$ g/l (Loos, 2015).

The 45 WFD Priority substances listed under Directive 2013/39/EC, amending the Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy.

Number	CAS number (1)	EU number (2)	Name of priority substance (3)	Identified as priority hazardous substance
(1)	15972-60-8	240-110-8	Alachlor	
(2)	120-12-7	204-371-1	Anthracene	X
(3)	1912-24-9	217-617-8	Atrazine	
(4)	71-43-2	200-753-7	Benzene	
(5)	not applicable	not applicable	Brominated diphenylethers	$X^{(4)}$
(6)	7440-43-9	231-152-8	Cadmium and its compounds	X
(7)	85535-84-8	287-476-5	Chloroalkanes, C 10-13	X
(8)	470-90-6	207-432-0	Chlorfenvinphos	
(9)	2921-88-2	220-864-4	Chlorpyrifos (Chlorpyrifosethyl)	
(10)	107-06-2	203-458-1	1,2-dichloroethane	
(11)	75-09-2	200-838-9	Dichloromethane	
(12)	117-81-7	204-211-0	Di(2-ethylhexyl)phthalate (DEHP)	X

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(13)	330-54-1	206-354-4	Diuron	
(14)	115-29-7	204-079-4	Endosulfan	X
(15)	206-44-0	205-912-4	Fluoranthene	
(16)	118-74-1	204-273-9	Hexachlorobenzene	X
(17)	87-68-3	201-765-5	Hexachlorobutadiene	X
(18)	608-73-1	210-168-9	Hexachlorocyclohexane	X
(19)	34123-59-6	251-835-4	Isoproturon	
(20)	7439-92-1	231-100-4	Lead and its compounds	
(21)	7439-97-6	231-106-7	Mercury and its compounds	X
(22)	91-20-3	202-049-5	Naphthalene	
(23)	7440-02-0	231-111-4	Nickel and its compounds	
(24)	not applicable	not applicable	Nonylphenols	X <sup>(5)</sup>
(25)	not applicable	not applicable	Octylphenols <sup>(6)</sup>	
(26)	608-93-5	210-172-0	Pentachlorobenzene	X
(27)	87-86-5	201-778-6	Pentachlorophenol	
(28)	not applicable	not applicable	Polyaromatic hydrocarbons (PAH) <sup>(7)</sup>	X
(29)	122-34-9	204-535-2	Simazine	
(30)	not applicable	not applicable	Tributyltin compounds	$X^{(8)}$
(31)	12002-48-1	234-413-4	Trichlorobenzenes	
(32)	67-66-3	200-663-8	Trichloromethane (chloroform)	
(33)	1582-09-8	216-428-8	Trifluralin	X
(34)	115-32-2	204-082-0	Dicofol	X
(35)	1763-23-1	217-179-8	Perfluorooctane sulfonic acid and its derivatives (PFOS)	X
(36)	124495-18-7	not applicable	Quinoxyfen	X
(37)	not applicable	not applicable	Dioxins and dioxin-like compounds	$X^{(9)}$
(38)	74070-46-5	277-704-1	Aclonifen	
(39)	42576-02-3	255-894-7	Bifenox	
(40)	28159-98-0	248-872-3	Cybutryne	
(41)	52315-07-8	257-842-9	Cypermethrin <sup>(10)</sup>	
(42)	62-73-7	200-547-7	Dichlorvos	
(43)	not applicable	not applicable	Hexabromocyclododecanes (HBCDD)	$\mathbf{X}^{(11)}$
(44)	76-44-8/	200-962-3/	Heptachlor and heptachlor epoxide	X
(45)	886-50-0	212-950-5	Terbutryn	

<sup>(1)</sup> CAS: Chemical Abstracts Service.

- (2) EU-number: European Inventory of Existing Commercial Substances (EINECS) or European List of Notified Chemical Substances (ELINCS)
- (3) Where groups of substances have been selected, unless explicitly noted, typical individual representatives are defined in the context of the setting of environmental quality standards.
- (4) Only Tetra, Penta, Hexa and Heptabromodiphenylether (CAS -numbers 40088-47-9, 32534-81-9, 36483-60-0, 68928-80-3, respectively).
- (5) Nonylphenol (CAS 25154-52-3, EU 246-672-0) including isomers 4-nonylphenol (CAS 104-40-5, EU 203-199-4) and 4-nonylphenol (branched) (CAS 84852-15-3, EU 284-325-5).
- (6) Octylphenol (CAS 1806-26-4, EU 217-302-5) including isomer 4-(1,1',3,3'-tetramethylbutyl)-phenol (CAS 140-66-9, EU 205-426-2).
- (7) Including benzo(a)pyrene (CAS 50-32-8, EU 200-028-5), benzo(b)fluoranthene (CAS 205-99-2, EU 205-911-9), benzo(g,h,i)perylene (CAS 191-24-2, EU 205-883-8), benzo(k)fluoranthene (CAS 207-08-9, EU 205-916-6), indeno(1,2,3-cd)pyrene (CAS 193-39-5, EU 205-893-2) and excluding anthracene, fluoranthene and naphthalene, which are listed separately.
- (8) Including tributyltin-cation (CAS 36643-28-4).
- (9) This refers to the following compounds: 7 polychlorinated dibenzo-p-dioxins (PCDDs): 2,3,7,8-T4CDD (CAS 1746-01-6), 1,2,3,7,8-P5CDD (CAS 40321-76-4), 1,2,3,4,7,8-H6CDD (CAS 39227-28-6), 1,2,3,6,7,8-H6CDD (CAS 57653-85-7), 1,2,3,7,8,9-H6CDD (CAS 19408-74-3), 1,2,3,4,6,7,8-H7CDD (CAS 35822-46-9), 1,2,3,4,6,7,8,9-O8CDD (CAS 3268-87-9)

10 polychlorinated dibenzofurans (PCDFs): 2,3,7,8-T4CDF (CAS 51207-31-9), 1,2,3,7,8-P5CDF (CAS 57117-41-6), 2,3,4,7,8-P5CDF (CAS 57117-31-4), 1,2,3,4,7,8-H6CDF (CAS 70648-26-9), 1,2,3,6,7,8-H6CDF (CAS 57117-44-9), 1,2,3,7,8,9-H6CDF (CAS 72918-21-9), 2,3,4,6,7,8-H6CDF (CAS 60851-34-5), 1,2,3,4,6,7,8-H7CDF (CAS 67562-39-4), 1,2,3,4,7,8,9-H7CDF (CAS 55673-89-7), 1,2,3,4,6,7,8,9-O8CDF (CAS 39001-02-0) 12 dioxin-like polychlorinated biphenyls (PCB-DL): 3,3',4,4'-T4CB (PCB 77, CAS 32598-13-3), 3,3',4',5-T4CB (PCB 81, CAS 70362-50-4), 2,3,3',4,4'-P5CB (PCB 105, CAS 32598-14-4), 2,3,4,4',5-P5CB (PCB 114, CAS 74472-37-0), 2,3',4,4',5-P5CB (PCB 118, CAS 31508-00-6), 2,3',4,4',5'-P5CB (PCB 123, CAS 65510-44-3), 3,3',4,4',5-P5CB (PCB 126, CAS 57465-28-8), 2,3,3',4,4',5-H6CB (PCB 156, CAS 38380-08-4), 2,3,3',4,4',5'-H6CB (PCB 157, CAS 69782-90-7), 2,3',4,4',5,5'-H6CB (PCB 167, CAS 52663-72-6), 3,3',4,4',5,5'-H6CB (PCB 169, CAS 32774-16-6), 2,3,3',4,4',5,5'-H7CB (PCB 189, CAS 39635-31-9).

- (10) CAS 52315-07-8 refers to an isomer mixture of cypermethrin, alpha-cypermethrin (CAS 67375-30-8), beta-cypermethrin (CAS 65731-84-2), theta-cypermethrin (CAS 71697-59-1) and zeta-cypermethrin (52315-07-8).
- (11) This refers to 1,3,5,7,9,11-Hexabromocyclododecane (CAS 25637-99-4), 1,2,5,6,9,10- Hexabromocyclododecane (CAS 3194-55-6),  $\alpha$ -Hexabromocyclododecane (CAS 134237-50-6),  $\beta$ -Hexabromocyclododecane (CAS 134237-51-7) and  $\gamma$  Hexabromocyclododecane (CAS 134237-52-8).

#### 2.3.2.2 The REACH Regulation

Regulation (EC) No 1907/2006<sup>12</sup> concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), (establishing a European Chemicals Agency, amending Directive 1999/45/EC and repealing Council Regulation (EEC) No 793/93 and Commission Regulation (EC) No 1488/94 as well as Council Directive 76/769/EEC and Commission Directives 91/155/EEC, 93/67/EEC, 93/105/EC and 2000/21/EC) aims to improve the protection of human health and the environment from the risks that can be posed by chemicals, while enhancing the competitiveness of the EU chemicals industry. It also promotes alternative methods for the hazard assessment of substances in order to reduce the number of tests on animals.

In principle, REACH applies to all chemical substances; not only those used in industrial processes but also in our day-to-day lives, such as in cleaning products, paints as well as in articles such as clothes, furniture and electrical appliances.

<sup>12</sup> http://eur\_lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2006:396:0001:0849:EN:PDF

REACH places the burden of proof on companies. To comply with the regulation, companies must identify and manage the risks linked to the substances they manufacture and market in the EU. They have to demonstrate how the substance can be safely used, and they must communicate the risk management measures to the users.

If the risks cannot be managed, authorities can restrict the use of substances in different ways. In the long run, the most hazardous substances should be substituted with less dangerous ones.

REACH establishes procedures for collecting and assessing information on the properties and hazards of substances.

Companies need to register their substances and to do this they need to work together with other companies who are registering the same substance.

The European Chemical Agency (ECHA) receives and evaluates individual registrations for their compliance, and the EU Member States evaluate selected substances to clarify initial concerns for human health or for the environment. Authorities and ECHA scientific committees assess whether the risks of substances can be managed.

Authorities can ban hazardous substances if their risks are unmanageable. They can also decide to restrict a use or make it subject to a prior authorisation.

#### Authorisation

The authorisation procedure aims to assure that the risks from Substances of Very High Concern are properly controlled and that these substances are progressively replaced by suitable alternatives while ensuring the good functioning of the EU internal market.

Substances with the following hazard properties may be identified as Substances of Very High Concern (SVHCs):

Substances meeting the criteria for classification as carcinogenic, mutagenic or toxic for reproduction category 1A or 1B in accordance with Commission Regulation (EC) No 1272/2008 (CMR substances) Substances which are persistent, bioaccumulative and toxic (PBT) or very persistent and very bioaccumulative (vPvB) according to REACH (Annex XIII)

Substances identified on a case-by-case basis, for which there is scientific evidence of probable serious effects that cause an equivalent level of concern as with CMR or PBT/vPvB substances

After a two-step regulatory process, SVHC may be included in the Authorisation List and become subject to authorisation. These substances cannot be placed on the market or used after a given date, unless an authorisation is granted for their specific use, or the use is exempted from authorisation.

Authorisations will be granted if the applicant can demonstrate that the risk from the use of the substance is adequately controlled. If not, an authorisation may still be granted when it is proven that the socio-economic benefits of using the substance outweigh the risks and there are no suitable alternative substances or technologies.

#### The Candidate List

The first step in the authorisation process is for the European Commission to decide on those substances that may have serious effects on human health or the environment and, therefore, the risks

resulting from their use must be properly controlled and the substances progressively replaced when possible.

A Member State or ECHA at the request of the European Commission can propose a substance to be identified as a Substance of Very High Concern (SVHC). If identified, the substance is added to the Candidate List (the Candidate List of 161 substances can be accessed at: <a href="http://echa.europa.eu/web/guest/candidate-list-table">http://echa.europa.eu/web/guest/candidate-list-table</a>), which includes candidate substances for possible inclusion in the Authorisation List (Annex XIV).

The inclusion of a substance in the Candidate List creates legal obligations to companies manufacturing, importing or using such substances, whether on their own, in preparations or in articles.

Recommendation for inclusion in the Authorisation List

ECHA prioritises the substances from the Candidate List to determine which ones should be included in the Authorisation List (Annex XIV of REACH) and therefore, subject to authorisation. ECHA regularly (at least every second

Year) submits recommendations to the European Commission, that will decide on the substances to be included in the Authorisation List.

Priority is normally given to substances with PBT or vPvB properties, wide dispersive use or high volumes. However, the regulatory effectiveness of the authorisation measure in protecting human health and the environment is also considered.

The draft recommendation includes, amongst other items, the following information:

- Sunset Date from which the placing on the market and the use of a substance is prohibited, unless an authorisation is granted or the use is exempt from authorisation
- Latest application date by which applications must be received if the applicant wishes to continue the placing on the market or use of the substance after the sunset date
- Review periods for certain uses, if any
- Uses exempted from the authorisation requirement, if any

The following table shows the substances that have been included in the Authorisation List (Annex XIV of REACH). Substances that have been recommended for inclusion in the Authorisation List (Annex XIV of REACH are shown in <u>Appendix 3</u> of the attached Annex to the current document.

#### Restrictions

Restrictions are a tool to protect human health and the environment from unacceptable risks posed by chemicals. Restrictions may limit or ban the manufacture, placing on the market or use of a substance.

A restriction applies to any substance on its own, in a mixture or in an article, including those that do not require registration. It can also apply to imports.

A Member State, or ECHA on request of the European Commission, can propose restrictions if they find that the risks need to be addressed on an EU-wide basis. ECHA can also propose a restriction on articles containing substances that are in the Authorisation list (Annex XIV).

The list of restrictions is in Annex XVII to REACH and includes all the restrictions adopted in the framework of REACH and the previous legislation, Directive 76/769/EEC. Each entry shows the substance or group of substances or the mixture, and the consequent restrictions conditions. The latest consolidated version of REACH presents the restrictions adopted until that date. Subsequent changes

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are included in the amending Commission regulations<sup>13</sup>. ECHA currently lists 105 restricted substances<sup>14</sup>, shown in Appendix 4 of the attached Annex to the current document.

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<sup>13</sup> http://echa.europa.eu/web/guest/regulations/reach/legislation

<sup>&</sup>lt;sup>14</sup> http://echa.europa.eu/addressing-chemicals-of-concern/restrictions/list-of-restrictions

## List of substances included in Annex XIV of REACH ("Authorisation List").

Name	EC Number	CAS Number	Sunset date	Latest application date	Exempted (categories of) uses
1,2-Dichloroethane (EDC)	203-458- 1	107-06-2	22/11/2017	22/05/2016	
2,2'-dichloro-4,4'-methylenedianiline (MOCA)	202-918- 9	101-14-4	22/11/2017	22/05/2016	
2,4 – Dinitrotoluene (2,4-DNT)	204-450- 0	121-14-2	21/08/2015	21/02/2014	
4,4'- Diaminodiphenylmethane (MDA)	202-974- 4	101-77-9	21/08/2014	21/02/2013	
5-tert-butyl-2,4,6-trinitro-m-xylene (Musk xylene)	201-329- 4	81-15-2	21/08/2014	21/02/2013	
Acids generated from chromium trioxide and their oligomers. Group containing: Chromic acid, Dichromic acid, Oligomers of chromic acid and dichromic acid	231-801- 5 236-881- 5	13530-68- 2 7738-94-5	21/09/2017	21/03/2016	
Ammonium dichromate	232-143- 1	7789-09-5	21/09/2017	21/03/2016	
Arsenic acid	231-901- 9	7778-39-4	22/08/2017	22/02/2016	
Benzyl butyl phthalate (BBP)	201-622- 7	85-68-7	21/02/2015	21/08/2013	Uses in the immediate packaging of medicinal products covered under Regulation (EC) No 726/2004, Directive 2001/82/EC, and/or Directive 2001/83/EC.
Bis(2-ethylhexyl) phthalate (DEHP)	204-211- 0	117-81-7	21/02/2015	21/08/2013	Uses in the immediate packaging of medicinal products covered under Regulation (EC) No 726/2004, Directive 2001/82/EC, and/or Directive 2001/83/EC.
Bis(2-methoxyethyl) ether (Diglyme)	203-924- 4	111-96-6	22/08/2017	22/02/2016	
Chromium trioxide	215-607- 8	1333-82-0	21/09/2017	21/03/2016	
Diarsenic pentaoxide	215-116- 9	1303-28-2	21/05/2015	21/11/2013	
Diarsenic trioxide	215-481- 4	1327-53-3	21/05/2015	21/11/2013	
Dibutyl phthalate (DBP)	201-557- 4	84-74-2	21/02/2015	21/08/2013	Uses in the immediate packaging of medicinal products covered under Regulation (EC) No 726/2004,

					Directive 2001/82/EC, and/or Directive 2001/83/EC.
Dichromium tris(chromate)	246-356- 2	24613-89- 6	22/01/2019	22/07/2017	
Diisobutyl phthalate (DIBP)	201-553- 2	84-69-5	21/02/2015	21/08/2013	
Formaldehyde, oligomeric reaction products with aniline (technical MDA)	500-036- 1	25214-70- 4	22/08/2017	22/02/2016	
Hexabromocyclododecane (HBCDD), alpha- hexabromocyclododecane, beta-hexabromocyclododecane, gamma- hexabromocyclododecane	221-695- 9 247-148- 4	134237- 50-6 134237- 51-7 134237- 52-8 25637-99- 4 3194-55-6	21/08/2015	21/02/2014	
Lead chromate	231-846- 0	7758-97-6	21/05/2015	21/11/2013	
Lead chromate molybdate sulphate red (C.I. Pigment Red 104)	235-759- 9	12656-85- 8	21/05/2015	21/11/2013	
Lead sulfochromate Yellow (C.I. Pigment Yellow 34)	215-693- 7	1344-37-2	21/05/2015	21/11/2013	
Pentazinc chromate octahydroxide	256-418- 0	49663-84- 5	22/01/2019	22/07/2017	
Potassium chromate	232-140- 5	7789-00-6	21/09/2017	21/03/2016	
Potassium dichromate	231-906- 6	7778-50-9	21/09/2017	21/03/2016	
Potassium hydroxyoctaoxodizincatedichromate	234-329- 8	11103-86- 9	22/01/2019	22/07/2017	
Sodium chromate	231-889-	7775-11-3	21/09/2017	21/03/2016	
Sodium dichromate	234-190-	10588-01- 9 7789-12-0	21/09/2017	21/03/2016	
Strontium chromate	232-142- 6	7789-06-2	22/01/2019	22/07/2017	
Trichloroethylene	201-167- 4	79-01-6	21/04/2016	21/10/2014	

Tris(2-chloroethyl)phosphate (TCEP)	204-118-	115-96-8	21/08/2015	21/02/2014
	5			

#### 2.3.2.3 The Marine Strategy Framework Directive (MSFD)

The Directive 2008/56/EC of 17 June 2008 establishing a framework for Community action in the field of marine environmental policy (Marine Strategy Framework Directive) establishes a framework within which Member States shall take the necessary measures to achieve or maintain good environmental status in the marine environment by the

Year 2020 at the latest. For that purpose, marine strategies shall be developed and implemented in order to (a) protect and preserve the marine environment, prevent its deterioration or, where practicable, restore marine ecosystems in areas where they have been adversely affected; (b) prevent and reduce inputs in the marine environment, with a view to phasing out pollution, so as to ensure that there are no significant impacts on or risks to marine biodiversity, marine ecosystems, human health or legitimate uses of the sea.

To assist Member States in the interpretation of what GES means in practice, the Directive sets out, in Annex I, eleven qualitative descriptors which describe what the environment will look like when GES has been achieved.. As regards contaminants, GES is considered to have been achieved under Descriptor 8 when "contaminants are at a level not giving rise to pollution effects"; and when "contaminants in fish and other seafood for human consumption do not exceed levels established by Community legislation or other relevant standards".

Marine strategies are to be developed by Member States taking into account inter alia indicative lists of elements set out in Annex III of the Directive. With regard to priority substances, Annex III includes contamination by hazardous substances, referring more specifically to the introduction of synthetic compounds (e.g. priority substances under Directive 2000/60/EC which are relevant for the marine environment such as pesticides, antifoulants, pharmaceuticals, resulting, for example, from losses from diffuse sources, pollution by ships, atmospheric deposition and biologically active substances), introduction of non-synthetic substances and compounds (e.g. heavy metals, hydrocarbons resulting, for example, from pollution by ships and oil, gas and mineral exploration and exploitation, atmospheric deposition, riverine inputs), and the introduction of radio-nuclides.

In 2010 the Commission produced a set of detailed criteria and indicators through the adoption of Commission Decision 2010/477/EU, on criteria and methodological standards on good environmental status of marine waters, in order to to assist Member States in the implementation of the Marine Directive. Under the current process of the review of GES criteria and methodological standards, the review with repect to MSFD Descriptor 8 is being performed by the MSFD Expert Network on Contaminants. With regard to contaminants GES criteria, the Expert Network has put forward the recommendation (EC JRC, 2015):

Establishment of an EU-wide minimum list of elements and/or parameters for assessing GES based on:

- WFD Priority Substances (including amendments)
- A clear and justified mechanism for excluding WFD priority substances from MSFD assessments where they are not relevant in the marine environment.
- -Other substances (marine region specific substances, selected through Regional Sea Convention mechanisms), or river basin specific pollutants) that might be relevant and would need to be monitored.

The overall recommendation is that the Commission Decision should not be prescriptive of substances, but of approach. Under MSFD, Member States shall consider WFD Priority Substances (WFD PS). However, some WFD PS might not be relevant for the marine environment and, consequently, should not be necessarily assessed in the marine environment (including coastal waters). Therefore, Member States can exclude the nonrelevant WFD PS for their own situation. The WFD PS exclusion process has to be clearly documented and justified based on predefined situations based on the chemical/physical properties of the contaminant (e.g. volatility and persistence in the marine environment),

monitoring data evidence and the significance of sources and inputs. Member States shall also consider pollutants relevant at regional level (Marine Region Specific Contaminants), selected on the basis of the information and knowledge gathered and approaches developed in Regional Sea Conventions (RSCs).

River Basin Specific Pollutants (RBSP) are considered under WFD in the transitional waters and coastal waters (1 nm). There might be a future provision to make a RBSP EQS non-compliance trigger further investigation in marine waters. Legacy pollutants are present in the marine environment. Their basic monitoring and assessment is needed to assess GES, even if direct mitigation measures cannot be provided.

#### 2.2.3 OSPAR

#### 2.2.3.1 The List of Subtances of Possible Concern - Recent developments

It is recalled that the OSPAR Hazardous Substances Strategy sets the objective of preventing pollution of the maritime area by continuously reducing discharges, emissions and losses of hazardous substances, with the ultimate aim of achieving concentrations in the marine environment near background values for naturally occurring substances and close to zero for man-made synthetic substances. The OSPAR Commission is implementing this strategy progressively by making every endeavour to move towards the target of the cessation of discharges, emissions and losses of hazardous substances by the Year 2020.

The OSPAR List of Substances of Possible Concern was adopted in 2002. It is a dynamic working list and is regularly revised as new information becomes available. This has resulted in substances being removed from the list. Some of the substances on the List have been included on the List of Chemicals for Priority Action.

In the work on the substances of possible concern, OSPAR is relying on progress on the evaluation of substances under the REACH Regulation and on the priorisation of substances under the Water Framework Directive. As a result the Dynamic Selection and Prioritisation Mechanism for Hazardous Substances (DYNAMEC) has been put on hold. The inclusion of substances with high environmental concerns into the REACH candidate list and under the Water Framework Directive is an important signal to OSPAR for its work on hazardous substances.

Within the process of reviewing the substances on the List of Substances of Possible Concern, the aim is to identify those substances which are of concern to the marine environment, and to which therefore the commitments of the Hazardous Substances Strategy should apply, but which are not covered adequately by the EC framework or some other international forum. This process has resulted in the grouping of the substances in four sections (A-D), to help identify those substances which warrant further work by OSPAR.

Section A includes substances which warrant further work by OSPAR because they do not meet the criteria for Sections B-D and substances for which, for the time being, information is insufficient to group them in Sections B-D. The substances listed in Section A are presented in <u>Appendix 5</u> of the current document.

Section B includes substances which are of concern for OSPAR but which are adequately addressed by EC initiatives or other international fora.

Section C includes substances which are not produced and/or used in the OSPAR catchment or are used in sufficiently contained systems making a threat to the marine environment unlikely

Section D includes substances which appear not to be "hazardous substances" in the meaning of the Hazardous Substances Strategy but where the evidence is not conclusive.

#### 2.2.3.2. The List of Chemicals for Priority Action

The List of Chemicals for Priority Action in its current form was adopted in 2002. There are currently 42 substances or groups of substances on the List of Chemicals for Priority Action as listed in the following table. OSPAR action is focused on the substances on Part A of the List. For each of these substances or groups of substances a background document has been prepared. Since 2002 the list has been revised to reflect removal of substances from this list, as well as the List of Substances of Possible Concern.

The Background Documents assess the situation for the substance and conclude on what actions OSPAR should take to move towards the cessation target. They are reviewed periodically, resulting in review statements or revised Background Documents, depending on whether the new information available affects the risk evaluation and the recommended actions.

## OSPAR List of Chemicals for Priority Action (Update 2013)

CAS No	Substances/Group of substances	Туре	Last Revision of Background document	Review statement on background document
A: CHEMIC	CALS WHERE A BACKGROUND DOCUM	ENT HAS BEEN OR IS BEIN	VG PREPARED	
	Cadmium	Metallic compound	2004 (Spain)	2010
	lead and organic lead compounds	Metal/organometallic compounds	2009 (Norway)	
	mercury and organic mercury compounds		2004 (UK)	2009
	organic tin compounds *	Organometallic compounds	2011 (The Netherlands)	
51000-52-3	neodecanoic acid, ethenyl ester	Organic ester	2011 (UK)	
1763-23-1	perfluorooctanyl sulphonic acid and its salts (PFOS) *	Organohalogens	2006 (UK)	2011
79-94-7	tetrabromobisphenol A (TBBP-A)		2011 (UK)	
87-61-6	1,2,3-trichlorobenzene		2005 (Belgium & Luxembourg)	2010
120-82-1	1,2,4-trichlorobenzene		2005 (Belgium & Luxembourg)	2010
108-70-3	1,3,5-trichlorobenzene		2005 (Belgium & Luxembourg)	2010
	brominated flame retardants		2009 (Sweden)	
	polychlorinated biphenyls (PCBs) *		2004 (Germany & Belgium)	2008
	polychlorinated dibenzodioxins (PCDDs) polychlorinated dibenzofurans (PCDFs)		2007 (Denmark & Belgium)	
	short chained chlorinated paraffins (SCCP)		2009 (Sweden)	
793-24-8	4-(dimethylbutylamino)diphenylamin (6PPD)	Organic nitrogen compound	2006 (Germany)	
115-32-2	Dicofol	Pesticides/Biocides/	2004 (Finland)	2008
115-29-7	Endosulfan	Organohalogens	2004 (Germany)	2008
	hexachlorocyclohexane isomers (HCH)		2004 (Germany)	2008
72-43-5	Methoxychlor		2004 (Finland)	2008
	pentachlorophenol (PCP)		2004 (Finland)	
1582-09-8	Trifluralin		2005 (Germany)	2012
23593-75-1	Clotrimazole	Pharmaceutical	2013 (France)	

732-26-3	2,4,6-tri-tert-butylphenol	Phenols	2006 (UK)	2009
	nonylphenol/ethoxylates (NP/NPEs) and related substances		2009 (Sweden)	
140-66-9	Octylphenol		2006 (UK)	2009
	certain phthalates: dibutylphthalate (DBP), diethylhexylphthalate (DEHP)♥	Phthalate esters	2006 (Denmark & France)	
	polyaromatic hydrocarbons (PAHs) §	Polycyclic aromatic compounds	2009 (Norway)	
	musk xylene	Synthetic musk	2004 (Switzerland)	
CAS No	Group of substances / substances	Туре	Ident	ified at <sup>†</sup>
B:CHEMIC SYSTEMS	CALS WHERE NO BACKGROUND DOCUL ‡	MENT IS BEING PREPARED	BECAUSE THEY ARE INTE	RMEDIATES IN CLOSED
4904-61-4	1,5,9 cyclododecatriene ‡	Aliphatic hydrocarbons	OSPAR 2002	
294-62-2	cyclododecane ‡		OSPAR 2002	
CAS No	Group of substances / substances	Type	Ident	ified at <sup>†</sup>
C: CHEMI	CALS WHERE NO BACKGROUND DOCU	MENT IS BEING PREPARED	BECAUSE THERE IS NO C	URRENT PRODUCTION OR
USE INTE	REST*			
59447-55-1	2-propenoic acid, (pentabromo)methyl ester	Organohalogens	OSPAR 2003	
36065-30-2	2,4,6-bromophenyl 1-2(2,3-dibromo-2-methylpropyl) *		OSPAR 2001	
85-22-3	pentabromoethylbenzene*		OSPAR 2001	
28680-45-7	heptachloronorbornene*			
2440-02-0	пертастногопогоогнене		OSPAR 2001	
2440-02-0 1825-21	pentachloroanisole*	Organohalogens (cont.)	OSPAR 2001 OSPAR 2001	
	pentachloroanisole*	Organohalogens (cont.)		
1825-21		Organohalogens (cont.)		
1825-21	pentachloroanisole*	Organohalogens (cont.)		
1825-21 -4	pentachloroanisole*  polychlorinated naphthalenes*, ††	Organohalogens (cont.)	OSPAR 2001	
1825-21 -4 1321-65-9	pentachloroanisole*  polychlorinated naphthalenes*, ††  trichloronaphthalene*	Organohalogens (cont.)	OSPAR 2001 OSPAR 2001	
1825-21 -4 1321-65-9 1335-88-2	pentachloroanisole*  polychlorinated naphthalenes*, ††  trichloronaphthalene*  tetrachloronaphthalene*	Organohalogens (cont.)	OSPAR 2001 OSPAR 2001 OSPAR 2001	

2234-13-1	octachloronaphthalene*		OSPAR 2001
70776-03-3	naphthalene, chloro derivs. *		OSPAR 2002
55525-54-7	3,3'-(ureylenedimethylene)bis(3,5,5- trimethylcyclohexyl) diisocyanate*	Organic nitrogen compound	OSPAR 2001
2104-64-5	ethyl O-(p-nitrophenyl) phenyl phosphonothionate (EPN)*	Pesticides/Biocides	OSPAR 2001
70124-77-5	flucythrinate*		OSPAR 2001
465-73-6	isodrin*		OSPAR 2001
2227-13-6	tetrasul*		OSPAR 2001
512-04-9	diosgenin*	Pharmaceutical	OSPAR 2001

#### Endnotes

The substances in this list were identified at the following OSPAR Commission meetings: OSPAR/MMC 1998: Agreement reference number 1998-16 (Annex 2 to the OSPAR Strategy with regard to Hazardous Substances);

(Note: When identifying the substances or groups of substances, OSPAR/MMC 1998 has not allocated CAS and EINECS registration numbers. Background documents adopted by the OSPAR Commission for these substances or groups of substances may indicate which substances have been addressed so far by OSPAR)

OSPAR 2000: Agreement reference number 2000-10;

OSPAR 2001: Agreement reference number 2001-2;

OSPAR 2002: Agreement reference number 2002-18;

OSPAR 2003: Agreement reference number 2003-19.

The identification of these substances and the consequent action required is explained in § 7.6 of the OSPAR 2002 Summary Record. In brief, these substances have rankings in terms of persistency, liability to bioaccumulate and toxicity which are of equal concern as the other substances on this list. However, to the best of OSPAR's knowledge, on the basis of information from industry, OSPAR accepts that this substance is produced and used exclusively as an <u>intermediate</u> in closed systems in the production of other substances, under conditions where the safeguards applying are sufficient to avoid reasonable concerns that discharges, emissions or losses of the substance could reach the marine environment. Therefore, every five

Years, commencing in 2003, Contracting Parties and, where appropriate, observers representing the chemicals industries should report to OSPAR:

- a. whether they have found any evidence that these chemicals are being produced, used or discharged without being subjected to safeguards to avoid reasonable concerns that discharges, emissions or losses of the substances could reach the marine environment, and, if so, what that evidence is, and what action (if any) has been taken;
- b. whether there have been any cases where applications have been made for approvals involving these chemicals, and, if so, what decision was taken.

\*The identification of these substances and the consequent action required is explained in § 4.13 of the OSPAR 2001 Summary Record. In brief, these substances have rankings in terms of persistency, liability to bioaccumulate and toxicity which are of equal concern as the other substances on this list. However, to the best of OSPAR's knowledge, there is no current production or use in the OSPAR states. Therefore, commencing in 2003 and every five

Years thereafter, or earlier, if information becomes available, Contracting Parties and, where appropriate, observers representing the chemicals industries should report to OSPAR:

a.whether they have found any evidence that these chemicals are being produced, used or discharged, and, if so, what that evidence is, and what action (if any) has been taken;

whether there have been any cases where applications have been made for approvals involving these chemicals, and, if so, what decision was taken.

- $\dagger\dagger$  Polychlorinated naphthalenes should be treated as a group of substances (OSPAR 02/21/1, § 7.7).
- ♦ PFOS is the highly persistent and toxic breakdown product of a number of perfluorooctanyl sulphonyl compounds. Several PFOS precursors have been selected on the OSPAR List of Substances of Possible Concern. The background document will identify these precursors and, if necessary, appropriate control measures will be proposed. CAS and EINECS numbers refer only to the acid form of PFOS.
- The following substances belonging to the group of polyaromatic hydrocarbons have been deselected from the OSPAR List of Substances of Possible Concern on the grounds that they do not meet the cut-off values for persistence in the Selection Criteria used in the Initial Selection Procedure adopted by OSPAR 2001 (*Reference Number: 2001-1*) and are therefore not considered to be a priority for action by OSPAR: naphthalene, 2-methyl- (CAS No. 91576); 1-phenanthrenecarboxylic acid, 1,2,3,4,4a,4b,5,6,10,10a-decahydro-1,4a-dimethyl-7-(1-methylethyl)-, methyl ester, [1R-(1.alpha.,4a.beta.,4b.alpha.,10a.alpha.)]- (CAS No. 127253); 1-phenanthrenemethanol, 1,2,3,4,4a,4b,5,6,7,9,10,10a-dodecahydro-1,4a-dimethyl-7-(1-methylethyl)- (CAS No. 127366); 7H-dibenzo[c,g]carbazole (CAS No. 194592); 13H-dibenzo[a,i]carbazole (CAS No. 239645); 1H-3a,7-methanoazulene, 2,3,4,7,8,8a-hexahydro-3,6,8,8-tetramethyl-, [3R-(3alpha,3abeta,7beta,8aalpha)]- (CAS No. 469614); 1-phenanthrenemethanol, 1,2,3,4,4a,4b,5,6,10,10a-decahydro-1,4a-dimethyl-7-(1-methylethyl)-, [1R-(1.alpha.,4a.beta.,4b.alpha.,10a.alpha.)]- (CAS No. 666842); cedrene- (CAS No. 11028425); 1-phenanthrenemethanol, tetradecahydro-1,4a-dimethyl-7-(1-methylethyl)- (CAS No. 13393936); 1-phenanthrenecarboxylic acid, tetradecahydro-1,4a-dimethyl-7-(1-methylethyl)-, methyl ester, [1R-(1alpha,4abeta,4balpha (CAS No. 19941287).
- The following substance belonging to the group of organic tin compounds has been deselected from the OSPAR List of Substances of Possible Concern on the grounds that it does not meet the cut-off value for persistence in the Selection Criteria used in the Initial Selection Procedure adopted by OSPAR 2001 (*Reference Number: 2001-1*) and is therefore not considered to be a priority for action by OSPAR: stannane, tributyl(1-oxododecyl)oxy- (CAS No. 3090366).

The following substance belonging to the group of polychlorinated biphenyls has been deselected from the OSPAR List of Substances of Possible Concern on the grounds that it does not meet the cut-off value for persistence in the Selection Criteria used in the Initial Selection Procedure adopted by OSPAR 2001 (*Reference Number: 2001-1*) and is therefore not considered to be a priority for action by OSPAR: 1,1'-biphenyl, 4,4'-dichloro- (CAS No. 2050682).

OSPAR 2006 agreed to deselect the following substances belonging to the group of certain phthalates: "DIDP (1,2-Benzenedicarboxylic acid,di-C9-11-branched alkyl esters, C10-rich (CAS N° 68515-49-1)), DIDP (di-"isodecyl"phthalate (CAS N° 26761-40-0)), DINP (1,2-Benzenedicarboxylic acid, di-C8-10-branched alkyl esters, C9-rich (CAS N° 68515-48-0)) and DINP (di-"isononyl" phthalate (CAS N° 28553-12-0)). They are not PBT substances for the reasons set out in the Agreement 2004-13 available on the OSPAR website (see OSPAR 2006 Summary Record, OSPAR 06/23/1 paragraph 8.3).

#### 2.2.4 HELCOM

It is recalled that the 1992 HELCOM Convention included in its Annex I a list of intrinsic chemical properties and characteristics allowing the identification of hazardous substances, as well as a preliminary list of harmful compounds. Thus, Part I of Annex I established that the identification and evaluation of compounds should be based on persistency, toxicity and other noxious properties, and on the tendency for bio-accumulation. Part II and III of Annex I set up two lists of harmful compounds, based on the described properties. Part II concerned substances which should be partially banned or prohibited (DDT and its derivatives DDE and DDD as well as PCBs and PCTs); and part III involved pesticides and biocides that should be minimised or whenever possible banned as listed in the table below.

#### HELCOM Convention substances to be minimised or whenever possible banned

	<b>CAS-number</b>		
Acrylonitrile	107131		
Aldrin	309002		
Aramite	140578		
Cadmium-compounds	-		
Chlordane	57749		
Chlordecone	143500		
Chlordimeform	6164983		
Chloroform	67663		
1,2-Dibromoethane	106934		
Dieldrin	60571		
Endrin	72208		
Fluoroacetic acid and	7664393, 144490		
derivatives			
Heptachlor	76448		
Isobenzane	297789		
Isodrin	465736		
Kelevan	4234791		
Lead-compounds	-		
Mercury-compounds	-		
Morfamquat	4636833		
Nitrophen	1836755		
Pentachlorophenol	87865		
Polychlorinated terpenes	8001501		
Quintozene	82688		
Selenium-compounds	-		
2,4,5-T	93765		
Toxaphene	8001352		

Furthermore, the 1998 HELCOM Recommendation 19/5 sets as the objective with regard to Hazardous Substances to prevent pollution of the Convention area by continuously reducing discharges, emissions and losses of hazardous substances towards the target of their cessation by the

Year 2020, with the ultimate aim of achieving concentrations in the environment near background values for naturally occurring substances and close to zero for man-made synthetic substances

It is further recalled that Recommendation 19/5 in Appendix 2 listed potential substances of possible concern. The list included about 300 substances meeting the criteria for selection, assessment and prioritisation according to the strategy to implement HELCOM Recommendation 19/5. Through a dynamic selection and prioritization mechanism ranking the listed substances by their degree of concern, a list of 42 selected compounds for immediate priority action was compiled (HELCOM Recommendation 19/5, Appendix 3), requiring the initial identification of their sources and pathways to the marine environment (using information from monitoring and research) as well as the assessing of the caused risks in order to determine relevant measures to minimize pollution.

More recently, the 2010 HELCOM Recommendation 31E/1, superseding HELCOM Recommendation 19/5 and taking into account the 2007 Baltic Sea Action Plan (BSAP) which includes specific measures for priority hazardous substances, set a new Strategy to implement the HELCOM Objective for hazardous substances to move towards the target of the cessation of discharges, emmissions and losses of hazardous substances, to achieve the Baltic Sea in good environmental status by 2021.

According to Recommendation 31E/1 the work of HELCOM is thence focused on a List of Priority Hazardous Substances, (Appendix II of Attachement 2 of Rec. 31E/1), as shown in the list below (corresponding to the BSAP list of Hazardous substances of specific concern to the Baltic Sea), with the aim to develop measures preventing pollution of the marine environment.

#### HELCOM List of Priority Hazardous Substances (HELCOM Rec. 31E/1)

- 1. Dioxins (PCDD), furans (PCDF) & dioxin-like polychlorinated biphenyls
- 2a. Tributyltin compounds (TBT)
- 2b. Triphenyltin compounds (TPhT)
- 3a. Pentabromodiphenyl ether (pentaBDE)
- 3b. Octabromodiphenyl ether (octaBDE)
- 3c. Decabromodiphenyl ether (decaBDE)
- 4a. Perfluorooctane sulfonate (PFOS)
- 4b. Perfluorooctanoic acid (PFOA)
- 5. Hexabromocyclododecane (HBCDD)
- 6a. Nonylphenols (NP)
- 6b. Nonylphenol ethoxylates (NPE)
- 7a. Octylphenols (OP)
- 7b. Octylphenol ethoxylates (OPE)
- 8a. Short-chain chlorinated paraffins (SCCP or chloroalkanes, C10-13)
- 8b. Medium-chain chlorinated paraffins (MCCP or chloroalkanes, C14-17)
- 9. Endosulfan
- 10. Mercury
- 11. Cadmium

The HELCOM List of Priority Hazardous Substances is to be kept under review and updated based on monitoring and other scientific data and internationally accepted methods and criteria.

Under Reccomendation 31E/1 a List of substances of possible concern is to be further developed and completed on the basis of the list of potential substances of concern contained in Recommendation 19/5 and Annex I of the Helsinki Convention.

Under the HELCOM Rec. 31E/1 guiding principles of the new Strategy to implement the HELCOM Hazardous Substances Objective is included the progressive replacement of hazardous substances by suitable alternative substances or technologies, where these are economically defendable and technically available. Emissions, discharges and losses of new hazardous substances are to be avoided by applying the principles of sustainable chemistry (defined in the Strategy, based on the OECD, as the design, manufacture and use of efficient, effective, safe and more environmentally benign chemical products and processes).

Furthermore, sound chemicals management has to be applied, including precautionary risk management measures based on the intrinsic properties of chemicals, using internationally agreed systems for the classification and labelling of chemicals.

Under the Rec 31E/1 the definition of hazardous substances is broadened to include substances that are very persistent and very bioaccumulative. In addition, substances that are Carcinogenic, Mutagenic or toxic to Reproduction (CMR) meeting the criteria for classification in category 1a or 1b in the UN Global Harmonised System for classification and labelling of chemicals (GHS). A third category includes other substances or groups of substances identified on a case by case basis as causing probable serious effects to human health and the environment, of an equivalent level of concern as those above (e.g. endocrine disruptors).

Regarding the updating of the HELCOM list of substances for priority action (under HELCOM Rec. 19/5) the new Strategy implies that the Commission will take into consideration the REACH Regulation criteria for the identification of persistent, bioaccumulative and toxic substances, and very persistent and very bioaccumulative substances (specified in Annex XIII of the REACH Regulation).

#### 3. Occurrence of Substances of concern in the Mediterranean marine environment

#### 3.1. Thematic Assessment of Hazardous Substances in the Mediterranean - MED-POL data

This section reflects the information presented in the last Thematic Assessment Report on Hazardous Substances in the Mediterranean coastal environment (UNEP/MAP/MED POL, 2011), which was based on the MED POL database, and which also commented on possible pollution trends (not necessarily statistically supported). The above mentioned Report covered the full period of MED POL Phase III and Phase IV, until 2010 as shown in the following table, and incorporated additional scientific information included in the four sub-regional assessment reports, which were prepared by MAP in the framework of the gradual application of the Ecosystem Approach for the management of human activities in the Mediterranean (UNEP/MAP, 2011).

The above Report used in principle, only representative data, basically considering the existence of sufficiently large datasets. In this respect, only trace metals were found suitable for trend assessment at the regional level in sediments and trace metals and organo-chlorinated compounds in marine biota. In the case of biota, while the MED POL Database includes a large number of marine species, the said Report focused on the bivalve *Mytilus galloprovincialis* and the benthic fish *Mullus barbatus*, as they are the more common and widely analysed species in the region.

As regards the specific parameters of the Report, they were selected on the basis of those considered of priority concern and taking into account the total number of samples and the geographical and temporal coverages. Thus, the selection included the trace metals Cd, Hg, Pb, Zn and Cu, and the families of DDTs and PCBs, as the more representative persistent organic pollutants (POPs). The Drin's (Aldrin, Endrin and Dieldrin), HCB and Lindane were also considered.

In order to harmonize the database, all concentrations were recorded in  $\mu g$  g<sup>-1</sup> or ng g<sup>-1</sup> dry weight. In cases where they were reported in other units or in wet weight basis, they were converted accordingly. In summary, after selection and harmonization of data, a total of 34738 observations, corresponding to more than 400 stations monitored during the MED POL Phases III and IV (1999-2010) were included in the Assessment. In addition median concentration values were depicted, as they were considered to constitute a better representation of the left-skewed populations studied.

Available data of hazardous substances in the MED POL Database (2001-2010)

	Sediments		Biota		
Country	Trace Metals	Organics	Trace Metals	Organics	
Albania			2001 - 2007	2001 – 2007	
Croatia	2002 – 2005, 2009	2009	2000 - 2005, 2009	1999 – 2005, 2009	
Cyprus			1999, 2001, 2004, 2005	1999 – 2007	
Egypt	2006, 2009, 2010	2006, 2009, 2010	2006, 2009, 2010	2009, 2010	
France	2006-2009	2006-2009	1997 - 2009	1999 – 2009	
Greece	1999, 2004, 2005, 2007	2004, 2005	1999, 2000, 2004 - 2007	2004, 2005	
Israel	1999 - 2009		1999 - 2009		
Italy	2001 - 2005	2001-2005	2001 - 2005	2001 – 2005	

Morocco	2007		1999 - 2007	2007
Slovenia		1999-2006, 2010	1999 - 2010	
Spain			2004 - 2007	2004 – 2007
Syria	2007		2007	
Tunisia	2006 - 2009		2001 - 2009	
Turkey	1999, 2003 - 2009		1999 2003 - 2009	2003 – 2009

#### Trace metals in sediments

An overview of the concentration ranges for Cd, total Hg, Pb, Zn and Cu in coastal sediments of the region is summarized in the following Table. As it is shown, Cd and Hg exhibit values several orders of magnitude lower than Zn, Pb and Cu being in an intermediate and similar position. Although the number of samples is far from being representative of the basin (the southern Central and Aegean-Levantine eco-regions are underrepresented) and the lithology of the continental shelf may influence the occurrence of trace metals in sediments, the values can be used as reference for comparison purposes.

In general, the Western Mediterranean is the region where concentrations are higher as a result of the intense human activities that can cause chemical contamination of coastal areas, followed by the Adriatic and the Aegean-Levantine basin. However, these values are in the lower range than those reported in previous assessments derived from MEDPOL I and II (UNEP, 1996).

Trace metals in sediments. Median (range) concentrations ( $\mu g g^{-1} dw$ )

Eco-region	Cd		HgT		Pb	Zn	Cu	
WESTERN MEDITERRANEAN	1.60 7.61)	(0.23-	0.16 12.6)	(0.02-	19.40 256)	(0.24- 50.10 731)	(1.0- 13.90 107)	(0.68-
ADRIATIC	0.21 18.5)	(0.01-	0.10 166.9)	(0.01-	9.83	(0.39- 67.00 980)	(5.0- 16.31 465)	(1.39-
CENTRAL MEDITERRANEAN	0.09 2.80)	(0.01-	0.05 6.00)	(0.00-	4.39 103.4)	(0.01- 32.66 205)	(0.02- 5.35 67.5)	(0.01-
AEGEAN- LEVANTINE	0.10 8.47)	(0.01-	0.15 6.70)	(0.00-	16.89 404)	(0.01-59.34 1505)	(0.02-18.52 962)	(0.01-

The analysis of trace metal levels by country reveals the occurrence of some stations with high values of Hg, Pb, Cu and Zn in Croatia. A wide range of values for all metals was found in Italy, particularly for Pb. Average values of Cd, Zn and Cu in the higher range were found in Morocco and Syria whereas values for Egypt and Israel were in the lower end. Values of Pb and Zn were also high in Greece, and of Zn and Cu in Turkey.

Besides urban and industrial inputs, rivers and streams are major contributors of metals of anthropogenic or natural origin to coastal areas. Moreover, land-based sources of specific trace metals are not only deposited in coastal sediments but may end up deep in marine canyons. Atmospheric deposition may also contribute to the contamination of deep sea sediments.

In the **Western Mediterranean**, major hotspots for trace metals, particularly Cd, Pb, Hg and Cu, are located in the areas of Marseilles-Fos and Toulon, in France, and Cartagena-Valle Escombreras-

Portman, in Spain, with important urban-industrial and mining areas, respectively. Sensitive areas are represented by the coastal lagoons (e.g. Berre, Thau and Mar Menor). The area influenced by the Rhone River discharges exhibited increased levels of all metals except Hg.

The levels of Pb are important along the Italian coast, especially in areas around the Gulf of Genova (e.g. Savona, La Spezia, Genova, Livorno) that also show evidence of Zn accumulation, while high levels of Pb are found in Naples. These levels can be linked to industrial and domestic wastes and harbour activities. Mercury levels are also high in the area of Messina and Palermo and Regio Calabria. The contributions related to tectonic sources are important near volcanic and geothermal sources as in the southern Tyrrhenian Sea. These contributions could explain natural important levels of mercury in some islands of the basin.

Significant concentrations of Zn, Cu and particularly Cd have been reported on the coast of Morocco (Tangier-Martil and Nador), whereas higher levels of Hg were found in Algeria (Algiers).

Trace metals have been extensively monitored in the **Adriatic Sea**. The Po River, draining the most industrialized part of northern Italy, is an important contributor in the area. High levels of Cd, Hg, Pb and Zn are found in the northern Adriatic, including the Venice and Grado Lagoons. The trace metals Hg and Pb are also critical contaminants in the Gulf of Trieste and Rijeka Bay. The occurrence of some discrete stations with high levels of Hg, Pb, Cu and Zn are found in Croatia, such as at Kastela and Martinska Bays. In Albania, Hg contamination from a former inland chlor-alkali plant is reflected in the sediments of Vlora Bay.

In the **Central Mediterranean** elevated levels of Hg have been measured in the Gulf of Taranto and in both the Tunisian and Italian coasts of the Strait of Sicily. However, an extensive study in the Ionian Sea revealed that Hg levels were generally comparable to those from other Mediterranean areas. The Tunis and Bizerta Lakes (Tunisia) also exhibit high contents of Pb. In the Hellenic coastal zone, the most elevated levels of such contaminants in sediments (e.g. Amvrakikos and Patraikos Gulfs) can be associated with the main sewage (domestic and industrial) outfalls. Very high values of Cd were found close to the city of Patras.

The analysis of representative trace metals in sediments of the **Aegean – Levantine basin** revealed the occurrence of high values of Cd and Zn in Iskenderun Bay (Turkey). Very high levels of Cd, Pb, Zn and Cu were found in Izmit Bay (Turkey). Concentrations of Pb were also high in Ismir Bay (Turkey). Most of the stations, particularly Ismir, Edremit and Candarli Bays (Turkey), exhibited moderate levels of Cu and Zn. Low to moderate trace metal levels were found in Israel, with high values of Hg and Zn at some stations (e.g. Haifa Bay). The level of Cd was elevated in the northern coast of Syria. Conversely, and with the exception of Hg, relatively low values were found in Egypt, around the mouth of the Nile River. The distribution patterns reflect anthropogenic sources originating from point and diffuse land-based sources, providing useful information on the identification of hotspots in the area although not fully comprehensive.

In Greece, monitoring of trace metals in sediments revealed a pollution gradient across the coastal areas, indicating different pollution fingerprints, with higher concentrations of Pb, Cu and Zn in the areas of influence of Athens, Thessaloniki and Kavala (Saronikos, Strymonikos and Kavala Gulfs).

### Trace metals in bivalves and fish

The mussel *Mytilus galloprovinciallis* has been the most widely used sentinel organism in the region. In the case of fish, *Mullus barbatus* (red mullet) has been extensively used for monitoring but only in the Aegean-Levantine region.

The summary of data corresponding to trace metals in mussels, shown in the following Table, provides the range of values found in each sub-region. The accumulation follows a similar trend as for sediments: Cd < HgT < Pb < Cu << Zn. Here, the reported values are, with the exception of hotspots,

of the same order of magnitude as those obtained during MED POL Phase I and II. As in the case of sediments, the median values can be used as a reference for a preliminary assessment of spatial trends and identification of hot spots.

Trace metals in Mytilus galloprovinciallis. Median (range) concentrations (ug g-1 dw)

Eco-region	Cd	HgT	Pb	Zn	Cu
WESTERN	0.66 (0.01-	0.13 (0.01-	2.00 (0.01-	130 (0.01-	6.30 (0.05-
MEDITERRANEAN	10.0)	7.4)	79.1)	5337)	114)
ADRIATIC	0.80 (0.03-	0.14 (0.01-	1.53 (0.07-	122 (5.7-	8.01 (0.51-
	2.73)	8.45)	67.5)	467)	81.6)
CENTRAL	0.43 (0.09-	0.16 (0.00-	0.81 (0.07-	87 (11.6-	9.32 (1.36-
MEDITERRANEAN	3.40)	7.00)	5.36)	565)	70.5)
AEGEAN-	0.75 (0.05-	0.08 (0.01-	2.28 (0.84-	131 (0.00-	7.84 (1.01-
LEVANTINE	5.27)	0.63)	5.97)	325)	112)

With the exception of Zn, which can be attributed to a major influence of the sediment lithology, the concentrations of trace metals in sediments and mussels exhibit a fair correlation. As it was observed for sediments, concentrations of trace metals in mussels are higher for the Western Mediterranean and the Adriatic Sea, although the data set is lacking information from several eastern and southern countries

In most cases, low or moderate levels were found, in particular for Cd, Hg and Zn, and to a lesser degree, for Pb and Cu. In this respect, Cd, Hg and Zn display a rather even distribution along the coasts of the different eco-regions, with a few hotspots in the coasts of Spain, in the areas of Cartagena (Cd) and Alboran (Zn), Italy, in the coasts of Sardinia (Cd) and Naples (Cd, Hg and Zn), and Turkey, in Ismir Bay (Cd). Hotspots for Hg are also found in Croatia (Rijeka Bay) and Greece (Patras). A large number of stations exhibit high values of Pb, usually associated with urban areas (e.g. Barcelona, Marseille, Genova, Fiumicino and Naples) and mining spots (Portman, Spain). High levels of Cu have been found in the Ligurian (e.g. Genova) and Tyrrhenian Seas (e.g. Fiumicino and Naples) as well as in the coast of Sardinia.

In addition to monitoring activities under the MED POL Programme, a large number of studies have been carried out in coastal enclosures and open areas of the Western Mediterranean, Adriatic and Aegean Sea. In the Western Mediterranean, a number of stations, such as those in the proximity of Genova, at the Ligurian Sea, and Naples have been found to be chronically polluted by all trace metals. In addition to these, certain sites can be considered as particular hot spots. High levels of Cd and Pb are found in the Sardinian coast (Portoscuso), Palermo and Cartagena-Portman. Higher levels of Hg and Pb were also found in coastal waters of the Tyrrhenian Sea (e.g. Fiumicino and Messina). All these stations are in proximity to mining, industrial and/or urban areas.

High levels of Pb are usually associated with direct river/urban sewage inputs. This accounts for levels found in mussels collected in the Marseille Gulf and Hyeres Bay (France), the mouth of the Llobregat River (NE Spain), also influenced by the Barcelona metropolis, Malaga, Fiumicino, etc.

Very high Hg concentrations were measured at five scattered sites along the Western Mediterranean basin (Portoscuso, Palermo and Maddalena-Sardinia Island, in Italy; Skikda in Algeria, and El Portus in Spain). There was also a predominance of high Hg values at Algiers Bay, along the north-west Italian coast (Piombino, Portoferraio) and in the French coast at Hyeres ouest.

Levels of Zn were also found to be high in the Alboran coast of Spain as well as in the southern coast of Italy (Calabria and Sicilia)

Trace metals are extensively monitored throughout the **Adriatic Sea** using mussels as bio-indicators, mostly confined in urban and industrial areas. Concentrations are moderate in the northern, NE and NW part of the basin. High concentrations of Pb and Cu were found in the Venice Lagoon and in the areas affected by the Po River discharges, consistent with urban/industrial discharges. The central, eastern and western Adriatic has also been extensively monitored. Concentrations of Pb and Hg are relatively high in the Mali Ston Bay (Croatia). High levels of Cd, Hg and Cu are found in the Kastela and Rijeka Bays (Croatia), due to the discharge of untreated urban and industrial wastewaters. In Albania, Pb and Cu concentrations above mean levels were found in mussels collected in Vlora Bay and close to Durres, a known dumping site of industrial residues.

The **Central Mediterranean** is not well represented in the MED POL Database. A few samples from south Italy and Greece are available. High levels of Hg were found in the Gulf of Patras (Greece) and moderately high levels of Cu were also found at the Sicily coast.

Trace metal analysis in biota in the **Aegean-Levantine region** exhibited low values in the case of *Mytilus galloprovinciallis*, with the exception of Cd, Pb and Cu in Turkey (Akçay and Ismir Bays) and Cd and Zn in Greece (Piraeus). In Israel, *Mactra corallina* was used as the indicator organism which exhibited relatively high values of Cd and Cu in the northern coast (Haifa). In Egypt, the clam *Donax trunculus* was the preferred indicator, exhibiting low concentrations that could just reflect a different accumulation rate of metals. Information is missing for the southern and eastern countries.

The red mullet (*Mullus barbatus*) showed rather uniform trace metal bioaccumulation through the region however certain stations exhibited high levels of Cd, Hg, Pb and Cu. The most significant feature is the uneven distribution of Cd, with significant hotspots in certain stations of the Greek Islands (Crete, Mytilini and Chios). High values of Hg and Pb were also found in Crete (Chania Bay) and Cyprus (Larnaca and Limassol Bays). The levels of Zn are also high in the Avramikos, Saronikos and Thermaikos Gulfs (Greece). The area of Mersin (Turkey) exhibits high values of Cu, while mullet collected in the northern coast of Syria exhibit high concentrations of Zn and Cu. However, the limited number of stations precluded any further consideration.

### Chlorinated compounds in bivalves and fish

Chlorinated pesticides have been extensively analysed in Mediterranean biota since the inception of MED POL (UNEP, 1990). Mussels and mullet have been the most widely studied organisms in the whole basin as part of many case studies published in the literature and assessed on the occasion of the implementation of the Stockholm Convention (UNEP, 2002). However, it has been only since the last decade that they have been systematically monitored, and data gathered in the MED POL Database, although data on mullet are limited to Cyprus and Turkey.

An additional number of individual studies are found in the literature. The levels reported for samples collected during the 90s exhibit much higher values than more recent ones. This may clearly reflect a decreasing trend resulting from the ban of chlorinated pesticides in the region.

As shown in the following Table, concentrations of Aldrin, Dieldrin, Lindane and Hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g<sup>-1</sup> range. Concentrations of DDTs were one order of magnitude higher, with p,p'-DDE being, in general, the predominant component, although recent inputs of DDT in some areas cannot be ruled out.

Concentrations are higher in the Aegean-Levantine region for aldrin and dieldrin, and in the Western Mediterranean for HCB and lindane. However, the highest values of HCB and lindane are found in Turkey and Albania, with a number of important hot spots. In the case of DDTs, their median values are similar for the four eco-regions, around 10 ng g<sup>-1</sup> dw, although exceptionally high values were found in the Adriatic, corresponding to Albania.

Chlorinated pesticides in Mytilus galloprovinciallis. Median (range) concentrations (ng g<sup>-1</sup> dw)

<b>Eco-region</b>	Aldrin	Dieldrin	НСВ	Lindane	Σ DDTs
WESTERN MEDITERRANEAN	0.34 (0.01- 11.10)	0.76 (0.01-4.54)	0.54 (0.07-8.60)	0.60 (0.01-20.95)	10.80 (0.24-322)
ADRIATIC	0.45 (0.001- 8.00)	0.37 (0.001- 7.70)	0.10 (0.005- 3.00)	0.24 (0.001-88.4)	11.00 (0.01-9779)
CENTRAL MEDITERRANEAN	1.63 (0.27-3.00)	1.0 (0.4-26.0)	0.15 (0.12-0.15)	0.11 (0.1-1.40)	10.24 (0.40-26.0)
AEGEAN- LEVANTINE	3.96 (0.44-22.11)	3.61 (0.04- 38.45)	0.25 (0.01- 26.52)		9.85 (1.01-173)

Despite the similarity of the mean DDT levels for the four eco-regions, available data indicate that contaminants are not uniformly distributed throughout the sub-regions. In the **Western Mediterranean**, areas of particular concern include estuaries (Rhône, Ebro), ports, bays and gulfs (Barcelona, Marseille-Fos, Bays of Algiers and Tunis, Naples, etc.). There is evidence that river inputs represent the most important source of pesticides entering the Western Mediterranean Sea.

In the Southern part of the W. Mediterranean, significant accumulation values of DDT were recorded at the Nador Lagoon (Morocco) and in Algiers Bay.

Coastal waters of the **Adriatic Sea** belong to unpolluted areas of chlorinated pesticides, with the exception of some stations from Albania. Concentrations up to 9779 ng g<sup>-1</sup> dw of total DDTs were found at the Durres and Vlora Bay, most probably due to the presence of stockpiles of obsolete pesticides in the country. Moderate concentrations of Lindane and DDTs were found in the Gulf of Trieste and the Marches region (Ancona), respectively.

Information on the **Central Mediterranean** is lacking, although the region seems relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the limited availability of data.

In the **Aegean-Levantine** eco-region, organochlorine compounds, mainly DDTs, were determined in mussels and red mullet across the Greek coastal waters and in some stations of Turkey and Cyprus. In all cases the concentrations of DDTs were quite low, although moderate concentrations of DDTs were present in a few stations of Turkey (Ismir Bay) and Greece (Amvrakikos Gulf), probably attributed to the intense agricultural activities in the area.

**PCBs** occur in the vicinity of industrial and urban sites, as well as in major river mouths. The geographical distribution of concentrations (7 ICES PCB congeners) in the indicator organism *Mytilus galloprovincialis* is shown in the following Table. The median values show the higher levels in the Adriatic where the samples from Albania are well above the average.

In the **Western Mediterranean**, the baseline levels are high and the sites most affected are the areas of Barcelona, Marseille and the Ligurian Sea, from Livorno to Nice, including the Genova harbour, and at the mouths of Rhone and Ebro Rivers. , Values up to 1500 ng g<sup>-1</sup> dw were recorded in one station of France. Rivers and wastewater discharges were the major sources of PCBs in western Mediterranean coastal areas.

PCBs in Mytilus galloprovinciallis. Median (range) concentrations (ng g<sup>-1</sup> dw)

Eco-region	CB138	CB153	Σ7CBs
WESTERN MEDITERRANEAN	4.20 (0.07-566)	6.18 (0.16-603)	12.6 (0.30-1500)
ADRIATIC	6.10 (0.1-350)	4.80 (0.05-85.5)	90.84 (2.23-875)
CENTRAL MEDITERRANEAN	5.00 (0.30-23.0)	4.27 (0.70-38.0)	
AEGEAN-LEVANTINE	0.49 (0.03-25.3)	0.77 (0.05-39.8)	2.61 (0.18-135)

Information for the **Central Mediterranean** is almost lacking but, apparently, the sub-region is relatively free of hotspots.

The **Adriatic Sea** has been extensively monitored for PCBs. It ranges from unpolluted to moderately polluted coastal areas. Concentrations are low along the western coast, with areas with high concentrations in the eastern bank, along the coasts of Croatia and Albania.

In the case of **Aegean-Levantine** spatial analysis is limited to Cyprus (fish) and Turkey (mussels and fish). The concentrations in mussels (Turkey) were rather low. The values for *Mullus barbatus* in the MED POL database can also be considered in the medium-low range, taking into account the higher accumulation capacity of fish with respect to mussels.

PCBs in marine biota across the Greek coastal environment, determined within the Greek monitoring system, were also low, the highest concentrations being observed in mussels collected from the Saronikos Gulf (industrial and urban effluents). On the other hand, bioaccumulation in fish revealed a homogeneous pattern indicating no point sources of pollution.

# **Temporal trends**

Temporal trends were preliminarily assessed in the Thematic Assessment Report on Hazardous Substances (UNEP/MAP/MED POL, 2011), taking into account the limited data available. The analysis focused mainly on the mussel *Mytilus galloprovincialis* (MG) and the fish *Mullus barbatus* (MB), with the exception of Israel, where the clam *Mactra corallina* (MC) was considered. For sediments, a few available time series from Israel (Haifa Bay) and France (Gulf of Lions) were considered.

As regards temporal trends for **trace metals in sediments**, a general downward trend of concentrations of trace metals is observed in sediment samples from the Haifa Bay, except for Cu. In the Gulf of Lions, the French RNO monitoring system has shown an apparent decrease of metals, particularly for Pb. Unfortunately, there is no information about the most polluted area of Marseille-Fos. On the contrary, concentrations of Hg, a critical contaminant in the Gulf of Trieste, do not show a decline, even 10 years after the closure of the Idrija mercury mine,.

As regards **temporal trends of trace metals biota**, in mussels, the country median values do not exhibit clear trends for trace metals, with the exception of particular situations such as Cd concentrations in Slovenia and in Morocco. However, in several cases there is an apparent decline of outlier values, such as in the case of Italy, which may reflect a general improvement on the hotspots.

Individual trends for trace metals at representative stations of the different sub-regions do not exhibit specific tendencies but it can be concluded that, in general, concentrations are relatively constant or declining. In the Western Mediterranean at a few stations, usually corresponding to those exhibiting the higher values (e.g. Marseille, Fos and Pombino), slightly increasing trends were observed, while

those of Genova and Naples show evidence of a decline. Data for trace metals in samples of mussels *Mytilus galloprovinciallis* collected in 21 stations in the period 1979-2006 within the French monitoring system (RNO) clearly show a general decline of concentrations during this time span.

In the Adriatic, trace metal levels did not show significant changes with time. However, general upward trends were observed in the stations exhibiting high values, particularly for Cd (e.g. Rijeka and Kastela Bays in Croatia and Durres and Vlora Bay in Albania). A long survey (1999-2010) performed by Slovenia in the Gulf of Trieste has shown that the concentrations of Hg in mussels have not declined during this period.

The coverage of Central Mediterranean and Aegean-Levantine regions is very limited. Levels of Cd, Hg and Pb in mussels decreased during the period 2001-2008 in the Bizerta Lake (Tunisia). In Israel, the levels found in clams during 2001-2010 in Haifa Bay show a significant increasing trend, despite the concentrations of trace metals in sediments revealing a consistent decline. In Turkey, red mullet collected in three stations in the south of the country (Mersin, Goksu and Tirtan) revealed upward trends of trace metals.

As regards temporal trends of **chlorinated pesticides in biota**, concentrations in mussels show clear evidence of a decline over time which is consistent with the banning of production and use of these compounds. The median values in mussels from Croatia and France exhibit clear decreasing trends, as well as the outlier values in the latter. The only exception seems to be Albania (e.g. Durres and Vlora Bay) which is known to keep stockpiles of obsolete chlorinated pesticides. The concentrations in mussels from Ismir Bay (Turkey), identified as a hot spot in the region, also exhibit a significant increase, probably due to a less efficient management of the existing regulations concerning the stocking of these obsolete pesticides. In general, the decreasing trend is faster for lindane and the other chlorinated pesticides than for DDT that appears to be consistent with the higher long-life of the latter.

The assessment of temporal trends of **PCBs** in biota is more problematic due to the lack of long-term consistent data mainly due to the change in concentration units (from Aroclor to individual congeners). The profiles obtained do not allow deriving any conclusion. However, the temporal trends for some individual stations generally reveal conservative or even increasing trends, probably reflecting an inefficient management of the existing regulations concerning their use and stockpiling.

In the Western Mediterranean, the stations showing relatively high levels of PCBs still exhibit increasing trends whereas for those more pristine sites trends seem to be stable or decreasing. For the other regions there is limited availability of data to allow drawing precise conclusions. Temporal trends from the Greek MED POL monitoring programme did not indicate a reduction of PCB levels despite the ban, indicating continuous inputs into the coastal environment.

As a conclusion from the above analysis, it was stressed that there is a need to establish monitoring programmes in many countries to fill the geographical data gaps, and ensure the continuation of existing temporal trend data. These programmes must be able to generate comparable and accurate data, taking into account the intrinsic variability of the environmental matrices considered.

Moreover, with the improvement and development of analytical techniques, the identification and quantification of emerging substances of potential concern for the marine environment because of their persistence, toxicity and bioaccumulation properties, is continuously increasing. They are believed to be ubiquitous but information on their occurrence in the Mediterranean is limited and should be improved.

#### 3.2 Scientific literature

A good basis in the scientific literature to undertake an assessment of the state of the Mediterranean marine environment is only available for the following substances or group of substances:

- Heavy metals (Cd, Pb, Hg)
- PAHs

- PCBs
- DDTs

The following substances or groups of substances are partially covered in the scientific literature:

- Organotin compounds (TBT)
- Dioxins and furans
- HCH
- Hexachlorobenzene

While for the rest of substances of concern very few data in the Mediterranean region are available in the scientific literature.

# 3.2.1 Metals and related compounds

### Heavy Metals (Cd, Pb, Hg) in sediments

Data about cadmium contents in marine sediments is scarce in the Mediterranean Sea, and concerns mostly sediments from the Northern Mediterranean. Roussiez et al. (2006) carried out a comprehensive study of the Gulf of Lions, where they found levels ranging from 0.22 to 0.82  $\mu$ g/g dw, being 0.36  $\mu$ g/g dw the average Cd concentration. In the Southern Evoikos Gulf, in the Aegean Sea, Cd values are slightly lower and ranged from 0.10 to 0.65 ppm (Angelidis and Aloupi, 2000). Lafabrie et al. (2007) also recorded Cd in marine sediments next to the Toscanian coast and the Corsica and Sardinia islands, being 0.40  $\mu$ g/g dw, 0.03  $\mu$ g/g dw and 0.07  $\mu$ g/g dw respectively. Finally, the Strait of Sicily has registered higher Cd levels between 0.1 and 1.6  $\mu$ g/g dw, being 0.7  $\mu$ g/g dw the average concentration (Böttcher et al., 2003).

Studies on lead levels in marine sediments have also been focused in the North Mediterranean. The Gulf of Lions (North Western Mediterranean) has registered concentrations ranging from 20.6  $\mu$ g/g dw up to 69.7  $\mu$ g/g dw with a mean concentration of 39.2  $\mu$ g/g dw (Roussiez et al., 2006). The Gulf of Taranto (Ionian Sea) recorded higher Pb values ranging between 44.7 to 74.8  $\mu$ g/g dw, although levels in sediments offshore dropped to 59.0  $\mu$ g/g dw. In the Southern Evoikos Gulf, Pb registered 13.8 to 19.4  $\mu$ g/g dw (Angelidis and Aloupi, 2000). Lafabrie et al. (2007) found 44.50 in the Toscanian coast, 4.67  $\mu$ g/g dw in sediments next to Corsica and 18.67  $\mu$ g/g dw next to Sardinia.

Data available about Mercury levels in Mediterranean sediments is commonly related to industrialised and densely occupied areas. Lafabrie et al. (2007) found Hg values up to 560 ng/g dw in the Toscanian coast, 20 ng/g dw in marine sediments in North-east of Corsica and 70 ng/g dw in Sardinia. Buccolieri et al. (2006) detected Hg concentrations in the Gulf of Taranto ranging from 40 to 410 ng/g dw in sediments near the coast and 70 ng/g dw in sediments offshore, in the centre of the gulf.

Concentrations of Hg in sediments offshore have also been studied. Levels in the Alboran Sea have been found to be around 56 ng/g dw. Very similar concentrations of about 44 ng/g dw Hg have been reported in the South Levantin Basin, the Ionian Sea and in the Levantin Basin (Ogrinc et al., 2007). An extensive study in the Strait of Sicily revealed that Hg levels ranged from 50 to 70 ng/g dw, with samples registering higher contents peaking up to 202 ng/g dw (Di Leonardo et al., 2006). Similarly, sediments of the Strait of Otranto reached 78 ng/g dw (Ogrinc et al., 2007). A distribution pattern for Hg can be observed from the available data, as higher Hg concentrations occur mostly in the coastal sediments, while the offshore sediments record lower Hg contents.

It is difficult to provide <u>background levels</u> of heavy metals so as to easily assess whether sediments are polluted and their degree of contamination. Metal natural levels may fluctuate from one geographical area to another depending on the type of substrate and the physical characteristics of the soil along with other chemical properties (i.e. amount of organic matter, carbonates) (Boughriet et al., 2007; Choi et al., 2006).

Many studies have shown that sediments can provide evidence of human impact on the environment since they present a historical record of human activity and pollution history (Vesely et al., 1993). In this case, Cadmium, Mercury and Lead reference values in the Mediterranean Sea have been estimated from different studies having collected sediment cores throughout the whole Mediterranean area.

During 1994-1995 the RNO collected five cores within the French Mediterranean coasts. For Mercury, background levels were estimated to an average value of 100 ng/g dw taking into consideration chemical fractions (i.e. organic matter, carbonates) and sediment grain-size (RNO, 2001). Similarly, other studies have reported reference values up to 38 ng/g dw for the Strait of Sicily (Di Leonardo et al., 2006) and ranging 100-200 ng/g dw in the Gulf of Naples (Baldi et al., 1983). In the same way, 0.15  $\mu$ g/g dw was established as the Cadmium limit concentration in non-polluted sediments of the French coasts (RNO, 2001) as well as for the whole Mediterranean Sea (Whitehead, 1984, in RNO, 2001). Data from a survey by IFREMER revealed lower Cd background values ranging 0.06-0.08  $\mu$ g/g dw in coastal areas from Morocco (RNO, 2001). For Lead, sediments registering levels higher than 30  $\mu$ g/g dw were considered as polluted in the French coastal area (RNO, 2001). Previously, concentrations of 25  $\mu$ g/g dw had been established as reference values (Whitehead, 1984, in RNO, 2001). Again, data from the Moroccan coastal area reported lower background levels of 17-21  $\mu$ g/g dw (RNO, 2001).

Taking into consideration current levels found in the Mediterranean Sea and published data concerning background concentrations, it may be suggested that Hg pollution occurs mostly in the coastal sediments, while Hg values in offshore areas are within the natural range values. For Cadmium, all the study sites described in the present report were far beyond the established background levels in the Mediterranean Sea. In the case of Lead, all coastal sediments reveal pollution levels except for the Southern Evoikos Gulf, whose Pb concentrations lay within background concentration ranges. Sediments distant from coasts also report natural Pb levels.

### Heavy Metals (Cd, Pb, Hg) in biota

Mussels are worldwide recognized as pollution bioindicators and they are used in mussel watch programs since they accumulate pollutants in their tissues at elevated levels in relation to pollutant biological availability in the marine environment. However, in the Mediterranean Sea, research studies concern mostly the North-western basin, and scarce data has been identified in the scientific literature concerning Hg levels in mussels.

Kljakovic-Gaspic et al. (2007) monitored the Blue Mussel (Mytilus galloprovincialis) in the Mali Ston Bay, located on the eastern Adriatic coast, from 1998 to 2005. The content of trace metal concentrations in the edible tissue of mussels (averages Cd:  $1.15~\mu g/g$  dw.; Hg:  $0.15~\mu g/g$  dw.; Pb:  $1.09~\mu g/g$  dw.) fell in the range of values usually found in low to moderately contaminated marine coastal areas, although according to EU and WHO legislation and guidelines, consumption of the edible tissue of the mussels was not harmful for humans. Analysis of temporal trends during the 7 years of monitoring showed that metal concentrations had not changed over time.

Similarly, a monitoring survey carried out during the 2001-2005 period in the Croatian coast using the blue mussel as an indicator species, determined that Pb and Hg were significantly elevated in the urban and industrial areas, while Cd was more uniformly distributed across the monitored sites, being also high in mussels from rural areas located far away from anthropogenic sources of pollution. The majority of values were yet below the maximum thresholds for fresh seafood. Again, metal concentrations had not changed during the five year-period (Kljakovic-Gaspic et al., 2007).

Blue Mussel individuals sampled in the Gulf of Taranto revealed Cd levels ranging from 0.23 up to 0.95  $\mu$ g/g dw between 1999 and 2000, slightly lower than those registered in the Adriatic Sea, while Pb concentrations were higher, ranging from 1.19 to 4.49  $\mu$ g/g dw (Cardellicchio et al. 2008).

In contrast, the Tyrrhenian Sea has recorded lower metal contents in the same species, analysed in 2000. Cadmium ranged between 0.32 and 0.49  $\mu g/g$  dw while Pb levels ranged from 1.67 up to 2.49  $\mu g/g$  dw. These values appear to be particularly low when comparing to highly polluted areas such as the Venice Lagoon, where Cd and Pb levels have been recorded to reach 4.64 and 8.26  $\mu g/g$  dw respectively (Conti and Cecchetti, 2003).

Mussels from the Turkish-Aegean Sea also appear to be more polluted, since trace metal values in M. galloprovincialis from 2002-2003 are more elevated. Cadmium ranged between 0.04 and 0.52  $\mu$ g/g ww (approximately 0.20 – 2.6  $\mu$ g/g in a dry weight basis using the "5" OSPAR Commission converting factor), while Pb ranged from 0.49 to 1.72  $\mu$ g/g ww (2.45 – 8.6  $\mu$ g/g dw) (Sunlu, 2006).

Concerning top predators, significant amounts of data are available concerning trace metal concentrations in different organs and tissues from predators at the top of the food chain in the Mediterranean Sea area.

Storelli et al. (2005) analysed heavy metals in Bluefin Tuna (*Thunnus thynnus*) and Swordfish (*Xiphias gladius*) caught in the Ionian Sea in 1993. In both species, metal levels in livers were approximately twofold than those found in muscle tissues, except for cadmium which showed considerably higher concentrations in livers. Comparing the two species, a significant variation in certain metal concentrations was observed. Mean mercury, lead and cadmium concentrations in muscle tissues and in livers were about two to nine times higher in bluefin tuna than in swordfish. Bluefin tuna appeared hence more contaminated than swordfish. In contrast to swordfish, tuna appeared to be more able to concentrate large amounts of mercury, cadmium and lead, demonstrating a potential as a bioindicator of pollution of the open sea ecosystems. In comparison with data reported by other authors, mean levels of Cd and Pb in Bluefin tuna were similar to those reported in the same species from the Mediterranean Sea. In addition, Cd and Pb concentrations occurring in muscle tissues of swordfish were also comparable to levels already reported.

Similarly, comparable mercury concentrations have been reported in bluefin tuna of similar size from the Tyrrhenian Sea, while in specimens of a larger size noted much higher values, as Hg concentrations in fish increase with body size (Storelli et al. 2005).

Regarding <u>cetacean species</u>, Monaci et al. (1998) studied trace metal concentrations in different tissues of striped dolphin (*Stenella Coeruleoalba*) from the Spanish coasts and the Ligurian and Tyrrhenian Seas between 1987 and 1994. Mean <u>concentrations of Hg</u> found in liver and other tissues were very high (mean for all liver samples of 976  $\mu$ g/g dw), but were in accordance with other reports from the same geographical area. As Hg accumulates with age in all tissues, it may reach concentrations of several hundreds of  $\mu$ g/g (wet weight basis) in dolphins of 30-40 years. The highest concentration found was 5,441  $\mu$ g/g dw in liver.

On the other hand, Hg levels in dolphins from the Ligurian and Tyrrhenian area were significantly higher than those from the Spanish area. In this case, the pattern of bioaccumulation of Hg found in the tissues of striped dolphins stranded along the Spanish and Italian coasts suggested the existence of two populations, distinct in terms of spatial niche and position in the food chain, presumably exposed to different dietary levels of these elements as a result of different man-made and natural inputs in the two geographical areas.

In addition, striped dolphins of the Mediterranean seem to have substantially higher concentrations of Hg than those of the Pacific and Atlantic. Mercury mean values of 205  $\mu$ g/g ww have been reported in liver of dolphins captured near the Japanese coasts (Monaci et al., 1998). Moreover, liver concentrations of Hg in dolphins stranded on the Mediterranean coasts of France have been found to be 6-7 times higher than those in dolphin stranded on the Atlantic side of the same country. In line with these statements, other studies have found mean concentrations of 668  $\mu$ g/g ww in 13 dolphins stranded on the Mediterranean coasts of France (Monaci et al., 1998).

More recent studies concerning Hg in cetaceans' species have also shown elevated levels. Capelli et al. (2007) studied Hg, Cd and Pb content of six cetacean species in the Ligurian Sea, where these species find their breeding and feeding needs, in order to assess anthropogenic pressure on this rich faunal biodiversity.

Mercury concentrations were largely variable: low values were found in *B. physalus*, *P. macrocephalus*; higher concentrations were observed in *Z. cavirostris* and *S. coeruleoalba*, although it must be pointed out that Hg levels in *S. coeruleoalba* were lower than those observed by Monacci et al. more than one decade before; finally, extremely high levels in *G. griseus* and one *T. Truncatus* were stated.

In the same species, the low range concentrations corresponded to the younger individuals, indicating the strong effect of age on Hg accumulation. The highest concentrations found in G. griseus and T. truncatus reached more than  $100 \mu g/g$  dw Hg in muscle. Other values in literature are comparable and even higher to those results, with the maximum ranging from 156 to  $334 \mu g/g$  dw. in T. truncatus and

from 123 to 1580  $\mu$ g/g dw in *G. griseus* and (Capelli et al., 2007). The liver was again the organ where the highest mercury concentrations were detected, exceeding 2,000  $\mu$ g/g dw.

When comparing to the Atlantic populations, Hg concentrations in muscle and liver of specimens from the Atlantic were lower. *S. coeruleoalba* and *T. truncatus* are the most frequently studied species. Higher Hg concentrations in Mediterranean individuals are frequently detected in *T. truncatus* when comparing to the Atlantic ones (Capelli et al., 2007).

Between 1987 and 1994, <u>Cd concentrations</u> found in tissues of dolphins from Ligurian and Spanish Mediterranean waters were slightly lower than those reported in previous studies. Tissue levels of Cd are influenced by dolphin diet, which varies with circumstances and availability. Cephalopods, an important dietary item for striped dolphins, are efficient accumulators of Cd and are therefore the dolphins' main source of exposure to this metal (Monaci et al., 1998).

Spatial trends observed in Cd concentrations in dolphins from the two analysed areas, the Ligurian Sea and the Spanish coasts, were similar to those of Hg. Dolphins from the Tyrrhenian and Ligurian Seas revealed higher Cd contents with respect to those from the Spanish coastal area, which may be due to greater Cd contamination from man-made sources in the former areas, or to the fact that the Italian dolphins come closer to the coast than Balearic dolphins, since the water is deep close to the Italian coasts (Monaci et al., 1998). Cardellicchio et al. (2002) studied *S. coeruleoalba* caught in 1991 from the South Italian waters, and found Cd levels up to 0.04 and 1.50  $\mu$ g/g ww in muscle and liver respectively (0.16 and 6  $\mu$ g/g in a dry weight basis using the Becquer et al. (1995) 0.25 converting factor). As it can be observed, Cd values from the Monaci et al. (1998) and Cardellicchio et al. (2002) studies are similar, as they concern the same species in the same geographical area and the same timeframe.

Regarding the study of Capelli et al. (2007), contemporary Cd levels in cetaceans from the Ligurian Sea have been reported to be in several cases below detection limit, and are usually low (between 0.01 and 0.82  $\mu$ g/g dw) except for the liver and kidney of *G. griseus* (38 and 71  $\mu$ g/g dw, respectively). Cadmium levels in *S. coeruleoalba* are still elevated and very similar to concentrations reported in the research studies previously described.

It has been reported that kidney is the critical organ for Cd accumulation. As suggested by other authors, Capelli et al. (2007) have also attributed the high Cd values measured in *G. griseus*, *Z. cavirostris*, and *S. coeruleoalba* to the big consumption of squids, generally rich in cadmium. *T. truncatus* shows renal Cd levels lower than *S. coeruleoalba*, as a result of the different amount of cephalopods in their diet.

Concerning <u>Pb levels</u>, Capelli et al. (2007) found that about 50% of the Pb values measured fell below the detection limit; values were low and ranged between 0.04 and 0.74  $\mu$ g/g dw, except for an anomalous high range value (2.7  $\mu$ g/g dw) in the liver of *G. griseus*. These values can be compared to data from more than one decade before. Cardellicchio et al. (2002) found slight higher Pb levels of 0.48  $\mu$ g/g dw in muscle and 0.88  $\mu$ g/g dw in liver in *S. Coeruleoalba* species from the South Italian coasts.

# 3.2.2 Organic tin

Organotins (OTs) have been widely used during the last decades as biocides, agricultural pesticides, polymer stabilisers, wood preservatives, and catalysts in a number of industrial processes. Nevertheless, their main source in the marine ecosystem is related to their use as antifouling paint biocides (Diez et al., 2002). OTs degrade slowly in the environment and in particular in sediment, and their high persistence and ecotoxicological risk has been widely documented since their discovery in Arcachon Bay (France) in the early 1980s. They are responsible for many deleterious effects to nontarget aquatic life (i.e. imposex, oyster malformation, mussel larvae mortality) at very low concentration levels (few ng/kg) (Diez et al., 2002).

Several countries have banned its use and restricted its application to large vessels. The French authorities banned the use of antifouling paints containing tributyltin (TBT) on boats less than 25 m long in 1982 and similar regulations on the usage of TBT in paints came into effect few years later in the European Union in 1989 (Diez et al., 2002). Following the retail ban on TBT-based antifouling

paints in the late 1980s in most of the developed countries, monitoring programmes are currently carried out in order to assess the effectiveness of present regulations.

Diez et al. (2002) performed a study concerning OT levels in <u>sediments</u> from the Northern and Southern areas of the Spanish Mediterranean Sea, between 1995 and 2000. OT concentrations in sediments from this area varied widely depending on the location. Results for the 1995 sampling survey revealed that butyltin compounds were predominant in the whole area of study and the highest levels were found in the Barcelona commercial harbour with average levels of 4,487 ng/g for TBT, although maximum concentrations TBT peaked up to 18,700 ng/g.

Data from 2000 revealed that most of the locations in the Barcelona commercial harbour increased compared to the concentrations obtained 10 yr before.

A study carried out by Martinez-Lladó et al. (2007) concerning TBT levels in the Barcelona harbour showed that the most enriched sediments were found in Port Vell, with levels ranging 326–4,702 ng/g dw.

In the Alboran Sea, OT concentrations in sediments of nine harbours and twelve sites have also been determined. The Almería harbour was the only commercial harbour and showed the highest values for OTs among the ports located into the southern basin, with maximum levels reaching 2,135 ng/g for TBT.

Similarly, OTs were measured again in 38 sediments collected in harbours of the Western Mediterranean Sea and domestic and industrial sewage disposal sites (Diez et al., 2003). The highest levels of TBT were detected in commercial harbours associated with inputs from large vessels (mean value: 2000 ng/g as Sn). Moreover, relatively high TBT values were also detected in fishing and recreational boating areas (mean level. 400 ng/g as Sn). Phenyltin derivatives occurred at lower levels than butyltins.

Furthermore, a comparative study of the OT occurrence in the sewage sludge disposal sites offshore Barcelona and Almeria (domestic) and into the Tarragona harbour (industrial) showed that domestic primary sewage sludge effluents can also contribute to coastal OT pollution (100 ng/g of TBT as Sn), although sewage sludge is a secondary source of OT contamination in the coastal environment in comparison to commercial harbours (Diez et al., 2003).

OTs were also measured in sediments of Toulon Bay in France during 2008-2009 (Pougnet et al, 2014) and the highest TBT concentrations were 2700 ng/g.

Concerning the Levantin Basin, a review from the Egyptian Mediterranean Sea reported levels in sediments from the Alexandrian coast in the range of 35–975 ng Sn/g dw, with the highest levels reported in the harbour and levels from sediments collected in Alexandria port ranging 1–2,067 ng Sn/g dw (Barakat, 2004). The high relative concentrations of TBT found in the sediments indicated that degradation processes in the sediments are minor, due to the anoxic sedimentary conditions at the sampling sites or to relatively recent inputs of TBT to these sites (Barakat, 2004).

In Malta, butyltins in superficial sediments also determined that the two main local sources of marine contamination by TBT were ship-repairing dockyards and marinas. In sediments concentrations ranged 30-1,500 ng/g Sn dw. At TBT levels found in local harbours, several sublethal biological responses were observed (i.e. imposex, inhibition of enzymatic processes), evident at TBT concentrations below the environmental quality standard (20 ng TBT/L) (Axiak et al., 2000).

The occurrence and seasonal variation of butyltin species were studied in seawater along the Mediterranean coasts of Turkey (Yemenicioglou and De Mora, 2009). OTs distribution in seawater was also studied along the Valenciana coastal waters (Marti et al, 2011) and TBT concentrations in Port of Denia were found above the corresponding EQS-MAC value.

<u>Marine bivalves</u> are among the most studied organisms and have also been chosen as sentinel organisms in TBT pollution monitoring programmes. A few reports are available concerning the accumulation of organotin compounds in fishes.

Morcillo et al. (1996) performed a study concerning TBT content in mussels (*Mytilus galloprovincialis*) from the Barcelona, the Masnou and the Sant Carles harbours in 1995. The

maximum concentration of organotin compounds was found in molluscs collected in Masnou marina, where the mean concentration of total organotin compounds was 5,444 ng/g dw as Sn.

In the Aegean Sea, TBT values averaging 17.1 ng/g www ere recorded in *M. galloprovincialis* collected between 2002-2003, similar or even lower than levels reported worldwide (Chandrinou et al., 2006). In contrast, in the Egyptian coast, marine organisms were found to accumulate TBTs to levels considerably greater than those present in the surrounding water. However, with a cessation of inputs, TBT concentrations have been calculated to be lost in a relatively short time of the order of 1 year. Mussels and clams collected in the Alexandria harbours were analyzed and exhibited concentrations of 97–420 and 93–320 ng/g www of TBT, respectively (Barakat, 2004).

TBT body burden was quantified in the gastropod Hexaplex trunculus collected from the Bizerta channel in Tunisia between 2002 and 2010 and a decrease of TBT levels over time was observed (Lahbib et al, 2011).

Concerning <u>OT</u> contents in <u>fish species</u> Morcillo et al. (1996) also studied organotin compounds in grey and red mullet (*Liza aurata* and *Mullus barbatus*) collected in 1995 in harbours (grey mullet) and offshore (red mullet). With regard to fish analysis, the concentration of total organotin compounds was considerably lower.

OTs levels were also determined in the sea mullet, *Mugil cephalus*, collected along the northern Mediterranean coast of Morocco as well as in the south Mediterranean coast of Spain (Hassani et al., 2006). TBT was the predominant compounds in the analyzed samples, reaching higher concentrations in liver than in muscle. Concerning the Spanish coast, the study demonstrated the absence of pollution in fish in the Spanish coast, although the presence of TBT in some sediment samples reflected its elevated use in the past. In Morocco, all harbour sites showed high levels of contamination, correlating well with traffic and with high maritime activity. (Hassani et al., 2006).

In the Alexandria coast, specimens of *Triglaparus lastovizo* (a demersal fish) and *Diplodus sargaus* (a pelagic fish) were analysed. Result values, ranging from 161 to 250 ng/g ww for *T. lastovizo* and from 155 to 212 ng/g ww for *D. sargaus* appeared to be within the range of levels also found in other marine areas such as the Italian coast as well as the Baltic and North Seas (Said et al., 2006).

Borghi and Porte (2002) determined organotin contamination in deep-sea ecosystems, which have been rarely studied. Several <u>deep-sea fish</u> species (*Mora moro, Lepidion lepidion, Coryphaenoides guenteri, Alephocephalus rostratus* and *Bathypterois mediterraneus*) were collected between 1000 and 1800 m depth in the NW Mediterranean and analysed to determine butyltin and phenyltin compounds. Organotin levels differed greatly among species. Total butyltin residues were up to 175 ng/g ww, and they were comparable to levels found in coastal fish collected along the Catalan coast. The highest butyltin and phenyltin residues were recorded in the liver of *M. moro* (174 and 1,668 ng/g ww as Sn, respectively) followed by *L. lepidion* (43 and 260 ng/g wet wt as Sn), both species from the *Moridae* family. The lowest residues were detected in *A. rostratus* (5 ng/g ww in the liver).

Interestingly, deep-sea fish contained much higher levels of phenyltins (up to 1,700 ng/g ww, and particularly TPT (up to 1,430 ng/g ww), than previously reported concentrations in shallow-water organisms. The obtained results confirmed the long-range transport of OTs to the deep-sea environments, and the subsequent exposure of fish inhabiting nonpoint source areas. The use of TPT in agriculture or as an antifouling agent, its transport to the deep-sea environment associated to particulate matter, and its nonbiodegradable nature in the food chain may account for the high residue levels detected in deep-sea organisms (Borghi and Porte, 2002).

Regarding OTs in top predators, Kannan et al. (1995) studied TBT and its breakdown products (MBT) and (DBT) in bottlenose dolphin (*Tursiops truncatus*), bluefin tuna (*Thunnus thynnus*) and blue shark (*Prionace glauca*). Samples were collected from the Italian coast of the Mediterranean Sea in 1992–1993. Concentrations of total butyltin in the liver of dolphin ranged 1,200–2,200 ng/g ww and were an order of magnitude higher than in the blubber (48–320 ng/g wet wt). TBT was the predominant butyltin species in the blubber while DBT accounted for a higher proportion in the liver of dolphins. Butyltin concentrations in bluefin tuna were lower than those in dolphins, again with TBT highest in the muscle and DBT in the liver. Finally, levels of BTs in blue sharks were lower than those in dolphin

and tuna, with kidney having the highest concentrations. TBT was the predominant form of butyltin derivatives in all the tissues of shark.

On the other hand, Focardi et al., 2000 determined concentrations of TBT and degradation products, (MBT) and (DBT), in the liver and kidney of striped dolphins (*Stenella coeruleoalba*) and bottlenose dolphins (*Tursiops truncatus*) found stranded along the western Italian (Tyrrhenian Sea) and Greek coasts in the period 1992-1994. Butyltin (BT) compounds were detected in almost all the samples analyzed and were higher in the kidney than in the liver. The highest BT concentrations were found in *S. coeruloealba* ranging 780-8,050 ng/g ww in kidney and 15-1,020 ng/g ww in liver. Bottlenose dolphins had lower BT concentrations than striped dolphins according to values found by Kannan et al. (1995) ranging 1,020 – 2,010 ng/g ww, although levels still remained elevated (liver: 27-43 ng/g ww; kidney 1,024-2,014 ng/g ww). Of the breakdown products, DBT was predominant in most liver samples and MBT was more abundant in kidney. The greater polarity of this BT compound with respect to DBT and TBT could explain the high concentrations in the kidney.

### 3.2.3 Polycyclic Aromatic Hydrocarbons

PAHs are formed mainly as a result of pyrolytic processes, especially the incomplete combustion of organic materials during industrial and other human activities, such as processing of coal and crude oil, combustion of natural gas - including heating - vehicle traffic, cooking and tobacco smoking, as well as in natural processes such as carbonization. The compositional pattern of pyrolytic PAHs (dominance of parent PAH with M.W>202), is characteristic of PAH mixtures formed during high temperature combustion (pyrolysis) of fossil fuels (Laflamme and Hites, 1978; Sporstol et al., 1983). On the other hand, unburned fossil fuels contain mainly low MW PAHs (1-3 rings) and their alkylated derivatives as their most abundant constituents (Neff, 1979). The latter compounds are known to degrade more severely than the high-molecular weight, mostly pyrolytic PAHs, through physical-chemical and microbial processes. As a consequence, an apparent predominance of pyrolytic PAHs is commonly observed in the marine sediments, unless outstanding petroleum-related inputs have occurred.

Investigations on PAH levels in the Mediterranean marine environment have been focused on sediments, especially in the North Western Mediterranean and the Adriatic Sea (Albaiges et al., 1984; Grimalt et al., 1984; Lipiatou and Saliot, 1991, 1992; Bouloubassi and Saliot, 1991, 1993a&b; Tolosa et al., 1996; Lipiatou et al., 1997 and references therein), while there are only few studies for the eastern Mediterranean (Gogou et al., 2000; Hatzianestis and Sklivagou, 2002a,b; Tsapakis et al., 2005, Botsou & Hatzianestis, 2012, Parinos et al, 2013) In some areas, some temporal trends are also available, as a result of the French and Italian monitoring networks. In general, PAH pollution in the Mediterranean is widespread detected in coastal areas, clearly influenced by urban and industrial emissions to air and water. Atmospheric inputs are the main source of pollution in the open sea.

In <u>sediments</u>, research has been focused on ports, coastal lagoons, river mouths and coastal enclosures closed to urban centres. Accordingly, only few data is available in the open sea for deep sediments. Although the total number of measured individual PAHs is not always the same, in general higher levels are usually detected in harbours, especially in Alexandria, Trieste and Toulon (PAH values higher than 1  $\mu$ g/g). High levels of PAHs have also been measured in the Gulf of Taranto, the Izmit Bay and Elefsis bay, where a significant industrial and oil refining activity is located. On the other hand, open sea sediments exhibit total PAHs concentrations from 0.1 to 1  $\mu$ g g<sup>-1</sup>

Measured levels in <u>biota</u> (especially in mussels) around the Mediterranean have also been reviewed in the scientific literature. PAH distribution was also studied during RNO and SIDIMAR campaigns and during Mytilos and Mytimed projects (Galgani et al, 2011). Maximum levels of PAHs in biota are usually lower than those reported in sediments. The highest levels (up to 46700 ng/g dw) have been detected for mussels and fish in the Egyptian coasts (El-Sikaily et al., 2002; El Deeb et al., 2007). Levels of up to 3000-4000 ng/g dw have been detected in French and Italian coasts, while in other regions they are generally below 1000 ng/g dw.

In both the Western and Eastern Mediterranean sites, the relative abundance of pyrolytic compared to petrogenic PAHs was slightly elevated at the deep sampling sites compared to the shallow ones (Tolosa et al., 1996; Gogou et al., 2000). Such a trend has been previously observed in several marine

environments and most likely results from: I) the preferential preservation of PAH released from combustion processes which are known to resist to degradation during atmospheric transport and sedimentation (Baker et al., 1991; Behymer and Hites, 1988; Wakeham, 1996), II) the preferential association of pyrolytic PAH with fine particles, which can be transported along long distances from the coast (Boehm and Farrington, 1984; Readman et al., 1984), and III) finally, this trend suggests that the atmospheric deposition could be the major source of pyrolytic PAHs in the open marine sites. The latter statement is further supported by the distribution profiles of PAHs in background aerosol samples, collected over the Mediterranean Sea (Simo et al., 1997; Gogou et al., 1996), which are similar to the one observed for the majority of the open Mediterranean Sea sediments.

### 3.2.4 Organohalogen compounds

### **PCBs**

Polychlorobyphenyls (PCBs) are ubiquitous global pollutants which can be often found in different environmental compartments, although their use and production in Europe has presently been severely restricted since 1987. They have been used in numerous industrial applications, such as dielectric fluids, insulators, additives and preservatives (Castells et al., 2008; Borrell and Aguilar, 2007).

Many data is available in the Mediterranean Sea region, and extensive reviews have been published by Tolosa et al. (1995) and by Gómez-Gutiérrez et al. (2007). This latter review considered concentrations in <u>sediments</u> from Mediterranean remote areas as a reliable approximation to background values, and adopted the 1 to 5 ng/g dw (Aroclor eq.) interval as a reference for non polluted PCB sediments in the Mediterranean Sea.

The main problem when comparing and interpreting the available data is that different scientists measure and report varying numbers of PCB congeners. The reported sum of PCBs can correspond to as little as seven or as many as 60 congeners or even to technical mixtures equivalents such as arochlors

Highest concentrations in the Mediterranean Sea were reported closed to urban and industrial areas (e.g. from Marseille, Barcelona, Naples) as well as in river discharges (e.g. from the Rhône, Ebro and Nile). Castells et al. (2008) found average levels for 12 PCB congeners in sediments from the Barcelona coastal area varying between 2.33 and 23.51 ng/g dw, with the most elevated concentration reaching 44.00 ng/g dw, although other studies have reported much higher PCB concentrations up to 2,224 ng/g dw in the same area (Gómez-Gutiérrez et al., 2007). The Marseille's coastal sediments have also registered very high PCB levels, varying between 14 and 15,815 ng/g dw. In the South Western Basin, PCBs have been found up to 323 ppb in sediments from Oran, Algeria, while in the Eastern Basin, values from the Alexandria coastal area ranged from 0.1 up to 96 ng/g dw.

Bays and Gulfs have also been largely studied. PCBs in the Naples and Pozzuolo Bays (Tyrrhenian Sea) have registered elevated concentrations up to 3,200 ng/g dw. In the Aegean Sea high values are also found, in Thessaloniki and Thermaikos Gulf reported up to 299 ng/g dw and the Piraeus and Saronikos Gulf reached 775 ng/g dw.

Finally, important PCB levels are also found in areas of river discharge. In the Northern Basin, the Rhone and Ebro Prodeltas recorded 1 to 472 ng/g dw of PCBs and 6 to 200 ng/g dw respectively, while the Po River registered lower concentrations from 13 to 129 ng/g dw. Higher levels have been reported from the Nile River, in the South Eastern Basin, varying between 53 and 1500 ng/g dw (Gómez-Gutiérrez et al., 2007).

When comparing registered levels in the Mediterranean coastal areas to the estimated background values, it appears that very high values are found in the nearshore areas, and most of them are considered as hotspots for PCBs. The Mediterranean Sea region is hence considered as one of the major PCB polluted zones worldwide (Borrell and Aguilar, 2007).

However, decreasing concentration gradients have also been found in transects offshore from these sources, clearly observed in studies concerning PCB concentrations in the suspended particulate matter from coastal and open Western Mediterranean waters. In a recent study covering the whole Western basin, a spatial gradient was observed from the continental shelf (3.5 - 26.6 pg/kg) towards the open

sea (1.7-6.6 pg/kg) and a relatively important enrichment (8.4 pg/kg) in open sea stations located in higher productivity frontal zones (UNEP/GEF, 2002).

Due to their chemical stability and their lipophilic character, PCBs persist in the environment and accumulate in <u>organisms</u>, especially in top predators of the food chain. However, according to some studies on temporal PCB trends in different matrices (mussels, sediment, vertebrates) from the north-western Mediterranean, levels might be tending to decline (Borrell and Aguilar, 2007)

In this sense, an attenuated decreasing trend was observed by the French <u>bivalve</u> monitoring IFREMER/RNO network for coastal pollution during the period 1979–1998 (Borrell and Aguilar, 2007). The French study showed that the decline in chlorinated pesticide concentrations followed the order: DDT > HCHs >> PCB. The PCB decrease was hence not as marked as for the other organochlorines. Likewise, Villeneuve et al. (1999) showed a decrease in PCB concentrations in the blue mussel (*Mytilus galloprovincialis*) from the period 1973-1974 to the 1988-1989. Range values dropped from 18-668 ng/g ww to 4-126 ng/g ww during the fifteen year-period. In the Balearic coasts, similar low range values of 1.6-6.6 ng/g ww were also found in mussels analysed between 1996 and 2000 (Deudero et al., 2007).

Similarly, PCB levels in *Merluccius merluccius* from the Adriatic Sea have been shown to slightly decrease between 1993 and 2003 from 1,380 ng/g to 943 ng/g lipid weight (Storelli et al., 2004), although the remaining levels are still high and the declining trend was not statistically significant. Such values are in accordance to the PCB concentrations in other fish species from the same marine region. Between the years 2000 and 2002, PCB in *Scyliorhinus canicula* from the South Adriatic reached high levels ranging from 500 up to 2,351 ng/g lipid weight (Storelli et al., 2006). In the same line, specimens of *Dicentrarchus labrax* from the Strait of Messina sampled in 2004 reported elevated PCB levels ranging from 63,200 to 109,400 ng/g lipid weight (Lo Turco et al., 2007).

Concerning levels in top predators, information regarding organochlorine compounds in tunas is, in general, scarce and particularly limited in Mediterranean Sea. Storelli et al. (2008) carried out a comparison between PCB levels in juvenile bluefin tunas in the Ionian Sea. Even though their result values were rather low (average 15.92 ng/g ww), recent studies in the Mediterranean region show that adult tuna specimens may reach range values of 224 to 1,660 ng/g ww in the same region and up to 262 ng/g ww in specimens from the whole Mediterranean. Thus, the comparison with literature data showed that concentrations found by Storelli et al. (2008) were markedly lower than those detected in specimens from the Tyrrhenian Sea, Ionian Sea and western Mediterranean Sea, which was attributed to biological parameters such as sex, age and size. Different studies performed in the Mediterranean region highlight that, in general, concentrations found in tuna from the Mediterranean are higher than those caught in other marine areas (e.g. Pacific Ocean), which lay in the low range of the Mediterranean Sea concentrations (Storelli et al., 2008).

On the other hand, concentrations of PCB in bottlenose dolphin blubber (*Tursiops truncatus*) from the North Western Mediterranean have also found to be very high, although they point to a decrease in levels. However, and as in other matrices, PCB decline in bottlenose dolphin between 1978 and 2002 has been found to be much less pronounced than that of other organochlorine pesticides (i.e. DDT). Similar results from other studies suggest that there is a steady source of these chemicals in the Mediterranean ecosystem (Borrell and Aguilar, 2007).

# Dioxins and Furans

Relatively little research has been carried out in the Mediterranean Sea concerning PCDD/Fs, although studies focusing on these compounds are gradually increasing. Some data is yet available regarding PCDD/Fs in sediments for Italy and Spain, especially concerning coastal lagoons, and also regarding biota. In general, the North-western basin appears to be the most PCDD/Fs contaminated part of the Mediterranean Sea.

Most studies have focused on levels in sediments from the Catalan coast. Concentrations in the range of 0.06 to 8.14 ng/g dw (0.40-39.24 pg/g dw WHO-TEQ) have been measured in 45 bottom marine sediments collected from 10 rivers of Catalonia flowing into the Mediterranean Sea in 2000 (Eljarrat et al. 2001a). The total WHO-TEQ (TEQ-PCDD/F + TEQ-PCB) values ranged between 0.43 and 42.8

pg/g dw. Therefore, the safe sediment value of 20 pg TEQ/g dw was exceeded for some of the samples analysed, corresponding to the Besós, Francolí, and Llobregat areas.

A similar study was performed by the same authors concerning coastal sediments from different Spanish pollution hotspots, such as the Tarragona and Almeria harbours or the mouths of the Besós and Llobregat Rivers near Barcelona (Eljarrat et al., 2005). Sediments were collected in 2002 and treated in order to quantify PCDD/Fs contents. When comparing both studies (Eljarrat et al. 2001 and Eljarrat et al. 2005), result values appear to be similar and even slightly higher in 2002.

In 2002, levels ranged from 0.1 to 48 pg TEQ/g dw for PCDDs and PCDFs, the total (TEQ-PCDD/F+TEQPCB) ranging between 0.3 and 75 pg TEQ/g dw. The suggested sediment quality guideline was again exceeded for some samples in 2002. From the 15 sediments analysed, four samples of the Besós and Tarragona areas exceeded the safe value set at 20 pg TEQ/g dw (Eljarrat et al., 2005).

Castro-Jimenez et al, 2013, were studied the accumulation of dioxins in deep-sea crustaceans, fish and sediments from the Blanes submarine canyon in North-Westren Mediterranean sea.

High contamination was detected in samples collected at the Besós as well as in sediments near Tarragona attributed to the high industrial impact of this area. (Eljarrat et al., 2005).

Munschy et al. (2008) performed a study concerning <u>PCDD/Fs levels</u> in <u>mussels</u> (*Mytilus galloprovincialis* and *M. edulis*) in the French coastal areas between 1981 and 2005. The results provide evidence of a general decrease in PCDD/F concentrations in the French coastal marine environment over the 24-year period at almost all study sites, with elevated levels remaining in Toulon. This overall decrease probably reflects the general drop in PCDD/F emissions in Europe since 1980. The lowest contamination level (3.77 pg/g dw) was determined in mussels from Corsica (0.052-0.089 pg WHO-TEQ/g ww). This concentration could be considered as a contemporary background reference level for French coasts and is in the range of the reference level proposed by for mussels from the North Sea (0.1–0.2 pg TEQ/g ww).

Rotllant el al. (2006) performed a study on PCDD/Fs contents in rose shrimp (*Aristeus antennatus*), a deep-sea species living at depths between 600 and 2500m. Specimens from the Western basin (Catalan and Balearic coasts) and from the Eastern basin (Western and Eastern Ionian) were collected and analysed in 2001. The rose shrimp was chosen as a biological indicator to assess contamination in deep environments from the Mediterranean Sea on account on its broad longitudinal and bathymetric distribution throughout the whole Mediterranean basin.

A tendency of PCDD/F levels to increase with depth was also observed. PCDD/F levels in shrimps increased from 153 to 416 pg/g lipid weight in the Balearic Sea (western Mediterranean) and from 61 up to 206 pg/g lipid weight in the western Ionian Sea (eastern Mediterranean). Thus, these results allowed reporting the presence of PCDD/Fs in deep-sea organisms and showed that deep-sea environments are not as undisturbed as might be supposed (Rotllant et al., 2006).

Concerning <u>fish species</u>, Bayarri et al. (2001) carried out a study regarding PCDD/Fs content in anchovy (Fam. *Engraulidae*), mackerel (Fam. *Scombridae*) and red mullet (*M. Barbatus*) from the Adriatic Sea. In general, PCDD and PCDFs contamination levels were found to be low, ranging from 0.33 to 0.50 pg/g ww PCDD and 0.71-1.53 pg/g PCDFs for anchovy, 0.32-0.53 pg/g ww PCDD and 2.49-3.38 pg/g ww PCDFs for mackerel and finally 0.29-0.60 pg/g ww PCDD and 0.99-1.49 pg/g ww PCDFs. As it can be observed, PCDFs analytical contributions were higher than those of PCDD.

I-TEQ results were greater for those species at higher levels in the trophic web (mackerel > red mullet > anchovy), although the higher fat content of these species should also be taken in account for part of the greater fresh weight-based PCDD/Fs measured. Contamination levels fell within 0.23 and 1.07 pg TEQ/g ww in the aforesaid species. Moreover, PCDD/Fs in species from the northern area were in general greater than those from the central and southern areas. Thus, these species showed a trend towards higher contamination levels associated with areas showing increased anthropogenic impact (Bayarri et al., 2001).

Levels of PCDD/Fs residues found in the analyzed fish specimens were of the same order as those observed in most developed countries. These results were hence comparable with PCDD/Fs

concentrations observed in fat sea fish from the Netherlands (between 6.65 and 11 pg TEQ/g fat). In Japan, coastal and marketing fatty sea fish has been reported to be contaminated with 0.33 and 0.87 pg TEQ/g ww, and levels of 0.6 pg TEQ/g ww have also been determined in mackerel from United Kingdom. However, higher dioxin levels have been found in sea fish from Germany (34–43 pg TEQ/g fat) or Sweden (1.8–9.0 pg TEQ/g ww) (Bayarri et al., 2001).

PCDD/Fs concentrations studied in twelve wild fish species and two farmed fish species (Miniero et al, 2014) in Italy were found quite lower than the tolerable maximum levels established in the European Union.

Jimenez et al. (1999) studied liver samples from five cetacean species found along the Italian shore of the Tyrrhenian, Adriatic, and Ligurian Seas within the period 1987-1992. Striped dolphin (Stenella coeruleoalba), bottlenose dolphin (Tursiops truncatus), Risso's dolphin (Grampus griseus), pilot whale (Balaenoptera physalus), and long-finned pilot whale (Globicephala melaena) were the target species. Total PCDD and PCDF levels ranged from 13 up to 112 pg/g ww. The highest levels were found both in striped dolphins with a maximum value of 112 pg/g ww and in Risso's dolphins with a maximum value of 91 pg/g ww. In contrast, the lowest levels were recorded in bottlenose dolphins. Regarding the contribution of PCDDs and PCDFs to total levels, in general a higher percentage contribution was found from PCDFs, which in some cases contributed up to 77%. Generally, the most abundant congener was OCDD, accounting for between 23 and 54% of the total PCDD and PCDF concentrations. The contribution of congener OCDF, (range of 6-44%) was also noteworthy. Total calculated TEQs ranged from 1 to 6 pg/g ww. The species exhibiting the highest TEQs was the striped dolphin with a maximum level of 7 pg/g, followed by the Risso's dolphin with a maximum level of 6 pg/g. Total TEQ levels were lower in the rest of the species studied; the lowest value observed was found in bottlenose dolphins with a concentration of 1.14 pg/g. It must also be pointed out that the major contributors to WHO-TEOs values in all studied species were coplanar congeners of PCBs rather than PCDD/Fs compounds.

Sampling areas (Ligurian, Ionian, and Adriatic) were also compared, and individuals from the Adriatic Sea appeared to have the lowest PCDD and PCDF levels; PCDD/Fs concentrations were higher in individuals from the Ligurian Sea).

Fossi et al. (2004) investigated the presence of PCDD/Fs in blubber of nine striped dolphin *Stenella coeruleoalba* sampled in the Mediterranean Sea (southern Tyrrhenian Sea – Aeolian area) in 2002. Total PCDD/Fs levels reached 73.91 pg/g ww, in accordance with the concentration range of 13-112 pg/g ww found previously by Jimenez et al. (1999 The equivalent WHO-TEQs values were 18 pg/g ww, much lower than those obtained in the previous study when co-planar PCB congeners were also taken into account. Regarding the contribution of PCDDs and PCDFs to total PCDD/F levels, it was found the same percentage contribution from both PCDDs and PCDFs. The lowest contribution to total TEQs came from PCDFs, while the contribution of PCDDs and co-planar PCBs was almost the same.

These results were also compared to populations from the Ligurian and Ionian Seas. In this case, the striped dolphins from the Aeolian area appeared to be less exposed to PCDD/Fs and other organochlorine compounds than specimens from the other Mediterranean areas (Fossi et al., 2004), what was also in accordance to previous studies (Jimenez et al., 1999).

# **Brominated flame retardants**

Brominated flame retardants (BFRs), and in particular polybrominated diphenyl ethers (PBDEs) are high production volume chemicals, popularly used as additive flame retardants by the polymer industry. They are ubiquitous environmental and can be magnified in fish tissue (Law et al., 2006).

Although these compounds are usually are considered as 'emergent pollutants', it has been observed that currently there is no very active research in this area. In the Mediterranean, all the studies identified have been published during the last 15 years.

In <u>sediments</u>, three different coastal areas of the Spanish Mediterranean coasts (n=15) where sampled in 2002 by Eljarrat et al. (2005). A total of 40 PBDEs where included in the analyses, and measured concentrations ranged between 2.7 and 134 ng/g dw, being the BDE-209 the main congener detected.

PBDEs were also studied in sediments from Monastir bay (Tunisia) and only thy BDE-47 congener was detected (Nouira et al, 2013).

The <u>biota</u> has focused the effort of most of the identified studies. PBDEs were studied in mussels collected from Apulia region (Giandomenico at al, 2013), while the maximum concentrations of PBDEs have been detected in cetaceans (in particular dolphins) sampled around the Italian coasts (Pettersson et al., 2004). The BDE-47 was the most prevalent congener in all the studies.

Comparing to other regions, Bodiguel et al. (2008) found out that concentrations of PBDEs in European hakes (*Merluccius merluccius*, *L*.) from the Gulf of Lions were up to 13.6 higher than hakes of similar size from the Bay of Biscay (Atlantic). On the other side, Johansson et al. (2006) observed higher levels of PBDEs in mussels sampled in the English Channel (France), than in a Mediterranean coastal lagoon.

PBDEs were also studied in NW Mediterranean deep-sea organisms (Koening et al, 2013).

In fact, the rate and temporal trends at which PBDE and other organohalogen compound concentrations rise and fall in the environment and in humans is still a source of much research interest. Indeed, it is not yet clear how future trends for these compounds could be anticipated, given the different environmental behaviour of the PBDE congeners and current changes in their industrial applications and in their regulatory measures. Data on the temporal trends in biological samples of the PBDEs are still relatively sparse and these studies require good sample sets of archived samples (Johansson et al., 2006). However, it must be noted that inputs of 'emergent' pollutants can be much higher than 'classic' organohalogen.

# **PFOS**

Perfluorinated compounds (PFCs), and in particular perfluoroctane sulfonate (PFOS), have also been identified as priority pollutants in the marine environment. In the Mediterranean, very few data on the occurrence of these compounds in the marine environment has been identified. Kannan et al. (2002) reported concentrations of PFCs in cormorants, marine mammals and fishes from the Italian coasts, from specimens sampled between 1991 and 1998. PFCs were also detected in filter-feeding molluscs collected in 2008 and 2010-2012 along the French coasts (Munschy et al., 2013, 2015).

Higher levels of PFOS were detected in livers of dolphins (up to 940 ng/g ww.), followed by whales, tuna and swordfish. Another study in the same area (Corsolini et al., 2008) found that the concentrations of PFOS in swordfish were below detection limits (1.5 ng/g ww.).

# Chlorinated paraffins

Very little research has been carried out in the Mediterranean region concerning these substances. Castells et al. (2008) found total CP concentrations varying from 210 to 2,090 ng/g dw. in marine sediments from the Barcelona (Spain) coastal environments.

### **DDTs**

Despite the ban on the production and use of DDTs, elevated levels of contamination are still present in the Mediterranean Sea, which has been considered one of the marine regions with the highest levels of PCB and tDDT worldwide (Borrell and Aguilar, 2007). At present time, inputs have been reported still reaching the Mediterranean coastal environment, mainly by atmospheric deposition of DDT and DDT leaching from agricultural soils, followed by discharges into estuarine areas that would help maintaining DDT presence in the coastal environment (Said et al., 2008).

Gómez-Gutiérrez et al. (2007) carried out a comprehensive review of reported DDT contents in <u>sediments</u> of the Mediterranean region, including main hotspots under the influence of densely occupied areas, and compared them to background values estimated from remote areas of the Mediterranean Sea As it can be seen, high concentrations in sediments near urban and mouth river areas were found in different Mediterranean sub-basins. In the Western Basin, the Rhone and Ebro prodelta registered DDTs levels from 1 to 472 ng/g and 6 to 200 ng/g respectively; concurrently, the coastal areas of Marseille and Barcelona recorded concentrations up to 225 ng/g and 195 ng/g. In the Ligurian Sea, sediments from the mouth of the Po River reached 129 ng/g, while in the Tyrrhenian

Sea, the bay of Naples and Pozzuolo recorded up to 312 ng/g. Finally, in the South Western Basin, an average of 40 ng/g DDTs was found in Alger (Algeria).

However, the highest DDT values have been found in the Eastern Basin. Sediments in the delta of the Nile River registered very high concentrations (up to 1,500 ng/g) in the early 90s, although rather low values were recently recorded in sediments of Burullu Lake (Nile prodelta) with a maximum concentration of 17.39 ng/g (Said et al., 2008). In contrast, very high DDT levels were again found in the coastal sediments of Alexandria reaching 826 ng/g, which resulted from diffuse agricultural runoff and agricultural untreated wastes. In the Aegean Sea, the Piraeus Bay registered similar elevated concentrations up 1,406 ng/g for DDTs in the mid 70s, although recent studies have also pointed out high concentrations in this area.

It must also be pointed out that despite the high and widespread occurrence of DDT in the Mediterranean Sea, a general decline in marine sediments and several segments of the marine biota along the north-western Mediterranean coasts has been generally observed between the 1960s and 2000s (Borrell and Aguilar, 2007). A decreasing temporal trend in sediment concentrations is also quite evident from the review carried out by Gomez-Gutierrez et al. (2007).

Many data is available concerning DDT concentrations in biota; however, it primarily concerns the western Mediterranean Sea. High levels are still found in mussels, fish and top predators, but a decreasing trend is also generally observed. This negative trend is consistent with the chemicals history of use. DDT was particularly used after the 1950s and its production increased exponentially until the 1970s. Manufacture ceased in the western world in the 70s, due to environmental concerns which led to restrictive legislation. Such regulations included most countries bordering the Mediterranean Sea. However, DDT and their derivatives are still being used as precursors of dicofol in some manufacturing plants (Spain) and, besides, the existing stockpiles of obsolete pesticides are poorly controlled or unregulated (Borrell and Aguilar, 2007).

The blue mussel *Mytilus galloprovincialis* has been frequently used as a bioindicator for pollution in the Mediterranean Sea. It was used between 1973 and 1974 in a contaminant survey carried out in the North Western Mediterranean basin; at that time, data reported on synthetic chlorinated hydrocarbons had showed very high concentrations of total DDT compounds in soft tissues of mussels, up to a maximum of 1,212.6 ng/g ww. Interestingly, DDT concentrations in the samples were generally higher than DDE and DDD metabolites (low *p,p'*-DDE/DDTs ratio), which was due to direct exposure to the pesticide (Villeneuve et al., 1999). While in 1973-1974 range values of total DDT were 18 – 668 ng/g ww with an average of 146 ng/g ww, in 1988–1989 the concentrations of DDTs had dropped to an average value of 26 ng/g ww and ranged between 4 and 126 ng/g ww (see **Error! Reference source not found.**). These data indicate a decline of DDT concentrations in the coastal environment of NW Mediterranean since the early 1970s, which is in agreement with the ban on DDT. The decreasing average factor is 5.6 between 1973-1974 and 1988-1989 (Villeneuve et al., 1999).

In the Balearic coasts, levels ranged between 1.6 to 6.6 ng/g ww (Deudero et al., 2007); higher contents have yet been detected in the Gulf of Naples reaching an average value of 177.2 ng/g lipid weight (range 32.1-308.8 ng/g lipid weight), also considerably higher than those reported for similar species from the coastal areas of Norway, Japan, China and the Philippines, subject to high anthropogenic impact (Naso et al., 2005).

Wide differences in DDT range values characterize fish species in the Mediterranean Sea. Low DDT levels are found in the *Dicentrarchus labrax* species in the Strait of Messina (Lo Turco et al., 2007), while much higher levels have been reported in mussels or bluefin tuna in the same area. Slightly higher values were recorded in *Clarries* sp. and *Oresochromus niloticus* in the Egyptian Mediterranean Sea (Said et al., 2008). In contrast, *Scyliorhinus canicula* from the South Adriatic Sea reflected high values with a mean of 1,170.9 ng/g lipid weight and reaching up to 1,875.0 ng/g lipid weight (Storelli et al., 2006).

However, decreasing trends have also been found within fish species. Storelli et al. (2004) showed a substantial reduction of DDT contents in codfish (*Merluccius merluccius*) from the Adriatic over a period of 10 years running from a maximum value of 1132 ng/g lipid weight basis in 1993 to a minimum of 618 ng/g lipid weight in 2003. The decline of concentrations of these contaminants in the

marine environment can be ascribed to the restrictions introduced at the beginning of the 1970s in different countries of Europe. The p,p'-DDE compound had the highest influence on the total DDT. The p,p'-DDE/DDTs ratio was 0.9.

Concerning DDT contents in top predators, the comparison performed by Storelli et al. (2008) between DDT levels in juvenile bluefin tunas (*Thunnus Thynnus*) in the Ionian Sea and other available data allowed concluding a similar pattern to the PCB concentrations (see PCB Section above). Their result values were low (13.06 ng/g ww) but were also in contrast with recent studies concerning the Mediterranean region. Adult tuna individuals may reach range values of 61 to 110 ng/g in the Ionian Sea and up to 178 ng/g in the Mediterranean area, clearly exceeding concentrations found in other regions such as the Pacific Ocean. The p,p'-DDE/DDTs ratio of juvenile tunas was rather high (0.9).

On the other hand, Corsolini et al. (2008) analysed organochlorine compounds in muscle samples of swordfish (*Xiphias gladius*) in the South Tyrrhenian Sea. DDT values in muscle and liver tissues where high, of 155 ng/g ww ( $\pm$ 125) and 309 ng/g ww ( $\pm$ 273) respectively. Among the DDTs, p,p'-DDE was largely the most abundant isomer. Consequently, the *p,p* '-DDE/ $\Sigma$ DDTs ratio was 0.75, which is in accordance with other contemporary results (Storelli et al., 2008).

A rigorous analysis of DDT in bottlenose dolphins blubber between 1978 and 2002 in different areas from the Western Mediterranean coasts allowed to observe high concentrations overall (6,500–548,800 ng/g lipid weight basis). However, a declining temporal trend was observed between 1978 and 2002 for total DDT concentrations, which decreased by a factor of 23.7 (see Error! Reference source not found.). As in other study cases, an increment of the DDE/tDDT ratio was also observed (Borrell and Aguilar, 2007).

High *p,p*'-DDE/DDTs ratios have been generally found in contemporary study cases (Corsolini et al., 2008; Storelli et al., 2008; Borrell and Aguilar, 2007). This ratio acts as an indicator of temporal trends for the DDTs, and high values (over the critical threshold of 0.6) may indicate that DDT contamination in specimens may not be due to present use but to remote use of DDT in agricultural activity. Thus, the ratios obtained (0.9 for Storelli et al. (2008) and 0.75 for Corsolini et al. (2008)) should indicate that the Mediterranean Sea does not receive new inputs of DDTs (Corsolini at al., 2008; Storelli et al., 2008).

It must be taken into consideration when interpreting the former indicator that p,p'-DDE is very stable in the environment and is the major metabolic product of DDT accumulating in the lipid fraction of fish carcasses, what would unavoidably contribute to the augmentation (and skew) of the ratio. Moreover, it is well-known that several countries are still using DDT-based pesticides even within the European Community, where DDT is permitted for uses other than plant protection, and that due to its high persistence and long-range transport potency, DDT can also reach the Mediterranean area from other parts of the world where it is still used intensively (Corsolini et al., 2008). However, despite the former considerations, the high p,p'-DDE/DDTs ratios may indicate that no significant new inputs of DDT have occurred in the north-western Mediterranean in recent decades, or are negligible in comparison to pre-1970s inputs (Borrell and Aguilar, 2007).

# **Hexachlorobenzene**

Hexachlorobenzene (HCB) was first introduced in 1945 as fungicide for seed treatments of grain crops, and used to make fireworks, ammunition, and synthetic rubber. Today it is mainly a by-product in the production of a large number of chlorinated compounds, particularly lower chlorinated benzenes, solvents and several pesticides. HCB is emitted to the atmosphere in flue gases generated by waste incineration facilities and metallurgical industries (UNEP/GEF, 2002). HCB has a relatively high bioaccumulation potential and long half-life in biota.

In a review of POPs levels in Mediterranean sediments, Gomez-Gutierrez et al. (2007) collected available data on HCB concentrations from 361 samples since the 70s, especially from NW Mediterranean, the Tyrrhenian and Adriatic Seas. Higher concentrations are reported in harbours and coastal lagoons (0.1-2800 and 0.3-2400 ng/g dw, respectively), comparing to background levels in deep sea sediments (0.04-0.8 ng/g dw). HCB concentrations in areas influenced by river discharges ranged between 0.05-39.4 ng/g dw (with highest levels in the Ebro and Rhone river mouths), while

levels in urban areas were 0.01-60 ng/g dw, with peak concentrations in Venice (up to 2400 ng/g dw)Error! Reference source not found..

According to the review of Gomez-Gutiérrez (2007), HCB concentrations in Mediterranean sediments did not show a clear temporal trend, although data available for the last period (2000 to present) was in the low range of concentrations. The lack of data in the period of 1970-1980, when its agrochemical application was more important greatly limits the analysis. Furthermore, the fact that HCB is also generated inadvertently as a by-product and/or impurity in several chemical processes or incomplete combustion could also modify any possible temporal trend (Gomez-Gutiérrez, 2007).

HCB is also widely distributed in Mediterranean marine <u>biota</u>. higher levels have been reported in marine mammals, followed by fish and mussels. Villeneuve et al. (1999) reported concentrations of HCB in mussels from NW Mediterranean coasts one or two orders of magnitude higher in 1973-1974 than in 1988-1989 (0.08-1.9 ng/g dw). In the NW Mediterranean HCB levels were also measured by Sole et al. (1994) during the 80s in the Ebro Delta (up to 1.8 ng/g ww) and from 1996 to 2000 in the Balearic coasts (0.1-1.1 ng/g ww) (Deudero et al., 2007). HCB have also been recently surveyed in the Gulf of Naples (Naso et al., 2005).

In fishes, significant levels of HCB (35-215 ng/g l.w.) have been reported in the Ebro Delta (Pastor et al. 1996) and along Spanish coasts (46.5 - 173 ng/g l.w.) (Serrano et al., 2008). Concentrations measured in codfish (M. merluccius) from the Adriatic dropped from 14.7 to 8.6 ng/g l.w. between 1993 and 2003 (Storelli et al., 2004). Lower concentrations (0.12-0.67 ng/g ww) were reported for deep sea fishes (Porte et al., 2000).

Decreasing temporal trends have also been observed in bottlenose dolphins (*T. truncatus*) from the Spanish coasts, where HCB mean concentrations in blubber decreased from 2260 to 0.46 ng/g l.w. between 1987 and 2002 (Borrell and Aguilar, 2007).

HCB was also found in monk seals (*M. monachus*) from Western Sahara and Greece (Borrell and Aguilar, 2007). Mediterranean individuals presented significantly higher levels of HCB concentrations (as well as tPCB, and DDTs) than their counterparts from the Atlantic.

# Lindane (γ-HCH)

"Lindane", which is essentially pure  $\gamma$ -HCH, has been of the most widely used insecticides in the world. Its insecticidal properties were discovered in the early 1940s. It controls a wide range of sucking and chewing insects and has been used for seed treatment and soil application, in household biocidal products, and as textile and wood preservatives (UNEP/GEF, 2002). Lindane and other HCH isomers are relatively persistent in soils and water, are much less bioaccumulative than other organochlorines because of their relatively low liphophilicity.

According to the UNEP/GEF (2002) review, HCHs in <u>sediments</u> were detected during the 80's in coastal sediments from the Western, Central and Eastern part of the Mediterranean, with mean values of 0.5-2.5 ng/g. In the 90s, levels of 0.02-0.94 ng/g dw ( $\Sigma$ HCHs) were reported in the Ebro prodelta, 10-51 ng/g dw ( $\gamma$ -HCH) in Abu Quir and El-Mex Bays, and 7-140 ng/g dw ( $\alpha$ + $\gamma$ -HCH) in Thermaikos Gulf (UNEP/GEF, 2002). Said et al. (2008) reported levels of nd-22.8 ng/g dw ( $\gamma$ -HCH) in sediments of Lake Burullus (Nile Delta).

In <u>biota</u>, a large survey of mussels performed during 1995-99 in the French Atlantic and Mediterranean coasts (over 700 samples) have shown mean values of 0.8/3.6 ng/g dw and 0.5/2.0 ng/g dw for  $\alpha/\gamma$ -HCHs, respectively (UNEP/GEF, 2002). According Villeneuve et al. (1999), average concentrations of lindane in mussels from NW Mediterranean coasts decreased from 49 to 2 ng/g dw (0.8-3.1) between 1973-1974 and 1988-1989. Some negative trends were also observed in mussels from the Balearic coasts between 1996 and 2000 (Deudero et al. 2007).

An extensive survey of *Mullus barbatus* carried out between 1986 and 1991 in 8 coastal stations of the Aegean Sea revealed concentrations of  $\alpha$  and  $\gamma$ -HCH of 0.1-0.5 ng/g ww and 0.6-3.5 ng/g ww, respectively (UNEP/GEF, 2002). Higher concentrations (0.03 – 8.6 ng/g ww) have recently been reported in fishes from Lake Burullus, Nile Delta (Said et al., 2008). In the Adriatic Sea,  $\gamma$ -HCH concentrations in codfish decreased from 7.6 to 3.7 ng/g l.w. between 1993 and 2003 (Storelli et al.,

2004), while in Spanish coasts levels of 19.8 - 37 ng/g l.w. have recently been reported in *S. aurata* (Serrano et al., 2008).

#### 3.2.5 Other contaminants

# Nonyl- and Octyl-phenols

Nonyl- and Octyl-phenols (NP and OP) are the starting material in the synthesis of alkylphenol ethoxylates (APEs), first used in the 60s. These compounds are highly effective cleaning agents or surfactants that have been widely used in a number of industrial sectors including textiles, pulp and paper, paints, adhesives, resins and protective coatings. Alkylphenols can also be used as plasticizers, stabilisers for rubbers, lube oil additives, and the alkylphenol phosphite derivatives can be used as UV stabilisers in plastics (UNEP/GEF, 2002).

Limited information on the occurrence of NP and OP in the Mediterranean region is available. Some studies have been reported in freshwater, sewage sludge (UNEP/GEF, 2002) and coastal waters (Marti et al, 2011, Sanchez-Avila et al, 2012)

<u>Sediment</u> samples were analysed in the Venice Lagoon, being the sum of NP, NP1EO and NP2EO in the range 150-13700 ng/g dw (Marcomini et al., 1990). In the Barcelona harbour, Diez et al. (2006) measured levels of 3.8-77 ng/g dw of NP.

In <u>biota</u>, NP, OP and their ethoxylates were detected in edible molluscs, cuttlefishes and squids, caught from 15 harbours along the Italian coast in 1997. NP reached the maximum concentration of 696 ng/g ww in the squids from the central Adriatic Sea. Levels were lower in mussels and clams (246-270 ng/g fresh tissue). OP generally occurred at levels 30 times lower than NP (Ferrara et al., 2001). A similar study on 8 edible marine species from the Adriatic Sea (Ferrara et al. 2005), determined levels of NP in the range 118-399 ng/g ww and 9.5-1431 ng/g ww in crustaceans and fish, respectively. OP was found at respective levels of 2-7-4.7 and 0.3-3.8 ng/g ww in crustaceans and fish.

Alkylphenols were also measured in red mullet (*M. barbatus*) collected from different sampling sites in the NW Mediterranean (Martin-Skilton et al., 2006). Levels of NP were 280-83100 ng/g bile, and OP levels 10-340 ng/g bile. Evidences of significant alterations in the endocrine system of red mullets were detected.

Corsi and Focardi (2002) determined levels of p-NP in fishes ( $Zosterisessor\ ophiocephalus$ ) in 3 sites of the Orbetello lagoon (Italy) during 1998 and 1999. Mean levels were between 0.12 and 1.41 ng/g ww.

Bouzas et al, 2011 determined levels of alkylphenols in edible bivalves from the Spanish coast. Mean NP values were 14-95 ng/g w/w.

#### Phthalates 4 2 2

They encompass a wide family of compounds. Dimethylphthalate (DMP), diethylphthalate (DEP), dibutylphthalate (DBP), benzylbutylphthalate (BBP), di(2-ethylhexyl)phthalate (DEHP)(C24H38O4) and dioctylphthalate (DOP) are some of the most common. DBP and in particular **DEHP** have been identified as common priority substances for the marine environment. They are widely used as plasticizers, insect repellents, solvents for cellulose acetate in the manufacture of varnishes and dopes. Vinyl plastic may contain up to 40% DEHP (UNEP/GEF, 2002).

Phthalates in coastal seawater have been reported by Marti, et al, 2011 and Sanchez-Avila et al, 2012, whereas no studies have been identified addressing phthalates occurrence in the Mediterranean sediments or biota. Only some unpublished data on DEHP levels (6-9  $\mu$ g/g dw) in sediments located off-shore Barcelona (Spain) are reported in the UNEP/GEF review (2002).

# **Pharmaceuticals**

Pharmaceuticals are a class of emerging environmental contaminants. The occurrence of these compounds in aquatic ecosystems has received increasing attention in recent years due to their pseudo-

persistancy nature (resulting from their continuous discharge into the environment) and potential biological activity. A lot of data has revealed that they can have a negative impact on living organisms and ecosystems. European Union established regulatory guidance to assess the presence of pharmaceuticals in the aquatic environment (Directive 2013/39/EU amending Directives 2000/60/EC and 2008/ 105/EC) (European Comission, 2013) as regards priority substances in the field of water policy. For the Mediterranean region there are only few studies concerning their distribution in coastal marine waters and sediments (Nodler et al, 2014, Moreno-gonzalez, 2015).

### 3.3 Overview

In the following table an attempt is made to summarize the main general findings and trends regarding the occurrence of pollutants in the Mediterranean marine environment, according to the information and data reviewed within this report. It must be noted that a deeper analysis of the identified information, and additional data (e.g. from national monitoring programs) may modify this preliminary overview.

• The <u>lack of comprehensive data</u> to properly assess the state of pollution in the Mediterranean region is a well known limitation, which has been emphasized in previous assessments of the Mediterranean environment (Civili and Jeftic, 1987; UNEP/MAP, 1996; Carvalho and Civili, 2001; EEA, 2006). Data is not homogeneously available in time and space. Most of the information has been published after the 90s, very few temporal trends are available, and data is mainly concentrated in the north western basin (although some significant amount of information is also available for the Aegean and Egyptian coasts).

Overview of the occurrence of pollutants in the Mediterranean marine environment according to the reviewed information.

	Availability of data		Indications from available temporal trends <sup>b</sup>		Maximum reporte levels in:				
	Sed.	Biota	Mainly in <sup>a</sup> :	Sed.	Biota	Matrix	Where		
Metals and related compounds									
Cadmium	Regular	Good	NW/ADR/AEG	NA	Upw.	Cetaceans	NW		
Lead	Regular	Good	NW/ADR/AEG	NA	NA	Mussels	ADR		
Mercury	Regular	Good	North & SLE	NA	Upw.	Cetaceans	NW		
Organic tin	Poor	Regular	NW/ADR/SLE	Upw. / Downw.	NA	Sed / biota	NW / ALB		
РАН	Good	Regular	North & SLE	NC	NC/Downw.	Sediments	NW / SLE		
Organohalogen									
PCBs	Good	Good	North & SLE	NC	Downward	Cetaceans	NW		
Dioxins & Furans	Poor	Regular	NW/ADR	NA	NC/Downw.	Cetaceans			
BFRs	Very poor	Poor	NW	NA	NA	Cetaceans	NW		
PFOS	Very poor	Very poor	NW/ADR	NA	NA	Cetaceans	NW		
Chlorinated paraffins	Very poor	No data	NW	NA	NA	NA	NA		
DDTs	Good	Good	North & SLE	Downward	Downward	Cetaceans	NW		
HCB	Regular	Poor	NW/ADR	NC	Downward	Cetaceans	NW		
	Very								
Lindane	poor	Poor	NW/ADR/AEG	NA	Downward	Sediments	SLE		
Other organic									
Nonyl/Octyl-	Very	Very							
phenols	poor	poor	NA	NA	NA	NA	NA		
Phthalates	Very	Very	NA	NA	NA	NA	NA		

	poor	poor					
	Very	Very					
Pharmaceuticals	poor	poor	NA	NA	NA	NA	NA

NA: Not Available information to be defined.

- Heavy metals (Cd, Pb, Hg), PAHs, PCBs and DDTs concentrates the majority of available data, especially in biota (mussels). Information is also available to partially assess the occurrence of organotins, dioxins and furans, and hexachlorobenzene, while for other 'emergent' priority pollutants, like BFRs, PFOS, chlorinated paraffins, alkylphenols or phthalates, very few data or no data has been identified in the Mediterranean.
- All the considered pollutants for which a certain amount and diversity of data is available, are found to be <u>widely distributed</u> in the Mediterranean environment.
- Regarding temporal trends, the 'old' organohalogen (DDT, PCB, HCB, lindane,...) generally show downward trends in measured concentrations, especially in biota. This is consistent with the phase out and restriction of use of these substances. Non intentional emissions like PAHs or dioxins and furans show also some indications of downward trends in biota, but this is not so evident everywhere. Upward trends in concentrations have only been reported in some cases for heavy metals (Cd, Hg; according to MED POL preliminary data), and TBTs (which would be expected to progressively decrease in the forthcoming years due to restrictions on its use as antifouling paints). Other emergent pollutants might have experienced upward trends during the last years, but not enough data is available to this respect.
- Generally, the <u>higher levels</u> of pollutants are detected in sediments and biota of coastal areas, mainly close to urban centres, harbours and river mouths. However, due to the persistence and liability to bioaccumulate of most of the considered substances, the maximum concentrations are usually reported in <u>cetaceans and top predators</u> (e.g. whales, dolphins, sharks, or tuna). These levels have been mainly reported in the NW basin; unfortunately very few data is available for these species in the eastern basin. Higher levels of PAHs are reported in sediments, which is consistent with the ability that fishes have to metabolize these compounds. The higher levels of lindane in sediments might be related with its relatively low liphophilicity.
- Another consideration to be highlighted is that when the concentrations of pollutants are compared with other regions, several authors conclude that the Mediterranean environment is more affected by the input of pollutants, which is usually attributed to its enclosure conditions. This has been mainly observed for PCBs, DDTs, and dioxins and furans. On the other hand, other pollutants like BFRs or PFOS appear to occur at similar or lower concentrations than in other areas.

Finally, it must be noted, that the potential risk that the actual occurrence of the considered pollutants may pose on the environment or the human health has not been addressed within this report. However, the main evidences in the Mediterranean have been observed for the imposex effect as a consequence of TBT pollution. In the Adriatic, no risk to human health was observed from the exposure to Cd, Pb and PCB through consumption of different seafood. In contrast, the mercury exposure due to consumption of certain fish species indicated that human health risk might be of concern (Storelli, 2008).

# 4. Emission of substances of concern in Mediterranean countries

# 4.1Sources of data

The key source of emission data at regional level is the National Baseline Budget (UNEP/MAP, 2002), which includes emission data to air and water for SAP priority pollutants in all MAP countries. The

<sup>&</sup>lt;sup>a</sup> NW: North western basin (including NWE, TYR, ION); ADR: Adriatic; AEG: Aegean; North (NW&ADR&AEG); SLE: South Levantin East; ALB: Alboran.

<sup>&</sup>lt;sup>b</sup> Upw.: Upward; Downw.: Downward; NC: not clear

baseline year was 2003, with an update published in 2008 (Releases, emissions and sources of pollutants in the Mediterranean region

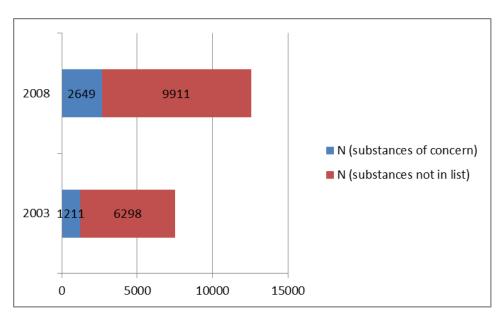
An assessment of 2003-2008 trends, MED POL 2011). In the following sections, an analysis of available data in the NBB is provided, with an emphasis on the priority pollutants identified above. However it should be noted that the database may be considered to be only partial at best (since two countries had not reported when the 2008 update was made, including Greece, the country with the largest number of records in the 2003 data). As was already noted in 2003, even where data are complete, the NBB (and other emission registers) can only ever cover the proportion of the priority substances that are actual and point source releases from industrial installations. In reality, many of the substances addressed here are emitted from non point sources, for example pesticides in agriculture and anti-fouling agents used in shipping; or from environmental resevoirs, such as banned products which are no longer used.

Countries may have also their own national inventories, or participate in other regional initiatives, like the European Pollutant Emission Register (EPER) , which is similar to the NBB, but do not include all the same substances.

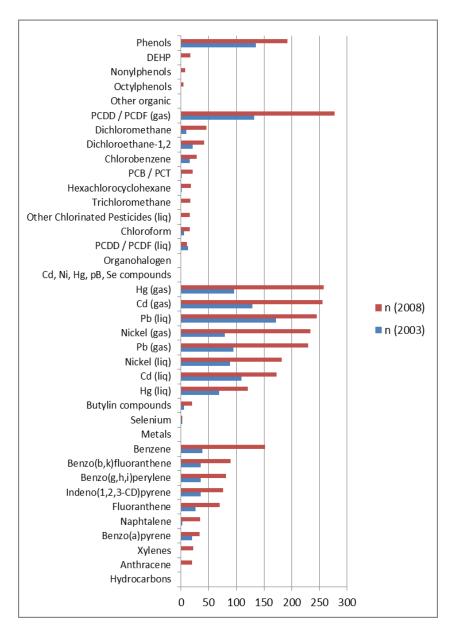
# 4.2The National Baseline Budget: available data

The NBB data was updated in 2008 but this data was not available at the time of the initial assessment and definition of the lists of substances of concern. For the present report, the 2008 NBB data is analysed in the context of the revised list of substances of concern (which for example includes the newly listed POPs and other WFD substances added since 2008).

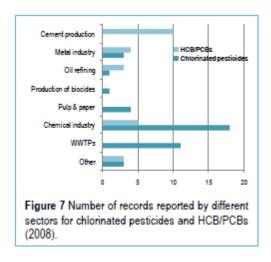
There were a total of 103 substances reported in the NBB database in 2008, compared with 75 in 2003; of these 103, 46 substances are listed as 'substances of concern' following the discussion above. In terms of numbers of records contained in the database for each chemical, 21% of the total records in 2008 relate to these substances of concern, compared with 16% in 2003 (Figure below). Similarly to 2003, the majority of records in NBB concern other categories of pollutant such as nutrients which are not considered in this assessment.



Among the chemicals that are listed as 'substances of concern', the NBB data suggests that there are relatively few that have very high numbers of records, and these are mostly in the metals category (Fig below). The earlier caveat applies however, as this does not necessarily mean that they are the most commonly emitted, rather that they are the most commonly recorded and reported in the NBB. As previously mentioned, the organohalogen pesticides are not likely to feature strongly in the NBB data (the single digit records for these have been removed from the chart below to improve presentation).



The assessment of trends between 2003 and 2008 that was conducted in 2011 identified the main sectors contributing to the records for HCBs/PCBs and chlorinated pesticides – primarily the chemical industry, WWTP and cement production (Figure 7 below).



### 5. Review of Emission Limit Values

### **5.1 HELCOM Recommendations**

Since the original report on priority contaminants, which refers to Recommendations up to and including HELCOM Recommendation 27/1, adopted 8 March 2006, on Limitation of Emissions into Atmosphere and Discharges into Water from Incineration of Waste, the following Recommendations relating to estallibhsing limits from pollution from land based sources have been adopted (please note this list does not include Recommendations relating to limits for nutrient pollution):

Rec 31E-4, adopted 20/05/2010, on Proper handling of waste/landfilling, which establishes leaching limit values for wastes for As, Ba, Cd, Cr total, Cu, Hg, Mo, Ni, Pb, Sb, Se, Zn, Chloride, Fluoride, Sulphate, Phenol index, DOC, and TDS; and establishing limit values for inert wastes (TOC, BTEX, PCBs, mineral oil and PAHs).

Rec 31E-3 adopted on 20/05/2010 on Cadmium in fertilizers, recommending governments to set a limit value for the content of cadmium in fertilizer;

Rec 29-1, adopted 05/03/2008 on Reduction of Emissions from Crematoria, establishing limit values for dust, CO and Hg.

Rec 28E-8, adopted on 15/11/2007 on Environmentally friendly practices for the reduction and prevention of emissions of dioxins and other hazardous substances from small-scale combustion, which (among others) recommends that the Contracting Parties develop in 2008 specific efficiency requirements and emission limit values for small scale combustion appliances

### **5.2 OSPAR Decisions and Recommendations**

Since the previous report, which covered OSPAR Recommendations up to and including -OSPAR Recommendation 2000/3 on Emission and Discharge Limit Values for the Manufacture of Emulsion PVC (e-PVC) from Vinyl Chloride Monomer, the convention has adopted the following recommendations:

OSPAR Recommendation 2002/1 on Discharge Limit Values for Existing Aluminium Electrolysis Plants, supplementing Recommendation 98/2 with discharge limit values to water for PAH (6 chemicals, Fluoranthene, Indeno(1,2,3-cd)pyrene, Benzo(k)fluoranthene, Benzo(a)pyrene, Benzo(b)fluoranthene and Benzo(ghi)perylene)

OSPAR Recommendation 2006/3 on Environmental Goals for the Discharge by the Offshore Industry of Chemicals that Are, or Which Contain Substances Identified as Candidates for Substitution, which recommends that Contracting Parties phase out the discharge of offshore chemicals that are identified as candidates for substitution.

In relation to the revised list of 'Substances of Concern', selenium and phenols are the only new substances to be included in limit values (HELCOM Recommendation 31E-4 for waste leachates) – the other substances already had applicable limits in the original 2008 list.

# 5.3 Stockholm and Basel Conventions

While the Stockholm Convention does not establish Emission Value Limits for the chemicals listed in Annex A with specific exemptions or in Annex B for restriction, it establishes a mechanism to evaluate the continued need for the allowed uses, based on Party submissions of production and use of exempted substances. These exist namely for DDT, endosulfan, lindane, and PFOS. The Stockholm and Basel Conventions address POPs wastes as well as production and use, and in this context have established a Low POPs limit of 50ppm for most of the listed chemicals. Wastes with lower concentrations than this level are not considered 'POPs wastes' and are therefore not subject to the strict requirements for final destruction of POPs.

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# Appendix 1 – List of Rotterdam Convention Chemicals

(back to section on Rotterdam Convention)

Chemicals listed in Annex III of the Rotterdam	CAS number
Convention	CAS number
2,4,5-T and its salts and esters	93-76-5 (*)
Alachlor	15972-60-8
Aldicarb	116-06-3
Aldrin	309-00-2
Azinphos-methyl	86-50-0
Binapacryl	485-31-4
Captafol	191906.00
Chlordane	57-74-9
Chlordimeform	6164-98-3
Chlorobenzilate	510-15-6
DDT	50-29-3
Dieldrin	60-57-1
Dinitro-ortho-cresol (DNOC) and its salts (such as ammonium salt, potassium salt and sodium salt)	534-52-1
Dinoseb and its salts and esters	88-85-7 (*)
EDB (1,2-dibromoethane)	106-93-4
Endosulfan	115-29-7
Ethylene dichloride	107-06-2
Ethylene oxide	75-21-8
Fluoroacetamide	640-19-7
HCH (mixed isomers)	608-73-1
Heptachlor	76-44-8
Hexachlorobenzene	118-74-1
Lindane (gamma-HCH)	58-89-9
Mercury compounds, including inorganic mercury compounds, alkyl mercury compounds and alkyloxyalkyl and aryl mercury compounds	
Monocrotophos	6923-22-4
Parathion	56-38-2
Pentachlorophenol and its salts and esters	87-86-5 (*)
Toxaphene (Camphechlor)	8001-35-2
Tributyl tin compounds	1461-22-9, 1983-10-4, 2155-70-6, 24124- 25-2, 4342-36-3, 56-35-9, 85409-17-2
Dustable powder formulations containing a combination of benomyl at or above 7%, carbofuran at or above 10% and thiram at or above 15%	137-26-8, 1563-66-2, 17804-35-2
Methamidophos (Soluble liquid formulations of the substance that exceed 600 g active ingredient/l) Replaced by listing of Methamidophos in 2015 (entry into force September 2015)	10265-92-6
Methyl-parathion (Emulsifiable concentrates (EC) at or above 19.5% active ingredient and dusts at or above 1.5% active ingredient)	298-00-0

Phosphamidon (Soluble liquid formulations of the substance that exceed 1000 g active ingredient/l)	13171-21-6
Actinolite asbestos	77536-66-4
Anthophyllite	17068-78-9, 77536-67-5
Amosite asbestos	12172-73-5
Crocidolite	12001-28-4
Tremolite	77536-68-6
Commercial octabromodiphenyl ether (including Hexabromodiphenyl ether and Heptabromodiphenyl ether)	36483-60-0, 68928-80-3
Commercial pentabromodiphenyl ether (including tetrabromodiphenyl ether and pentabromodiphenyl ether)	32534-81-9, 40088-47-9
Perfluorooctane sulfonic acid, perfluorooctane	1691-99-2, 1763-23-1, 24448-09-7,
sulfonates, perfluorooctane sulfonamides and	251099-16-8, 2795-39-3, 29081-56-9,
perfluorooctane sulfonyls	29457-72-5, 307-35-7, 31506-32-8, 4151-
	50-2, 56773-42-3, 70225-14-8
Polybrominated Biphenyls (PBBs)	13654-09-6, 27858-07-7, 36355-01-8
Polychlorinated Biphenyls (PCBs)	1336-36-3
Polychlorinated Terphenyls (PCTs)	61788-33-8
Tetraethyl lead	78-00-2
Tetramethyl lead	75-74-1
Tris(2,3 dibromopropyl)phosphate	126-72-7

(back to section on Rotterdam Convention)

## **Appendix 2 – List of Basel Convention chemicals**

(back to Basel Convention section)

<b>Convention Annex</b>	Wastes listed
Annex I – Categories of	[Y1-Y18 are waste streams e.g. clinical wastes or from various
wastes to be controlled	manufacturing sectors – not listed as they do not refer to specific
	chemicals; apart from:
	Y10 Waste substances and articles containing or contaminated with
	polychlorinated biphenyls (PCBs) and/or polychlorinated terphenyls
	(PCTs) and/or polybrominated biphenyls (PBBs)]
	Y19 Metal carbonyls
	Y20 Beryllium; beryllium compounds
	Y21 Hexavalent chromium compounds
	Y22 Copper compounds
	Y23 Zinc compounds
	Y24 Arsenic; arsenic compounds
	Y25 Selenium; selenium compounds
	Y26 Cadmium; cadmium compounds
	Y27 Antimony; antimony compounds
	Y28 Tellurium; tellurium compounds
	Y29 Mercury; mercury compounds
	Y30 Thallium; thallium compounds
	Y31 Lead; lead compounds
	Y32 Inorganic fluorine compounds excluding calcium fluoride
	Y33 Inorganic cyanides
	Y34 Acidic solutions or acids in solid form
	Y35 Basic solutions or bases in solid form
	Y36 Asbestos (dust and fibres)
	Y37 Organic phosphorus compounds
	Y38 Organic cyanides
	Y39 Phenols; phenol compounds including chlorophenols
	Y40 Ethers
	Y41 Halogenated organic solvents
	Y42 Organic solvents excluding halogenated solvents
	Y43 Any congenor of polychlorinated dibenzo-furan
	Y44 Any congenor of polychlorinated dibenzo-p-dioxin
	Y45 Organohalogen compounds other than substances referred to in this
	Annex (e.g. Y39, Y41, Y42, Y43, Y44)

(back to Basel Convention section)

### Appendix 3 - Recommendations for inclusion in Annex XIV of REACH Regulation (Authorisation List)

Name	EC Number	CAS Number	Recommendation Year	Status
Aluminosilicate Refractory Ceramic Fibres (Al-RCF) are fibres covered by index number 650-017-00-8 in Annex VI, part 3, table 3.1 of Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures, and fulfil the three following conditions: a) oxides of aluminium and silicon are the main components present (in the fibres) within variable concentration ranges b) fibres have a length weighted geometric mean diameter less two standard geometric errors of 6 or less micrometres (µm) c) alkaline oxide and alkali earth oxide (Na2O+K2O+CaO+MgO+BaO) content less or equal to 18% by weight	-	-	5 (draft: 2013; final: February 2014)	Recommended for inclusion in Annex XIV
4-(1,1,3,3-tetramethylbutyl)phenol, ethoxylated (4-tert-OPnEO) [covering well-defined substances and UVCB substances, polymers and homologues]	-	-	5 (draft: 2013; final: February 2014)	Recommended for inclusion in Annex XIV
Diazene-1,2-dicarboxamide (C,C`-azodi(formamide)) (ADCA)	204-650- 8	123-77-3	5 (draft: 2013; final: February 2014)	Recommended for inclusion in Annex XIV
N,N-dimethylformamide (DMF)	200-679-	68-12-2	5 (draft: 2013; final: February 2014)	Recommended for inclusion in Annex XIV
Zirconia Aluminosilicate Refractory Ceramic Fibres (Zr-RCF) are fibres covered by index number 650-017-00-8 in Annex VI, part 3, table 3.1 of Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures, and fulfil the three following conditions: a) oxides of aluminium, silicon and zirconium are the main components present (in the fibres) within variable concentration ranges b) fibres have a length weighted geometric mean diameter less two standard geometric errors of 6 or less micrometres (µm). c) alkaline oxide and alkali earth oxide (Na2O+K2O+CaO+MgO+BaO) content less or equal to 18% by weight	-	-	5 (draft: 2013; final: February 2014)	Recommended for inclusion in Annex XIV

N,N-Dimethylacetamide (DMAC)	204-826-	127-19-5	4 (draft: 2012; final: January 2013)	Recommended for inclusion in Annex XIV
Cobalt dichloride	231-589- 4	7646-79- 9	3 (2011)	Recommended for inclusion in Annex XIV
Cobalt(II) dinitrate	233-402-	10141- 05-6	3 (2011)	Recommended for inclusion in Annex XIV
Cobalt(II) sulphate	233-334-2	10124- 43-3	3 (2011)	Recommended for inclusion in Annex XIV
Cobalt(II) diacetate	200-755- 8	71-48-7	3 (2011)	Recommended for inclusion in Annex XIV
Cobalt(II) carbonate	208-169- 4	513-79-1	3 (2011)	Recommended for inclusion in Annex XIV
Alkanes, C10-13, chloro (Short Chain Chlorinated Paraffins)	287-476- 5	85535- 84-8	1 (2009)	Recommended for inclusion in Annex XIV

## **Appendix 4 - ECHA List of Restrictions**

Entry no	Substance / group of substances / mixture	EC Number	CAS Number	Consolidated text	Appendix	New amendment (EU Regulation)	Q&A	Standards
1	Polychlorinated terphenyls (PCTs)	-	-	Page 217				
2	Chloroethylene (Vinyl chloride)	200-831-0	75-01-4	Page 217				
3	Liquid substances or mixtures, which are regarded as dangerous in accordance with Directive 1999/45/EC or are fulfilling the criteria for any of the following hazard classes or categories set out in Annex I to Regulation (EC) No 1272/2008: (a) hazard classes 2.1 to 2.4, 2.6 and 2.7, 2.8 types A and B, 2.9, 2.10, 2.12, 2.13 categories 1 and 2, 2.14 categories 1 and 2, 2.15 types A to F,(b) hazard classes 3.1 to 3.6, 3.7 adverse effects on sexual function and fertility or on development, 3.8 effects other than narcotic effects, 3.9 and 3.10,(c) hazard class 4.1,(d) hazard class 5.1.	-	-	Page 217				EN 14059:2002
4	Tris (2,3 dibromopropyl) phosphate	-	126-72-7	Page 218				
5	Benzene	200-753-7	71-43-2	Page 219				
6a	Asbestos fibres: Crocidolite	-	12001-28-4	Page 219	Appendix 7			
6b	Asbestos fibres: Amosite	-	12172-73-5	Page 219	Appendix 7			
6c	Asbestos fibres: Anthophyllite	-	77536-67-5	Page 219	Appendix 7			
6d	Asbestos fibres: Actinolite	-	77536-66-4	Page 219	Appendix 7			
6e	Asbestos fibres: Tremolite	-	77536-68-6	Page 219	Appendix 7			
6f(1)	Asbestos fibres: Chrysotile	-	12001-29-5	Page 219	Appendix 7			

6f(2)	Asbestos fibres: Chrysotile	-	132207-32-0	Page 219	Appendix 7		
7	Tris(aziridinyl)phosphinoxide	208-892-5	545-55-1	Page 220			
8	Polybromobiphenyls,Polybrominatedbiphenyls (PBB)	-	59536-65-1	Page 220			
9a	Soap bark powder (Quillaja saponaria) and its derivatives containing saponines	273-620-4	68990-67-0	Page 220			
9b	Powder of the roots of Helleborus viridis and Helleborus niger	-	-	Page 221			
9c	Powder of the roots of Veratrum album and Veratrum nigrum	-	-	Page 221			
9d	Benzidine and/or its derivatives	202-199-1	92-87-5	Page 221			
9e	(e) o-Nitrobenzaldehyde	209-025-3	552-89-6	Page 221			
9f	Wood powder	-	-	Page 221			
10a	Ammonium sulphide	235-223-4	12135-76-1	Page 221			
10b	Ammonium hydrogen sulphide	235-184-3	12124-99-1	Page 221			
10c	Ammonium polysulphide	232-989-1	9080-17-5	Page 221			
11a	Volatile esters of bromoacetic acids: Methyl bromoacetate	202-499-2	96-32-2	Page 221			
11b	Volatile esters of bromoacetic acids: Ethyl bromoacetate	203-290-9	105-36-2	Page 221			
11c	Volatile esters of bromoacetic acids: Propyl bromoacetate	-	35223-80-4	Page 221			
11d	Volatile esters of bromoacetic acids: (d) Butyl bromoacetate	242-729-9	18991-98-5	Page 221			
12	2-naphthylamine	202-080-4	91-59-8	Page 222			
13	Benzidine	202-199-1	92-87-5	Page 222			
14	4-Nitrobiphenyl	202-204-7	92-93-3	Page 220			

15	4-Aminobiphenyl xenylamine	202-177-1	92-67-1	Page 222			
16a	Lead carbonates: Neutral anhydrous carbonate (PbCO3)	209-943-4	598-63-0	Page 222			
16b	Lead carbonates: Trilead-bis(carbonate)-dihydroxide 2PbCO3-Pb(OH)2	215-290-6	1319-46-6	Page 222			
17a	Lead sulphates: (a) PbSO4	231-198-9	7446-14-2	Page 222			
17b	Lead sulphates: PbxSO4	239-831-0	15739-80-7	Page 222			
18	Mercury compounds	-	-	Page 223			
18a	Mercury	231-106-7	7439-97-6	Page 223	No 847/2012	entry 658 entry 659	
19	Arsenic compounds	-	-	Page 224		entry 660 entry 661	
20	Organostannic compounds	-	-	Page 226		entry 157	
21	Di-µ-oxo-di-n-butylstanniohydroxyborane/ Dibutyltin hydrogen borate C8H19BO3Sn (DBB)	401-040-5	75113-37-0	Page 228			
22	Pentachlorophenol	201-778-6	87-86-5	Page 228			
23	Cadmium	231-152-8	7440-43-9	Page 228		entry 662 entry 158 entry 824 entry 825	

24	Monomethyl — tetrachlorodiphenyl methane Trade name: Ugilec 141	-	76253-60-6	Page 232			
25	Monomethyl-dichloro-diphenyl methane Trade name: Ugilec 121,Ugilec 21	-	-	Page 233			
26	Monomethyl-dibromo-diphenyl methane bromobenzylbromotoluene, mixture of isomers Trade name: DBBT	-	99688-47-8	Page 233			
27	Nickel	231-111-4	7440-02-0	Page 233		entry 663	EN 1811:2011 EN 1811/AC:2012 EN 12472:2005+A1:200 EN 16128:2011
28	Substances which appear in Part 3 of Annex VI to Regulation (EC) No 1272/2008 classified as carcinogen category 1A or 1B (Table 3.1) or carcinogen category 1 or 2 (Table 3.2) and listed as follows: — Carcinogen category 1A (Table 3.1)/carcinogen category 1 (Table 3.2) listed in Appendix 1 — Carcinogen category 1B (Table 3.1)/carcinogen category 2 (Table 3.2) listed in Appendix 2	-	-	Page 234	Foreword Appendix 1 Appendix 2Appendix 11	entry 156	
29	Substances which appear in Part 3 of Annex VI to Regulation (EC) No 1272/2008 classified as germ cell mutagen category 1A or 1B (Table 3.1) or mutagen category 1 or 2 (Table 3.2) and listed as follows: — Mutagen category 1A (Table 3.1)/mutagen category 1 (Table 3.2) listed in Appendix 3 — Mutagen category 1B (Table 3.1)/mutagen category 2 (Table 3.2) listed in Appendix 4	-	-	Page 234	Foreword Appendix 3 Appendix 4Appendix 11	entry 156	
30	Substances which appear in Part 3 of Annex VI to Regulation (EC) No 1272/2008 classified as toxic to reproduction category 1A or 1B (Table 3.1) or toxic to reproduction category 1 or 2 (Table 3.2) and listed as	-	-	Page 234	Foreword Appendix 5 Appendix 6Appendix	entry 156	

	follows: — Reproductive toxicant category 1A adverse effects on sexual function and fertility or on development (Table 3.1) or reproductive toxicant category 1 with R60 (May impair fertility) or R61 (May cause harm to the unborn child) (Table 3.2) listed in Appendix 5 — Reproductive toxicant category 1B adverse effects on sexual function and fertility or on development (Table 3.1) or reproductive toxicant category 2 with R60 (May impair fertility) or R61 (May cause harm to the unborn child) (Table 3.2) listed in Appendix 6				11		
31a	Creosote,wash oil	232-287-5	8001-58-9	Page 235			
31b	Creosote oil, wash oil	263-047-8	61789-28-4	Page 235			1
31c	Distillates (coal tar), naphthalene oils,naphthalene oil	283-484-8	84650-04-4	Page 235			
31d	Creosote oil, acenaphthene fraction, wash oil	283-484-8 292-605-3	90640-84-9	Page 235			
31e	Distillates (coal tar), upper,heavy anthracene oil	266-026-1	65996-91-0	Page 235			
31f	Anthracene oil	292-602-7	90640-80-5	Page 235			
31g	Tar acids, coal, crude,crude phenols	266-019-3	65996-85-2	Page 235			
31h	Creosote, wood	232-419-1	8021-39-4	Page 235			
31i	Low temperature tar oil, alkaline, extract residues (coal), low temperature coal tar alkaline	310-191-5	122384-78- 5	Page 235			
32	Chloroform	200-663-8	67-66-3	Page 236			
34	1,1,2-Trichloroethane	201-166-9	79-00-5	Page 236			
35	1,1,2,2-Tetrachloroethane	201-197-8	79-34-5	Page 236			
36	1,1,1,2-Tetrachloroethane	-	630-20-6	Page 236			

37	Pentachloroethane	200-925-1	76-01-7	Page 236			
38	1,1-Dichloroethene	200-864-0	75-35-4	Page 236			
40	Substances classified as flammable gases category 1 or 2, flammable liquids categories 1, 2 or 3, flammable solids category 1 or 2, substances and mixtures which, in contact with water, emit flammable gases, category 1, 2 or 3, pyrophoric liquids category 1 or pyrophoric solids category 1, regardless of whether they appear in Part 3 of Annex VI to that Regulation or not.	-	-	Page 237		entry 665	
41	Hexachloroethane	200-666-4	67-72-1	Page 238			
43	Azocolourants and Azodyes	-	-	Page 238		entry 666	EN ISO 17234-1:2010 EN ISO 17234-2:2011 EN 14362-1:2012 EN 14362-3:2012
45	Diphenylether, octabromo derivative C12H2Br8O	-	-	Page 239			
46a	Nonylphenol C6H4(OH)C9H19	246-672-0	25154-52-3	Page 239		entry 667	
46b	Nonylphenol ethoxylates (C2H4O)nC15H24O	-	-	Page 239		entry 667	
47	Chromium VI compounds	-	-	Page 240			EN 196-10:2006
48	Toluene	203-625-9	108-88-3	Page 241		entry 668	
49	Trichlorobenzene	204-428-0	120-82-1	Page 241			
50a	Polycyclic-aromatic hydrocarbons (PAH): Benzo[a]pyrene (BaP)	-	50-32-8	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671	IP 346 ISO 21461:2012 EN 16143:2013
50b	Polycyclic-aromatic hydrocarbons (PAH): Benzo[e]pyrene (BeP)	-	192-97-2	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671	

50c	Polycyclic-aromatic hydrocarbons (PAH): Benzo[a]anthracene (BaA)	-	56-55-3	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
50d	Polycyclic-aromatic hydrocarbons (PAH): Chrysen (CHR)	-	218-01-9	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
50e	Polycyclic-aromatic hydrocarbons (PAH): Benzo[b]fluoranthene (BbFA)	-	205-99-2	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
50f	Polycyclic-aromatic hydrocarbons (PAH): Benzo[j]fluoranthene (BjFA)	-	205-82-3	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
50g	Polycyclic-aromatic hydrocarbons (PAH): Benzo[k]fluoranthene (BkFA)	-	207-08-9	Page 241	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
50h	Polycyclic-aromatic hydrocarbons (PAH): Dibenzo[a,h]anthracene (DBAhA)	-	53-70-3	Page 242	No 1272/2013 No 326/2015	entry 669 entry 670 entry 671
51a	The following phthalates (or other CAS and EC numbers covering the substance): Bis (2-ethylhexyl) phthalate (DEHP)	204-211-0	117-81-7	Page 242	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 984 entry 985
51b	The following phthalates (or other CAS and EC numbers covering the substance): Dibutyl phthalate (DBP)	201-557-4	84-74-2	Page 243	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 984

						entry 985
51c	The following phthalates (or other CAS and EC numbers covering the substance): Benzyl butyl phthalate (BBP)	201-622-7	85-68-7	Page 243	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 984 entry 985
52a	The following phthalates (or other CAS- and EC numbers covering the substance): Di-"isononyl" phthalate (DINP)	249-079-5 271-090-9	28553-12-0 68515-48-0	Page 243	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 748 entry 984
52b	The following phthalates (or other CAS- and EC numbers covering the substance): Di-"isodecyl" phthalate (DIDP)	247-977-1 271-091-4	26761-40-0 68515-49-1	Page 243	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 748 entry 984
52c	The following phthalates (or other CAS- and EC numbers covering the substance): Di-n-octyl phthalate (DNOP)	204-214-7	117-84-0	Page 243	No 326/2015	entry 672 entry 673 entry 674 entry 675 entry 676 entry 748 entry 984
54	2-(2-methoxyethoxy)ethanol (DEGME)	203-906-6	111-77-3	Page 243		
55	2-(2-butoxyethoxy)ethanol (DEGBE)	203-961-6	112-34-5	Page 244		

56	Methylenediphenyl diisocyanate (MDI) including the following specific isomers	247-714-0	26447-40-5	Page 244		entry 677	EN 14387:2004+A1:2008
56a	4,4'-methylenediphenyl diisocyanate	202-966-0	101-68-8	Page 244			
56b	2,4'-Methylenediphenyl diisocyanate	227-534-9	5873-54-1	Page 244			
56c	c) 2,2'-Methylenediphenyl diisocyanate	219-799-4	2536-05-2	Page 244			
57	Cyclohexane	203-806-2	110-82-7	Page 245			
58	Ammonium nitrate (AN)	229-347-8	6484-52-2	Page 245		entry 678 entry 679 entry 680	
59	Dichloromethane	200-838-9	75-09-2	Page 246			
60	Acrylamide	201-173-7	79-06-1	Page 249			
61	Dimethylfumarate (DMF)	210-849-0	624-49-7	Page 249			
62a	Phenylmercury acetate	200-532-5	62-38-4		No 848/2012		
62b	Phenylmercury propionate	203-094-3	103-27-5		No 848/2012		
62c	Phenylmercury 2-ethylhexanoate	236-326-7	13302-00-6		No 848/2012		
62d	Phenylmercury octanoate	-	13864-38-5		No 848/2012		
62e	Phenylmercury neodecanoate	247-783-7	26545-49-3		No 848/2012		
63	Lead and its compounds	231-100-4	7439-92-1	Page 249			
64	1,4-Dichlorobenzene (p-dichlorobenzene)	203-400-5	106-46-7		No 474/2014		

#### Appendix 5 - OSPAR List of Substances of Possible Concern

#### (back to OSPAR section)

#### Section A

Substances which warrant further work by OSPAR because they do not meet the criteria for Sections B-D and substances for which, for the time being, information is insufficient to group them in Sections B-D

Checked against ECHA (REACH) registrations database (on 20 February 2015) and found to <a href="https://example.com/have-been registered">have</a> been registered

CAS No.	Chemical name	Type
67124-09-8	2-propanol, 1-(tert-dodecylthio)-	Aliphatic hydrocarbon
25321-09-9	benzene, bis(1-methylethyl)	Aromatic hydrocarbon
84852-15-3	phenol, 4-nonyl-, branched	Phenol
50-28-2	Oestradiol (EC name: estradiol) (IUPAC Name estra-1,3,5(10)-triene-3,17-diol)	Hormones
53-16-7	Oestron (EC name: estrone) (IUPAC Name 3-hydroxyestra-1,3,5(10)-trien-17-one)	Hormones
25973-55-1	phenol, 2-(2H-benzotriazol-2-yl)-4,6- bis(1,1-dimethylpropyl)- EC name 2-(2H-benzotriazol-2-yl)-4,6-ditertpentylphenol	Phenol
3081-01-4	1,4-benzenediamine, N-(1,4- dimethylpentyl)-N'-phenyl- (EC and IUPAC name: N-(1,4-dimethylpentyl)-N'-phenylbenzene-1,4-diamine)	Organic nitrogen compound
1163-19-5	benzene, 1,1'-oxybis[2,3,4,5,6- pentabromo- (EC number: bis(pentabromophenyl) ether) (IUPAC Name: 1,1'-oxybis(pentabromobenzene)	Organohalogen
4378-61-4	dibenzo[def,mno]chrysene-6,12-dione, 4,10-dibromo-	Organohalogen
39489-75-3	phenol, 2,4-dichloro-5-nitro-, carbonate(2:1) (ester)	Organohalogen
85535-84-8	alkanes, C10-13, chloro (EC name: Alkanes, C10-13, chloro)	Organohalogen
78-00-2	lead, tetraethyl- (EC number: tetraethyllead) (IUPAC Name: tetraethylplumbane)	Organometallic compound
129-00-0	Pyrene	PAH
83-32-9	acenaphthylene, 1,2-dihydro- (EC number: acenaphthylene) (IUPAC Name: 1,2-dihydroacenaphthylene)	РАН
120-12-7	Anthracene	PAH
128-69-8	perylo[3,4-cd:9,10-c'd']dipyran-1,3,8,10- tetrone (EC Name: perylene-3,4:9,10-tetracarboxylic dianhydride) (IUPAC Name: isochromeno[4',5',6':6,5,10]anthra[2,1,9-def]isochromene-1,3,8,10-tetrone)	РАН
9016-45-9	Nonylphenolethoxylate EC name Poly(oxy-1,2-ethanediyl),.alpha(nonylphenol)-omegahydroxy-	Phenol
4051-63-2	[1,1'-bianthracene]-9,9',10,10'-tetrone, 4,4'-diamino-	PAH
5510-99-6	phenol, 2,6-bis(1-methylpropyl)- (EC name: di-sec-butylphenol, mixed isomers)	PAH
26140-60-3	Terphenyl	PAH
38640-62-9	naphthalene, bis(1-methylethyl)- (EC & IUPAC name: bis(isopropyl)naphthalene)	PAH
3147-75-9	phenol, 2-(2H-benzotriazol-2-yl)-4-(1,1,3,3-tetramethylbutyl)-	Phenol
registered	nst ECHA (REACH) registrations database (20 February 2015) and	
65294-17-9	methylium, tris[4-(dimethylamino)phenyl]-, salt with 3-[[4-	Organic nitrogen compound
68015-60-1	benzenesulfonic acid, 2-amino-, (1- methylethylidene)di-4,1-phenylene ester	Organic nitrogen compound
68083-48-7	2-butanone, O-[[[[1,3,3-trimethyl-5-[[[[(1-methylpropylidene)amino]oxy]carbonyl]a	Organic nitrogen compound
68844-77-9	Astemizole	Organic nitrogen compound
128-83-6	9,10-anthracenedione, 1-amino-2-bromo-4-[(4-methylphenyl)amino]-	Organic nitrogen compound
79-95-8	phenol, 4,4'-(1-methylethylidene)bis[2,6- dichloro-	Organohalogen
87-82-1	benzene, hexabromo-	Organohalogen
	benzene, pentabromomethyl-	Organohalogen

CAS No.	Chemical name	Туре
95-95-4	phenol, 2,4,5-trichloro-	Organohalogen
634-66-2	benzene, 1,2,3,4-tetrachloro-	Organohalogen
1336-36-3	1,1'-biphenyl, chlorinated	Organohalogen
2051-24-3	1,1'-biphenyl, 2,2',3,3',4,4',5,5',6,6'- decachloro-	Organohalogen
2437-79-8	1,1'-biphenyl, 2,2',4,4'-tetrachloro-	Organohalogen
7012-37-5	1,1'-biphenyl, 2,4,4'-trichloro-	Organohalogen
33979-03-2	1,1'-biphenyl, 2,2',4,4',6,6'-hexachloro-	Organohalogen
68-90-6	Benziodarone	Organohalogen
101-76-8	benzene, 1,1'-methylenebis[4-chloro-	Organohalogen
128-63-2	pyrene, 1,3,6,8-tetrabromo-	Organohalogen
335-57-9	heptane, hexadecafluoro-	Organohalogen
355-42-0	hexane, tetradecafluoro-	Organohalogen
423-50-7	1-hexanesulfonyl fluoride, 1,1,2,2,3,3,4,4,5,5,6,6,6-tridecafluoro-	Organohalogen
559-11-5	2-propenoic acid, 2,2,3,3,4,4,5,5,6,6,7,	Organohalogen
3278-89-5	benzene, 1,3,5-tribromo-2-(2- propenyloxy)-	Organohalogen
13654-09-6	1,1'-biphenyl, 2,2',3,3',4,4',5,5',6,6'- decabromo-	Organohalogen
26447-49-4	Hexabromododecane	Organohalogen
32534-81-9	benzene, 1,1'-oxybis-, pentabromo deriv.	Organohalogen
32536-52-0	benzene, 1,1'-oxybis-, octabromo deriv.	Organohalogen
36861-47-9	bicyclo(2.2.1)heptan-2-one, 1,7,7- trimethyl-3-[(4-methylphenyl)methylene]-	Organohalogen
38521-51-6	benzene, pentabromo(bromomethyl)-	Organohalogen
41604-19-7	1,1'-biphenyl, 4-bromo-2-fluoro-	Organohalogen
41999-84-2	benzene, 1,4-dichloro-2,5- bis(dichloromethyl)-	Organohalogen
53742-07-7	1,1'-biphenyl, nonachloro-	Organohalogen
52434-90-9	1,3,5-triazine-2,4,6(1H,3H,5H)-trione, 1,3,5-tris(2,3-dibromopropyl)-	Organohalogen
65925-28-2	benzene, 1-[2-(2-chloroethoxy)ethoxy]-4-(1,1,3,3-tetramethylbutyl)-	Organohalogen
75-74-1	lead, tetramethyl-	Organometallic compound
427-45-2	stannane, fluorotris-p-chlorophenyl-	Organometallic compound
50-32-8	benzo[a]pyrene	PAH
53-70-3	dibenz[a,h]anthracene	PAH
56-49-5	benz[j]aceanthrylene, 1,2-dihydro-3- methyl-	PAH
56-55-3	benz[a]anthracene	PAH
57-97-6	benz[a]anthracene, 7,12-dimethyl-	PAH
82-05-3	7H-benz[de]anthracen-7-one	PAH
191-24-2	benzo[ghi]perylene	PAH
192-97-2	benzo[e]pyrene	PAH
198-55-0	Perylene	PAH
206-44-0	Fluoranthene	PAH
207-08-9	benzo[k]fluoranthene	PAH
218-01-9	Chrysene	PAH
92-24-0	Naphthacene	PAH
116-66-5	1H-indene, 2,3-dihydro-1,1,3,3,5- pentamethyl-4,6-dinitro-	PAH
132-65-0	Dibenzothiophene	PAH
189-55-9	benzo[rst]pentaphene	PAH
189-64-0	dibenzo[b,def]chrysene	PAH
191-07-1	Hexabenzobenzene	PAH
191-07-1	dibenzo[def,mno]chrysene	PAH
191-26-4	dibenzo[def,p]chrysene dibenzo[def,p]chrysene	PAH
191-30-0		
192-65-4	naphtho[1,2,3,4-def]chrysene benzo[c]phenanthrene	PAH PAH

CAS No.	Chemical name	Туре
215-58-7	benzo[b]triphenylene	PAH
217-59-4	Triphenylene	PAH
224-41-9	dibenz[a,j]anthracene	PAH
1705-85-7	chrysene, 6-methyl-	PAH
335-128-8	chrysene, 1-methyl-	PAH
5522-43-0	pyrene, 1-nitro-	PAH
53-19-0	benzene, 1-chloro-2-[2,2-dichloro-1-(4- chlorophenyl)ethyl]-	Pharmaceutical
50-53-3	Chlorpromazine	Pharmaceutical
50-63-5	1,4-pentanediamine, N(4)-(7-chloro-4- quinolinyl)-N(1),N(1)-diethyl-, phosphate	Pharmaceutical
54-05-7	1,4-pentanediamine, N4-(7-chloro-4- quinolinyl)-N1,N1-diethyl-	Pharmaceutical
58-38-8	10H-Phenothiazine, 2-chloro-10-[3-(4- methyl-1-piperazinyl)propyl]-	Pharmaceutical
69-23-8	1-Piperazineethanol, 4-[3-[2-(trifluoromethyl)-10H-phenothiazin-10-	Pharmaceutical
146-56-5	1-Piperazineethanol, 4-[3-[2-(trifluoromethyl)-10H-phenothiazin-10-	Pharmaceutical
440-17-5	10H-phenothiazine, 10-[3-(4-methyl-1- piperazinyl)propyl]-2-(trifluoromethyl)-,	Pharmaceutical
749-13-3	4'-fluoro-4-[4-hydroxy-4-(3- trifluoromethylphenyl)piperidino]butyroph	Pharmaceutical
1257-78-9	1,2-ethanedisulfonic acid, compd. with 2- chloro-10-[3-(4-methyl-1-	Pharmaceutical
2062-78-4	1-[1-(4,4-Bis[4-fluorophenyl]-butyl)-4- piperidinyl]-1,3-dihydro-2H-benzimidazol-	Pharmaceutical
3759-07-7	10(9H)-acridinepropanamine, N,N,9,9- tetramethyl-, [R-(R*,R*)]-2,3-	Pharmaceutical
4394-00-7	niflumic acid	Pharmaceutical
4757-55-5	10(9h)-acridinepropanamine, n,n,9,9- tetramethyl-	Pharmaceutical
10331-57-4	[1,1'-biphenyl]-2,2'-diol, 5,5'-dichloro	Pharmaceutical
22832-87-7	1H-imidazole, 1-[2-(2,4-dichlorophenyl)-2-[(2,4-dichlorophenyl)methoxy]ethyl]-,	Pharmaceutical
57648-21-2	1-butanone, 4-[4-(2,3-dihydro-2-thioxo-1	Pharmaceutical
59467-70-8	4H-imidazo[1,5-a][1,4]benzodiazepine, 8- chloro-6-(2-fluorophenyl)-1-methyl-	Pharmaceutical
67969-69-1	1-octanesulfonamide, N-ethyl-1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-	Pharmaceutical
26864-56-2	4-piperidinol, 1-[4,4-bis(4-fluorophenyl	Pharmaceutical
29098-15-5	benzoic acid, 2-[(2,6-dichloro-3-methylp	Pharmaceutical
52468-60-7	piperazine, 1-[bis(4-fluorophenyl)methyl]- 4-cinnamyl-	Pharmaceutical
25154-52-3	phenol, nonyl-	Phenol
1138-52-9	phenol, 3,5-bis(1,1-dimethylethyl)-	Phenol
2668-47-5	[1,1'-biphenyl]-4-ol, 3,5-bis(1,1- dimethylethyl)-	Phenol
17540-75-9	phenol, 2,6-bis(1,1-dimethylethyl)-4-(1- methylpropyl)-	Phenol
104-40-5	phenol, 4-nonyl-	Phenol
120-95-6	phenol, 2,4-bis(1,1-dimethylpropyl)-	Phenol
3846-71-7	phenol, 2-(2H-benzotriazol-2-yl)-4,6- bis(1,1-dimethylethyl)-	Phenol
21150-89-0	phenol, 4-(1,1-dimethylethyl)-, hydrogen phosphate	Phenol
50849-47-3	benzaldehyde, 2-hydroxy-5-nonyl-, oxime	Phenol
57-63-6	17-ethynylestradiol	Hormones
72-33-3	17alpha-ethynyl-1,3,5[10]-estratriene-3,17beta-diol 3-methyl ether	Hormones
630-56-8	pregn-4-ene-3,20-dione, 17-[(1- oxohexyl)oxy]-	Hormones
52179-28-9	propanoic acid, 2-[4-(2,2- dichlorocyclopropyl)phenoxy]-2-methyl-,	Organic ester
26999-29-1	phosphorodithioic acid, O,O-diisooctyl ester	Organic ester
527-20-8	benzenamine, 2,3,4,5,6-pentachloro-	Organic nitrogen compound
93-46-9	1,4-benzenediamine, N,N'-di-2- naphthalenyl-	Organic nitrogen compound
139-60-6	1,4-benzenediamine, N,N'-bis(1-ethyl-3- methylpentyl)-	Organic nitrogen compound
494-03-1	2-naphthalenamine, N,N-bis(2- chloroethyl)-	Organic nitrogen compound
1606-67-3	1-pyrenamine	Organic nitrogen compound
1691-99-2	1-octanesulfonamide, N-ethyl-1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-	Organic nitrogen compound
5285-60-9	benzenamine, 4,4'-methylenebis[N-(1- methylpropyl)-	Organic nitrogen compound
13680-35-8	benzenamine, 4,4'-methylenebis[2,6- diethyl-	Organic nitrogen compound

CAS No.	Chemical name	Type
15114-15-5	9,10-anthracenedione, 4,8-diamino-2-(4- ethoxyphenyl)-1,5-dihydroxy-	Organic nitrogen compound
19774-82-4	methanone, (2-butyl-3-benzofuranyl)[4-[2-(diethylamino)ethoxy]-3,5-diiodophenyl]-,	Organic nitrogen compound
29312-59-2	benzenamine, 4-(2,6-diphenyl-4- pyridinyl)-N,N-dimethyl-	Organic nitrogen compound
52740-90-6	2-anthracenecarboxamide, 1-amino-N-(3- bromo-9,10-dihydro-9,10-dioxo-2-	Organic nitrogen compound
64381-97-1	1,4-benzenediamine, N,N,N'-tris(1- methylpropyl)-	Organic nitrogen compound

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