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Scenario analysis of pollutants loads to European regional seas for the year 2020. Part II: Assessment of priority chemicals – an example with three pilot substances

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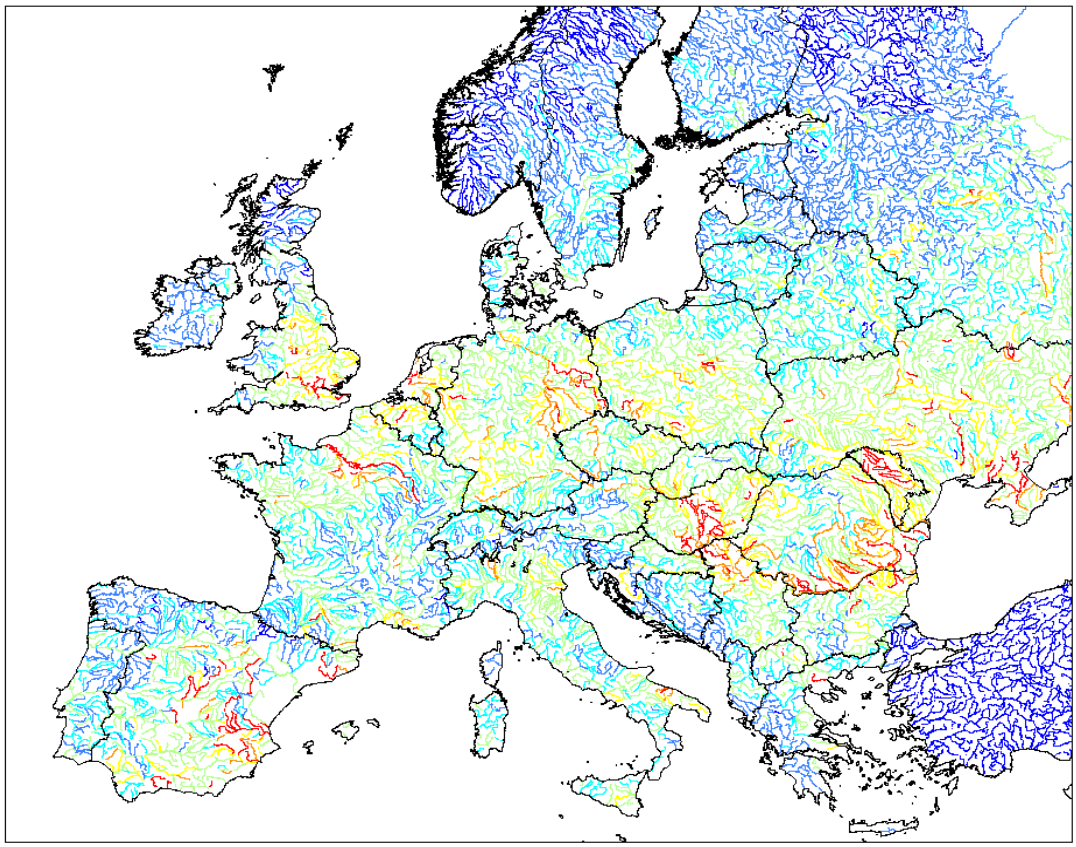

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Executive Summary

In order to support the implementation of the Marine Strategy Framework Directive, DG Environment and the Joint Research Centre joined to carry out a study on the expected cumulative impact of existing EU environmental legislation on the quality of the marine environment, with specific reference to the case of aquatic discharges to European seas. The assessment, considering regional seas as final receiving water bodies, focused mainly on trends and options for reduction of inland-based emissions of nutrients and chemicals. Therefore, the results of this study are useful not only for the implementation of the Marine Strategy Framework Directive, but also to other policies developed by the EU to control emissions to water bodies from a variety of sources (e.g. the Water Framework Directive, the Nitrates Directive, the Urban Waste Water Treatment Directive).

The results of a few scenario analyses affecting emissions to the European regional seas up to the year 2020 are presented for convenience in two separate EU reports. The first one deals with the assessment of loads of Nitrogen and Phosphorus, while present one focuses on three chemicals taken as pilot substances - Lindane, Trifluralin and Perfluorooctane sulfonate (PFOS). The scenarios were agreed with stakeholders at DG ENV following some preparatory meetings. They do not intend be exhaustive, but examples of what can be further achieved making use of the modelling and database developed during the different phases of the project.

Part I: Policy options and alternative measures to mitigate land based emission of nutrients (Nitrogen and Phosphorus)

The study on nutrient loads was divided in three phases. The first one was focusing on data collection and model development. The second phase dealt with the retrospective assessment for the years 1985-2005, including the collection of all relevant data and a trend analysis for nutrient loads to pave the way for the scenario development and evaluation that was the focus of the third present part of the study. First, a Business as Usual (BAU) scenario was defined and then three types of mitigation scenarios were developed and tested by addressing

i) the collection and treatment of point sources (UWWD and PFREE scenarios),

ii) a change in European human diet (WHO and WCRF scenarios)

iii) the management of manure application in Europe (MANU scenario).

Scenarios were run with the GREEN model, which uses input from anthropogenic activities (agriculture, industries, wastewater) to calculate the load of nitrogen (N) and phosphorus (P) for the whole Europe on a sub-catchment basis. The dual objective was to assess changes of land based nutrient loads in European regional seas following the recommendation of the Marine Strategy Framework Directive by providing at the same time an assessment of impacts of actions affecting inland based nutrient emission.

All mitigation measures were assessed using as reference both the year 2005 and the Business as Usual scenario (BAU) including change in population count and distribution and considering the status-quo in wastewater treatment for 2020. It also includes prospects for food production and prediction for crop and nutrient requirements.

Results obtained on the mitigation of nutrient emitted as point sources combined several parameters as the changes in population density, the increase of connection rate to the sewage network, the upgrading of wastewater treatment plants. They clearly emphasized the gap between EU-15 and new Members States.

For EU 15, implementation of the Directive 91/271/EEC (UWWD scenario) mostly results in the upgrading of existing treatment plants with basic treatment to more stringent treatment of nitrogen and phosphorus, leading to a significant decrease of point sources emission for nitrogen (from -7% for the Netherland up to -50% in Ireland). Results for phosphorus are also very significant (with a decrease up to 63% in Belgium) when combined with a ban of phosphates and other phosphorus compounds in household laundry detergents (PFREE scenario) as suggested by the Commission proposal COM (2010) 597 amending Regulation (EC) No 648/2004.

For the new Member States, the full implementation of the UWW-Directive leads to an important transfer of nutrient sources from non-collected emission (scattered dwelling) to point-sources emission (connected to sewers). It tends to limit the impact of the UWWD scenario and leads to a significant increase of point source as for example for Romania (+54%N; +34%P) or for Slovenia (+73%N; +51%P) accompanied with complete reduction (>90%) of scatter dwelling emissions. It is important to note in this context that scattered dwellings are a major source of groundwater contamination.

Options to mitigate nutrient emitted as diffuse sources include the change in European human diet (WHO and WCRF scenarios) and the management of manure application in Europe (MANU scenario). The WHO and WCRF scenarios, considering progressive decrease of beef and pork meats consumption and an increase of vegetal proteins in human diet, have a low impact on nitrogen and phosphorus diffuse sources. This is partially explained by the storylines of these two scenarios, which, according to the agri-economic prevision of the CAPRI (Common

Agricultural Policy Regional Impact) model, consider a significant decrease in meat consumption in Europe, but at the same time an important increase of meat export outside Europe necessary for farms to be economically sustainable. In the more stringent WCRF scenario, the sum of anthropogenic diffuse emissions is decreased by 4 % for nitrogen and 3% for phosphorus at the scale of EU-27. This clearly highlights the necessity to consider simultaneously human meat intake and meat production in order to achieve significant decrease of nutrient emissions from animal breeding.

While previous environmental assessments put emphasis on a change in food *consumption* as an efficient way to reduce nitrogen input to the environment in Europe, this report suggests that a more realistic scenario analysis should consider both agricultural *production* and *trade*. Indeed, it is shown that the *production* of meat in Europe will be essentially preserved even in the presence of a drastic decrease in European *consumption* of meat due to a large increase of meat export towards other countries.

The third type of mitigation scenario tested in this study concerned an optimized distribution of animal manure. The MANU scenario leads to a further decrease of nitrogen diffuse sources, with even a shift in the nitrogen source apportionment for several basins in Europe. This scenario also emphasizes a significant decrease in the application of mineral nitrogen, with evident benefits also due to the continuously increasing price of nitrogen fertilisers.

The assessment of nutrient loads to European seas made use of the GREEN model, which was previously shown to be appropriate for the estimation of nutrient fluxes based on a simplified representation of the processes involved in transport and retention. The following table summarizes the results of the scenarios implementation for each European sea:

Nitrogen loads *(1000 tons of N/yr, min and max simulated)*	Baltic	North Sea	Atlantic	Black Sea	Mediterranean
REF	475 - 687	872 - 1420	707 - 1190	434 - 635	697 - 982
BAU	488 - 714	902 - 1469	733 - 1244	458 - 671	731 - 1011
UWWD	483 - 709	836 - 1403	691 - 1202	446 - 659	693 - 972
WHO	486 - 712	893 - 1454	729 - 1236	457 - 670	730 - 1008
WCRF	481 - 701	873 - 1419	717 - 1208	455 - 665	721 - 993
MANU	394 - 540	692 - 1059	591 - 932	400 - 554	616 - 805

Phosphorus loads *(1000 tons of P/yr, min and max simulated)*	Baltic	North Sea	Atlantic	Black Sea	Mediterranean
REF	27 - 35	46 - 64	37 - 48	31 - 38	61 - 83
BAU	27 - 35	47 - 66	38 - 50	33 - 40	64 - 86
UWWD	26 - 34	35 - 53	31 - 43	29 - 36	54 - 75
PFREE	22 - 30	28 - 47	27 - 39	25 - 33	49 - 70
WHO	27 - 35	47 - 65	38 - 49	33 - 40	64 - 85
WCRF	27 - 35	47 - 65	38 - 49	33 - 40	64 - 85
MANU	27 - 37	48 - 69	38 - 51	34 - 49	63 - 81

(Nitrogen and phosphorus loads have been calculated for a range of 21 years hydrological conditions. Results are provided with a Min-Max loads simulated over the 21 years simulation period)

In the horizon 2020, the BAU scenario simulated an increase of both nitrogen and phosphorus loads exported to European seas. This increase is more significant for nitrogen especially when combined with high hydrological condition (high flow).

When comparing the mitigation of nutrient inputs with the concomitant change in nutrient loads, it is clearly demonstrated that point source mitigation measures are the most effective option. Indeed, a change in point sources emissions could be directly linked to a change in nutrients exported to European seas once aquatic retention is taken into account, while mitigation of diffuse nutrient inputs are submitted to both the terrestrial and aquatic attenuation. Beyond this first result, scenarios efficiency has to be considered with respect to the source apportionment of each nutrients (N and P), in order to estimate it capacity for mitigating nutrient emissions. Scenarios results differ for Nitrogen and Phosphorus.

For phosphorus, the rate of removal in WWTPs has considerably improved during the last decades with about 90 % of P removal now achieved for the most advanced WWTPs. Considering that human emissions of phosphorus are now stabilized to a value close to physiological releases, additional mitigation could only come from a complete ban of phosphorus in laundry detergents (PFREE scenario), which will result in the most significant reduction of phosphorus export to European seas.

In the case of nitrogen, the source apportionment indicates that nitrogen flowing to European sea is widely dominated by agricultural (diffuse) sources. Consequently, while scenarios targeting nitrogen point-sources are more efficient they enable only a low reduction of the overall amount nitrogen loads. The most important decrease of nitrogen output is related to the optimization of animal manure application.

The mitigation options to be selected might lead to very different (even opposite) effects on nutrient loads to European seas and the study showed the importance of a simultaneous assessment of both nitrogen and phosphorus emissions and exports.

Part II: Assessment of priority chemicals – an example with three pilot substances

The second part of the analysis focused on a European-scale assessment of chemical loads to regional seas. For chemicals, a major difficulty comes from the limited availability of data on the location of sources and the extent of emissions. In this study, we have used statistical information from EUROSTAT plus data available from the European Monitoring and Evaluation Programme (EMEP) and a number of literature studies. However, the sources of information are in general limited, which represents the main obstacle for the evaluation of pollutant loads originating from the European continent.

Due to the extraordinarily high number of chemical products on the market, which potentially could be discharged to the sea after being used or released to the environment during manufacturing, the assessment was not intended to be exhaustive, but rather to develop a methodology for the identification of hot-spots in Europe and its capability for the estimation of chemical loads to European coastal waters under different scenarios.

In order to perform prospective scenarios for Europe, the concentrations and loads of three test-case chemicals were evaluated firstly for the following baseline years: 1995 and 2005 for the insecticide Lindane; 2003 for the herbicide Trifluralin; and 2007 for the industrial pollutant Perfluorooctane Sulfonate (PFOS). Then, a set of different scenarios was defined and the assessments made using either a "direct" or an "inverse" modelling approach.

The "direct" method is based on a priori available information about chemical emissions and answers the question "Where do chemicals go after being emitted?". In this part of the study, the "direct" version of the spatially resolved Multimedia Assessment of Pollutant Pathways in the Environment (MAPPE) screening model was applied in the scenario case-studies of Lindane and Trifluralin, additionally supplemented by a simple non-spatial box model application.

The "inverse" approach tries to answer the question "Where do pollutants come from?". Inverse models can support large scale assessments of source apportionment by estimating emission factors at regional, river basin or continental scale in relation to the population density or other

proxies. This approach was employed in the backward tracking of PFOS emissions from pan-European riverine measurements. Then, on the basis of the estimated average emission factor for Europe, the spatial GIS analyses made possible to evaluate the scenarios for the annual load of PFOS to European seas.

The scenarios for the three pilot chemicals considered are formulated to the time horizon of year 2020 assuming different types of legislative measures (for example business as usual, ban, phase out, etc.) or aiming at specific targets (as per total and disaggregated load to European seas or possible "cleaning-up" of soil in Europe). Details about the scenarios are provided separately for each test case.

The summary of the scenario results for chemical substances is given below. When considering the output of the scenario analyses, it is important to take into account that the project aimed at testing the applicability of the modelling platform. Moreover, since limited data on emissions are available certain assumptions had to be made, thus restricting the use of the present scenario results only for screening purposes.

1. Lindane (γ-HCH)

Lindane is a relatively well known insecticide officially banned in the European Union since 1995. Some Lindane emissions, however, are still occurring due to releases from stockpiles or other sources, which are difficult to quantify. Lindane was selected as a pilot substance in this study because of the availability of data about past emissions and measured environmental concentrations.

Air emissions of post-ban use of γ–HCH are quite difficult to be estimated although these sources may significantly affect the current environmental concentrations (Breivik et al., 2004). A European inventory of Prevedouros et al., (2004) suggests that ca. 135000 tons were applied over the period 1970–1996 with the major contributions originating from France, Spain and the Netherlands. Besides, other authors estimated that approximately 650 tons were emitted in 1998 and the contribution of each European country was calculated (Breivik et al., 2004). However, at present the only comprehensive and reliable source of emission data at European scale is the European Monitoring and Evaluation Programme (EMEP), providing official data on the atmospheric emissions by country (CEIP, 2009).

The current study relies on Lindane air emissions estimated by EMEP (www.emep.int). Accordingly, the air emissions of γ-HCH in the Northern hemisphere were estimated to be 432

tons in 2005 including 71 t in North America, 68 t in Central America, 200 t in Southeast Asia and 92 t in Europe. In addition, the European continent received an extra deposition of ca. 2% of emissions coming from sources originated in North America and China (Gusev et al., 2006).

Then, based on the officially reported information for the amount and spatial pattern of the air emissions in Europe and trans-continental long range transport, the following atmospheric scenarios for Lindane were analyzed:

- **BAU** – (Business as Usual) no change of air emissions; keeping 2005 level of emissions up to the time horizon of 2020 (about 92 tons are supposed to be emitted in 2005);
- **trend** – a scenario continuing the emissions decline observed in the period 2000-2005, as a result of the regulation process started in 1995; accordingly, the emissions in 2020 equal to 45.6 t/y (49.6% of 2005 level);
- **linear** – a generic scenario which respects the regulations and assumes a gradual linear reduction of emissions starting in 2005 and ending in 2011 with 23 t/y (25% of the 2005 level of emissions); the choices of 2011 as an end year of measures and the percentage of emission reduction were provisional; no change of emissions after 2011;
- **ban** – a scenario consistent with the regulation acts considering a fast exponential reduction of the European emissions in the period 2005-2011; emissions in 2011 are supposed to be equal to 5.4 t/y (the quantity arriving in Europe by the Long Range Atmospheric Transport according to 2005 data); as for the linear scenario, the selection of 2011 as an end year of measures is provisional; from 2011 to 2020 no European emissions, but the scenario accounts 5.4 t/y intercontinental atmospheric transport from North America and Southeast Asia using 2005 data as a background level (the last available data from EMEP (Gusev et al., 2006).

After the description of possible scenarios for air emissions, the next step was to specify the corresponding emissions to the other environmental media. According to UNEP (Breivik and Wania, 2002), about 59% of the total amount of Lindane is used for soil treatment, while seed treatment, which presumably yields lower emissions to air, accounts for 34%. Furthermore, Lindane is generally applied in liquid formulations (mostly wettable powders) and only a minor fraction is used in the solid state (dusts, powders, and granules). On this basis, a mode of division of total Lindane emissions by 17.5% to the atmosphere, 80% to the agricultural soil, and 2.5% to freshwater is assumed by Breivik and Wania, (2002) and Vizcaino and Pistocchi, (2010). Practically, the equivalent approach was used in the EMEP modeling applications for Lindane

(www.emep.int). Therefore, the same fractioning of Lindane emissions was adopted up to the year 1999 and only atmospheric emissions from 2000 onwards since the ban of Lindane in EU for agriculture use.

The results of the present scenario study on Lindane for EU27 plus Norway, Switzerland, Croatia, Serbia, the western Balkan countries and Turkey allowed us to conclude following:

- The comparison with the OSPAR data (OSPAR Commission, 2011) or with data used to force a 3D model of North Sea (Ilyina et al., 2008) showed that MAPPE model produced consistent results for the riverine load of gamma-HCH to European seas which eventually could differ from the other estimates by not more than a factor of two;
- The model assessed European sea loads of 745 tons for 1995 (based on the official emission data provided by EMEP) appears to be reduced by 98.3% in 2005, ten years after the start of the EU regulations for γ-HCH;
- In 2020, under the BAU scenario, a Lindane sea load of ca.12.5 tons per year would be expected;
- The trend and ban scenarios support a reduction of the load to the European seas in 2020 by 74% and 95%, respectively, when compared to the BAU estimate;
- The discharge of Lindane under BAU scenario is affecting mainly the European coast of the Atlantic Ocean (49 % of the total for Europe), Mediterranean (27 %) and Black seas (19 %), while in the case of the ban scenario the Black sea (43 %) is the main recipient, followed by Mediterranean (19 %) and Baltic (17%) seas and Atlantic Ocean (16%).

2. Trifluralin

Trifluralin (a priority substance under the Water Framework Directive) is an herbicide banned in EU countries since 2008. Presently, very little information on Trifluralin emissions is available at pan-European scale, and the only comprehensive dataset for the 25 EU Member States is provided by EUROSTAT with reference to the year 2003. Besides, this study assumed that the contribution of the long-range atmospheric transport and of UWWTP effluents is a negligible source of Trifluralin pollution. Thus, the scenario study of Trifluralin sea load focuses only on the emissions to European soil.

In order to assess the soil emissions of Trifluralin, the method of Pistocchi et al. (2009) was applied using EUROSTAT data (although this approach tends to overestimate the use of plant

protection products). This method assumes that when a certain class of pesticides is applied, then likely only a single substance from that class is used everywhere across Europe as a representative for this pesticide class. Accordingly, the **BAU** scenario considers as input for Trifluralin the data on the entire class of dinitroaniline herbicides (8 substances including Trifluralin) for which EUROSTAT reported 6174 tons applied to arable crop land in Europe during 2003 (average use of 1.56 kg/km^2/y with a regional variability ranging from 0.01 to more than 20).

Furthermore, the study investigated the potential impact of the complete ban of Trifluralin applications. In the **ban** scenario the emissions were assumed to drop, in the period 2004-2010, from the typical BAU application for 2003 to an amount of 0.005 kg/km^2/y, taken as an approximation towards zero emissions from 2011 onwards for each of the EU25 countries.

In addition, despite of the uncertainty of the data about Trifluralin applied to the soil, a specific **partial effectiveness** scenario was analysed aiming to assess how much the soil emissions should be reduced in order to ensure that the annual sea load remains lower than a given limit taken here as one third of the BAU estimate.

The modelled scenarios for Trifluralin indicated that:

- According to the BAU scenario based on EUROSTAT usage data, in 2020 the sea load of Trifluralin, considered as representative for the entire group of dinitroaniline herbicides, is estimated to be ca. 61.7 tons, the same as for the reference year 2003;
- The complete ban scenario forecasts ca. 0.07 t/y sea load and in practice eliminates the concern about the discharge of Trifluralin to the European seas in a time-frame of one year due to degradation in soil;
- Under the available data used in BAU scenario, the European coastal areas of the Atlantic ocean and North sea receive the higher fractions of the European sea load, 29.5% and 22%., respectively, followed by the Baltic (19%) and Mediterranean (17.5%) seas. However, it is worth stressing again that these estimates are built on incomplete emission inventory considering only EU25 countries.
- In the partial effectiveness scenario the total sea load of Trifluralin is expected not to exceed one third of the BAU load when the application of dinitroaniline herbicides to soil is reduced at least by 66% for the EU25 countries across Europe.

3. Perfluorooctane sulfonate - PFOS

Perfluorinated compounds, including Perfluorooctane Sulfonate (PFOS), are chemicals produced for their non-stick and water repellent properties. They have been used during the last 50 years both in industry and as components of consumer products in the manufacture of coatings for cookware and clothing, stain resistant carpets, food packaging, fire-fighting foams, paints, and adhesives, with additional uses in the photo-, electronics-, and aerospace industries.

Unlike Lindane and Trifluralin, which are multimedia chemicals, PFOS can be regarded as a single-medium molecule primarily related to the water compartment. Actually, PFOS's high solubility and its persistence make it a virtually conservative and instantly a water-transported substance. Since PFOS is environmentally persistent, bioaccumulative and potentially harmful, it was listed as chemical for regulation within the Stockholm Convention and was banned in the European Union in 2007.

The study employed the approach of a backward tracking of PFOS emissions from riverine measurements as described by Loos et al. (2009) and Pistocchi and Loos (2009) who consider the atmospheric deposition of PFOS as a negligible diffuse source that could be disregarded. Accordingly, it was found that PFOS emissions correlate rather well with river basin population. Thus, for PFOS an average European emission factor of 27.4 µg/day per capita was estimated. The latter is fairly consistent with previously found estimates of 40 µg/day/inhabitant in Bayreuth (Germany) and 57 µg/day/person for Switzerland. Then, the average emission factor for Europe was used as a basis of a GIS model able to calculate European maps of PFOS river water concentrations and load to seas.

In the study, the **BAU** scenario for PFOS is referring to 2007 as baseline year. Additionally, the sea load of PFOS was assessed by considering a **scenario of 50% reduction** of emissions and also answering the question by what percentage the emissions should be decreased in order to guarantee that the total sea load stays below a given threshold, "a **scenario targeting a sea load of 1 ton per year"**.

The scenario results allow concluding that:

- Based on the average emission factor of 27.4 µg/inhabitant/day and the map of population density in Europe, when BAU scenario was supposed the concentrations of PFOS in surface water vary from less than 0.001 to more than 10000 ng/L with a mean equals to 7.1 ng/L for Europe;

- Under BAU scenario conditions the total sea load of PFOS from Europe is estimated to be, on average, 5.8 tons per year. Practically, the model foresees a half of this amount if a cut of emissions is assumed to take place as from the scenario of 50% reduction;
- The highest load of PFOS to marine coastal waters according to BAU scenario comes from the Danube river (followed by the Rhine) exporting annually more than 1 ton. Accordingly, Black Sea receives ca. 27.4% from the total load of PFOS to European seas;
- The spatial analyses anticipate that the total annual load of PFOS to European seas will decrease below the target value of 1 t/y only when the current emissions across to European countries are diminished at least by 84%.

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1. Introduction

The previous phase of the study about the expected cumulative impact of existing EU environmental legislation on the quality of the marine environment extensively debated the concepts and limitations currently hampering the assessment of chemicals fate at European scale (Pistocchi, 2009). Then, in order to perform retrospective and prospective chemical scenarios, the second year report specified reference concentrations and loads for three test-case contaminants in Europe: insecticide Lindane with reference to 1995 and 2005, herbicide Trifluralin considering year 2003 as baseline and the industrial pollutant Perfluorooctane Sulfonate (PFOS) with background in 2007. The purpose of the assessments was to obtain an overall European screening picture, to characterize the spatial fate and distribution of the chemicals of concern, as well as to identify possible hot spots in Europe and to estimate potential chemical loads to European seas (Pistocchi, 2009).

In general, the assessment of chemical fate and sea load at continental scale is typically performed on the basis of fate and transport models which use data about chemical emissions to different environmental media and convert them into predicted concentrations, fluxes and loads (Pistocchi, 2008; see also the references given thereby). In turn, the chemical fate and transport models make use of either a "direct" or an "inverse" modeling approach with the direct being a traditional one.

The direct models are based on *a priori* available information about chemical emissions and are intended to answer the question "Where do chemicals go after being emitted?". Thus, when data for chemical emissions and locations of sources could be retrieved from existing European databases, the model predictions of spatial distribution of chemicals prove to be sufficiently accurate for screening purposes (Pistocchi et al, 2010a). For that reason, the direct version of the spatially resolved **M**ultimedia **A**ssessment of **P**ollutant **P**athways in the **E**nvironment (**MAPPE**) screening model (Pistocchi, 2008; Pistocchi et al, 2010b) was applied in the case studies of Lindane (Pistocchi, 2009; Vizcaino and Pistocchi, 2010) and Trifluralin (Pistocchi et al., 2009).

Oppositely, the inverse modeling deals with the question "where do pollutants come from?" and is based on measurements of chemical concentrations in river network. Inverse models may support large scale assessment of the surface water quality by estimating chemical emission factors at regional, river basin or continental scale in relation to the population density or another proxy.

As a result, the earlier stage of the study motivated the applicability of the inverse modeling technique for PFOS. For this chemical the emission estimates are still poor and sparse, however, relatively extensive monitoring data are available from pan-European campaigns (Loos et al., 2009). Then, the potential of the inverse applications was used for backward tracking of PFOS emission factors from the existing riverine measurements (Pistocchi and Loos, 2009). Besides, the inverse approach was used recently in an investigation of fate of pharmaceuticals and personal care products emitted to the European river network (Pistocchi et al, 2011a).

Moreover, the direct and inverse model calculations of the environmental concentrations of chemicals prove to be sufficiently correct for screening at continental scale and generally the models demonstrated error levels within one order of magnitude (Pistocchi, 2009).

However, the accurate spatial prediction of chemicals fate at European level is still critically dependent on the knowledge of exact amounts and spatial distribution of pollutant emissions. For instance, the reliable information on pesticide emissions is expected to become available in the coming years following the implementation of the Framework Directive on the Sustainable Use of Pesticides 2009/128/EC, and the Regulation on the Statistics of Plant Protection Products, 185/2009 of the European Parliament and the Council. Once such information is available, the pesticides environmental concentrations and consequently sea loads might possibly be calculated more precisely.

Thus, considering the current data limitations, in the case of Lindane and Trifluralin it was decided to assess short and especially long term scenarios for chemical loads to European seas not only with the spatial direct MAPPE model but also using a simplified transient (non-spatial) box model. The latter consider entire European continent as one single unit; however, capitalize on the availability of maps of environmental parameters from spatial models to account for the variety of removal rates that govern chemical fate. This enables providing not only one single estimation of sea loads, but also the range of their variability.

For PFOS potentially a direct model can be used also to simulate a certain scenarios when their emissions are previously estimated.. However, as it will be discussed in a subsequent chapter of this report, actually the hazardous substance PFOS does not require scenario simulation using direct models due to its relatively simple fate Therefore, in the study the scenarios for PFOS will be assessed using the already available inverse spatial model for chemical fate in the stream network.

2. Formulation of scenarios for chemicals

Taking into account the discussions and conclusions made in the second year report (Pistocchi, 2009) as well as the recommendations provided by the communications with DG ENV afterwards, the likely order of magnitude of emissions to different environmental compartments is the critical point to define possible trend of chemical load in time and space.

For instance, Figure 1(a) shows a generic example of different types of tendency of the evolution of emissions that one may wish to consider once a chemical is subjected to restrictions or ban. Then, for a given emission scenario to a certain medium – for example a moderate gradual reduction of air emissions as presented in Figure 1(b), the spatial intra- and inter-media transfer of chemicals will provide the range of variability of the mass present in the different environmental compartments. These can be seen as "environmental breakthrough curves" which indicate the time required for emissions to be "flushed off" through the surrounding world.

Hence, in the light of the thoughts and considerations provided in the previous chapter, the scenarios for the expected impact of EU legislation in terms of the aquatic discharge from land to European seas have been formulated by taking into consideration the following aspects:

a. Time horizon

In general, the scenario simulations should be conducted using as initial condition the chemical mass or concentrations in a particular starting year when the measures (ban, phase-out, restriction, etc.) are enforced. Then, the reference time interval for the scenarios is specified from that moment until 2020 assuming different emission targets to be reached (for instance complete ban or partial restriction, etc.) and considering variants of the effectiveness of the implementation of the EU policies (for example gradual, linear or exponential reduction of emissions, etc.).

b. Type of action

The type of action taken is a key issue in order to reduce chemical loads. A few options are proposed to be considered in the study:

- **Business as Usual** (**BAU**) - no action to be taken, the reference (current) chemical emissions continue to be released without any changes;
- **Complete ban** - no use of a given chemical in any form or no direct emissions to any environmental media. However, indirect emissions resulting from atmospheric long range transport or leaching from contaminated sites could still contribute to ambient pollution;

therefore, this type of action eventually entails a quantification of additional emissions, which may be problematic to specify;

- **Phase out** – a cease of chemical emissions is assumed to occur following a gradual linear or fast exponential temporal decrease with a given speed or within a specified time period;
- **Partial** – a restriction of chemical use is assumed to be partially effective and the cease of emissions is successful only to some extent. The case is introduced as an alternative of ban scenario in order to consider more "realistically" the future trends of chemical loads.

c. Target

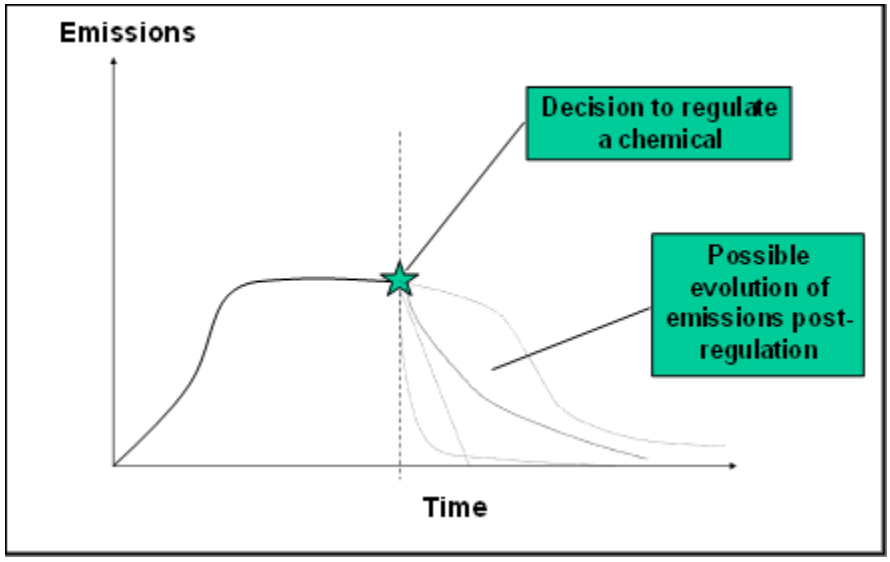
As a target of the actions here are considered:

- Spatial dimension - the desired effect of the actions simulated by scenarios could be pan-European or regional one.;
- Optimal measure - the modelling exercises should help answering the question "what is the best action to reach a given target?". In this case, several variants should be tested in order to identify when a *priory* fixed target will be achieved.

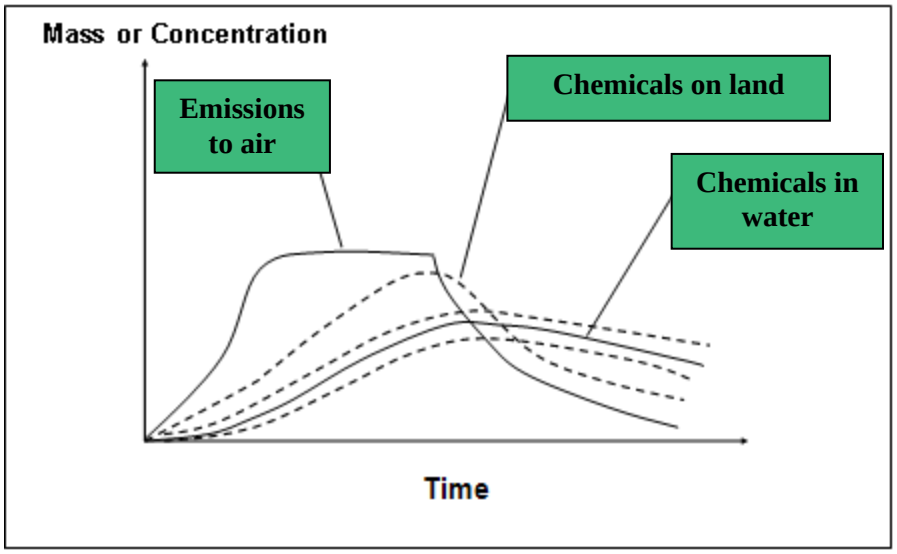
However, the ultimate goal of all investigated scenarios was to identify the plausible and realistic measures able at reducing chemical loads to European coastal water.

The scenarios for the three pilot chemicals considered in the present study are summarized in the Table 1. They were formulated and subsequently investigated on the basis of the above features accounting the specific reference conditions and the time horizon of year 2020 as well as the different types of measures or spatial targets.

Lastly, it is worth to stress that in PFOS scenarios since the inverse modeling approach is used only a reduction of current level of emissions is considered. Then, clearly the PFOS sea loads will change in the same proportion as the emissions. However, it will be of interest to assess which European rivers will experience PFOS concentrations below a given threshold, or which zones of Europe will contribute substantially to the sea load of this chemical.



(a) - Different evolutions of emissions after decision for regulation



(b) - Temporal evaluation in different environmental compartments

Figure 1 – A conceptual scheme of scenario reasoning.

Chemical	Target	Type of action	Time horizon
Lindane	Reduction of environmental concentrations and sea load under different policy measures; Entire EU or region specific.	Business as usual; Complete ban (plus trans-continental air transport); Partial effectiveness (linear or gradual reduction of emissions)	Baseline 1995 and a reconstruction for 2005 2020
Trifluralin	Reduction of environmental concentrations and sea load under different policy measures; Entire EU or region specific; Total sea load to remain below a provisional limit (21t/y).	Business as usual; Complete ban; Partial effectiveness (reduction of emissions by a given proportion).	Baseline 2003; 2020
PFOS	Reduction of river concentrations and sea load under different policy measures; Entire EU or region specific; European rivers with concentration above warning limit of 30 ng/L; Total load to European seas to decrease below a tentative threshold (1 t/y).	Business as usual; Complete ban; Partial effectiveness (reduction of emissions by a given proportion).	Baseline 2007; 2020

Table 1 - Summary of the chemical scenarios considered.

3. Modelling tools to run chemical scenarios

The section presents briefly the tools used in the project to run the formulated chemical scenarios. As was commented before, for PFOS no further development of the already available "inverse" modeling tool is needed (Pistocchi and Loos, 2009). However, this is not the case when the "direct" modeling technique has to be applied, since apart of the existing by now European version of MAPPE geo-reference model (Pistocchi, 2008), a new box modeling tool was built for pesticides as explained below in more details.

In general, MAPPE Europe model (Pistocchi, 2008; Pistocchi et al., 2010b) is a GIS based direct model which provides a user-friendly way to simulate steady-state fluxes and concentrations of chemical pollutants emitted by industrial activities or other point or diffuse sources.

The target contaminants, potentially considered by MAPPE model, are organic compounds such as Persistent Organic Pollutants (POPs) (*e.g.* polychlorinated biphenyls (PCBs), dioxins, furans, etc.) as well as pesticides, pharmaceuticals, volatile organic compounds, or other industrial, agricultural and house used chemicals. The model output consists of annual fate maps in terms of chemical concentrations, deposition rates, fluxes to other environmental compartments, etc.; the latter could be used for screening of hot spots or hazard zones at continental scale with spatial resolution 1 km^2.

However, in order to develop an unsteady tool to run the scenarios under "direct modeling scheme", an additional zero-dimensional time-dependent model was built up similarly to (Scheringer et al, 2000). This transient non-spatial box model is described shortly below (for details see Pistocchi et al, 2011b); it enables drawing the time trend of the average chemical distribution at large scale – e.g. at country, regional, continental or global scale, taking into account the spatial variability of environmental processes.

The non-spatial time-dependent box model considers for each environmental compartment a general mass balance equation in the form (Pistocchi et al, 2011b):

$$V\frac{dC}{dt} = emission - kCV$$

where C is concentration, V is the compartment bulk volume, "*emission*" includes direct emissions and transfer from other media, and k is the overall removal rate. For example, this conceptualization was used to develop Earth box model to quantify the timely trend of DDT in

different environmental media for a period of few decades (Schenker et al, 2008; Pistocchi et al, 2011b) including a forecast up to 2050.

Similarly, the present chemical scenarios are performed using the time series of emissions and the 5^{th}, 50^{th} and 95^{th} percentiles of chemical removal rates found by aggregating and averaging MAPPE model results for the European continent. This enables to forecast the "median situation" and its variability in terms of 5 and 95% confidence intervals.

Besides, this formulation accounts for the uncertainty introduced by the application of the continental scale averages when do not consider the exact location of the chemical emissions. The same approach is applied to specify the intermedia transfer exchange - e.g. the atmospheric deposition or soil run-off, but for simplicity neglecting possible co-variations. For instance, to compute the confidence interval of the air deposition, we consider only the variability of the air deposition rate which was applied only to the median of the chemical mass in the atmosphere. Apparently this type of calculations can be readily implemented in a spreadsheet form practically at no computational cost.

Lastly, the inverse version of MAPPE model (Pistocchi and Loos, 2009) assumes that the concentrations in river network reflect the chemical emissions and removal processes in the environment e.g. degradation, volatilization, settling, dilution, etc. Then, at steady state the chemical mass flow at a given river cross section should be equal to the sum of all emissions upstream, each reduced by the removal occurring along the stream network. Mathematically this can be written as:

$$C \times Q = \sum_{i \in catchment} E_i \exp\left(-\frac{\ln 2}{DT50} t_i\right)$$

where E_i is the chemical mass discharge [MT^{-1}] emitted at the generic i-th location of the catchment, t_i [T] is the time spent by the chemical in water from the i-th emission location to the river cross section of measurement, and C [ML^{-3}] is the observed concentration and Q [L^3T^{-1}] is the river water discharge at the same cross section. Respectively, DT50 [T] is the total removal half life in the stream network. The time t_i is the line integral of the inverse of water velocity along the flow trajectory passing through the location of emission E_i, and is numerically computed in geographic information systems through a "flow length" function weighted by the inverse of flow velocity (for details see Pistocchi and Loos, 2009 or Pistocchi et al., 2011a).

4. Lindane (gamma-HCH) scenarios

Lindane is a relatively well known chemical that poses limited concern presently, due to the extensive regulation underwent in Europe and globally during the last years. The reason to be selected as a pilot priority substance in this study was the relatively high number of data available allowing to evaluate the models and to define alternative scenarios. Although Lindane is officially banned in the European Union since 1995, some emissions still occurring due to releases from stockpiles and other sources, as discussed in Breivik et al. (2004). These are difficult to be quantified and may be an ideal subject of a scenario study.

4.1 Background information

Hexachlorocyclohexane (HCH) is an organochlorine compound largely used as an insecticide since the 1940s in the whole world for all kinds of crops, but also for non-agricultural purposes (as seed treatment or solvent use) as reported by the World Health Organization (Vizcaino and Pistocchi, 2010). HCH has two commercial forms: technical HCH with a content of 8-15% of γ-HCH, and Lindane, with 99% of γ-HCH. The technical HCH was phased out during the 1970s and 1980s and its usage has recently been reported to be ceased globally. Therefore, since the 1980s, Lindane is considered as the dominant source of γ–HCH (Vizcaino and Pistocchi, 2010).

The application of Lindane is mainly to soil as pesticide, but its volatility and a long atmospheric lifetime lead to a global transport. The major process controlling removal of Lindane from soils is volatilization. Due to its physico-chemical properties, namely relatively high solubility and low polarity compared to other organochlorines (log*Kaw*=-3.68; log*Kow*=3.7) and higher persistence (*DT50*=43days in air and *DT50*=708days in water and soil), γ-HCH can be detected in all environmental compartments, including soil, water, sediments, air and biota, where it tends to accumulate (Vizcaino and Pistocchi, 2010). Lindane enters surface water as a result of runoff from agricultural land. When released to water, Lindane is not expected to volatilize significantly. Lindane is very stable in fresh as well as in salt water environments. It is removed from the water column through secondary mechanisms, such as bounding to particles, adsorption to the sediment or to fish through the gills, skin or ingestion (Vizcaino and Pistocchi, 2010).

Besides, by the reason of its negative toxic effect to humans and wildlife, Lindane usage was reduced since late 1970s and severe restrictions are currently applied. In particular, the substance is listed in Aarhus protocol on POPs on the Convention on Long-Range Trans-boundary Air Pollution (UNECE, 1998), the List of Chemicals for Priority Action of the OSPAR Convention

for the Protection of the Marine Environment of the Northeast Atlantic (OSPAR, 2007) and also in the Stockholm Convention as a substance to be eliminated (UNEP, 2007). In Europe the use of Lindane was banned or severely restricted since the mid-1990s (IHPA, 2006) and this chemical is listed also under the European Water Framework Directive as a priority hazardous substance (Decision 2455/2001/EC).

4.2 Scenarios for Lindane emissions

Following the general scheme adopted for the development of the chemical scenarios in this study and respecting the specifics of Lindane use and fate, first the atmospheric emissions are specified up to the year 2020.

Emissions of post-ban usage of γ–HCH are quite difficult to be estimated although these sources may significantly affect the current environmental concentrations (Breivik et al., 2004). The European inventory of Prevedouros et al. (2004) suggests ca. 135000 tons were applied over the period 1970–1996 with the major contributions originating from France, Spain and the Netherlands. Besides, other authors found that approximately 650 tons were emitted in 1998 and the contribution of each European country was calculated (Breivik et al., 2004).

At present, the only comprehensive and reliable source of emission information at European scale is the European Monitoring and Evaluation Programme (EMEP), providing the official data about the atmospheric emissions by country (CEIP, 2009). Another potential source is the expert estimates (for example Pacyna et al., 1999; and Denier van der Gon et al., 2007), however, the latter could be highly uncertain. According to (CEIP, 2009) Lindane emissions in Europe have experienced an important decrease in most countries since Lindane ban in 1995, and about a half of the emitted amount is accounted by France.

This study relies on Lindane emissions estimated by EMEP (www.emep.int) in the framework of the reporting and analysis of chemical data under the protocol on long range transport of chemicals of the Montreal convention. Examples of the typical spatial distribution of Lindane emissions to atmosphere in Northern hemisphere and Europe are presented in Figure 2 for years 2005 and 2008 respectively, taken from EMEP data (www.emep.int).

Another emission inventory for persistent organic pollutants in 2000, including γ-HCH, is made for UNECE European countries and based on data from the Convention on LRTAP (Denier van der Gon et al., 2007). The inventory was based on activity scenarios developed in the framework of the EU CAFE programme and the key source analysis of the projected emissions assuming full implementation of the UNECE protocols. Here it is worth mentioning that practically this

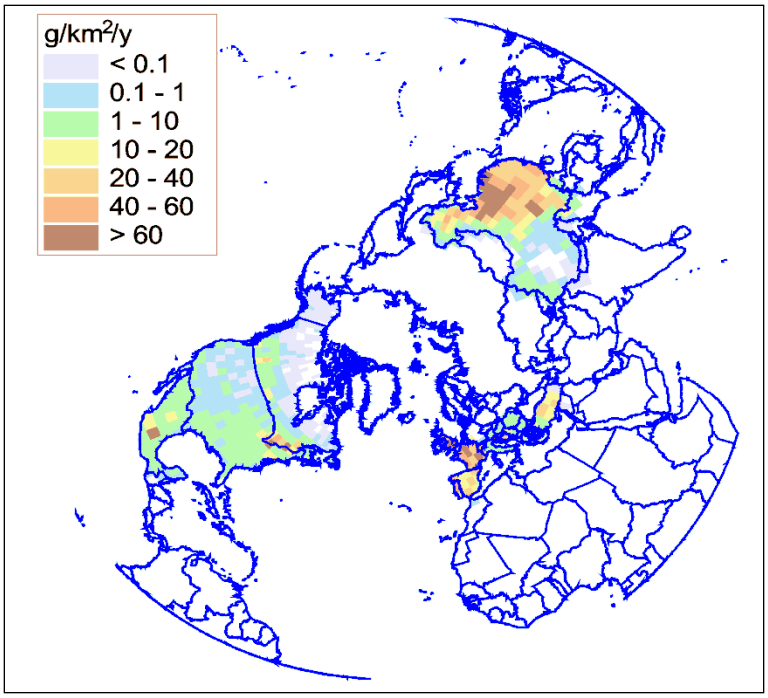
inventory gives emission values close to the official EMEP data apart for Italy (143.8 t/y versus EMEP's estimation of only 2.2 t/y). In addition, Denier van der Gon et al. (2007), presented emission projections for 2010, 2015, 2020 and forecasted that future HCH emissions will stabilize at ca. 255 t/y and will not change much over time because HCH use is reduced already before 2000 and the other measures are not affecting the currently allowed use of HCH.

However, according to recently available EMEP data (Gusev et al., 2006) the total emissions of γ-HCH in the Northern hemisphere (see Figure 2) were estimated to be 432 tons in 2005 including 71 t in North America, 68 t in Central America, 200 t in Southeast Asia and 92 t in Europe. In addition, the European continent received an extra deposition of ca. 2% of emissions coming from sources originated in North America and China (Gusev et al., 2006).

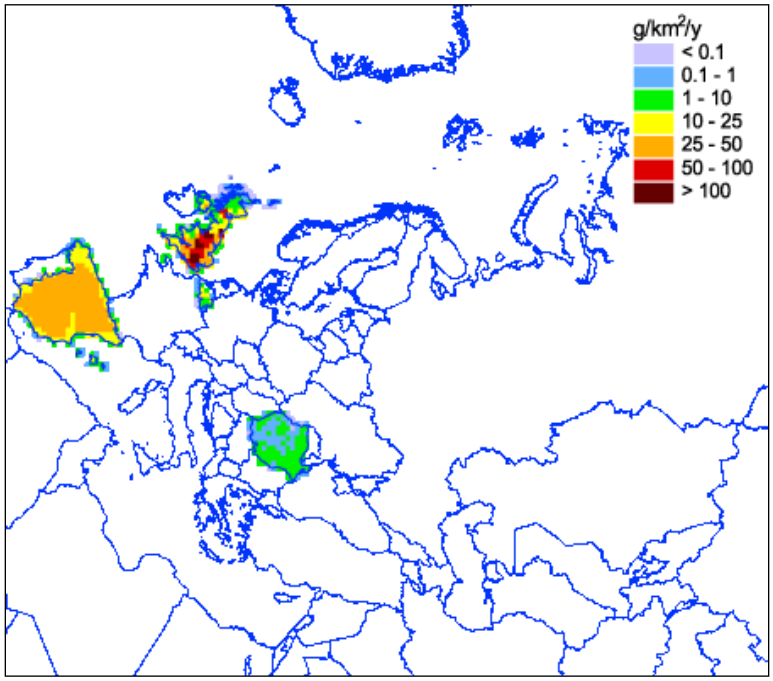
Besides, according to EMEP during the period 1990-2004 the emitted annual amounts of Lindane were reduced by 93% in Europe and only 11 European countries reported γ-HCH usage at least for one year in this period (Gusev et al., 2006). For example, in Belgium and UK the maximum contribution to total emissions is made by the solvent and other product use while in Croatia, Germany, Romania and Spain the agriculture was the dominant one. Practically the recent runs of the EMEP MSCE-POP model for γ–HCH for the period 2005-2008 were based on emission data for 2005 with small variations (Shatalov et al., 2010).

Based on the above information sources the possible scenario evolution of Lindane (γ-HCH) air emissions could be specified as follows (see the Figure 3 and also the Table 1):

- **BAU** – (Business as Usual) no change of air emissions; keeping 2005 level of emissions up to the time horizon of 2020 (about 92 t/y are supposed to be emitted in 2005);
- **trend** – a scenario continuing the emissions decline, observed in the period of 2000-2005, as a result of regulation process started in 1995 (see details on Figure 3b); accordingly, the emissions in 2020 equal to 45.6 t/y (49.6% of 2005 level);
- **linear** – a generic scenario which respects the regulations and assumes a gradual linear reduction of emissions starting in 2005 and ending in 2011 with 23 t/y (25% of the 2005 level of emissions); the choices of 2011 as an end year of measures and the percentage of emission reduction were provisional); no change of emissions in the period 2011-2020;

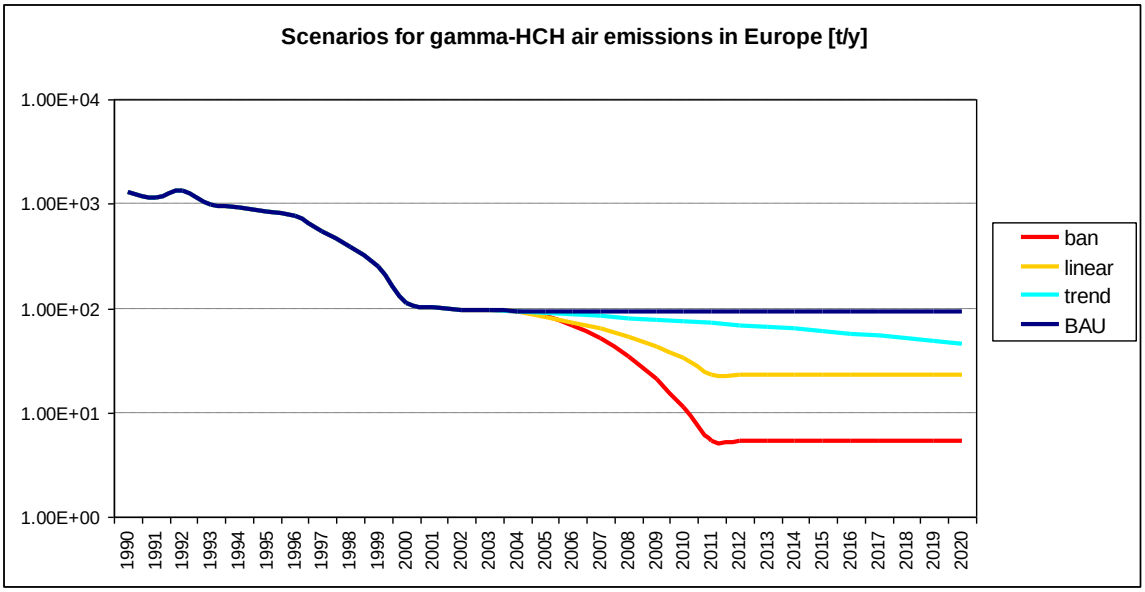


(a) – In Northern hemisphere during 2005 by EMEP, [g/km2/y]

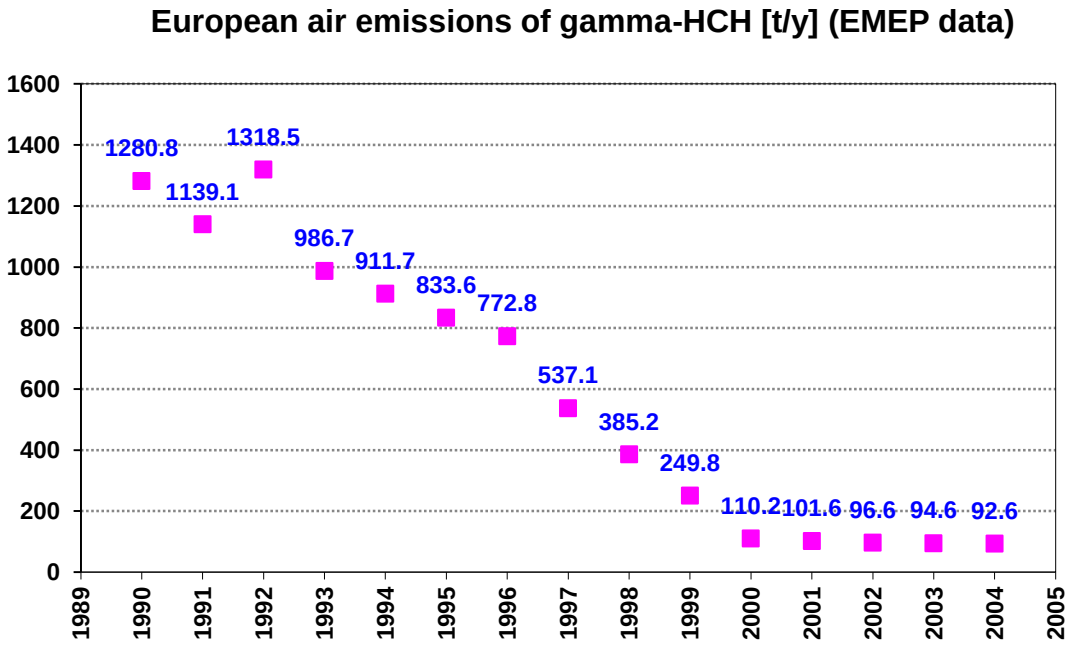


(b) – In Europe during 2008 by EMEP, [g/km2/y]

Figure 2 – Examples of spatial distribution of γ-HCH air emissions taken from EMEP (www.emep.int).



(a) - Possible evolution of Lindane air emissions during 2005-2020 time period in Europe. In the ban and linear scenarios the choice of 2011 as an end year of measures was provisional; no changes of air emissions 2011 onwards.



(b) – Estimates of Lindane quantities emitted to air during 1990-2004 period provided by official EMEP data

Figure 3 - European air emissions of Lindane (γ-HCH).

- **ban** – a scenario is consistent with regulation acts considering a fast exponential reduction of the European emissions in the period 2005-2011; emissions in 2011 are supposed to be equal to 5.4 t/y (the quantity arriving in Europe by the Long Range Atmospheric Transport according to 2005 data); as for the linear scenario the selection of 2011 as an end year of measures is provisional; from 2011 to 2020 no European emissions but the scenario accounts 5.4 t/y intercontinental atmospheric transport from North America and Southeast Asia using 2005 data as background emission level (the last available data from EMEP (Gusev et al., 2006).

After the specification of possible scenarios for air emissions, the next step was to stipulate the corresponding emissions to the other environmental media. In this context, there is a need to indicate the mode of release of Lindane and to specify the fractions emitted to the various environmental compartments.

For instance Denier van der Gon et al. (2007) showed that the relative contribution of source sectors to gamma-HCH emissions upon full implementation of the POP protocol by all UNECE–Europe countries is: Agriculture 18.5% and Solvent and product use (industrial) 85.5%.

Besides, according to UNEP (Breivik and Wania, 2002), about 59% of the total amount of Lindane are used for soil treatment, while seed treatment, which presumably yields lower emissions to air, accounts for 34%. Furthermore, Lindane is generally applied in liquid formulations (mostly wettable powders), and only a minor fraction is used in the solid state (dusts, powders, and granules). On this basis, a mode of division of total Lindane emissions by 17.5% to the atmosphere, 80% to the agricultural soil, and 2.5% to freshwater is assumed in POPCYCLING Baltic (Breivik and Wania, 2002) and MAPPE Europe (Vizcaino and Pistocchi, 2010) models.

To this end, the 17.5:80:2.5 fractions to air, soil and surface water was adopted in the present study up to year 1999, and only atmospheric emissions from 2000 onwards since Lindane was banned in EU for agriculture use in 1995 (see also the Figure 3b). Practically, the same approach was used in the EMEP modeling applications for Lindane (www.emep.int).

4.3 Overview of existing Lindane model applications

In order to validate the results of the models used in this study, the data collection and a short overview of already existing Lindane fate models at European scale were done.

Prevedouros et al. (2004) presented BETR multimedia fate model for Europe examining the fate of γ-HCH based on 1998's emission data. In accordance with the predominant wind direction, the

model predicted high air concentrations close to the major sources (France and Spain) as well as towards Central and Northeast regions. The atmospheric emissions supported max concentrations of 23 pg/m^3 with an estimated spatial variability of up to 2 orders of magnitude. The elevated soil concentrations found in Scandinavian region provided further evidence of the potential of increased scavenging by forests and subsequent accumulation by organic-rich terrestrial surfaces. The lowest soil concentrations are found in the South and East. A difference of factor of 100 in soil concentrations was evident, with a maximum of 15 pg/g in Belgium, the Netherlands and Northeast France. Since no background concentrations or historical release trends were taken into account, the concentrations for all compartments are likely underestimated. It was concluded that the measured and predicted concentrations fall in range of one order of magnitude (Prevedouros et al., 2004).

Besides, recent results about spatial distribution of γ-HCH within the EMEP region during 2008 are published by Shatalov et al., (2010), based on EMEP's MSCE-POP model simulations. Elevated γ-HCH air concentrations (50 pg/m^3 and above) were characteristic for UK and Spain. Moderate levels of air concentrations were obtained for the countries of the Western and Central Europe (5 – 20 pg/m^3). The countries of the Eastern Europe are characterized by particularly low γ-HCH air concentrations (about 5 pg/m^3), which is mainly connected with the absence of information on the emissions for this region. In addition, these authors reported the European spatial distribution of annual mean γ-HCH air deposition varying from 1 to 50 g/km^2/y and soil concentrations in the range 0.0001-0.07 ng/g (Shatalov et al., 2010).

However, the MSCE-POP model predictions of γ-HCH air concentrations for 2007 were somewhat lower for a few observation stations in Eastern Europe while at the same time, the measurements at the sites in Scandinavian countries were overestimated by the EMEP model (Gusev et al., 2009). The most significant differences in air concentrations account for a factor varying between 3 and 5. The reason can be triggered to uncertainties of amounts of Lindane emissions, and their spatial distribution or seasonal variations. Additional source of differences could be uncertainties in γ-HCH physical-chemical properties and their temperature dependence. For the remaining monitoring sites, the differences were about a factor of two (Gusev et al., 2009).

4.4 European Lindane box model - set up and verification

According to the description of the transient box model, generally presented in the 3rd section, the mass of γ-HCH in air at the end of a generic year *t* could be calculated as (Pistocchi et al., 2009; and Pistocchi et al., 2011b):

$$M(t) = M(t-1) \exp(-K) + E(t) * (1 - \exp(-K)) / K$$

where $E(t)$ is the air emission during year t, $M(t-1)$ is the mass of Lindane at the end of the previous year, and K is the overall air removal rate of Lindane. The latter was found using physico-chemical properties of Lindane and landscape and climate parameters by MAPPE Global model (Pistocchi et al., 2011b) from which we extracted the 5^{th} , 95^{th} and 50^{th} percentiles in order to account for European spatial variability of Lindane's fate. The retrieved removal rate coefficients (see Table 2) are well harmonized with those prescribed by the established fate and exposure models as USEtox (Rosenbaum et al., 2008) or IMPACT 2002 (Pennington et al., 2005). Further, the atmospheric deposition $D(t)$ during the year t is given by:

$$D(t) = (E(t) + M(t-1)) * Kdep$$

where $Kdep$ is the atmospheric deposition rate for Lindane (see Table 2). Then, we repeated the calculation using the time series of air emissions for the 5^{th}, 95^{th} and 50^{th} percentiles of K and $Kdep$ which enabled computing a "median" $M(t)$ and its 5 and 95% confidence interval .

Removal rate	5%ile	median	95%ile
Air, overall [$year^{-1}$]	40	396	4000
Air deposition [-] (as a fraction of the total air removal rate)	0.06	0.28	0.55
Soil, overall [$year^{-1}$]	0.36	1.96	11
Soil, removal by runoff [-] (as a fraction of the total soil removal rate)	0.02	0.1	0.5

Table 2 - Statistics of the γ-HCH environmental removal rates.

Similarly, the calculations could be performed for soil compartment, lumping in the emission term both direct emissions to soils and atmospheric deposition assuming for European continent that 94% of the deposition occurs on soil surface and 6% over the surface of water bodies (Pennington et al., 2005). In this case, we compute the overall soil removal rate the runoff removal rates, which yield the discharge from soils to the stream network, along the same logics used to compute the air deposition (see Table 2). Respectively, the load to European seas was found as a sum of direct emissions to surface water, diffuse discharge from soil and atmospheric deposition over the aquatic bodies.

In particular, the Lindane box model derives the concentrations in air, soil and surface water from mass into the corresponding media and their volumes, calculated on the basis of following data: surface area of Europe ($1.02*10^{12}$ m^2), average height or depth of different compartments (e.g.

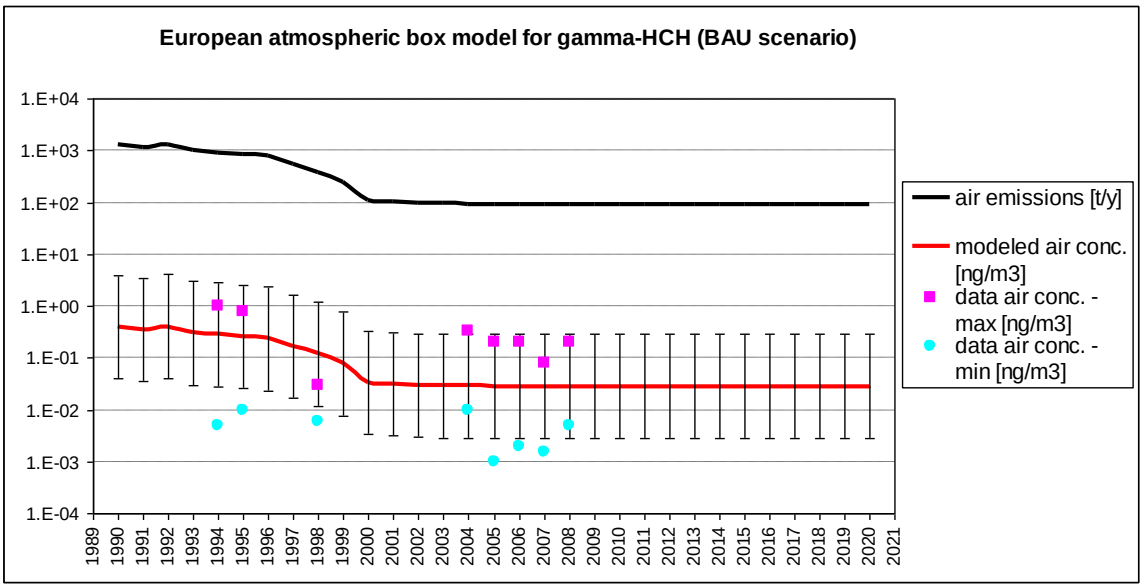
800m for air and 0.3m for soil; it is assumed a soil density of 1400kg/m^3), and total water amount (1-3*10^{12} m^3) annually discharged to the European seas given by Pennington et al., (2005) and Pistocchi et al., (2011a).

Then, the evidences that the developed European box model produces relevant results for γ-HCH concentrations are presented in Figure 4for air and in Figure 5for soil and surface water, respectively.

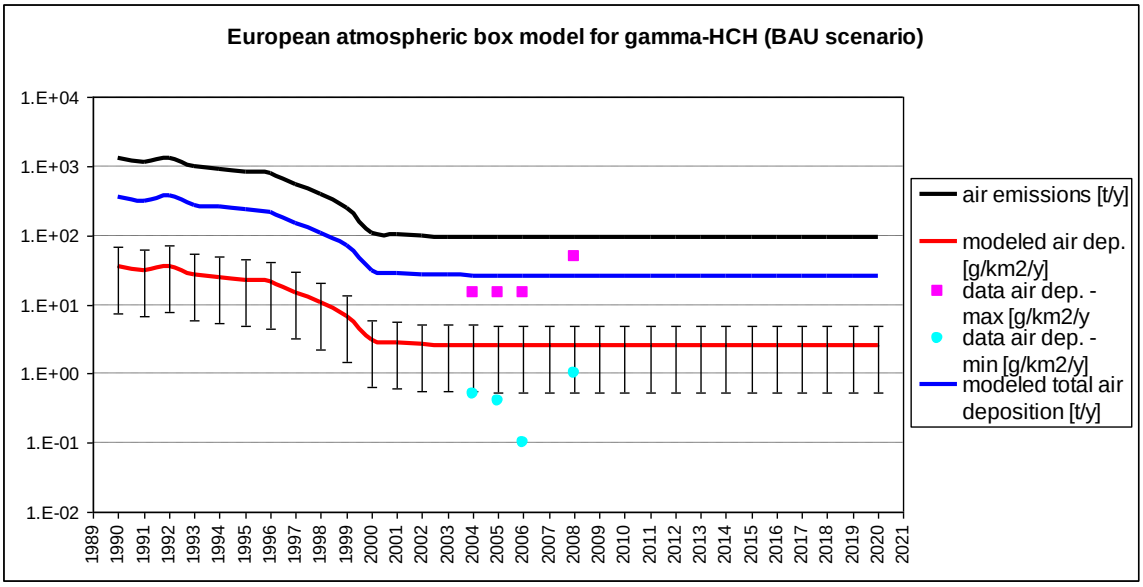
For example, in the case of air (see Figure 4**Error! Reference source not found.**) the referred data used to specify upper and lower limits of atmospheric concentrations and air deposition, are as follows: during 1994 – measurements plus outcome of the POPCYCLING-Baltic model (Breivik and Wania, 2002); in 1995 – results from MAPPE model (Vizcaino and Pistocchi, 2010); in 1998 – measurements and BETR model results (Prevedouros et al., 2004); in 2004-2006 and 2008 – results from MSCE-POP model (www.emep.int); and in 2007 – monitoring measurements (www.emep.int). Subsequently, by comparing against the measurements or model results could be concluded that the current box model, given its simplicity, is generating compatible results for Europe apart for 1998. However, as stated in (Prevedouros et al., 2004) BETR model outcome for air concentrations are likely underestimated observations and predicted concentrations fall in range of one order of magnitude compared to measurements, thus, close to the box model forecast. Latter is not surprising because the quoted observations or model results are for Baltic Sea region, known with lower atmospheric concentrations of γ-HCH in Europe (Breivik et al., 2004).

Moreover, the conclusion about good performance of the atmospheric part of the European box model could be extended to the soil and surface water compartments (see Figure 5). In this case the box model results are compared with MAPPE for 1995 and 2005 (Vizcaino and Pistocchi, 2010), by means of MSCE-POP for 2008 (Shatalov et al., 2010) only for soil concentrations and with the observations for surface water concentrations in river network around North Sea in 2001 (Ilyina et al, 2008).

As a final point, the work of Ilyina et al (2008) estimated the riverine input flux of γ-HCH to North Sea as a sum of discharge from the big continental rivers as: Elbe, Weser, Ems, Rhine, Meuse and Scheldt and British rivers: Thames, Welland, Humber, Tees, Tyne and Forth. It was concluded that in the period 1996-2001 the γ-HCH concentrations in rivers were generally reduced, thus in 2001, the range of measured surface water concentrations were 0.15-22 ng/L, which corresponds to the outcome of the Lindane box model: mean=3.18 ng/L; and a range 0.64-14.37 ng/L.

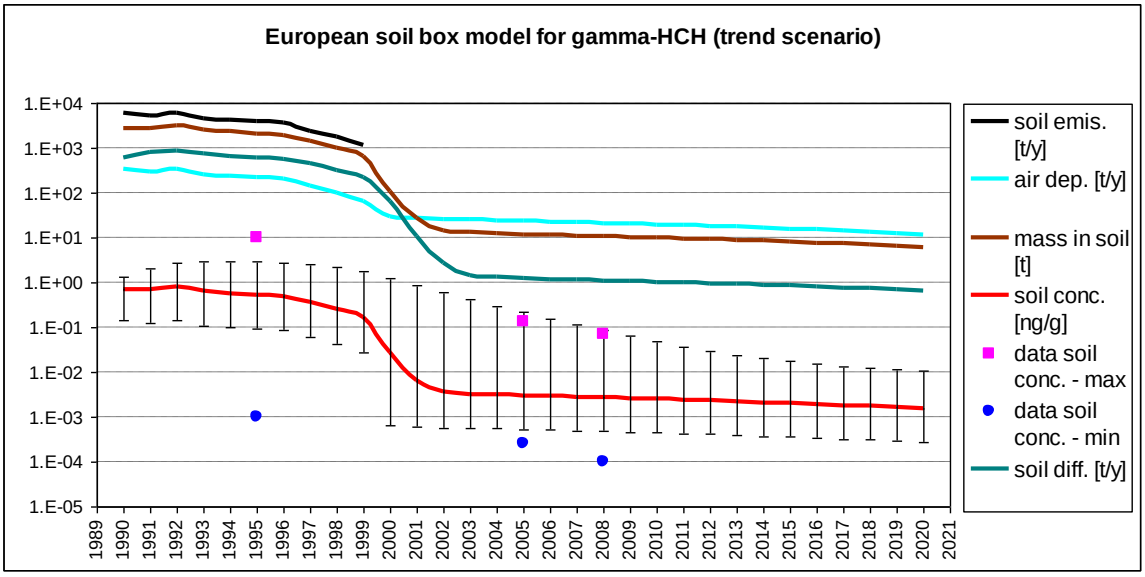


(a) – Annual average concentrations in air [ng/m^3]

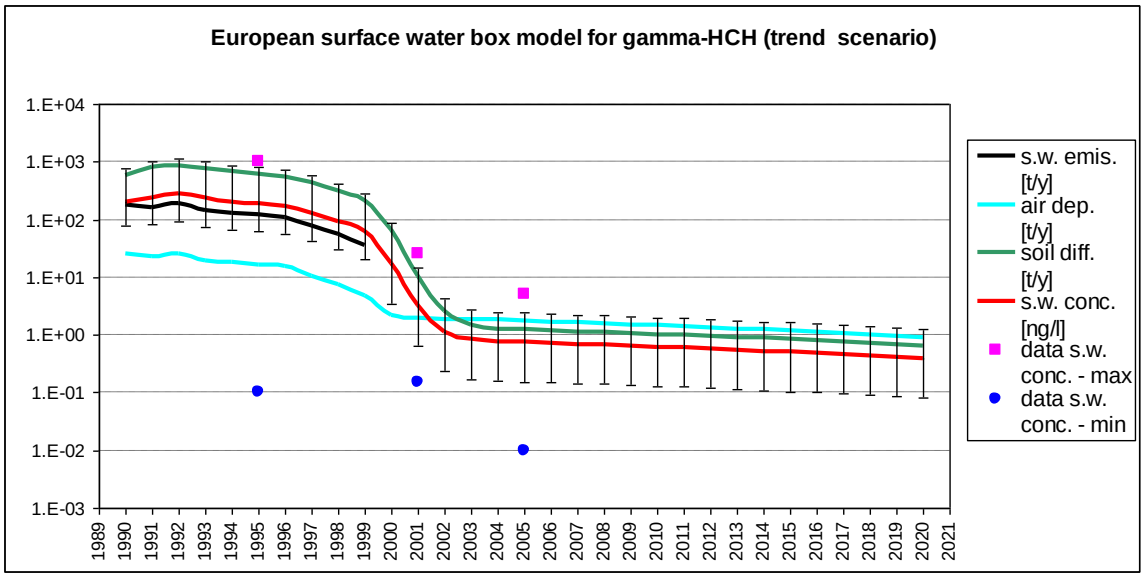


(b) – Annual mean atmospheric deposition [g/km^2/y]

Figure 4 – Results of European atmospheric box model for γ-HCH under BAU scenario. The comparative data used to specify the upper and lower limits of variability are taken as follows: during 1994 – measurements plus outcome of POPCYCLING-Baltic model (Breivik and Wania, 2002); in 1995 – results from MAPPE model (Vizcaino and Pistocchi, 2010); in 1998 – measurements and BETR model (Prevedouros et al., 2004); in 2004-2008 – results from MSCE-POP model (www.emep.int); and in 2007 – from EMEP observations (www.emep.int).



(a) – Annual mean soil concentration [ng/g] and mass [t/y] resulting from direct emissions to soil and atmospheric deposition.



(b) – Annual average surface water concentration [ng/L] formed by direct emissions, diffusion from soil and atmospheric deposition.

Figure 5 – Results of European soil and surface water box models for γ-HCH under trend scenario. The comparative data used to specify upper and lower limits are taken from: MAPPE model results for soil and surface water in 1995 and 2005 (Vizcaino and Pistocchi, 2010); EMEP MSCE-POP model (Shatalov et al., 2010) for soil concentration in 2008; and observations for surface water concentrations in 2001 (Ilyina et al, 2008).

4.5 Spatial model results for Lindane under different scenarios

This study employs as a main tool for gamma-HCH scenario runs the European version of MAPPE, a GIS multimedia chemical fate model (Pistocchi, 2008; Vizcaino and Pistocchi, 2010), to produce a quantitative spatial description of the behaviour of γ-HCH in Europe with an emphasis on continental surface waters.

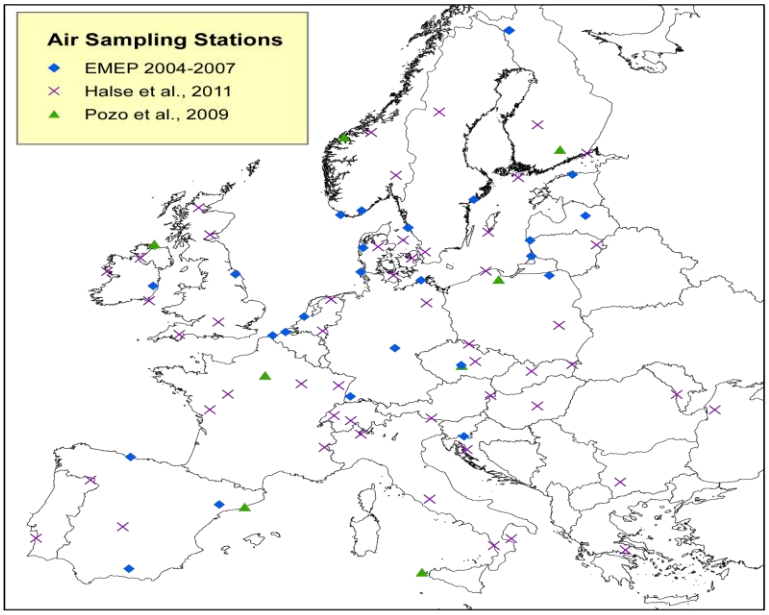
As a usual first step in modelling studies, a verification of MAPPE Europe model was done in order to evaluate the model performance and its capability to properly simulate the Lindane distribution and its environmental fate across the European continent.

For example, as reported in (Vizcaino and Pistocchi, 2010), MAPPE reproduces reasonably γ-HCH distribution and variation in atmosphere along the years 1995 and 2005. In general, the predicted air concentrations of γ-HCH are in good agreement with the measurements at the EMEP monitoring stations and with EMEP MSCE-POP model results and show a range 0.01-1 ng/m^3 (median=0.3) for year 1995, and 0.001-0.1 ng/m^3 (median=0.02) for 2005 (Vizcaino and Pistocchi, 2010).

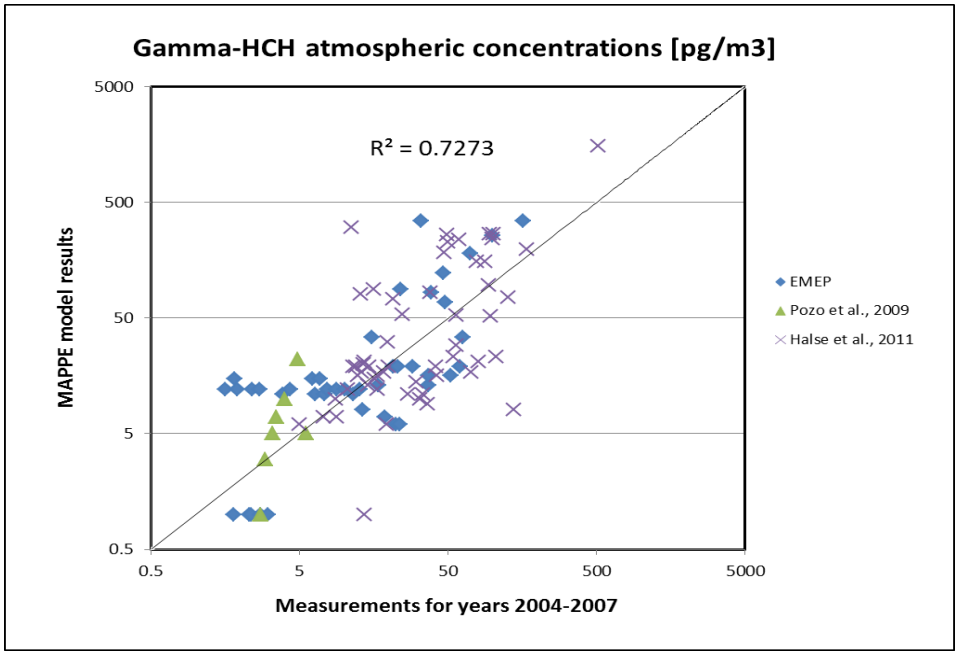
Additionally, we verified our results for atmosphere, obtained by MAPPE model under BAU scenario conditions, against reliable measurements with a good European coverage (see Figure 6) taken from EMEP data for the period 2004-2007 (www.emep.int) and also the observation data sets for years 2005 (Pozo et al., 2009) and 2006 (Helse et al., 2011). As can be seen the correlation between model results and measurements is really high (ca. R^2=0.74) which proves one more time the good model performance.

MAPPE soil results for 1995 (Vizcaino and Pistocchi, 2010) suggest that γ-HCH mass in soil is controlled by direct emissions to soil and follows agriculture intensity with high inter annual variations due to differences in emission along the year. The predicted annual average values of soil concentration of γ-HCH are in the range of 0.001-10 ng/g with a median ca. 0.1 ng/g. The uppermost values were found to be close to those measured in forest soils with higher organic carbon content (Vizcaino and Pistocchi, 2010).

In 2005, the spatial distribution of γ-HCH in soil is more homogeneous than in 1995, apart for the countries which are still using Lindane, due to the "smoothing" effect of atmospheric deposition that dilutes chemical mass over larger areas with lower gradients. The model results are spread in a range 0.0001-0.1 ng/g with a median of 0.001 ng/g, hence, 100 times less than in 1995 (Vizcaino and Pistocchi, 2010).



(a) – spatial distribution of sampling locations across Europe



(b) – correlation between MAPPE model atmospheric results obtained under BAU scenario conditions and observations; the other scenarios give lower correlations.

Figure 6 – Verification of atmospheric part of MAPPE model against reliable measurements with a good European coverage using EMEP data for the period 2004-2007 (www.emep.int) and passive samplers observations for 2005 (Pozo et al., 2009) and 2006 (Helse et al., 2011).

Furthermore, the soil compartment of MAPPE also was evaluated referring to "trend" scenario conditions against EMEP MSCE-POP model results for annual average Lindane mass in soil during 2008 (as shown on Figure 7). The comparison evidenced that MAPPE is likely to slightly overestimate the EMEP model estimates, in particular those giving the higher concentrations of Lindane in soil (more than 0.01 ng/g), but the coefficient of correlation (R^2=0.45) is still reasonably high to guarantee the worthy MAPPE model work. Possible reasons for the observed deviations between the model outcomes could be a different conceptualization of processes specifying the chemical fate in soil or simply alterations in the soil parameters in the both models.

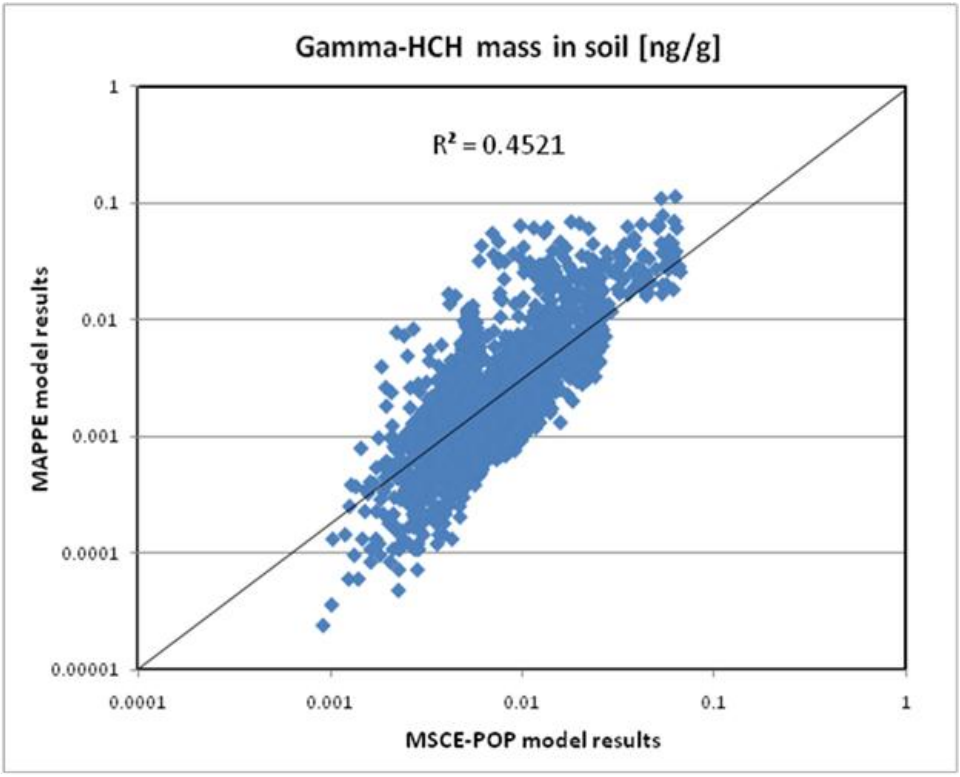


Figure 7 - Verification of soil compartment of MAPPE model referring to "trend" scenario conditions against EMEP MSCE-POP model results for annual average Lindane mass in soil during 2008.

Next, we focus on the surface water compartment results of MAPPE model. In fact, the gamma-HCH has been subject to extensive monitoring in European rivers as one of the priority hazardous substances under the WFD (Directive, 2000/60/EC). Consequently few databases exist presently

– for example EIONET, 2009; Catalonian Water Agency database, 2009; ARGE-Elbe monitoring programme, 1995-2005, showing also a seasonal distribution and a trend of decreasing of concentrations along the years.

The Lindane concentrations predicted by MAPPE for year 1995, when Lindane was still used in agriculture, are in a range 0.001-3000 ng/L while for 2005, assuming severe restrictions in Lindane use, the model yields to substantial reduction and the range is 0.001–500 ng/L(mean=13.8) according to Vizcaino and Pistocchi, (2010).

In Vizcaino and Pistocchi, (2010), the MAPPE estimates were compared with monitoring data for 1995 on River Elbe in Germany (25 sampling points) (ARGE-Elbe monitoring programme, 1995-2005). However, only for this data set a good correlation between the modelled and observed water concentrations was obtained (R^2=0.86), accompanied by the reasonable correspondence in terms of orders of magnitude (for most sampling locations the differences were within the factor of two; see Figure 3 from Vizcaino and Pistocchi, 2010).

Conversely, for 2005 neither the predicted surface water concentrations by Vizcaino and Pistocchi, (2010) nor those considering the present three scenarios correspond well to the observations at monitoring stations (405 samples distributed throughout the Europe). However, even being a bit higher comparing to observations, the model calculations provide mean values and variability ranges quite similar to measurements, in particular to EEA EIONET data base. Eventually, this discrepancy could be explained by one of the limitations of MAPPE model, namely, the lack of intra-annual variability of river water discharge since MAPPE uses as input the annual averages of the river discharge.

Thus, based on the fact that the MAPPE predictions for Lindane river concentrations lay generally in the range of measurements (EIONET, 2009; Catalonian Water Agency database, 2009; ARGE-Elbe monitoring programme, 1995-2005) we concluded that the surface water compartment of MAPPE model is performing also reasonably well, similarly to the atmospheric and soil ones.

After the MAPPE model verification, the gamma-HCH environmental distributions in Europe and sea load have been calculated firstly under the BAU scenario conditions, assuming only air emissions from Europe – ca. 86 tons and skipping the transcontinental atmospheric transport.

Apart of the BAU case, the trend scenario was also studied (in total 42.5 tons of Lindane released to air being a sum of the reduced European emissions and LRAT for 2005) and ban scenario (no discharge in Europe but 5.4 tons of extra atmospheric emissions originated from remote sources

with a background 2005's level taken into account; the additional emissions are assumed to be spread homogeneously over the entire European continent).

The resulting spatial maps of Lindane atmospheric concentrations and corresponding air deposition for the above three scenarios are presented in Figure 8. The results show, as expected, that the pollution by Lindane is reduced considerably towards the ban scenario direction. Logically, the higher levels of atmospheric concentrations and air deposition are observed near to the sources of elevated emissions. As a result, bigger mass of Lindane in soil (see Figure 9) and consequently a rising of surface water concentrations through the soil run-off can be observed, as highlighted in the Figure 10, representing an example of the Lindane surface water concentrations in Europe under BAU scenario.

In addition, the Figure 11 shows spatial changeability of the Lindane sea load depending on the different scenario runs.

Detailed statistical information about the range of variability, mean values and standard deviation of the simulated Lindane concentrations in air, soil and surface water as well as for Lindane atmospheric deposition and riverine sea load is shown in the Table 3.

Moreover, the analyses based on the spatial maps for Lindane discharge to surface water allow assessing the total and disaggregated loads to European coastal waters, as provided in the Table 4. It was found that , the European discharge of Lindane basically affects Atlantic Ocean (49 % of the total for Europe), Mediterranean (27 %) and Black (19 %) seas, under BAU scenario and predominantly the Black sea (43 % of the total), followed by Mediterranean sea (19 %), Baltic sea (17%) and Atlantic Ocean (16%), when the ban scenario is considered. Obviously, the Lindane load to regional seas reflects the pattern of the spatial distribution of the atmospheric deposition and mass in soil.

Furthermore, the spatial analyses show that, when the BAU scenario is enforced, a total sea load of ca.12.5 tons per year should be expected. However, for year 2020, the trend and ban scenarios forecast respectively a 74% and 95% sea load reduction comparing to the BAU case.

Scenario	Variable	Mean	St.dv.	Range
BAU	Air concentration [ng/m3] (annual mean)	2.3 e-2	6.7 e-2	3.1
	Air deposition [g/km2/y]	4.8	25.4	3412.0
	Mass in soil [ng/g] (annual mean)	1.3 e-2	3.3 e-2	3.2
	Surface water concentration [ng/L] (annual mean)	13.9	70.0	9511.1
	Riverine Load to seas [t/y]	1.8 e-2	0.1	2.3
Trend	Air concentration [ng/m3] (annual mean)	1.1 e-2	4.0 e-2	1.2
	Air deposition [g/km2/y]	1.3	7.0	851.4
	Mass in soil [ng/g] (annual mean)	3.9 e-3	1.1 e-2	0.9
	Surface water concentration [ng/L] (annual mean)	6.0	29.1	4152.1
	Riverine Load to seas [t/y]	4.4 e-3	3.0 e-2	0.6
Ban	Air concentration [ng/m3] (annual mean)	1.5 e-3	2.8 e-3	0.1
	Air deposition [g/km2/y]	0.3	0.7	39.3
	Mass in soil [ng/g] (annual mean)	7.9 e-4	1.1 e-3	4.3 e-2
	Surface water concentration [ng/L] (annual mean)	0.9	3.6	302.5
	Riverine Load to seas [t/y]	7.9 e-4	9.1 e-3	0.2

Table 3 - Statistics about the range of variability, mean values and standard deviation of the simulated concentrations of Lindane in air, soil and surface water and for Lindane atmospheric deposition and riverine load to European seas.

Besides, it is worth mentioning that Ilyina et al., (2008) estimated γ-HCH load of ca. 1 ton to North Sea in 2001; logically MAPPE forecasted lower amount of only 0.43 tons for the later year 2005 as shown in Table 4. Then, assuming that North Sea receives approximately a shear of about 6.8% from the total load to European seas (an average value found using data provided in the Table 4), it could be calculated a total European load of about 14.7 tons for year 2001. The latter well corresponds to the estimate of 12.5 tons produced by MAPPE respecting the BAU scenario circumstances (with a baseline in 2005) as well as to the box model forecast of 12.7 tons for 2001 (as presented in the next section).

In order to further evaluate MAPPE model results, an additional comparison with data for gamma-HCH load to maritime area of OSPAR Convention was done. For instance, the OSPAR Commission reported for year 2009 (OSPAR Commission, 2011) the following ranges of riverine inputs to Atlantic Ocean - lower=0.031 t/y and upper= 0.66 t/y and for North Sea - lower=0.064 t/y and upper=0.66 t/y. Then, comparing these values to MAPPE forecasts (see Table 4), we found that actually the model doubled the OSPAR data when the trend scenario is accounted (amongst the considered scenarios this should be the nearest to 2009 conditions) but at the same time MAPPE model well approximates OSPAR loads when considering the "ban" scenario conditions.

In conclusion, the results of the performed comparison with OSPAR data or the outcome of a sophisticated model for North Sea (Ilyina et al., 2008), allowed concluding that MAPPE model produced relevant estimates for the riverine load of gamma-HCH to European seas which eventually could differ from the real discharges by not more than a factor of two.

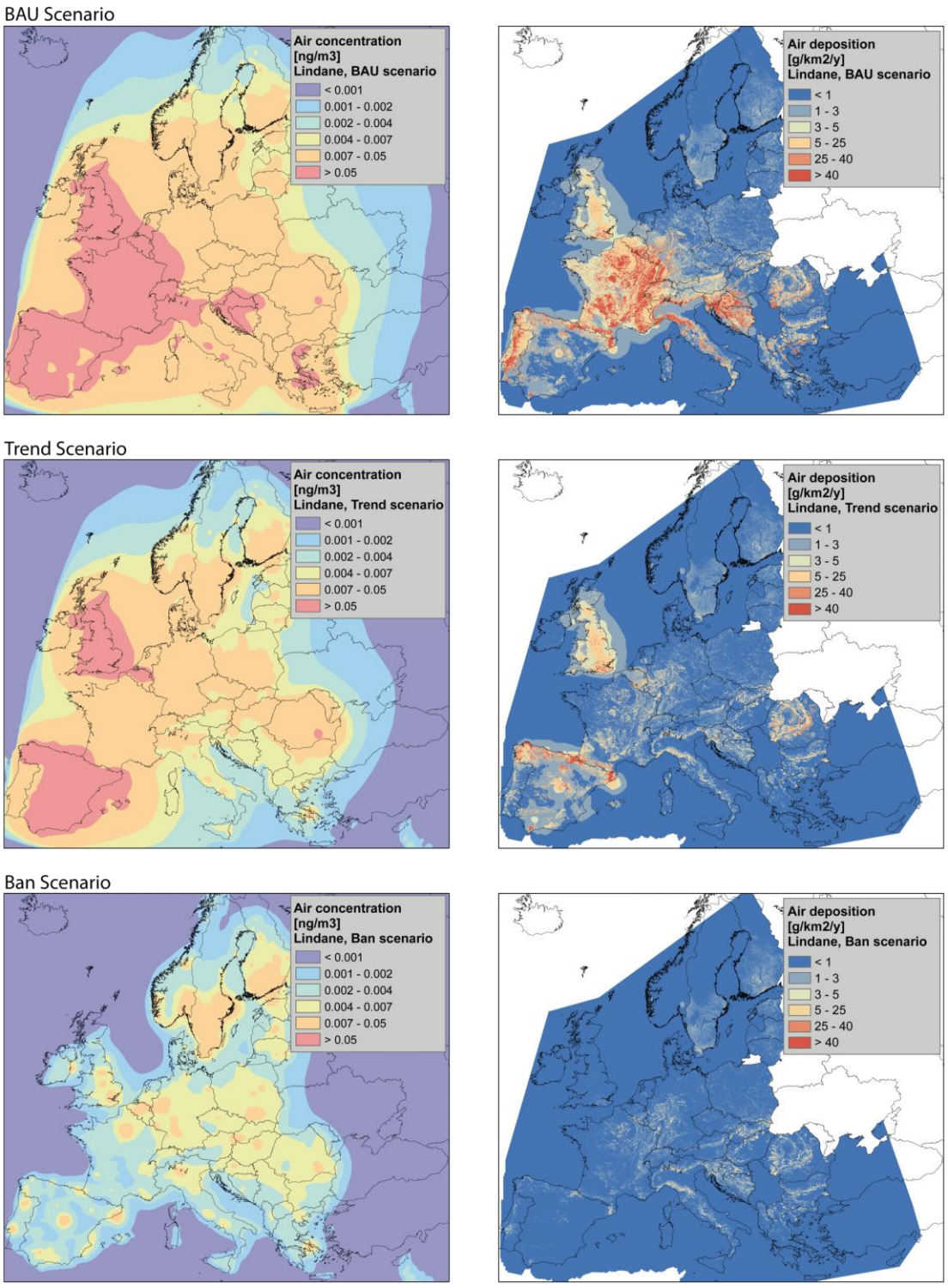


Figure 8 - Spatial maps of Lindane atmospheric concentrations and corresponding air deposition under BAU, trend and ban scenarios.

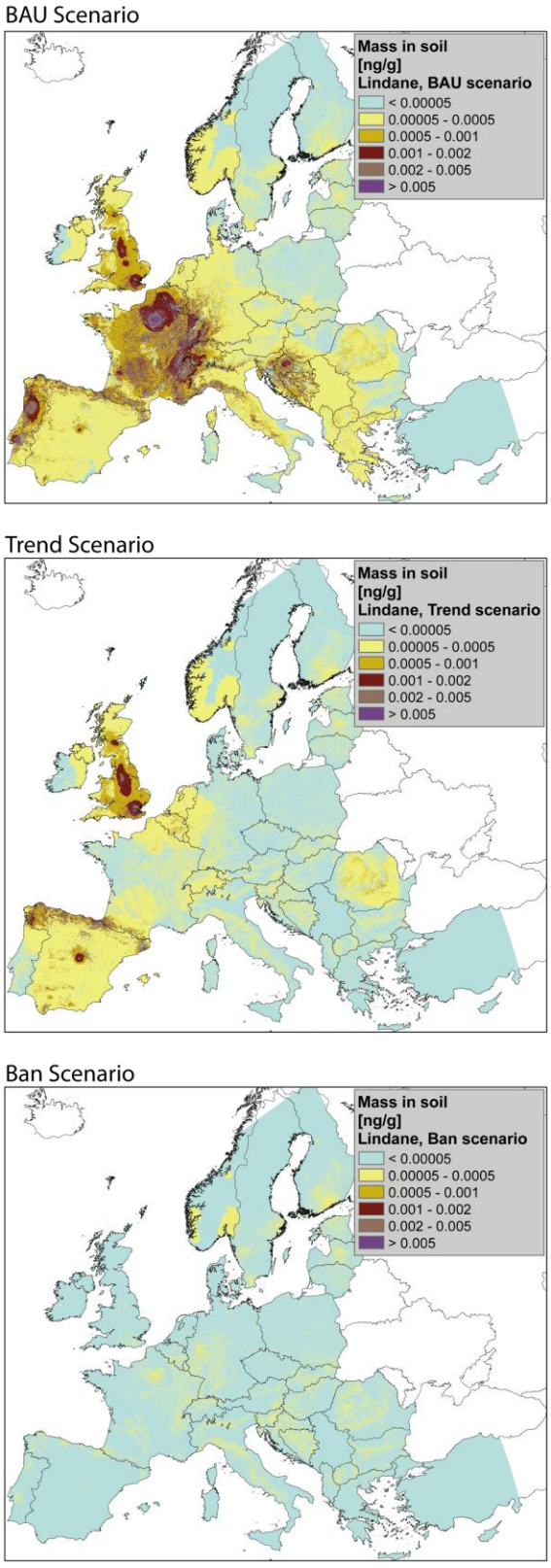


Figure 9 – Spatial maps of Lindane mass in soil under BAU, trend and ban scenarios.

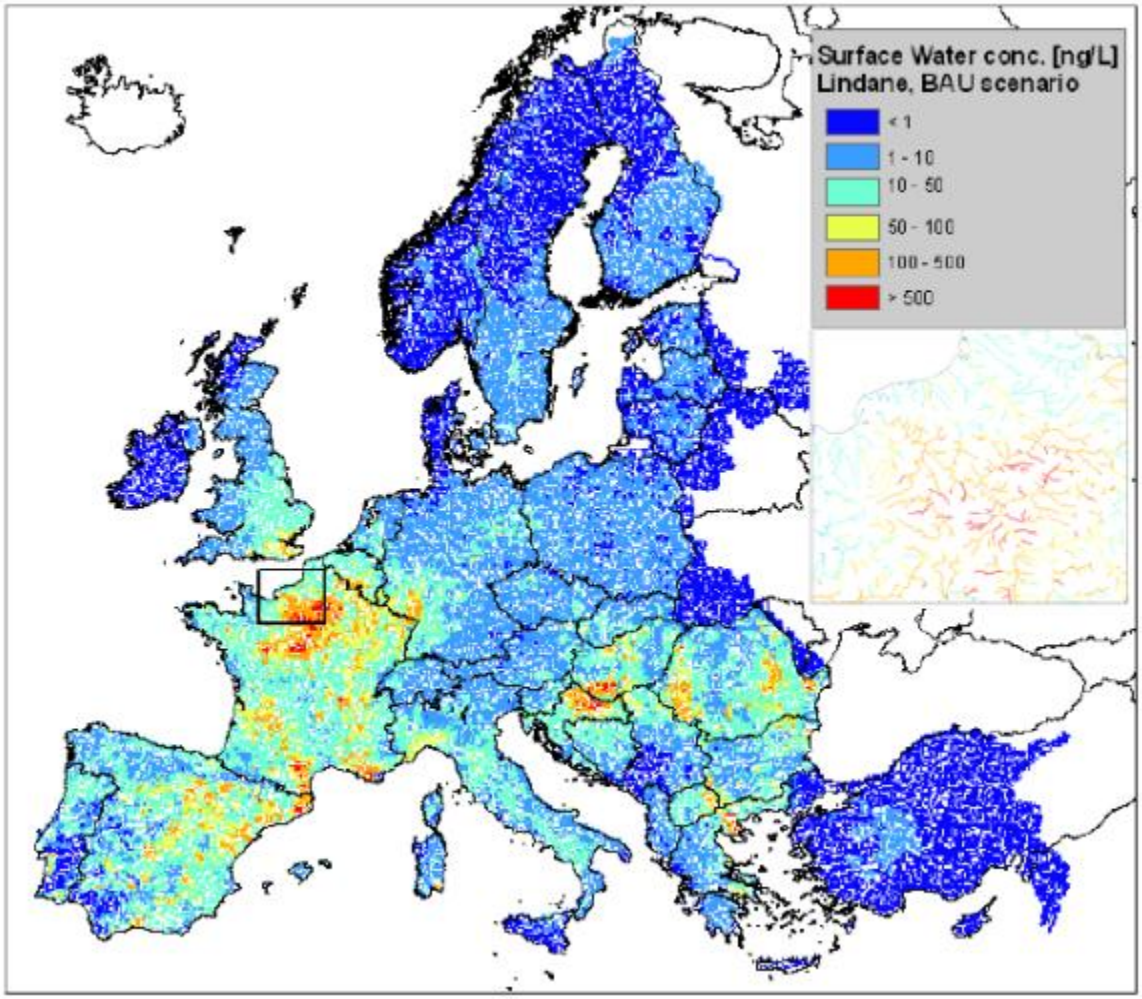
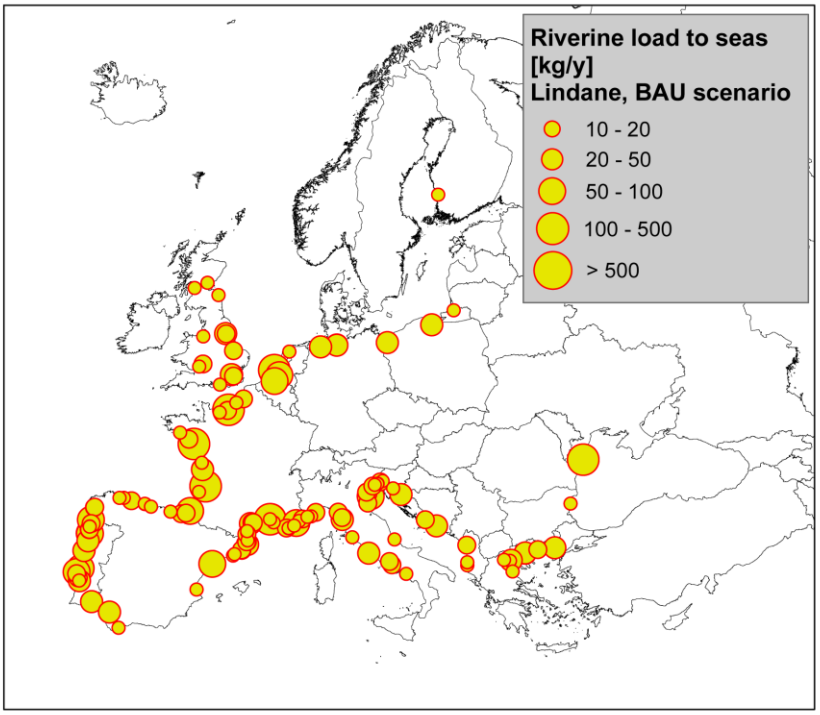
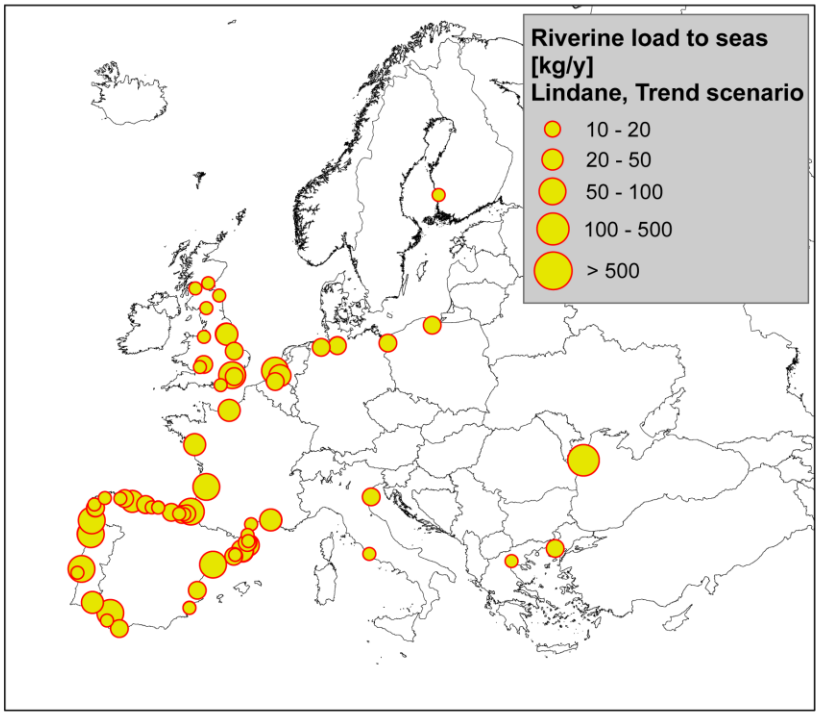


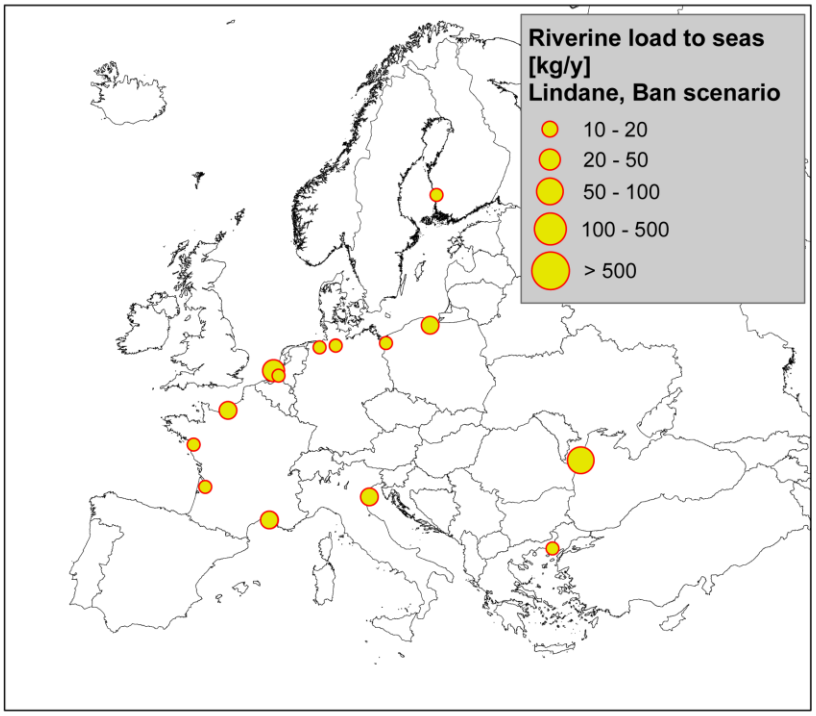
Figure 10 - An example of surface water concentrations of Lindane under BAU scenario.



(a) – BAU scenario



(b) - trend scenario



(c) – ban scenario

Figure 11 – Modelled estimates about spatial changeability of Lindane load to European seas depending on different scenarios

Load to seas [tons/year]	Scenario		
	BAU	Trend	Ban
Atlantic ocean	6.129	1.343	0.094
Baltic sea	0.2	0.134	0.101
Black sea	2.321	0.697	0.257
Mediterranean sea	3.405	0.735	0.115
Northern sea	0.431	0.396	0.03
Total	12.486	3.305	0.597

Table 4 – Modelled assessment of total and disaggregated load of Lindane to European coastal seas in 2020.

4.6 Results of Lindane scenarios by the box model

Examples of Lindane (γ-HCH) load to European seas produced by the box model as a result of direct emissions to surface water, atmospheric deposition and diffusion from soil are presented in the Figure 12 for the two "extreme" scenarios – BAU and complete ban.

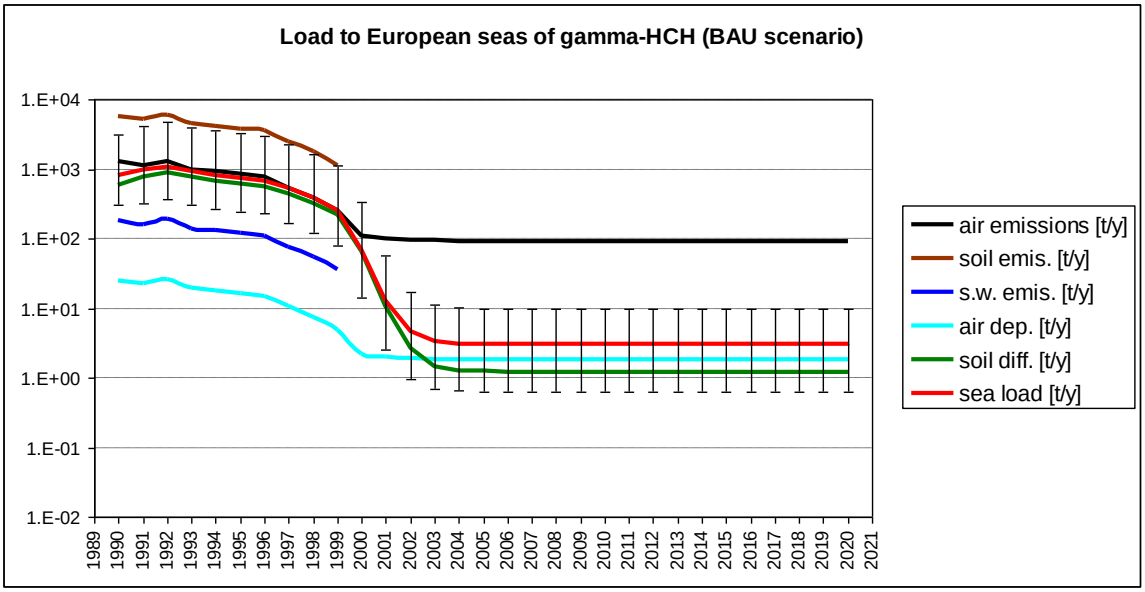
In both scenarios, up to 1999, the sea load is formed basically by the soil diffusion given the higher Lindane soil applications and the 5 times lower atmospheric and almost 2 orders of magnitude lower surface water emissions. On average, in the period 1990-1995, the total loads varied about 900 t/y (ca. 745 tons in 1995). Afterwards, the load is reduced 3-4 times since the phase out started after 1995 as a result of the application of EU regulations and reached ca. 256 t in 1999 (34.3% of 1995's load) but remains relatively high because of still existing agricultural applications. Onwards 2000, due to the null soil and surface water emissions the situation is significantly improved and the sea load diminishes to ca.3.1 t/y (with a range of 0.65-10 t/y) in 2004, as forecasted by the box model (an explanation for the observed differences between box and spatial model forecasts is given below).

According to BAU scenario from 2005 up to 2020 the sea load is practically constant (ca. 3 t per year). Besides, under BAU scenario the sea load is supported primarily by the stabilized atmospheric emissions originated from European or trans-boundary sources through the atmospheric deposition and secondly by soil diffusion.

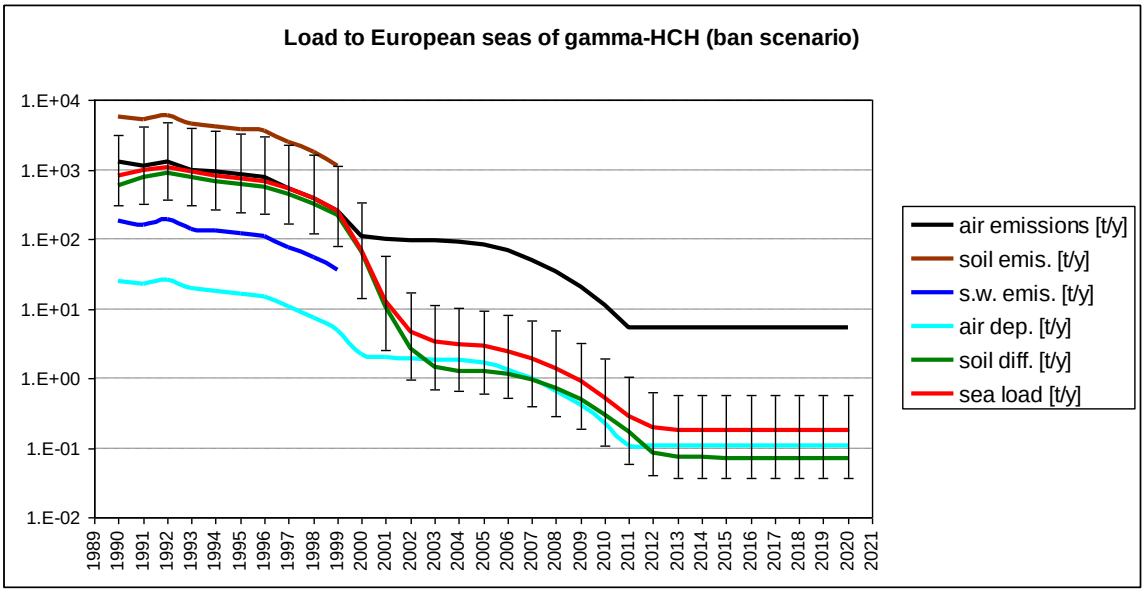
In the case of "ban" scenario, the process of reduction of the sea load continues further onwards 2005 and results to only 0.53 t in 2010 (almost 6 times less compared to the level of 2005). Contrary to BAU scenario, during the transition period of 2005-2010 the sea load is formed equally by air deposition and soil diffusion, but afterwards both BAU and "ban" scenarios demonstrated similar pattern with dominant "atmospheric feeding". However, in the ban scenario the stable final value of load (0.18 t/y) is reached in 2015; it is remaining the same up to 2020 (only 6% comparing to the load in 2005).

Clearly, the other two scenarios estimated sea loads between the BAU and ban forecasts - for example the 'trend" scenario gives 2.49 tons in 2010 and 1.54 t in 2020 while the "linear reduction" scenario calculates 1.24 tons in 2010 and 0.76 t onwards 2013.

However, it is important to stress that the spatial model forecasts are always closer to maximal estimates of the box model (instead of mean ones as expected) which indicates that projections based on average box model guess likely to underestimate 3-4 times the Lindane load to seas.

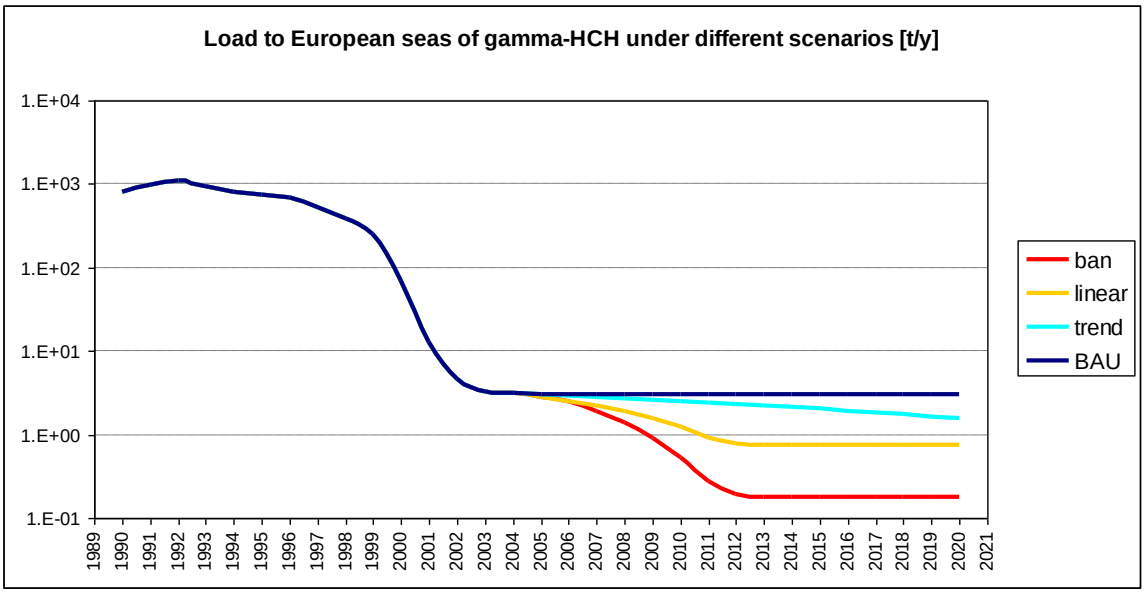


(a) - BAU scenario

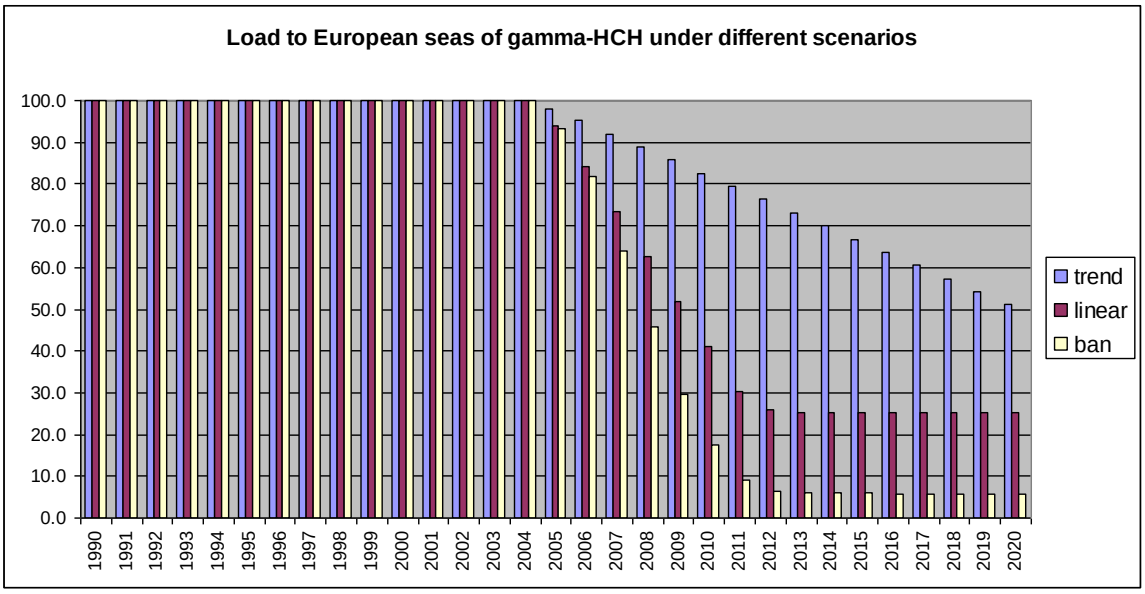


(b) – Ban scenario

Figure 12 - Modelled estimations of γ-HCH load [t/y] to European seas as a result of direct emissions to surface water, atmospheric deposition and diffusion from soil sources under two extreme scenarios: (a) – **BAU** - no change of air emissions; keeping 2005's level of emissions up to the time horizon of 2020 (92 tons are supposed to be emitted in 2005); and (b) - **ban** – a scenario is consistent with regulation acts considering a fast exponential reduction of the European emissions in the period 2005-2011; emissions in 2011 are supposed to be equal to 5.4 t/y (the quantity arriving in Europe by the Long Range Atmospheric Transport according to 2005 data); as for the linear scenario the selection of 2011 as an end year of measures is provisional; from 2011 to 2020 no European emissions but the scenario accounts for 5.4 t/y intercontinental atmospheric transport from North America and Southeast Asia using 2005 data as background emission level (the last available data from EMEP (Gusev et al., 2006).



(a) – Model forecast of absolute sea load in [t/y]



(b) – Relative effect of scenarios on sea load as a percentage of estimates for BAU [%]

Figure 13 – Comparison of modelled estimates for γ-HCH load to European seas under different scenarios.

Moreover, the scenario runs indicated (see Figure 12) that about 4-5 years are needed for the stabilization of the γ-HCH load to European seas assuming only atmospheric emissions. Besides, when the sea load is formed primarily by the atmospheric deposition it represents about 3.3% from the atmospheric emissions originated either from sources inside Europe or advected by the intercontinental atmospheric transport.

The summary of the γ-HCH scenarios, performed by the European box model, about the expected impact of EU legislation on the aquatic discharges from land to marine environment is presented on Figure 13a. The results allow concluding that in 2005, ten years after the start of EU regulation measures for the γ-HCH, the European load to seas, representing 745 t/y in 1995, is reduced of more than 2 orders of magnitude to only 3t/y.

The model results indicated that the decrease of sea load took place in two periods: firstly – from 1995 to 1999 with 65.7% decline, compared to 1995's level of emissions; secondly – during period of 2000-2005 a drop by 98.3% comparing to 1995 as the starting year of EU regulation for Lindane.

Lastly, the efficiency of the different Lindane scenarios in terms of percentage of load reduction compared to BAU scenario has been calculated (see Figure 13b). Obviously, the 'ban" option is the best working one because is supporting a reduction of sea load in 2020 to only ca.6% from the BAU forecast e.g. 94% efficiency. Next are the "linear reduction" scenario with 75% and the "trend" scenario via 49% efficiency, respectively.

5. Trifluralin scenarios

Trifluralin is an herbicide banned in EU countries, since 2008, due to its harmful characteristics. Moreover, it is a priority substance under the WFD. Presently, a very little information on Trifluralin emissions is available at pan-European level, and the only comprehensive dataset available for the 25 EU Member States is provided by ESTAT with reference to year 2003. However, this database allows estimating emissions only at the level of chemical class (a group of chemicals with similar properties), but not for individual substances, as discussed by Pistocchi and Bidoglio, 2010.

5.1 Background information

Trifluralin is a selective dinitroaniline herbicide used to control a wide range of annual grasses and broadleaf weeds; it is bioaccumulative and persistent and a possible human carcinogen subject of a transfer across the soil-water environment (Shirzadi et al., 2008). Trifluralin comprises relatively low water solubility but is highly toxic to fish (LC50=0.089 mg/L) and other aquatic organisms; it is absorbed by soil, resists to leaching, and can persist in soils for months; for instance in soil the half degradation time is DT50=57–126 days (Cooke et al, 2004).

The second phase of study concluded that scarce data on the use and emissions to the environment of individual pesticides exist at European level, and Trifluralin is no exception (Pistocchi, 2009). Presently, the only possibility to map the pesticides emissions in Europe relies on data available for the pesticides usage by chemical classes with reference to the years 1992-2003 (EUROSTAT, 2007). Updated information on pesticide emissions is expected to become available onwards 2013 after the implementation of the Framework Directive on the Sustainable Use of Pesticides (2009/128/EC) and the Regulation on the Statistics of Plant Protection Products (185/2009 of the European Parliament and the Council).

For that reason Pistocchi et al. (2009) proposed a method, based on the EUROSTAT data and the Corine Land Cover 2000 database (http://terrestrial.eionet.europa.eu/CLC2000), to estimate emissions of the individual substances, assuming that when a chemical class is utilized, then it is likely that only a single substance from that class is used everywhere across Europe as a representative for entire chemical class.

5.2 Scenarios for emissions

Appling the scheme described above and assuming that the contribution of the long-range atmospheric transport or UWWTP effluents are negligible sources of Trifluralin pollution, the study of Trifluralin load to sea focuses only on soil environmental compartment (Pistocchi et al., 2009).

Then, firstly the **BAU** scenario with a baseline year 2003 is formulated considering the above described approximation for Trifluralin emissions to soil. Obviously, this approach leads to a systematic overestimation of the use of any plant protection chemical; the more chemicals the class of substances includes, the larger will be the overestimation.

As for the dinitroaniline group of herbicides (8 chemicals including Trifluralin) EUROSTAT (2007) reports the use of 6174 tons in EU25 for 2003. An example map of spatial distribution of Trifluralin emissions to cultivated soil in 2003 is presented in the Figure 14. From this map it could be derived that an average amount of 1.56 kg/km^2/y of Trifluralin (range from 0.01 to more than 20 kg/km^2/y; the standard deviation equals to 3) is applied to arable crop land in EU25.

Apart from the BAU scenario, similarly to the Lindane case, the study investigates on annual basis, the impact of the partial **effectiveness** of legislative measures in terms to assess by which percentage the reference soil emissions should be reduced in order to ensure that the **annual sea load remains below a given limit** assumed to be one third of the estimated BAU load.

Furthermore, within complete **ban** scenario, the soil emissions are supposed to be dropped, in the period 2004-2010, from the typical BAU applications referenced for 2003 to a provisional small amount of 0.005 kg/km^2/y taken as an approximation towards zero emissions from 2011 onwards.

Lastly, despite of the high uncertainty in Trifluralin emissions, it is performed an additional specific exercise, called soil "**cleaning-up**", in order to assess the time period in terms of months, required for cleaning-up Trifluralin from European soil after the complete ceasing of the emissions to soil.

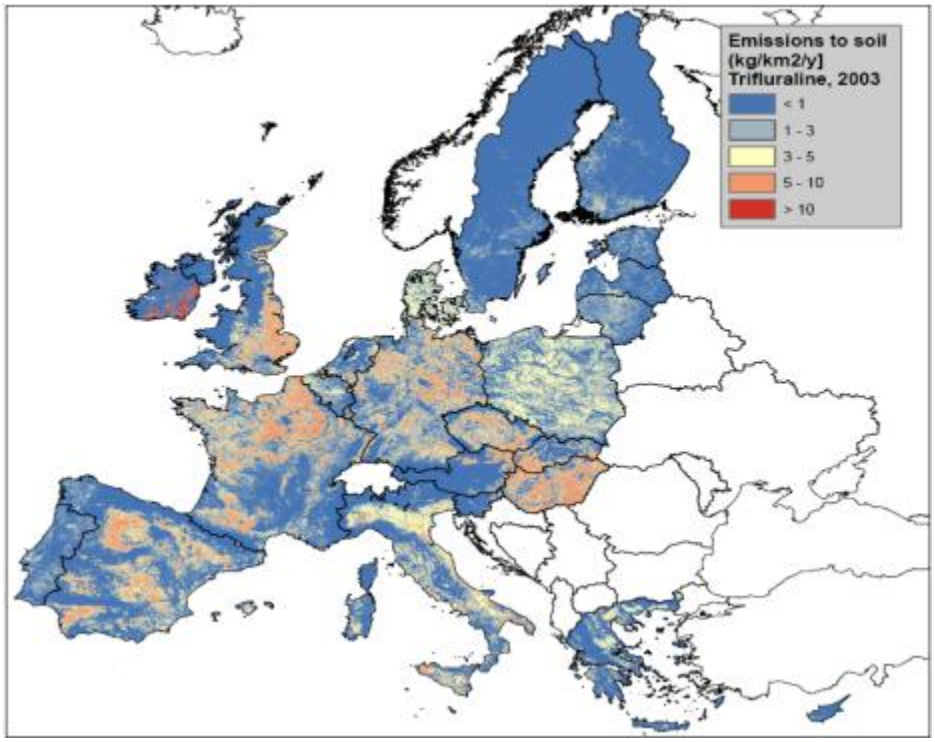


Figure 14 - Use of dinitroaniline herbicides in 2003 by EU25 considered as Trifluralin soil emissions in BAU scenario (an average use of 1.56 kg/km^2/y with a range from 0.01 to more than 20 kg/km^2/y, standard deviation=3).

5.3 Description of modelling tools for Trifluralin scenarios

Potentially, the Trifluralin scenarios could be run by means of the MAPPE model. However, to estimate the Trifluralin sea load, in reality only the soil part of MAPPE is needed in order to screen the Trifluralin mass in soil and the consecutive diffusion through run-off to streams and rivers (Pistocchi et al., 2009).

Actually, other authors also applied similar approach to study the spatial distribution of pesticides in the environment. For instance, Verro et al. (2002) used an integrated daily step model and GIS to predict the spatial distribution of pesticides in Lombardy region of Italy. Tiktak et al. (2004) applied a distributed model to compute the pesticides leaching to groundwater for Europe, while Schriever and Liess (2007) developed an indicator based on the runoff risk aiming to screen the pesticide pressure in Europe. Therefore, in the case of Trifluralin we decided to use a simplified version of MAPPE model which is based only on the soil mass balance equations and using built-in GIS functionalities.

In fact, the simplified version of MAPPE model, needed to run scenarios for Trifluralin, is already described in Pistocchi (2008) and Pistocchi and Bidoglio (2010). Thus, here we focus more on the details about the transient box modeling tool developed to produce average estimates of the sea loads in a faster way. However, even that the spatial and box models for Trifluralin are build on the same equations, the important difference between them is the lack of spatial variability in the latter as typical for box models.

Indeed, since the usage of Trifluralin as a plant protective substance and its physical-chemical properties ($\log K_{aw}$=-2.98 and $\log K_{ow}$=5.34) e.g. affinity to stay in soil and eventually to be discharged to surface water in dissolved or particulate phase through the diffusion runoff and its lower degradability(K_{deg_soil}= 0.009d^{-1} and K_{deg_water}=0.009d^{-1}) in particular in soil and water environment, it is possible to quantify the sea discharge of Trifluralin using only a transient soil model in a form of spatial or a single box models (Pistocchi et al., 2009). This makes the simplified soil model schemes very valuable for screening level assessments (Pistocchi et al., 2009).

Under these assumptions, the annual average mass M_{soil} [ML^{-2}] of a pesticide in soil, following a pulse emission in a generic year, can be computed as:

$$M_{soil}(t) = M(t-1)\exp(-Kt) + \frac{(1-\alpha)E_{soil}(1-\exp(-Kt))}{Kt}$$

where E_{soil} [$ML^{-2}T^{-1}$] is net pesticide emission to soil, assuming one single instant application during the year, α [-] is the portion of emissions drifted by wind or dripping from distribution equipment directly to surface water, K [T^{-1}] is the overall removal rate of a given pesticide in soils assumed to be constant throughout the year, and t is the averaging time period (one year in the present scenarios).

In the report the overall removal rate *K* of Trifluralin is considered as the sum of degradation, washout (runoff and leaching), erosion and volatilization; the range of variability of *K* was found, as in the case of Lindane, using MAPPE model: median=0.015d^{-1}; min=0.003d^{-1} and max=0.08d^{-1}.

Besides, assuming an equilibrium partitioning of the chemical between solid, liquid and gas fractions, the annual average concentration of chemical in liquid C_{liq} [ML^{-3}] and solid C_{sol} [ML^{-3}] phases, as a function of mass in the soil were calculated as:

$$C_{liq}(t) = \frac{M_{soil}(t)\theta}{h\,(K_d\,\rho + \theta + (\varphi - \theta)K_{aw})} \qquad C_{sol}(t) = \frac{M_{soil}(t)K_d\rho}{h\,(K_d\,\rho + \theta + (\varphi - \theta)K_{aw})}$$

where *h* is the soil thickness [L]; K_d = solid-liquid distribution coefficient [$M^{-1}L^3$]; ρ = soil bulk density [ML^{-3}]; θ = soil water content [-]; φ = soil porosity [-]; and K_{aw} is the none dimensional Henry's constant [-].

For screening purposes, the solid fraction is usually not considered in the quantification of chemical load to sea because at continental extent the sediments are slowly transported by the surface water (Pistocchi, 2009). Furthermore, the parameters *h*, θ and φ are assumed to be constant in time and space - *h*=0.3m, ρ=1400kg/m^3, θ=0.2 and φ=0.4 (Pistocchi et al, 2009). The drifted portion of soil emissions is usually accepted to be ca. 1% (Pistocchi, 2009). In the absence of more specific information, K_d is often represented as proportional to the octanol-water partition coefficient K_{ow} and the organic carbon *OC* [MM^{-1}] or organic matter content of soil (an average value of 20 g/kg was assumed for Europe), thus K_d=0.41x*OC*xK_{ow}.

Then, the Trifluralin discharge to stream-river network [$ML^{-2}T^{-1}$] and the consequent load to European seas [$ML^{-2}T^{-1}$], is simply the sum of liquid discharge from cultivated soil plus the wind-drifted part of the soil emissions, considered as additional emissions to surface water:

$$Load(t) = (C_{liq}(t) * Q + \alpha E_{soil}) * Area_{cropland}$$

where Q [mm/y] is the annual runoff from soil (median=150 mm/y; a range 61.5-920) and the area of cropped land is estimated to be ca. 1.256 millions square kilometers for Europe.

5.4 Results and discussion of Trifluralin scenarios

Under BAU scenario the spatial distribution of Trifluralin mass in soil and its diffusive discharge to river network, as shown in the Figure 15 and Figure 16, are calculated based on the approximate estimates for soil emissions during 2003. For instance, the average mass of Trifluralin in soil is 0.28 [kg/km^2] (standard deviation=0.58) while the diffusive discharge is characterized by mean value of 0.019 [kg/km^2/y] (standard deviation=0.042). Then, the riverine load is considered as a sum of diffusion runoff from soil and the additional emissions to the surface water, supposed to be equal to a loss of 1% from the amount emitted to the soil. As was expected the surface water discharge follows entirely the pattern of spatial distribution of Trifluralin application to the soil.

The spatial GIS model for Trifluralin also allows the calculation of disaggregated load to European seas as shown in the Figure 17 and Table 5. As can be seen the total sea load is estimated to be 61.74 tons under BAU scenario conditions, however, keep in mind that we considered as input data for Trifluralin the emissions for the entire class of dinitroaniline herbicides.

Besides, according to the presently available emission data, the European coastal zone of Atlantic ocean and North sea (see Table 5**Error! Reference source not found.**) receive the higher fractions of the European total sea load – 29.5 % and ca. 22%., respectively, followed by Baltic (19%) and Mediterranean (17.5%) seas. However, it worth stressing again that these estimates are built on incomplete emission inventory considering only EU25 countries.

In addition, the results from the specific scenario performed to identify the conditions supporting the sea load of Trifluralin below the hypothetical limit of 20.52 tons per year (assumed to be one third of the BAU load) are presented in Table 5, Figure 18 and Figure 19, respectively. Under this scenario, the average mass of Trifluralin in soil is 0.09 [kg/km^2] (standard deviation=0.27). The scenario indicates that the total sea load of Trifluralin will not exceed 21 tons per year when the BAU level of the emissions to soil are reduced at least by 66% across the EU25 states.

Furthermore, the technique of GIS spatial analyses was used to quantify the soil "cleaning up" in terms of time needed to reduce the pollution caused by the eventual accumulation of Trifluralin in the soil matrix. In this exercise the initial concentration of Trifluralin corresponds to the soil emissions in 2003. The obtained results are presented in the Table 6 according to the frequency of spatial distribution of different classes of Trifluralin mass in soil as a percentage of the total crop land in Europe. Then, the variation of the percentages of the concentration classes with the time in terms of months is giving an idea about the "cleaning-up" process. In general, the results show a fast Trifluralin elimination since 7 months after the application practically more than 99% of the European crop land is characterized by mass below the threshold value of 0.5 kg/km^2 (three times less than the average use of Trifluralin in BAU scenario).

To this end, the spatial model estimated 0.074 t/y of sea load of Trifluralin (see Figure 20) when the ban scenario was enforced. In this scenario, the soil emissions are supposed to be dropped in the period 2004-2010 from the typical BAU application of Trifluralin to a provisional amount of 0.005 kg/km^2/y taken as an approximation towards zero emissions from 2011 onwards.

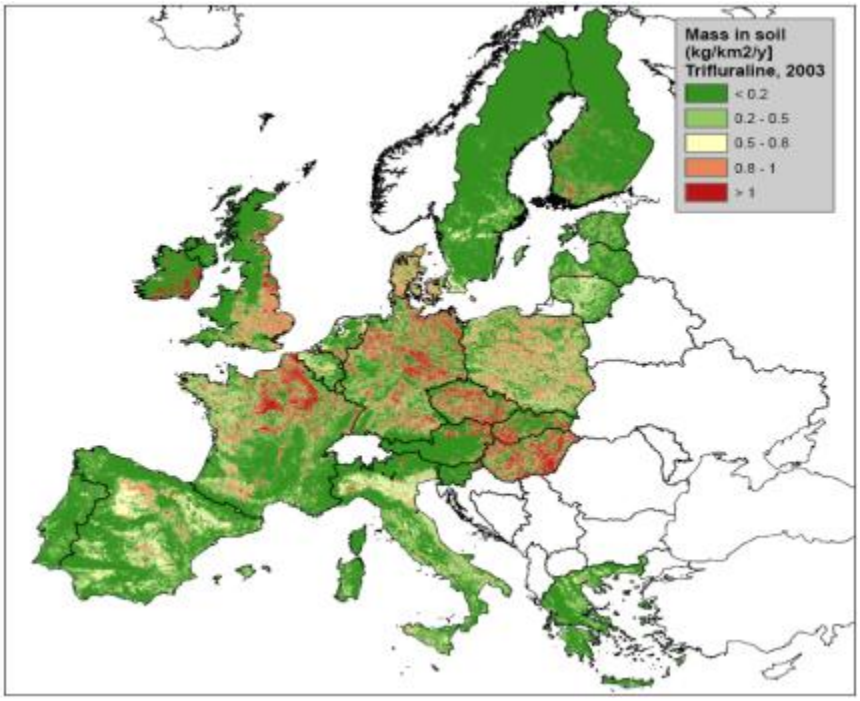


Figure 15 – Modelled results of BAU scenario for Trifluralin mass in soil [kg/km^2] with a reference conditions from year 2003.

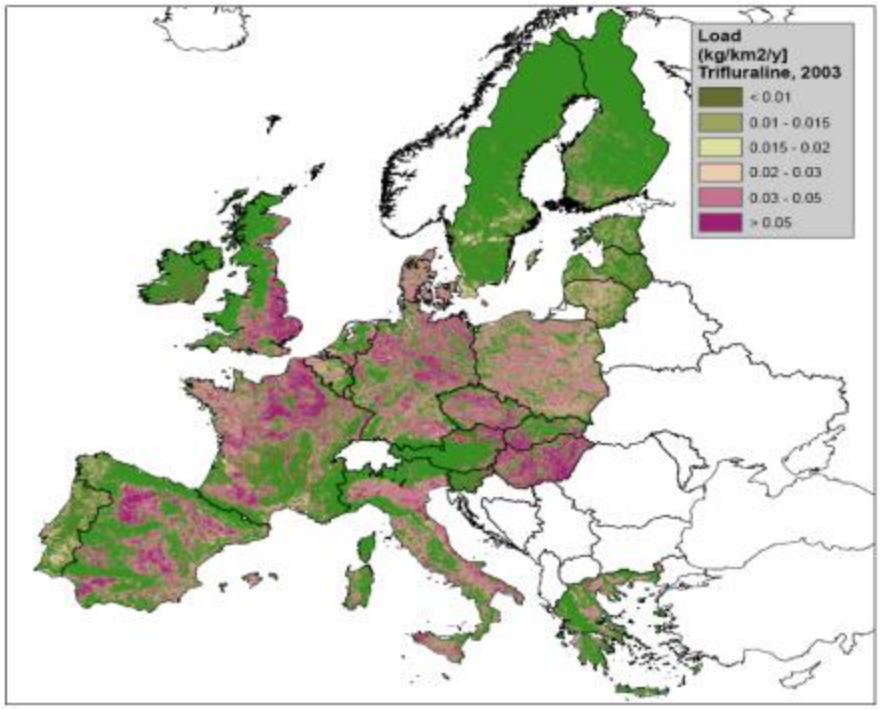


Figure 16 – Modelled results of BAU scenario for discharge of Trifluralin to surface water [kg/km2/y] with a reference conditions from year 2003 as a sum of diffusion runoff from soil and additional emissions to surface water supposed to be equal to 1% loss from soil emissions.

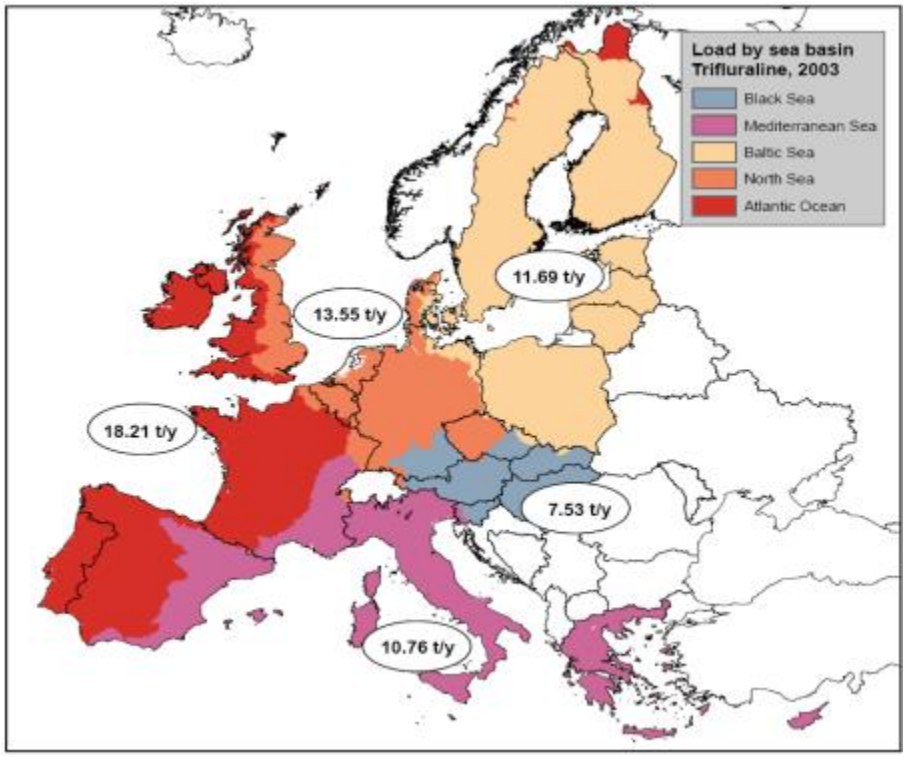


Figure 17 – Modelled results about the annual load of Trifluralin by a sea (in total 61.74 t/y) under BAU scenario with a baseline of 2003.

Load of Trifluralin to Europan seas [tons/year]	Scenarios			
	BAU		66% reduction	
	[tons per year]	% of the total	[tons per year]	% of the total
Atlantic ocean	18.21	29.50	6.05	29.48
Baltic sea	11.69	18.94	3.89	18.96
Black sea	7.53	12.19	2.5	12.18
Mediterranean sea	10.76	17.42	3.58	17.45
Northern sea	13.55	21.95	4.5	21.93
Total	61.74	-	20.52	-

Table 5 – Modelled annual load of Trifluralin to European seas under BAU and "66% emission reduction" scenarios. The model estimates are built on an incomplete emission inventory considering only EU25 countries and use as an input the emissions for the entire class of dinitroaniline herbicides from EUROSTAT data.

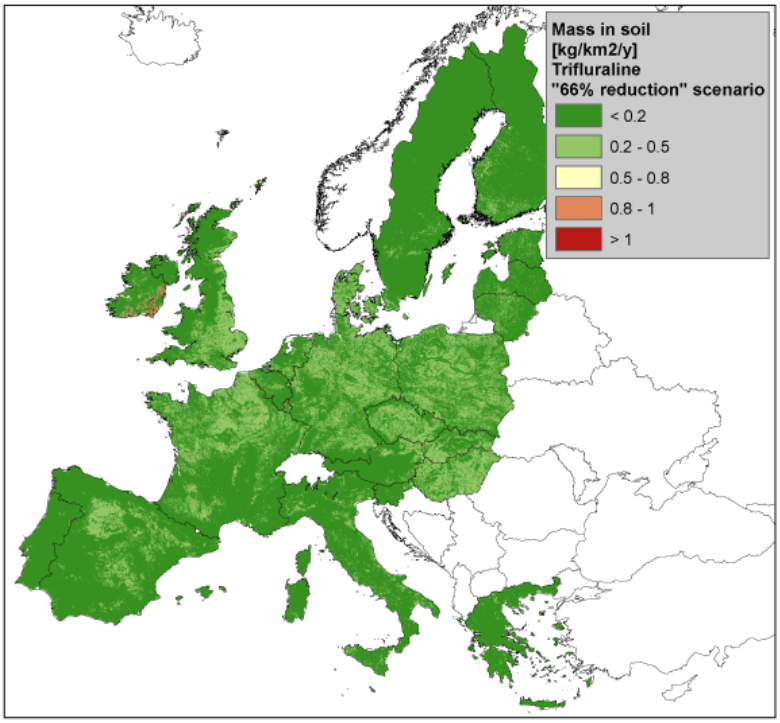


Figure 18 – Modelled results for Trifluralin mass in soil under the specific scenario assuming 66% reduction of BAU level of the emissions to soil across

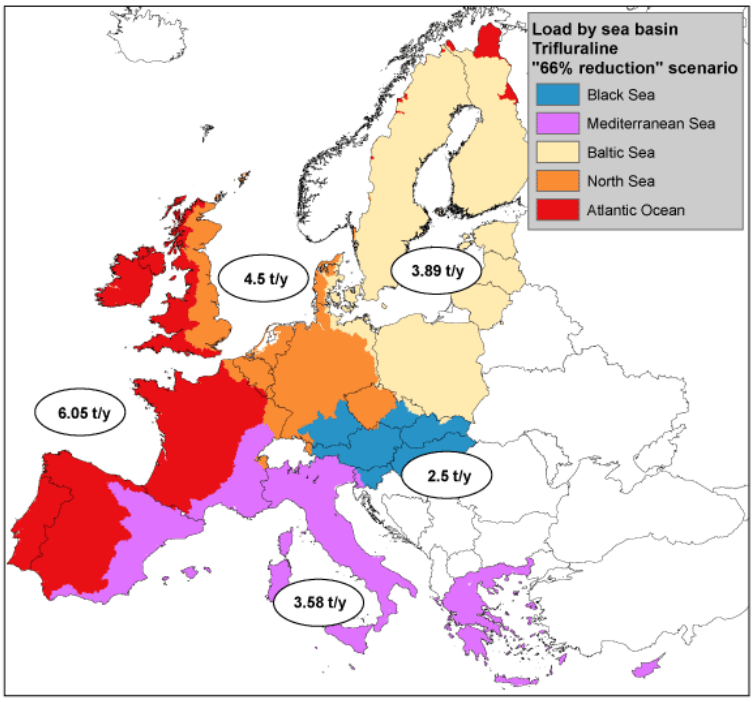


Figure 19 - Modelled results for Trifluralin load to seas (in total 20.52 t/y) under the specific scenario assuming 66% reduction of BAU level of the emissions to soil across Europe.

Classes of Trifluralin mass in soil [kg/km^2]	Frequency of spatial distribution as a percentage from total crop land [%]							
	emissions to soil	after month(s)						
		01	02	03	04	05	06	07
< 0.5	51.47	54.52	58.56	64.66	73.59	83.08	93.95	99.19
0.5 - 1	6.25	6.39	9.23	13.86	15.89	15.93	5.71	0.66
1 - 2	9.87	10.83	16.63	18.82	10.18	0.79	0.25	0.12
2 - 3	8.90	10.87	11.76	2.32	0.14	0.10	0.05	0.01
3 - 5	8.79	9.40	3.39	0.10	0.08	0.05	0.01	0.01
> 5	14.71	7.99	0.43	0.24	0.12	0.05	0.02	0.02

Table 6 – Modelled results for temporal variability, in months after application to soil, of different classes of distribution of Trifluralin mass in soil as a percentage of the total crop land in Europe. The initial spreading respects BAU scenario conditions.

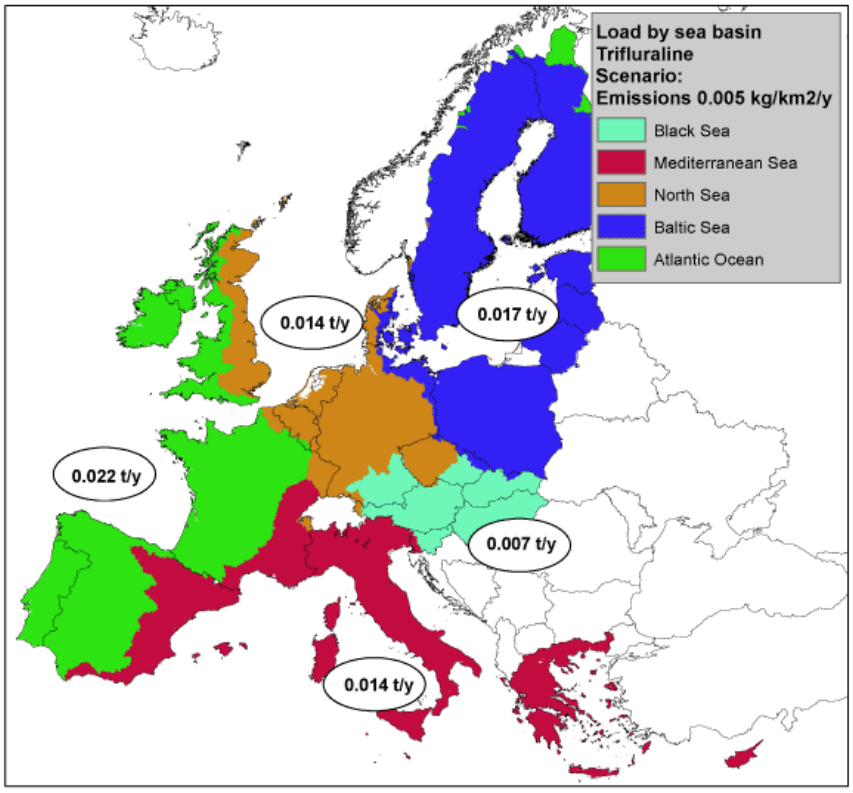


Figure 20 – Modelled estimates about the Trifluralin annual load to European seas (in total 0.074 t/y) when the ban scenario is enforced. In ban scenario the soil emissions are supposed to be dropped in the period 2004-2010 from the typical BAU application of Trifluralin to a provisional amount of 0.005 kg/km^2/y taken as an approximation towards zero emissions from 2011 onwards for each of the EU25 countries.

Besides, the ban scenario indicates that the cease of Trifluralin emissions to soil practically reduces the concern for sea load to negligible levels in a time frame of one year as found with the models used here on the basis of estimates for Trifluralin's persistence (confirmed also by the already described "cleaning-up" exercise).

According to the simplified box model for Trifluralin, it was found that more than 99% of the sea load of Trifluralin is formed by the wind-drifted portion of the soil emissions considered as additional "direct" surface water emissions (confirmed also by the spatial model simulations). Furthermore, since the degradation processes, less than 0.5% of the amount of Trifluralin applied to soil annually contributes to the soil run-off and sea load. For that reason, unlike Lindane case study, it is not possible to see eventual range of sea load depending of the spatial variability of Trifluralin removal rates in Europe.

Additionally, the Trifluralin box model estimates for Europe an average concentration 18.4 ng/L in surface water assuming baseline conditions of 2003 year. In fact, this corresponds to the measurements published for some European rives as follows: a range from 2 to 45 ng/L during 2004 in Spain (Planas et al., 2006); around 30 ng/L in Tuscany surface water and between 20 and 300 ng/L for 2000 in Kalamas river in Greece (Konstantinou et al., 2006); and from 10 to 75 ng/L in the period 2000-2003 in surface water samples taken in northern part of Greece (Vryzas et al., 2009).

As a final point, the box model simulated under the BAU scenario on average 28.2 tons annual sea load of Trifluralin, considering this substance as a representative chemical of dinitroanaline herbicides. The latter represents ca.46% from the estimation found by of the spatial GIS model which confirms again that the box model tends to underestimate at least by half the pesticides sea load.

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6. PFOS scenarios

Perfluorinated compounds are chemicals produced for their non-stick and water repellent properties. They have been used during the last 50 years both in industry and as components of consumer products in the manufacturing of coatings for cookware and clothing, stain resistant carpets, food packaging, firefighting foams, paints, and adhesives, with additional uses in the photo-, electronics-, and aerospace industries.

6.1 Background information and definition of scenarios

During the last years there is a growing concern about Perfluorinated compounds, including Perfluorooctane Sulfonate (PFOS), because they are globally distributed, environmentally persistent, bioaccumulative, and potentially harmful (Pistocchi and Loos, 2009; see also the references given inside). For that reason PFOS has been listed as chemical for regulation within the Stockholm Convention and was banned in the European Union in 2007 for most applications (Directive 2006/122/EC). Contrary to classical POPs, PFOS is primarily emitted to surface water and accumulates in seawater which represents its major and most important final fate sink.

Unlike Lindane and Trifluralin, which are multimedia chemicals, PFOS can be regarded as a single-medium molecule primarily triggered to the water compartment. Its high solubility and its persistence make it a virtually conservative and instantly water-transported substance. The consequence of this behavior is that, while Lindane and Trifluralin can be stored in soil and later released gradually after the ceasing of emissions, PFOS, will be still washed out from old products even its if emissions already stopped.

Moreover, taking into account unknown PFOS emissions at continental scale, what becomes useful for decision makers, according to us, is to specify, firstly, the recent emissions and corresponding sea load. In the report this exercise is entitled as **BAU** scenario. This scenario is based on the available in-house observation data with a baseline of 2007 according to the recent pan-European measurement campaign done by the IES of JRC (Loos et al., 2009). Secondly, in addition to BAU scenario, the sea load of PFOS is assessed under variants of emissions considering for example a scenario of **"50% reduction**" or answering the question **"to what extent the emissions have to be cut in order to guarantee that the sea load remains below a given threshold?"**

The present report employs the approach and results of the backward tracking of PFOS's emissions from riverine measurements, as described by Pistocchi and Loos (2009). The latter are used as a basis in a GIS model able to calculate European maps of river water concentrations and PFOS load to seas. Besides, following the assumptions made in the same publication this model application considers the atmospheric deposition of PFOS as negligible diffuse source which could be not accounted.

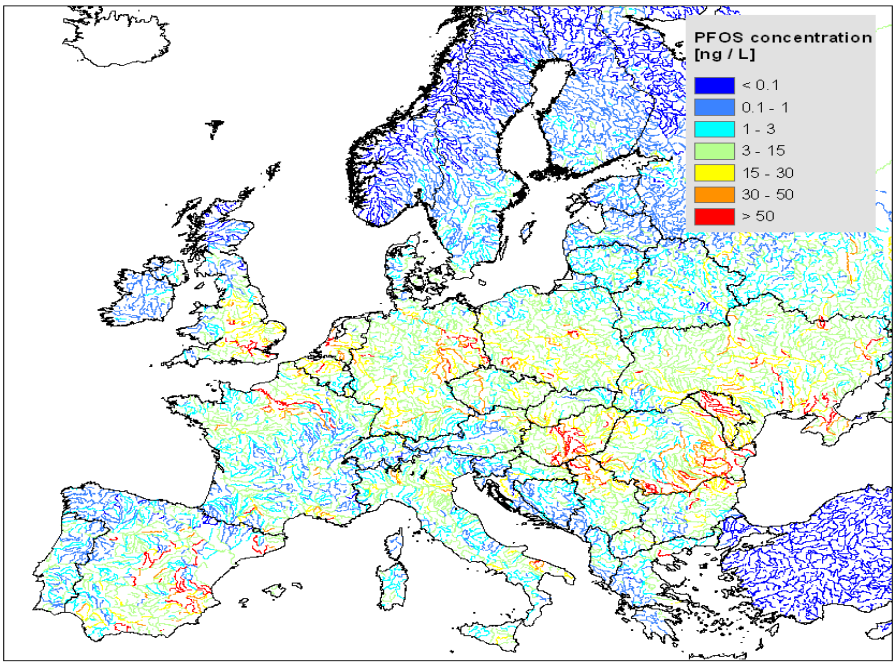
6.2 Results and discussion of PFOS scenarios

Pistocchi and Loos (2009), estimated the aqueous PFOS emissions across Europe using a broad data set of observed river water concentrations (Loos et al., 2009) to ensure a more comprehensive and representative coverage of the European continent. Besides, they reported that primarily the waste water treatment plant effluents are the major cause for PFOS pollution while the "non-point sources" such as urban runoff or atmospheric deposition play relatively minor role.

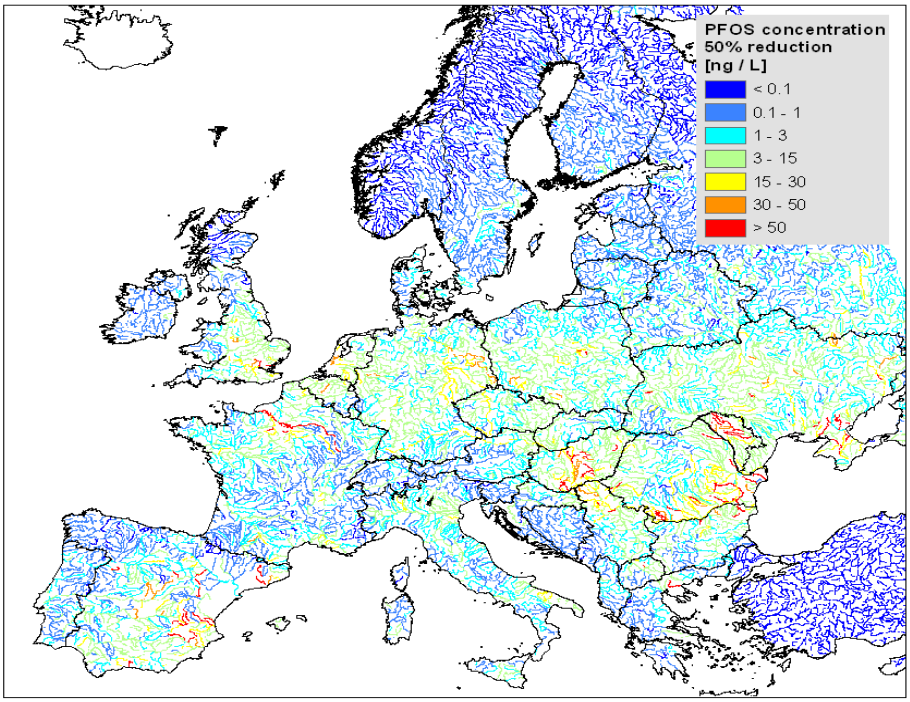
In addition, Pistocchi and Loos (2009) proved that PFOS emissions correlate rather well with river basin population. Thus, for PFOS an average European emission factor of 27.4 µg/day per capita was estimated. The latter is fairly consistent with previously found estimates of 40 µg/day per capita in Bayreuth (Germany), and 57 µg/day per capita in Switzerland. Then, on the basis of the guessed average PFOS emission factor for Europe(Pistocchi and Loos, 2009), we computed maps of concentrations in the European river network.

Under BAU scenario (see Figure 21a), the PFOS concentrations diverge from less than 0.001 to more than 10000 ng/L with an average equals to 7.1 ng/L for Europe (see also Table 7). For example, the model simulates PFOS concentrations for Scheldt River between 0.001-849 ng/L (mean=23, stdv=38), in Rhine River within the range 0-6000 ng/L (mean=12.4, stdv=92) and in Danube river between 0-50000 ng/L (mean=19.6, stdv=203). However, the match between observed and computed concentrations, as shown in the Supporting Information of Pistocchi and Loos (2009), is within a factor of 10, but so big deviations happen not so frequently. Therefore, the accuracy of the developed PFOS model could be considered as satisfactory for the purposes of the present screening application.

In addition, the Figure 21b shows also the improvement of environmental conditions as a result of the application of scenario "50% reduction" of PFOS emissions; not surprisingly the mean value is diminished 2 times to only 3.6 ng/L (see also Table 7).

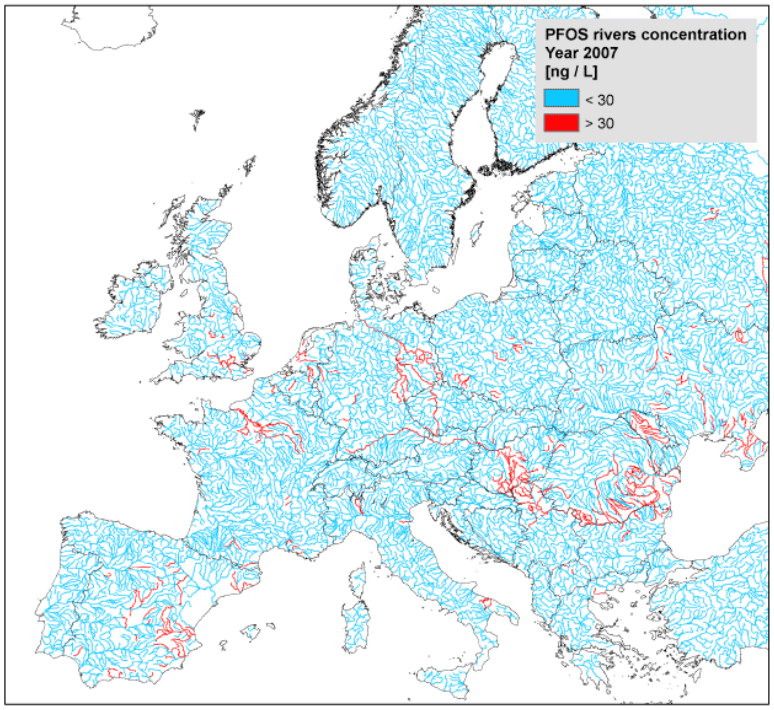


(a) – BAU scenario with baseline of year 2007

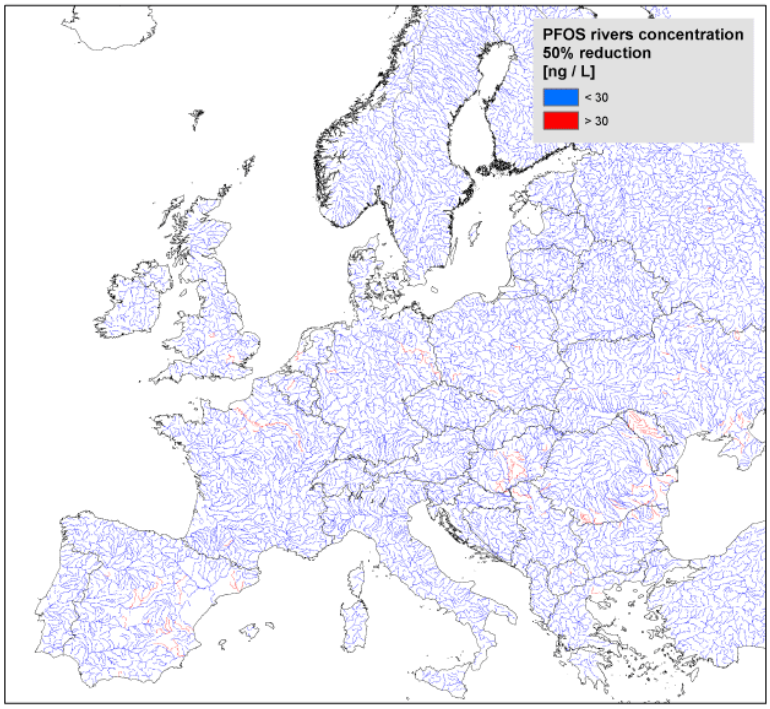


(b) – Scenario "50% reduction" of emissions

Figure 21 – Modelled maps of PFOS river concentrations under different scenarios.

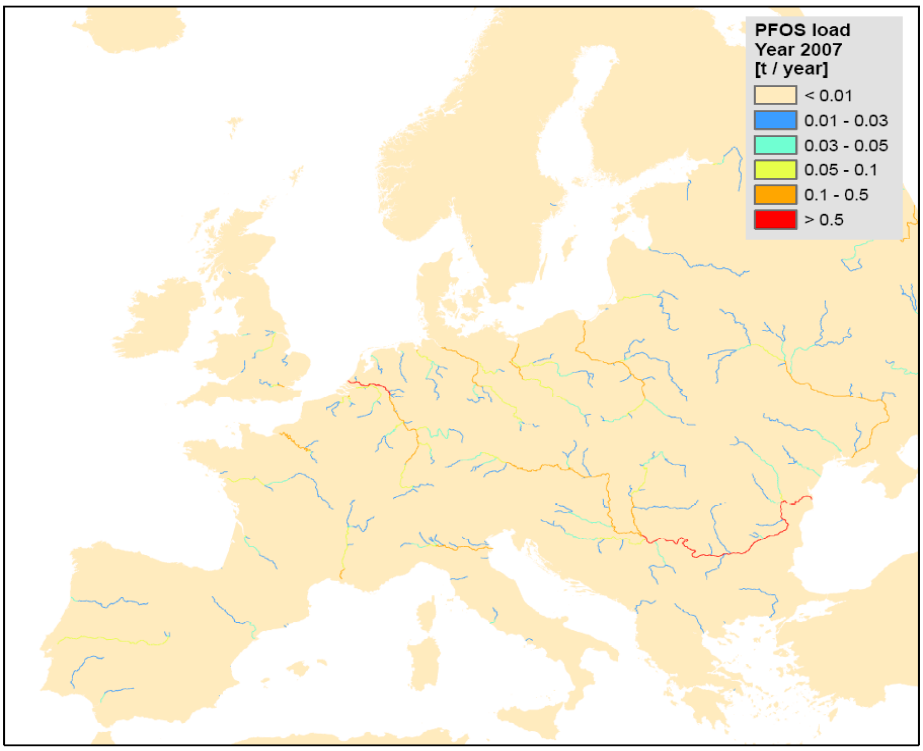


(a) - Under BAU scenario with baseline of 2007 year

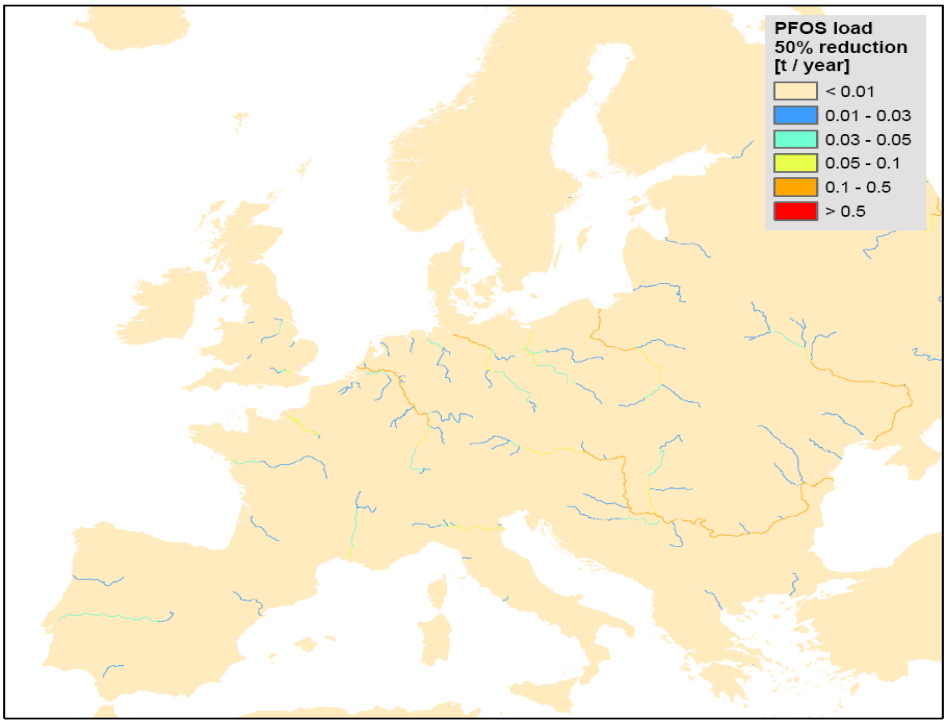


(b) - Under the scenario of "50% reduction" of emissions

Figure 22 – Modelled maps of European rivers expressing PFOS concentrations above warning value of 30 ng/L (specified by a pan-European monitoring campaign; see Loos et al., 2009).



(a) – BAU scenario with baseline of year 2007



(c) – Scenario "50% reduction" of emissions

Figure 23 – Modelled maps of PFOS release to the surface water under different scenarios.

Scenario	Variable	Mean	St.dev.	Range
BAU	Surface water concentration [ng/L] (annual mean)	7.1	113.7	51725.0
	Release to surface water [t/y]	2.7 e-4	9.6 e-3	0.9
50% reduction	Surface water concentration [ng/L] (annual mean)	3.6	56.9	25862.0
	Release to surface water [t/y]	5.2 e-4	9.3 e-3	0.5

Table 7 - Statistics about the mean values, standard deviation and variability range of the simulated PFOS concentrations in European surface water and for the load of PFOS to European seas.

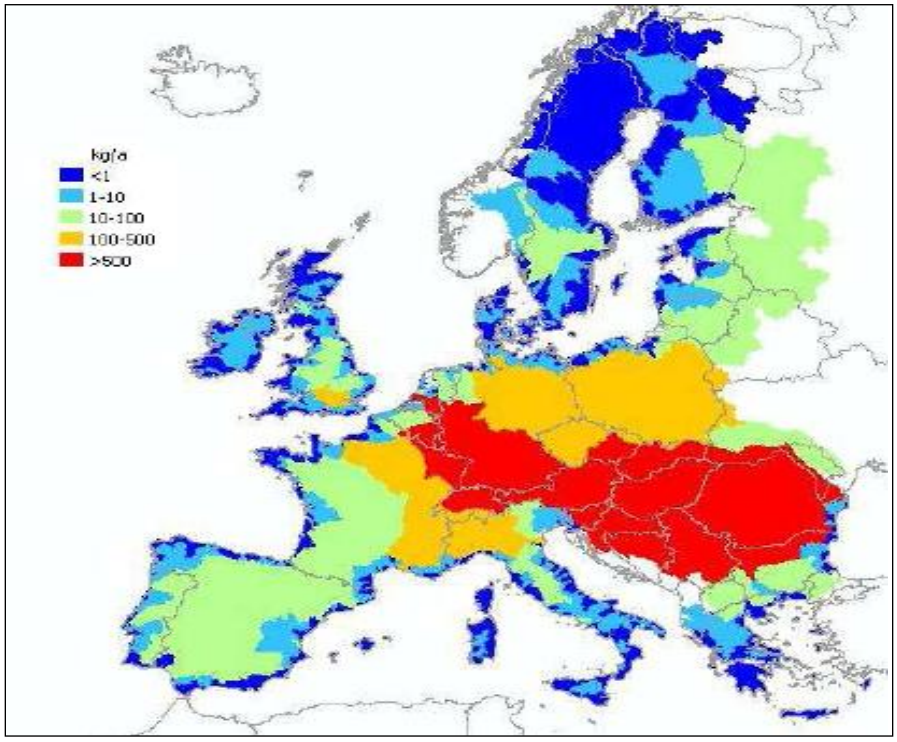


Figure 24 – Modelled map of PFOS sea load [kg/year] by the mainly contributing European river basins.

PFOS annual load to sea	BAU scenario (baseline 2007)	
	[t/y]	% of total
Atlantic ocean	1.240	21.2
Baltic sea	1.117	19.1
Black sea	1.597	27.4
Mediterranean sea	0.853	14.6
North sea	1.033	17.7
Total	**5.840**	**-**

Table 8 - Model estimated annual load of PFOS to European seas under BAU scenario with 2007 as a baseline year.

Besides, an indicative "warning limit" of 30 ng/L has been specified for PFOS on the basis of 90th percentiles of measured sampling concentrations by Loss et al. (2009). Then, the spatial analyses based on the concentration maps, allowed identifying the European rivers most contaminated by PFOS (defined here as the rivers with PFOS concentrations over the warning limit proposed) as shown in the Figure 22. Again, the environmental conditions are getting better beneath the "50% reduction" scenario.

Additionally, the difference between the BAU and "50% reduction" scenarios can be followed for the discharge (load) of PFOS to stream network as illustrated by the maps for Europe provided on the Figure 23. In the case of BAU scenario, the PFOS release to surface water vary in the interval 0.00005-0.9 t/y with an average for Europe of 0.0003 t/y (see also the Table 7).
Subsequently, the riverine loads of PFOS to European seas are presented for the main contributing river basins in the Figure 24 and disaggregated by sea in the Table 8.

It was found that amongst the European rivers Danube (followed by Rhine) delivers the largest amount of PFOS exporting annually more than 1 ton. For this reason, Black Sea is receiving the highest share, ca. 27.4%, from the total PFOS load to European seas.

In turn, the total European sea load of PFOS is estimated being ca. 5.84 tons per year under BAU scenario conditions. As expected a half of this amount will be projected when the "50% reduction" scenario is applied.

The spatial analyses allow also forecasting that the total annual load of PFOS to European seas will decrease below the target value of 1 ton per year only when the current emissions across to European countries are reduced at least by 84%.

Source	Year	River Basin	Country	Surface water Concentration [ng/L]		Load [kg/y]	
				Measurement	Model	Estimates	Model
Loos et al., 2008	2007	Po	Italy	7.52	6.78	211	190
Huset at al., 2008	2006	Glatt	Switzerland	43	11.1	10.4	4.2
Sanchez et al., 2010	2009	Ter	Spain	9.56	5.08	8.07	3.5
Ahrens et al., 2010	2007	Elbe	Germany	5.3	24.4	143	271

Table 9 – Evaluation of MAPPE model results using available data for PFOS surface water concentrations and riverine load to stream network or to seas.

As a final point, the results obtained by MAPPE model have been verified using published data for PFOS surface water concentrations and riverine load to stream network or to seas as provided in Table 9. For example in the case of Po River (Loos et al., 2008) the model forecasts well the PFOS river concentrations (the correlation is R^2=0.42, in total 21 samples) along with the sea load being practically measured at the nearest station to Adriatic Sea. However, as evident from the other comparisons, the model tends to underestimate about 2-3 times the concentration observations and load estimates for Ter (Sanchez et al., 2010) and Glatt (Huset et al., 2008) rivers but overestimates by factor of 2-3 those for Elbe river (Ahrens et al., 2010). Nevertheless, the MAPPE model produces in general total sea load of PFOS arising from European continent in the same range as the estimates for the perfluorinated compounds provided by another modeling exercise including the 14 major European rivers (McLachlan et al., 2007).

7. Conclusions

The present study investigates the possible impacts of different scenarios on the aquatic discharges of three pilot chemicals from land to marine environments in Europe. The reference concentrations and loads have been specified for three test-case contaminants: the insecticide Lindane with reference years 1995 and 2005; the herbicide Trifluralin, considering year 2003 as baseline; and the industrial pollutant Perfluorooctane Sulfonate (PFOS) with 2007 as background year. The specific purpose of the assessments is to characterize for the European continent the fate and spatial distribution of the chemicals of concern and to estimate future chemical loads to the European seas up to the time horizon of 2020. The major problem encountered in the study is the lack of adequate information on extent and location of chemical emissions in Europe.

1. Lindane

The model analyses of scenarios for Lindane show that:

- The comparison with OSPAR data and with the input data used to force a 3D model for North Sea allowed concluding that MAPPE model produced relevant estimates for the riverine load of gamma-HCH to European seas which eventually could differ from the other prognoses for the chemical discharges by not more than a factor of two;
- The European sea load of 745 tons in 1995, based on the official emission data provided by EMEP and on modeling estimations, appears to be reduced by 98.3% in 2005, ten years after the start of the EU regulations for γ-HCH;
- In 2020, under BAU scenario, an annual sea load of ca.12.5 tons is expected;
- The trend and ban scenarios for 2020 estimate a reduction of the annual sea load of Lindane by 74 % and 95 %, respectively, compared to the BAU case;
- Under BAU scenario the European discharges of Lindane affect the Atlantic Ocean (49 % of the total for Europe), Mediterranean (27 %) and Black (19 %) seas; in the case of ban scenario predominantly the Black sea (43 % of the total), followed by Mediterranean (19 %) and Baltic (17%) seas and Atlantic Ocean (16%).

2. Trifluralin

Based on the scenario results produced by the models it was found that:

- According to the BAU scenario, an annual load of Trifluralin of ca.61.7 tons is estimated in 2020. However, it should be kept in mind that this is an overestimation, because the aggregated emission data of EUROSTAT for the agriculture use of the entire group of dinitroaniline herbicides in EU25 have been considered as input data for Trifluralin model in 2003.
- The complete ban scenario forecasts ca. 0.07 t/y and in practice reduces the concern about loads of Trifluralin to European seas to a negligible level in a time-frame of one year due to degradation in soil as found with the models on the basis of data for Trifluralin's persisitance ;
- Under the available EUROSTAT emission data used in BAU scenario, the European coastal zone of the Atlantic Ocean and North Sea receive the higher fractions of the Trufluralin's sea load – 29.5% and 22%, respectively, followed by Baltic (19%) and Mediterranean (17.5%) seas. However, it is worth stressing that these estimates are built on incomplete emission inventory considering only EU25 countries.
- In the partial effectiveness scenario, the total sea load of Trifluralin is expected to decrease to one third of the BAU estimates when Trifluralin's emissions to soil in the EU25 countries across Europe are reduced at least by 66%.

3. PFOS

The analysis of scenarios shows that:

- Based on the average emission factor of 27.4 µg/inhabitant/day and the map of population density in Europe, the concentrations of PFOS in the European surface water under BAU scenario conditions vary from less than 0.001 to more than 10000 ng/L with a mean 7.1 ng/L for Europe;
- The highest sea load of PFOS according to BAU scenario comes from the Danube River (followed by Rhine) exporting annually more than 1 ton. For this reason, the Black sea is receiving the largest share, ca. 27.4%, of the total load of PFOS to European seas;

- Under BAU scenario conditions, the total sea load of PFOS from Europe is estimated to be, on average, 5.84 tons per year. The model forecasts approximately a half of this amount when a 50% reduction of emissions takes place;
- The spatial analyses anticipate that the total annual load of PFOS to European seas will decrease below the value of 1 t per year only when the current emissions across European countries are reduced at least by 84%.

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Abstract

In order to support the implementation of the Marine Strategy Framework Directive, DG Environment and the Joint Research Centre joined to carry out a study on the expected cumulative impact of existing EU environmental legislation on the quality of the marine environment, with specific reference to the case of aquatic discharges to the European seas.

The report describes a few scenario analyses affecting emissions to the European regional seas up to 2020 for Lindane, Trifluralin and Perfluorooctane sulfonate (PFOS) taken as pilot substances. The scenarios developed are agreed with stakeholders at DG ENV following some preparatory meetings. The scenarios do not intend be exhaustive, but examples of what can be further achieved making use of the modelling and database development made in the different phases of the project.

For Lindane, the model estimated European sea load of 745 tons per 1995, based on the official emission data provided by EMEP, appears to be reduced by 98.3% in 2005, ten years after the start of the EU regulations for γ-HCH. Besides, under the BAU scenario, a Lindane sea load of ca.12.5 tons per year should be expected. The trend and ban scenarios support, respectively, a reduction of the load to the European seas in 2020 by 74% and 95% when compared to the BAU estimate.

Aimed at Trifluralin, according to the BAU scenario, an annual load of ca.61.7 tones is estimated in 2020. However, this is an overestimation of sea load, because the aggregated emission data of EUROSTAT for the agriculture use of the entire group of dinitroaniline herbicides in for EU25 have been considered as model input data. The ban scenario forecasts ca. 0.07 t/y and in practice eliminates the concern about loads of Trifluralin to European seas to a negligible level due to degradation in soil.

Considering PFOS under BAU scenario conditions the total sea load from all European countries is estimated to be 5.8 tons per year. The model forecasts approximately a half of this amount when a 50% reduction of emissions takes place.

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