# A comparison of marine radionuclide dispersion models for the Baltic Sea in the frame of IAEA MODARIA program

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A B S T R A C T

Four radionuclide dispersion models have been applied to simulate the transport and distribution of $^{137}Cs$ fallout from Chernobyl accident in the Baltic Sea. Models correspond to two categories: box models and hydrodynamic models which solve water circulation and then an advection/diffusion equation. In all cases, interactions of dissolved radionuclides with suspended matter and bed sediments are included. Model results have been compared with extensive field data obtained from HELCOM database. In-ventories in the water column and seabed, as well as $^{137}Cs$ concentrations along 5 years in water and sediments of several sub-basins of the Baltic, have been used for model comparisons. Values predicted by the models for the target magnitudes are very similar and close to experimental values. Results suggest that some processes are not very relevant for radionuclide transport within the Baltic Sea, for instance the roles of the ice cover and, surprisingly, water stratification. Also, results confirm previous findings concerning multi-model applications.

*Keywords:*
Baltic Sea
$^{137}Cs$
Dispersion
Sediment
Box models
Hydrodynamic models

## 1. Introduction

Models play a major role in the cases of accidental releases of pollutants in order to obtain rapid assessment decision for countermeasures to minimize the potential impact on humans and the environment. Marine models were used to evaluate the transport and dispersion of oil, radionuclides and other pollutants both on short term predictions and also for long-term assessments of the impact to humans by the consumption of marine food as well as for the impact to the environment and biota. In relation to radionuclide dispersion modelling, major international exercises on modelling of transport and transfer of radionuclides in the marine environment were related to deep-sea dumping (CRESP/NEA, 1980s - early '90s; CRESP, 1996), disposal of intermediate and high-level waste in Arctic coastal seas (IASAP/IAEA, 1992–96; IAEA, 2003) and nuclear weapons testing in the South Pacific (Mururoa and Fangataufa assessment/IAEA, 1996–98; IAEA, 1998). During the recent decade several significant developments indicate that a new international modelling exercise can achieve significant progress: new developments in modelling (complex 3-D hydrodynamic models, optimized coding allowing implementation of complex models, techniques involving various scales and deterministic/statistical approaches, ecological modelling, dynamic transfer models etc), improved knowledge of oceanographic and atmospheric drivers, increased database of generic and specific parameters, new knowledge of chemical form-specific biogeochemistry and the effect of environmental change (e.g. ocean acidification) on the fate of radionuclides in the marine environment. The accident at the Fukushima Dai-ichi NPP in Japan in March 2011 resulted in significant releases to the marine environment, which prompted a large interest from modellers world-wide. Tracking contaminated seawater of defined origin can be used as a tool to validate

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oceanographic models and to follow ocean circulation over long periods and distances. Another example was the high deposition of the Chernobyl fallout over the Baltic Sea and the significant contamination of this semi-enclosed brackish sea area. The Baltic Sea is the best investigated and monitored marine environment worldwide. Within HELCOM (Helsinki Convention on the Protection of the Marine Environment of the Baltic Sea Area, www.helcom.fi) the group MORS (Monitoring of Radioactive Substances) established an internationally agreed monitoring network among the Contracting Parties in 1986 and collected all the data in a common data base. Therefore, this sea area would be a good example to test hydrodynamic marine models to simulate the dispersion and behavior of radionuclides.

The MODARIA[2] project, of the International Atomic Energy Agency (IAEA), was initiated to make progress in relation to the assessment of radioactive substances in the environment and its impact to man and biota. Working group 10 was dealing with modelling of marine dispersion and transfer of radionuclides accidentally released from land-based facilities. Different models developed in Member States should be applied to the accidental releases and discharges from the Fukushima Daiichi accident in the Pacific and to the accidental fallout deposition on the Baltic Sea from the Chernobyl nuclear power plant disaster in 1986. The latter case caused a significant long-lasting contamination in this semi-enclosed sea area, primarily with $^{137}$Cs and $^{134}$Cs. While the comparison of different models applied to the Fukushima accidental releases into Japanese coastal waters showed a high variability, four different models applied to the contamination in the Baltic Sea resulted in a fairly consistent pattern of $^{137}$Cs distribution, both in the water column and in the surface sediment layer. The models were of different nature and developed for different purposes.

This paper describes the results of the four models and compares the time and space distribution of $^{137}$Cs after five years of simulation. The objective of this benchmark exercise is to compare predictions and to further develop models for dispersion and transfer of radionuclides in the marine environment, which can be used for radiological and environmental impact assessment in support of decision making in case of accidental releases of radionuclides to the marine environment.

In spite of the large amount of radionuclide data generated for the Baltic Sea, mainly after Chernobyl accident, relatively few modelling studies on radionuclide transport have been carried out for this environment. A one-dimensional vertical dispersion model was used to explain the distribution of Cs isotopes in the water column of the central Bothnian Sea for the first six months after the accident (Ribbe et al., 1991). A box model was latter applied to $^{137}$Cs and $^{90}$Sr by Nielsen (1997). Very local applications within the Baltic Sea have been described by Kumblad et al. (2003) for $^{14}$C and by Erichsen et al. (2013) for isotopes of Ni, Cs and Th. A Lagrangian model was described by Toscano-Jiménez and García-Tenorio (2004), which was applied to Chernobyl $^{137}$Cs deposition over the whole Baltic but on a very limited time window. More recently, Monte (2014) has studied $^{137}$Cs transport with the box model implemented within MOIRA-PLUS decision system. A blind application was first carried out (using model default parameters) and, later, model output was improved by tuning parameters.

Some general information on the Baltic Sea marine system is given in Section 2 and, in the following section, the modelling exercise is described. Finally, results are discussed in Section 4.

## 2. The Baltic Sea

The Baltic is a shallow sea, with maximum and mean depths around 450 m and 50 m respectively, connected to the North Sea through the Danish Straits. A map of the Baltic, indicating locations of interest, is presented in Fig. 1.

Tides in the Baltic Sea are very small, with amplitudes smaller than 5 cm in most of the sea, due to its limited connection to the North Sea (Pugh, 1987). There is an excess of precipitation and river runoff over evaporation in the Baltic. Thus, there is an outflow of fresher, low salinity, water in the surface layer and a deep inflow of more dense water through the Belt Sea around Denmark. This communication is very shallow (sill depth about 18 m) and significant mixing between both water layers occurs. However, this results in a permanent halocline and thermocline in the Baltic Sea, which extend over the different basins. As a result, the average inflow of saline water from the North Sea via the Skagerrak and Kattegat into the Baltic is small (Pickard and Emery, 1982). Actually, there is a high-frequency exchange of water going on all the time, but it has almost no effect on the Baltic Sea, as the same water just goes back and forth. Only during very exceptional conditions influx and salt water intrusion events last long enough (over two weeks) to reach far enough into the Baltic Sea, not receding again. During such significant pulses, the Baltic Sea receives between 200 and 400 cubic kilometres of salty ocean water within a few weeks (Feistel et al., 2008; Matthäus, 2006; Nausch et al., 2014)[3]. Even infrequent pulses are sufficient to keep the Baltic Sea a saline body of water below the permanent halocline. Salinity decreases from the Belt Sea to the Gulf of Finland and the Bothnian Bay. Stratification is reduced with distance from the Baltic Sea entrance. Almost no salinity difference between bottom and surface water is apparent in the most northern areas of the Baltic Sea and the Gulf of Finland.

Prevailing winds in the Baltic Sea are from the west and southwest. Characteristic values of the wind speed 10 m over the sea surface are 8, 6, 5 and 7 m/s for winter, spring, summer and fall respectively (Leppäranta and Myberg, 2009). The annual mean wind speed is about 6 m/s.

The main sources of suspended particulate matter (SPM) into the Baltic Sea are river supply and primary production. Both sources are of the same order of magnitude. About $10^{10}$ kg (10 Tg) of SPM are annually introduced into the sea (HELCOM, 2001). It has been estimated (Toscano-Jiménez, 2013) that the mean SPM concentration in freshwater entering the Baltic from rivers is 20 g/m$^3$. This freshwater is introduced from the main rivers: Neva (2460 m$^3$/s), Vistula (1065 m$^3$/s), Neman (632 m$^3$/s), Oder (573 m$^3$/s), Kemijoki (562 m$^3$/s) and Angermanälven (489 m$^3$/s), where figures correspond to mean flows. But a large number of small rivers, more or less uniformly distributed along the coast must be considered as well. Mean freshwater supplies in several basins are listed in Table 1 (from Toscano-Jiménez, 2013).

In general, SPM concentrations in the Baltic are low and present low seasonal variability. Mean SPM concentrations in the surface layer are presented in Table 1 as well. They have been obtained from Secchi disk measurements (Hakanson et al., 2004). Absolute maximum concentrations (some 10 g/m$^3$) are found in the east of the Gulf of Finland, due to the large discharge of Neva River. Close to the seabed, SPM concentrations are some 1–2 g/m$^3$ larger than in the surface.

The occurrence of man-made radionuclides in the Baltic Sea has four main causes:

[2] Modelling and Data for Radiological Impact Assessments. Further information can be found here: http://www-ns.iaea.org/projects/modaria/default.asp?l=116.

[3] http://www.io-warnemuende.de/tl_files/forschung/meereswissenschaftliche-berichte/mebe93_2014-zustand-hc.pdf.

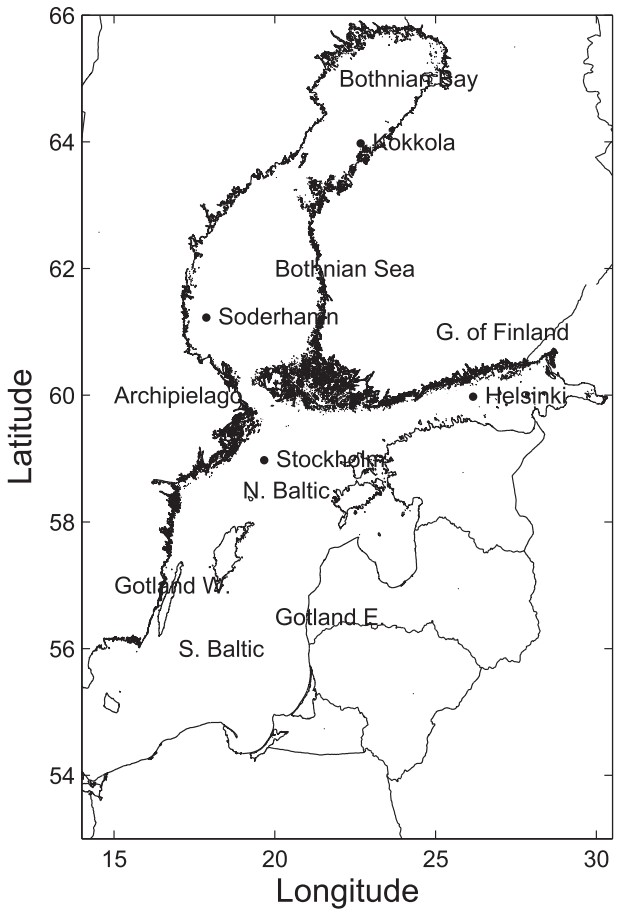


**Fig. 1.** Map of the Baltic Sea showing the different sub-basins considered in this study. Bullets indicate points where time series of radionuclide concentrations are obtained.

- fallout from atmospheric nuclear weapons tests,
- the accident in Chernobyl nuclear power plant in 1986,
- discharges from the two European facilities for reprocessing of spent nuclear fuel, at Sellafield in the UK and La Hague in France, and
- authorized discharges of radioactivity into the Baltic Sea occurring during the routine operation of nuclear installations.

The impact of non-nuclear facilities (e.g. hospitals, industry) on the radioactivity in the Baltic Sea is negligible and very local (Ilus and Ilus, 2000). Dumping of radioactive waste in three dumping sites is also a negligible source (HELCOM, 2013). The total and relative contributions of each source are given in Table 2 (HELCOM, 2013). It is obvious that Chernobyl fallout is the main source, accounting for 83% of the total input. It needs to be mentioned that also $^{134}$Cs was deposited by this fallout with a $^{134}$Cs/$^{137}$ Cs activity ratio of about 0.5, but due to its half-live of only about 2 years, this

**Table 1**
Mean freshwater supplies to several sub-basins (Toscano-Jiménez, 2013) and SPM concentrations (Hakanson et al., 2004).

| Basin | Supply ($m^3$/s) | SPM ($g/m^3$) |
|---|---|---|
| Bothnian Bay | 3104 | 5.0 ± 1.5 |
| Bothnian + Aland Sea | 2860 | 4.5 ± 1.5 |
| Gulf of Finland | 3556 | 5.5 ± 1.5 |
| Gotland + Northern + Southern Baltic | 4630 | 3.0 ± 1.5 |

**Table 2**
Total and relative $^{137}$Cs inputs to the Baltic Sea up to 2010 (HELCOM, 2013).

| Source | Total (TBq) | Relative contribution (%) |
|---|---|---|
| Nuclear weapon tests | 800 | 13 |
| Chernobyl fallout | 4700 | 83 |
| Sellafield and La Hague | 250 | 4 |
| Authorized discharges | 2.4 | 0.04 |

radionuclide was only detectable until the beginning of the 1990s (Nies and Wedekind, 1987).

Deposition from Chernobyl was evenly distributed and the highest contaminated areas were the Gulf of Finland and Bothnian Sea. A map of $^{137}$Cs activities in surface water (sampling depth less than 10 m) in October 1986 is presented in Fig. 2. The distribution also reflects the deposition on land on the drainage area of the Baltic Sea. This map has been constructed from interpolation of measurements (Gritchenko et al., 1989a,b; Nies, 1989).

## 3. Modelling

Models are briefly described in the Appendix. Very different models in structure have been applied. These are two box models (POSEIDON and NRPA model) and a three-dimensional hydrodynamic model accounting for density gradients and incorporating ice thermodynamics (THREETOX model). Finally, an intermediate approach, consisting of a two-dimensional depth-averaged hydrodynamic model forced with annual mean winds (USEV model), has been used as well. Hydrodynamic models calculate the current fields which are used to calculate the transport of radionuclides. An advection/diffusion equations is solved for this purpose, which incorporates additional terms accounting for radionuclide exchanges between water, suspended matter and bed sediments. In the case of box models, water fluxes between boxes are used in the conventional way. The main characteristics of the applied models are summarized in Table 3.

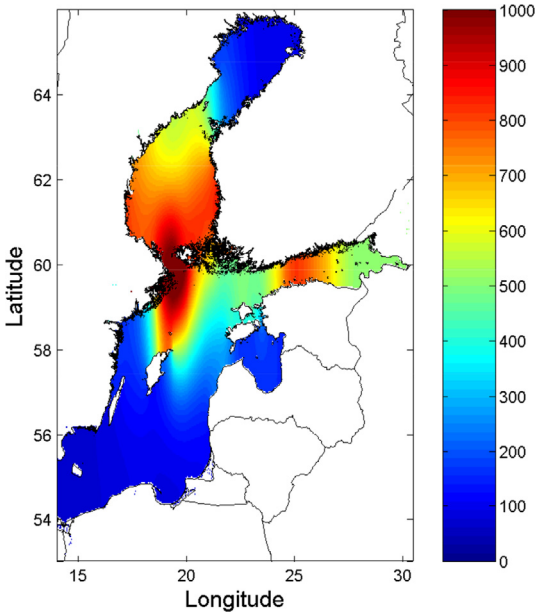


**Fig. 2.** $^{137}$Cs ($Bq/m^3$) in surface water of the Baltic Sea in October 1986 interpolated from measurements.

**Table 3**
Main model characteristics. [1]North Atlantic/Arctic Ocean Sea Ice Model (Karcher and Harms, 2000). [2]Swedish Meteorological and Hydrological Institute. [3]Ocean monitoring and forecasting program (www.myocean.eu). [4]Performed by the European Centre for Medium-Range Weather Forecasts.

| | USEV | THREETOX | NRPA | POSEIDON |
|---|---|---|---|---|
| Spatial resolution | 2 min | 2 min | 10 boxes | 98 boxes including 47 marine boxes and 16 river boxes for the Baltic Sea |
| Vertical levels | 1 | 20 $\sigma$-layers | 3 | 2 |
| Time resolution | 30 min | 16 s | 21.9 h | 1 year |
| Hydrodynamic forcing | Annual mean wind | Open boundary: T, S, water elevation and velocity from MyOcean[3] reanalysis. Atmospheric forcing from ERA[4] Interim, seasonal river discharges | NAOSIM[1] Arctic model (from RISØ) | Averaged currents from SMHI[2], 10 year reanalysis. River discharges |
| Ice dynamics | no | yes | yes for the Arctic, no for Baltic Sea | no |
| SPM | 4.5 mg/l (constant) | Sediment transport model for one fraction with $d_0 = 30$ μm | 1 mg/l (constant) | Averaged values from THREETOX |
| Interactions with solid phases | Kinetic model | Kinetic model | Distribution coefficient (equilibrium) | Distribution coefficient (equilibrium) |
| Biota | no | no | Concentration factor | Dynamic foodweb model |
| $^{137}$Cs source | Chernobyl (Fig. 2) | Chernobyl (Fig. 2) | Chernobyl (Fig. 2). Global deposition. Sellafield/La Hague | Chernobyl (Fig. 2). Global deposition. Sellafield/La Hague |

Models are started about 6 months after Chernobyl deposition and when the first investigation on the fallout distribution in the Baltic Sea was carried out, in October 1986 (Fig. 2; this map has been obtained from measurements in Gritchenko et al., 1989a, 1989b; Nies, 1989). This date is $t$=0, and five year long simulations are carried out. The same information is extracted from all the models to allow intercomparisons and comparisons with field data from HELCOM database. These data are annual $^{137}$Cs concentrations in the water column and bed sediments (mean values over several Baltic Sea sub-basins) and annual inventories in the water column and bed

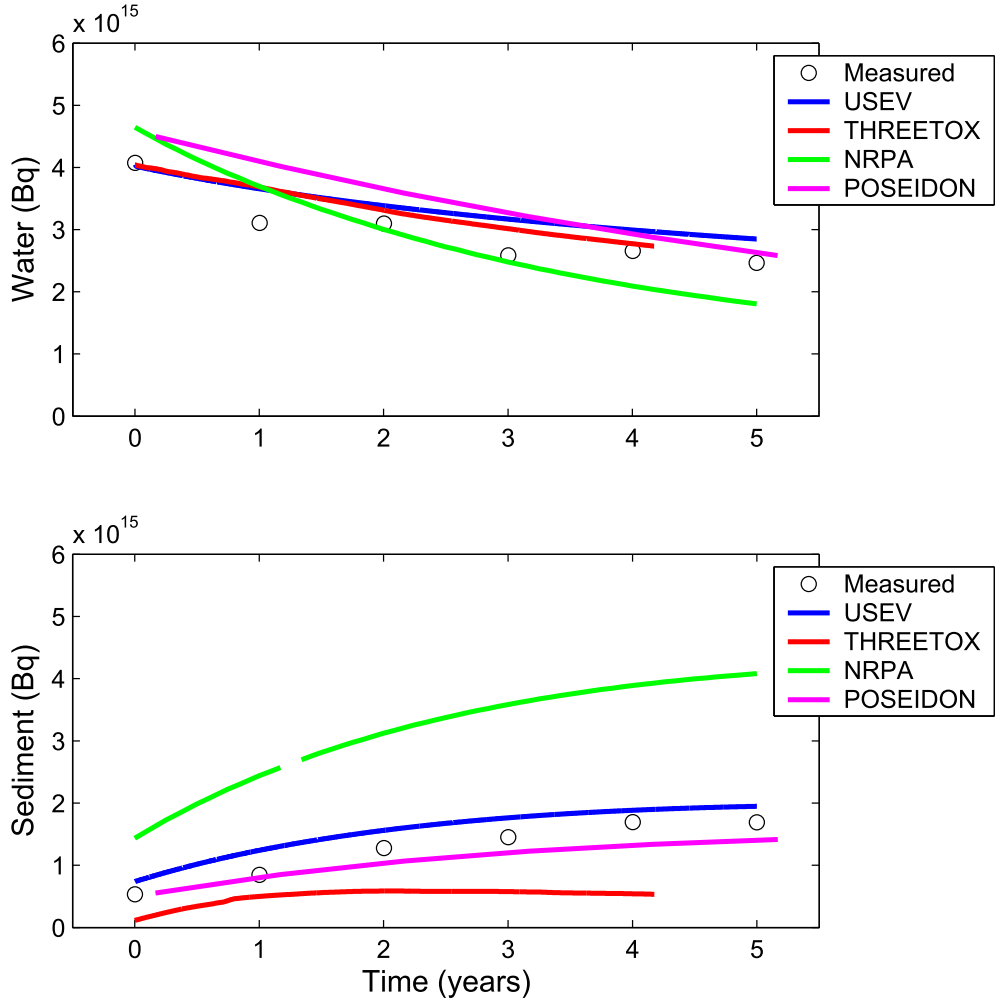


**Fig. 3.** Calculated $^{137}$Cs inventories in water and sediments, as well as values estimated from measurements. The time origin corresponds to October 1986.

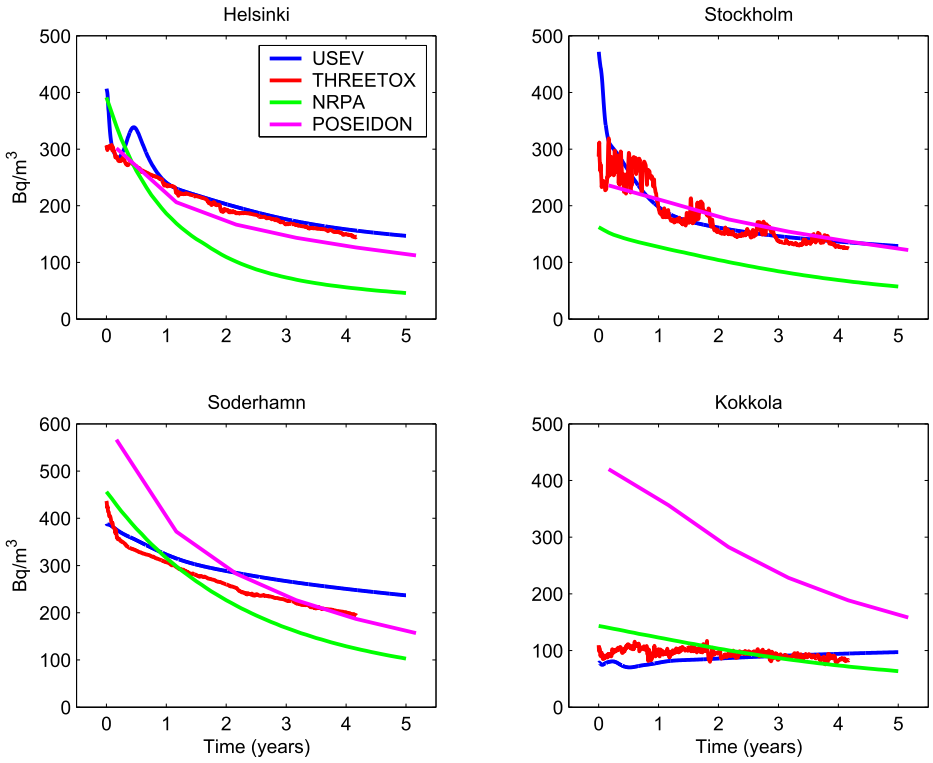


**Fig. 4.** Calculated $^{137}$Cs concentrations in water at points indicated in Fig. 1. The time origin corresponds to October 1986.

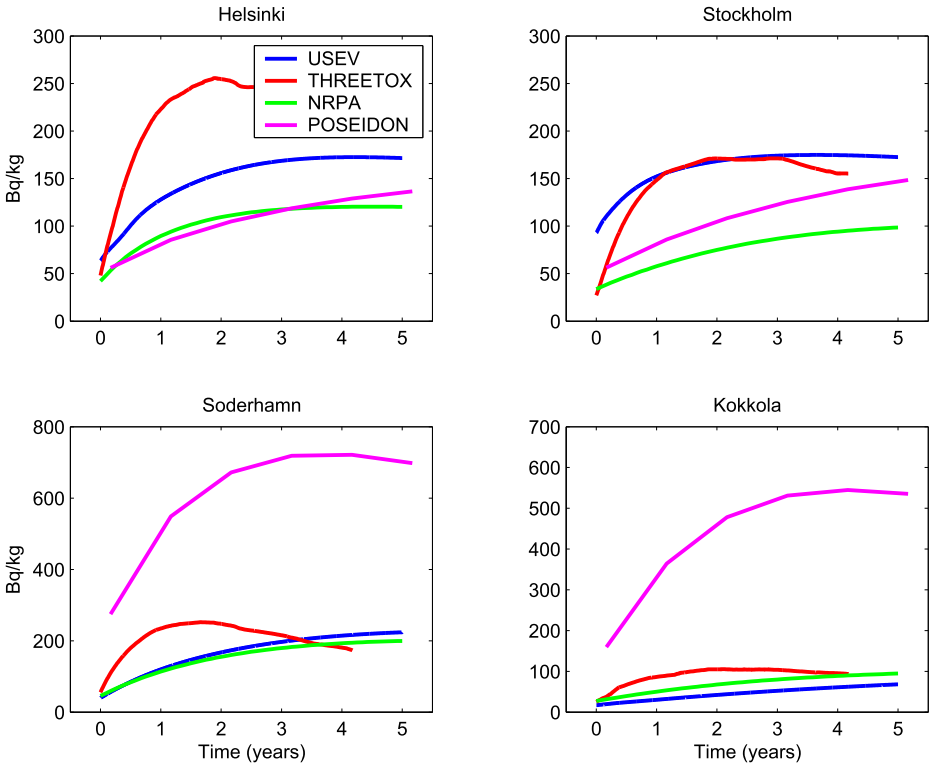


**Fig. 5.** Calculated $^{137}$Cs concentrations in bed sediments at points indicated in Fig. 1. The time origin corresponds to October 1986.

sediments estimated from measurements (Ikäheimonen et al., 2009). Additionally, time series of $^{137}$Cs concentrations in water and bed sediments have been produced for some locations in the Baltic, which are used for further model intercomparisons.

It must be pointed out that no calibration at all was made for POSEIDON, THREETOX and NRPA models. Instead, default parameters were used. Only in the case of USEV model, data on $^{137}$Cs inventories in the water column and seabed were used to

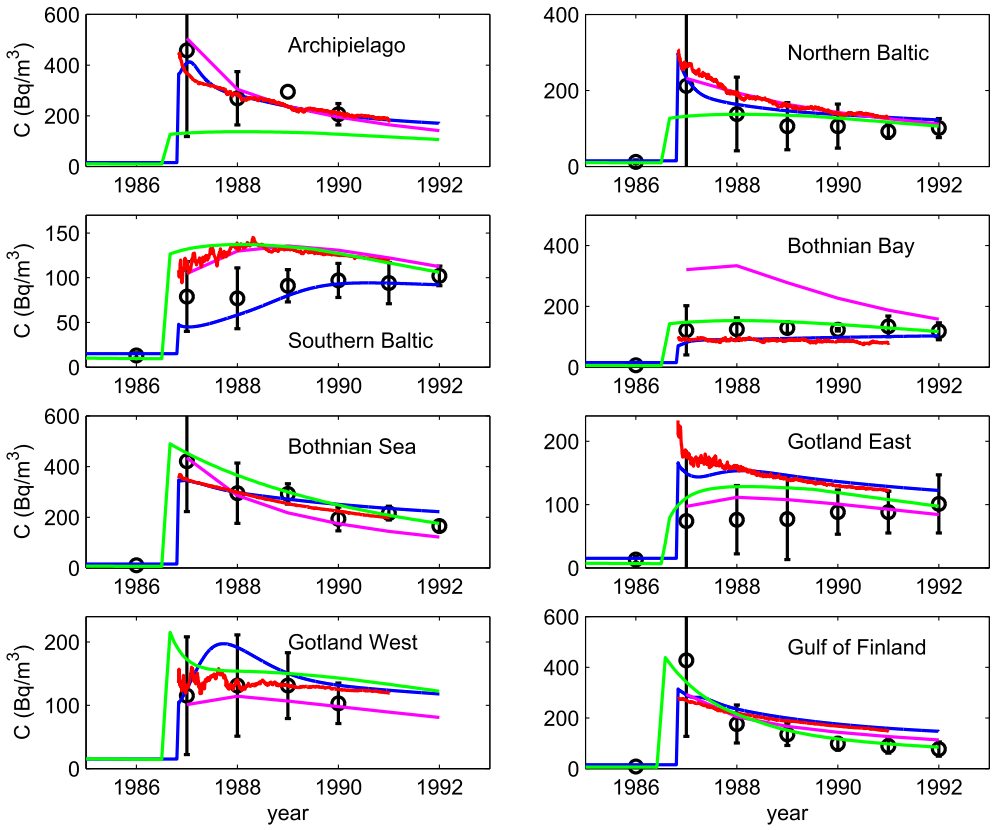


**Fig. 6.** Calculated and measured mean $^{137}$Cs concentrations in the dissolved phase in several basins. Errorbars are 1$\sigma$ standard deviations of the measured mean values. Only one sample was measured if errorbars do not appear. Blue: USEV, magenta: POSEIDON, red: THREETOX, green: NRPA. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

calibrate uptake/release processes, as described in appendix A.4.

## 4. Results and discussion

A comparison of the temporal evolution of $^{137}$Cs inventories in the water column and seabed sediments calculated by the four models is presented in Fig. 3. Values estimated from measurements (Ikäheimonen et al., 2009) are also presented. Water column inventory estimated from measurements decreases as $^{137}$Cs is progressively fixed to bed sediments, which leads to an increase in the seabed inventory. It can seen that calculated temporal trends by all models reproduce the observed behaviour. Moreover, results are very close for all models, specially for the water column.

Calculated time series of $^{137}$Cs concentrations in the water column and in bed sediments, for four points indicated in Fig. 1, are presented in Figs. 4 and 5 respectively. Except in Kokkola, all models predict an exponential decay of concentrations in the water column. Predicted concentrations are also very similar. In Kokkola, there is a difference between hydrodynamic (THREETOX and USEV) and box (NRPA and POSEIDON) models, which may be attributed to the different nature of both model types. While the latter ones again give a concentration decrease, essentially constant (or slightly increasing) $^{137}$Cs concentrations are produced by the hydrodynamic models. In the case of sediments, differences between models are larger than for the dissolved phase. Nevertheless, all models predict essentially the same temporal trends. It is also interesting to note that results from the two hydrodynamic models remain close: maximum differences in the predicted concentrations do not reach a factor 2, being much smaller in some locations.

Fig. 6 shows the predicted $^{137}$Cs concentrations in the dissolved phase in several basins of the Baltic Sea. The calculated results correspond to the mean value for the considered basin (i.e., mean values between all boxes or grid cells which cover the basin). Accordingly, field data (from HELCOM database) are also mean values for each basin and the errorbars are 1$\sigma$ standard deviations of such mean values. Observed temporal trends of $^{137}$Cs concentrations are generally reproduced by all models in all basins. An abrupt concentration increase is produced at the moment of the accident and, from this time on, a decrease in concentrations is observed in most basins due to horizontal and vertical dilution as well as to uptake on suspended matter in the water column. On the long-term, the decrease must be attributed to both the transport of radionuclides to seabed sediments and to the export of radionuclides from the Baltic Sea through the Kattegat and Skagerrak into the North Sea. This leads to slightly enhanced levels of $^{137}$Cs in these areas and in south of Norway (Nies et al., 2009; Michel et al., 2012)[4].

Results for bed sediments are presented in Fig. 7. Measurements have a much larger dispersion than in the case of water samples. Thus, model results generally lie within errorbars. Nevertheless, it is interesting to note that trends produced by all models are very similar. In this case there is an increase in concentrations due to the input of radionuclides from the water column. Moreover, even differences between predicted values are relatively small. Model results in Figs. 6 and 7 are mean values over a number of grid cells or boxes, thus differences between models could be reduced

[4] http://www.bsh.de/de/Meeresdaten/Beobachtungen/Radioaktivitaet/StSch4481Endbericht280709.pdf.

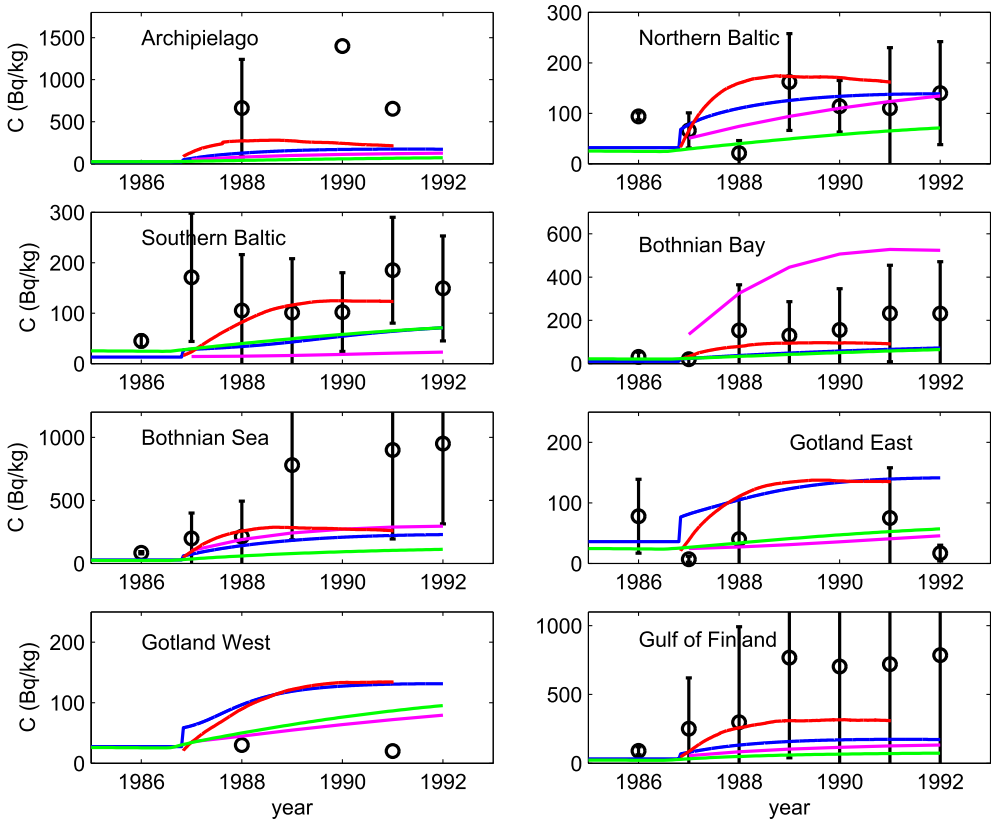


**Fig. 7.** As Fig. 6 but for bed sediments.

because of this averaging process. Nevertheless, differences between models in specific points (time series in Figs. 4 and 5) also remain relatively small. Consequently, we can conclude that effectively there is a considerable agreement between all model predictions.

Maps of calculated concentrations in water and bed sediments, for January 1991, by hydrodynamic models (THREETOX and USEV) are presented in Fig. 8. It is evident that calculated distributions are very similar, both for water and sediments. Only USEV model is producing slightly higher concentrations than THREETOX in the Bothnian Sea. It is of interest to observe how similar the sediment distribution maps are. The high concentrations produced by THREETOX in the eastern Gulf of Finland must be attributed to the high SPM concentrations in this area (see Section 2). USEV model uses an uniform SPM distribution, but THREETOX includes a SPM transport model. Thus these high concentrations are reproduced by TRHEETOX but not by USEV model. A high SPM concentration increases scavenging of radionuclides from the water column to bed sediments.

Models with very different structures (box and hydrodynamic models) have been applied to simulate the dispersion of $^{137}$Cs in the Baltic, including interactions of radionuclides with suspended matter particles and bed sediments. These interactions are also described in different ways: using an equilibrium distribution coefficient or using a dynamic approach. In spite of the differences between the applied models, results are very consistent. Very similar concentrations in water and sediments are predicted for the selected locations and basins, as well as for inventories in the water column and in the sea bed.

It has been stated (Monte et al., 2005, 2006) that a main factor of uncertainty in models is due to the difficulties of representing interactions of dissolved contaminants with the solid phases. In the intercomparison exercise described in Monte et al. (2006) for the Dnieper-Bug estuary, these difficulties did not affect model performances. Indeed, very coherent results were obtained. Such estuary is a relatively energetic environment, with significant currents. Thus, it was suggested (Monte et al., 2006) that due to this relatively fast water dynamics, water/sediment interactions were not significantly affecting transport and dispersion. However, currents in the Baltic Sea are not significantly larger than in the Dnieper-Bug estuary. Moreover, it is an almost closed and shallow water system with several deeper basins. Consequently, interactions of dissolved radionuclides with the solid phases should be significant. Nevertheless, in this environment, water/sediment interactions do not appear to be a significant source of discrepancy between models.

In any case, the Baltic Sea is a very complex marine system, with vertical stratification, significant horizontal density gradients, fresh water supplies and partially covered with ice, in particular in the Northern parts, the Gulf of Bothnia and the Gulf of Finland, during some months each year (which affects not only deposition events taking place during winter, which is not the case, but also has implications on water circulation itself). In spite of this, model results are consistent. Even in the case of hydrodynamic models, the USEV model constitutes a very simple approach in which all these processes are neglected. In contrast, they are included in the complex THREETOX model. Therefore, it can be concluded that they must not play a significant role in the redistribution of contaminants within the Baltic Sea (Fig. 8). Of course, this may not be the case in a different marine area. In addition, given the relatively short simulated times (5 years) and water residence time in the Baltic Sea (some 10–30 years according to Leppäranta and Myberg, 2009), exchanges of radionuclides with the Atlantic Ocean do not play a significant role. While THREETOX and USEV models only include Chernobyl deposition as $^{137}$Cs source (added over a pre-Chernobyl background), Sellafield and La Hague releases are considered in

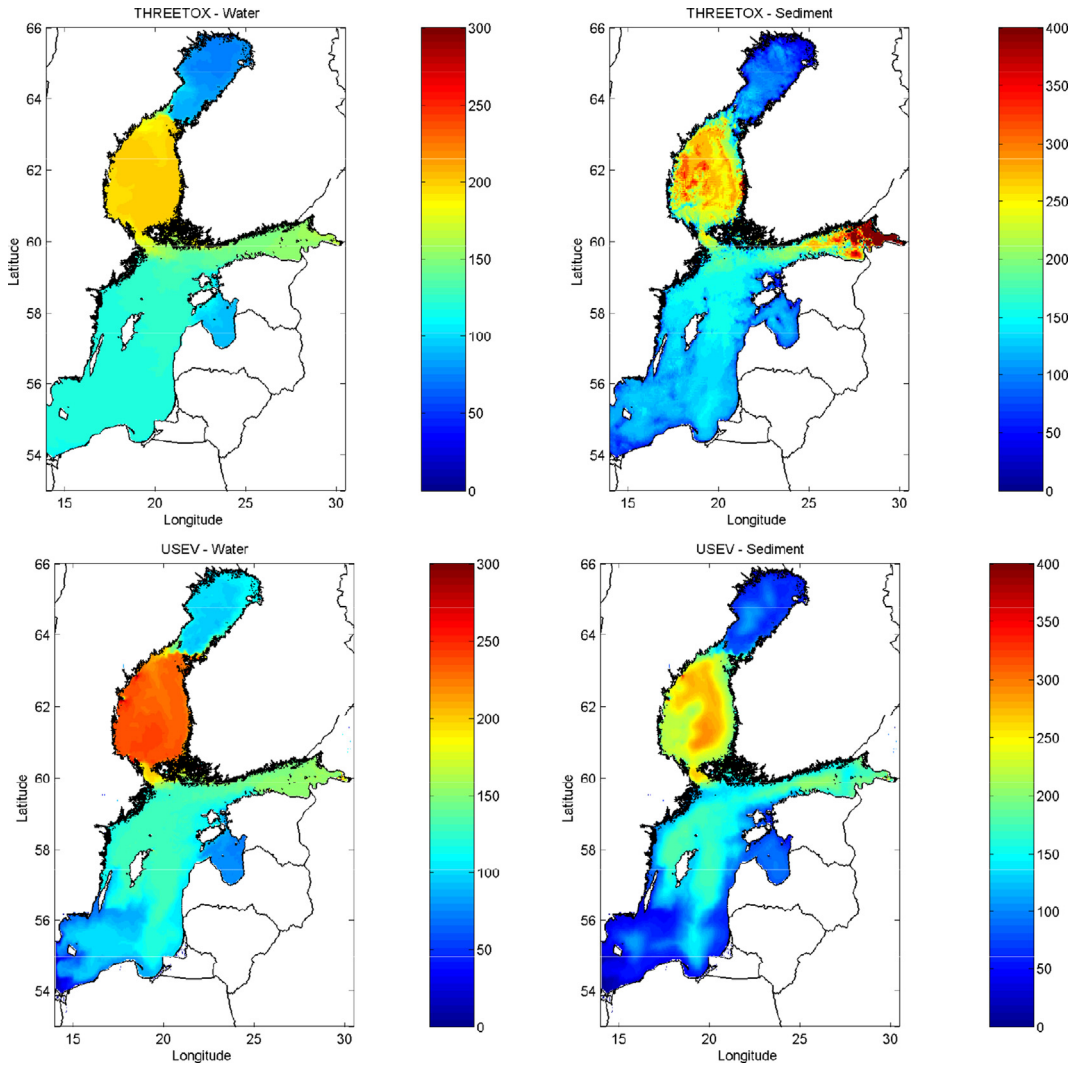


**Fig. 8.** Calculated concentrations for January 1991 in water (Bq/m$^3$) and sediments (Bq/kg) by hydrodynamic models THREETOX and USEV. Although THREETOX domain is slightly larger, maps are drawn over the same area (USEV domain) for a better comparison.

POSEIDON and NRPA models. From intercomparison of model results and comparisons with observations in Figs. 6 and 7, it becomes obvious that Chernobyl fallout is the dominant source, as commented in Section 2.

Significant work has been done by Monte et al. (2006) and Monte (2009) concerning multi-model applications. It has been claimed that, given a certain level of process understanding, different model structures and parameter values can be equally acceptable. Traditionally, it is supposed that an "ideal model" exists. This is an unique model, inherent to nature. Thus, different models are different realizations of the ideal model in view of the specific applications for which they were developed. Consequently, a multi-model approach can be accepted if and only if the different models are developed to solve problems of different kinds, for which different realizations of the ideal model can be appropriate. Monte et al. (2006) have found that this statement cannot be easily supported. Our results confirm this previous finding by Monte and co-workers. Models with very different structures and parameters have been applied to the same environmental problem and no criteria can be found to decide which could be the most appropriate one. In this sense, it is interesting to point out that models may perform differently depending on the target variable. For instance, one model may predict radionuclide concentrations in bed sediments in good agreement with measurements, but it may provide not so good results for the water column. For another model, the situation may be the opposite.

Of course, it should be kept in mind that, for a correct model comparison, the right question should be "asked" to each model. This is particularly relevant when box and hydrodynamic models are compared, and has already been pointed out by Iosjpe and Periáñez (2005): different model approaches can lead to comparable results if these results are extracted in the correct way.

Radionuclide concentrations in the Baltic Sea were available when this study was carried out. Thus, a real blind-test exercise was not possible. However, as commented before, no calibration was made for POSEIDON, THREETOX and NRPA models. Only in the case of USEV model, data on $^{137}$Cs inventories in the water column and seabed were used to calibrate uptake/release processes (appendix A.4). Thus, results of the present exercise should not have been significantly contaminated by the previous knowledge of data.

## 5. Conclusions

Four radionuclide dispersion models have been applied to simulate the transport and distribution of $^{137}$Cs fallout from

Chernobyl accident in the Baltic Sea. Models correspond to two categories: box models and hydrodynamic models. In all cases, interactions of dissolved radionuclides with suspended matter and bed sediments are included. Thus, models are very different in structure and parameters.

Model results have been compared with extensive field data obtained from HELCOM database. Inventories in the water column and seabed, as well as $^{137}$Cs concentrations along 5 years in water and sediments of several sub-basins of the Baltic have been used for model comparisons.

Two main points should be considered: i) the significant conceptual, numerical and parameterization differences between models and ii) the complexity of the Baltic Sea system. In spite of these two points, models results are rather close. Even for bed sediments, which have been recognized as a significant source of model discrepancy. The same temporal trends are predicted by the models for $^{137}$Cs inventories in the water column and sediments and for $^{137}$Cs concentrations in these two phases in a number of sub-basins. Values predicted by the models for the target magnitudes are very similar and close to experimental values. Generally speaking there is an increase in concentrations in bed sediments as radionuclides are scavenged from the water column, where concentrations slowly decrease, as it is obvious.

Results from this exercise suggest that some processes are not very relevant for radionuclide transport within the Baltic Sea, for instance the roles of the ice cover and, surprisingly, water stratification by the halocline and thermocline. It is also clear that Chernobyl fallout is the dominant $^{137}$Cs source into the Baltic Sea.

In addition, results confirm previous findings concerning multimodel applications. Models with very different structures and parameters have been applied to the same environmental problem and no criteria can be found to decide which could be the most appropriate one. Generally speaking, the model to be applied of course depends on the modelling purpose: for instance a fast assessment after an acute accidental release or a long-term radiological study, which could be considered as the two extreme cases. The horizon of the present exercise, i.e. time scale and spatial resolution of results (5 years and sub-basin level respectively), may be considered as an intermediate one. At this level, there is a significant agreement between box and hydrodynamic models for the present scenario. The discrepancy would probably increase as moving towards smaller scales, not properly solved by coarse box models. As moving towards longer time scales and larger domains, the situation can be hardly handled by complex hydrodynamic models, due to computational limitations, and box models might be the best choice.


## Acknowledgement

Work carried out in the frame of IAEA MODARIA (Modelling and Data for Radiological Impact Assessments) program. The authors are indebted to all members of MODARIA working group 10 for useful discussions held during group meetings. Work partially supported by EU FP7 EURATOM project PREPARE: "Innovative integrative tools and platforms to be prepared for radiological emergencies and post-accident response in Europe", Project No. 323287.


## A Model descriptions

### *A.1 NRPA model*

The present model uses a modified approach for compartmental modelling (Iosjpe et al., 2002, 2009; Iosjpe, 2006) which allows for dispersion of radionuclides over time. The box structures for surface, mid-depth and deep water layers have been developed based on the description of polar, Atlantic and deep waters in the Arctic Ocean and the Northern Seas and site-specific information for the boxes generated from the 3D hydrodynamic model NAOSIM (Karcher and Harms, 2000). Such structure is presented in Fig. 9.

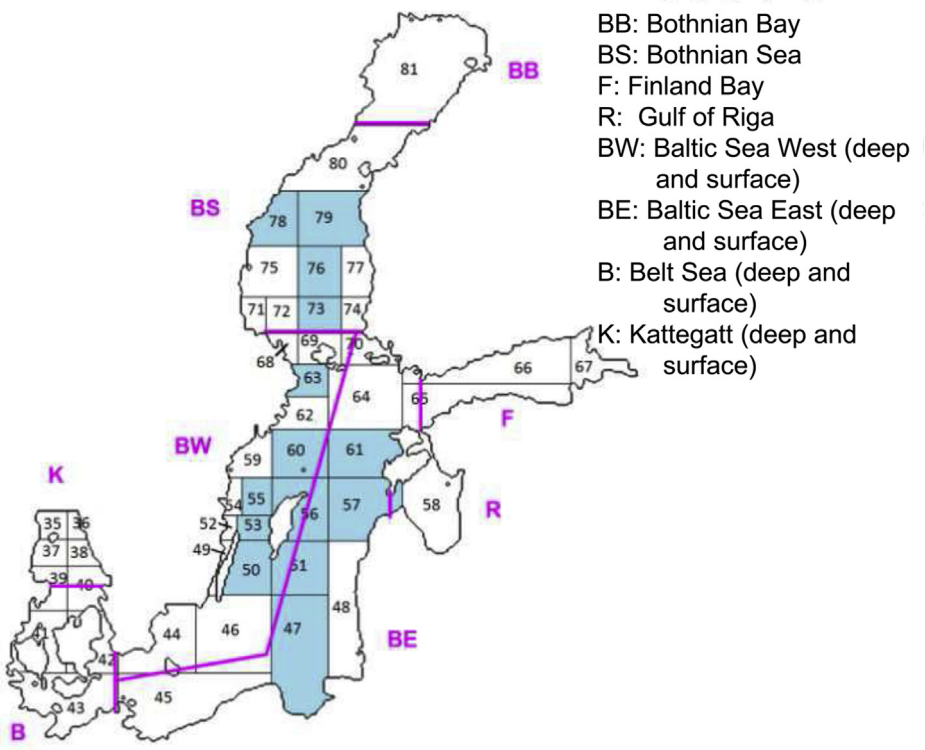


**Fig. 9.** POSEIDON and NRPA model box structures. Pink lines define NRPA model boxes and numbered boxes correspond to POSEIDON. Blue boxes are those divided into two water layers. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The box model includes the processes of advection of radioactivity between compartments, sedimentation, diffusion of radioactivity through pore water in sediments, particle mixing, pore water mixing and a burial process of radioactivity in deep sediment layers. Radioactive decay is calculated for all compartments. The contamination of biota is further calculated from the radionuclide concentrations in filtered seawater in the different water regions. Doses to the human are calculated on the basis from seafood consumptions, in accordance with available data for seafood catches and assumptions about human diet in the respective areas. Dose rates to biota are developed on the basis of calculated radionuclide concentrations in marine organisms, water and sediment, using dose conversion factors. Its structure is presented in Fig. 10.

described by fluxes of radionuclides due to advection and diffusion processes. The transfer of radionuclides in the bottom sediments is described by a three-layer model. The transfer of radioactivity from the upper sediment layer to the water column is described by diffusion and bioturbation. Radioactivity in the upper sediment layer migrates downwards by diffusion and by burial (caused by the ongoing settling of particles from overlying water). The upwards transfer of radioactivity from the middle sediment layer to the top sediment layer occurs only by diffusion. Burial causes an effective loss of radioactivity from the middle to the deep sediment layers, from which no upward migration occurs. The model is completed by the dynamical food web model and dose module to calculate human dose from exposure by different pathways.

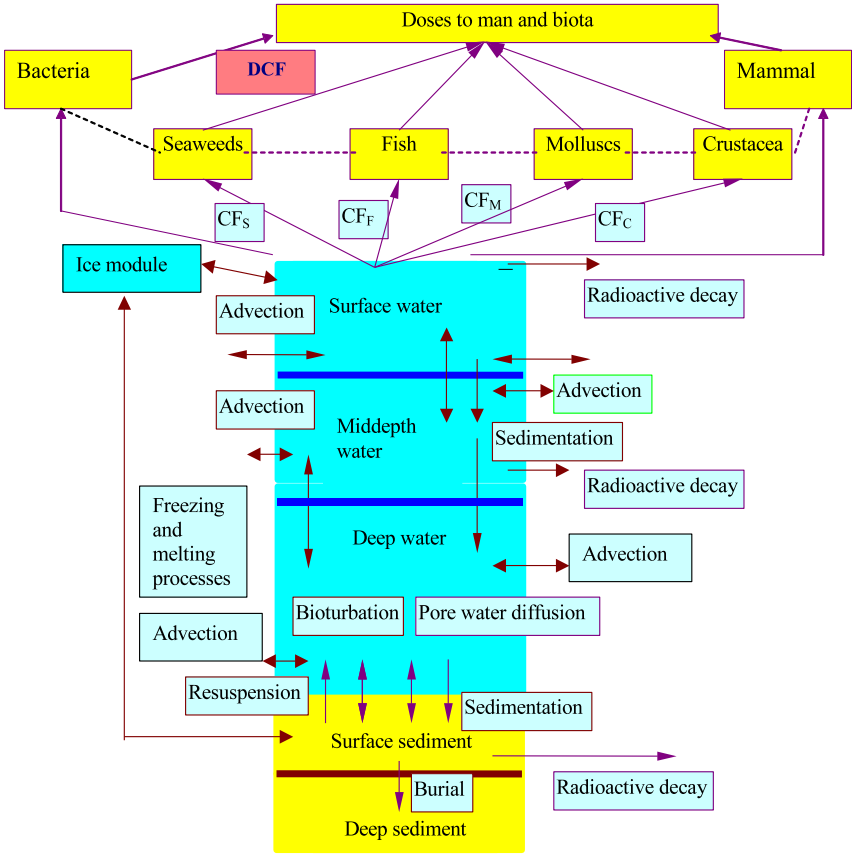


**Fig. 10.** NRPA model structure.

*A.2 POSEIDON*

The compartment model POSEIDON was developed and improved in several papers (Lepicard et al., 1998; Lepicard et al., 2004; Maderich et al., 2014). It is a model for radiological assessments of routine and accidental radioactivity releases into the sea. The POSEIDON compartments describing the water column are subdivided into a number of vertical layers also containing suspended matter. The model assumes equilibrium between dissolved and particulate radioactivity in the water column, described by a distribution coefficient. The radionuclide concentration for each compartment is governed by a set of differential equations including the temporal variations of concentration, the exchange with adjacent compartments and with the suspended and bottom sediments, radioactive sources, and decay. The exchanges between boxes are

The model was customized for the Baltic Sea as shown in Fig. 9. Volume and average depth for each new box was calculated based on the bathymetry of Baltic Sea provided by the Swedish Meteorological and Hydrological Institute (SMHI). The Baltic Sea compartments were connected with the North Sea compartments from MARINA project (CEC, 1990). Boxes with depth larger than 60 m were divided into 2 layers (surface and bottom) for a rough description of stratification in the Baltic Sea. These boxes were marked by blue in Fig. 9. Water fluxes between boxes were calculated by averaging over 10 years the three-dimensional currents provided by the SMHI. River runoff was also taken into account for the largest 16 rivers. Total river runoff was 484 $km^3$/year according to Leppäranta and Myberg (2009).

The simulations were carried out for the period 1945-2010. The sources of $^{137}Cs$ are global deposition from weapon testing,

deposition from Chernobyl accident and release from Sellafield and La Hague reprocessing plants. The global atmosphere deposition due to bomb tests was estimated for boxes 1-61 from Risø Research Reactor measurements and fallout for 62-81 boxes from the Leningrad Nuclear Power Plant measurements. The atmosphere deposition due to Chernobyl accident was taken into account according HELCOM (1995). The release of $^{137}$Cs from Sellafield (into box 15) and from La Hague (into box 26) was taken into account from HELCOM (2009).

### *A.3 THREETOX*

The modelling system THREETOX was developed for simulating the dispersion of radionuclides and other contaminants in the local and regional scales (Margvelashvily et al., 1997; Maderich et al., 2008; Johannessen et al., 2010). The system includes models for hydrodynamics, ice dynamics-thermodynamics and models for sediment and radionuclide transport (Fig. 11). The prognostic variables of the hydrodynamic model are the three components of the velocity fields, temperature, salinity, water surface elevation and kinetic energy of turbulence and its dissipation rate. The ice model predicts the ice drift, thickness and ice concentration. The interactions between water, ice and atmosphere are parameterized the by bulk aerodynamic formulae. The transport, deposition and resuspension of several fractions of sediments are calculated by the sediment transport model. The radionuclide concentration in solute, suspended sediments and the seabed is predicted by the radionuclide transport model. A one-step reversible model was used to describe the exchanges of radionuclides between water and sediments. The numerical algorithm was implemented on a horizontal curvilinear-orthogonal coordinate system.

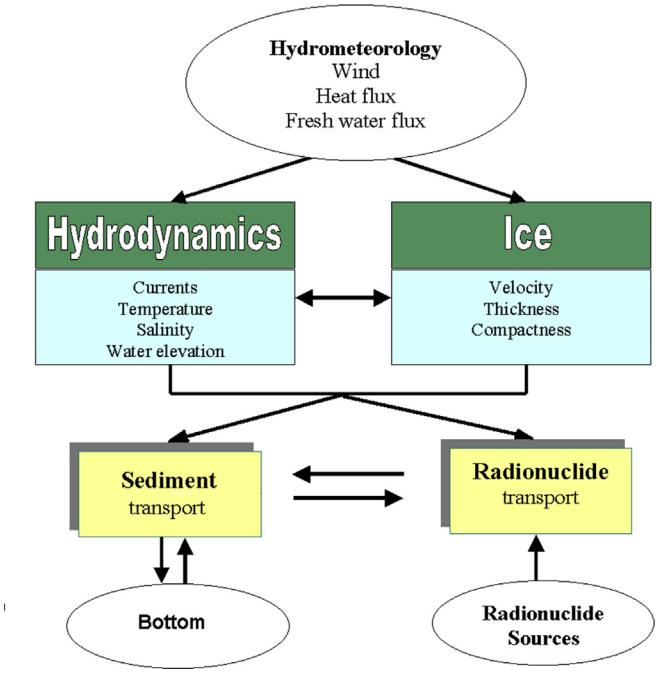


**Fig. 11.** THREETOX modelling system.

The THREETOX model was customized for the Baltic Sea. The bathymetry was obtained from the GEODAS database with 2 minute resolution, both in longitude and latitude. The bathymetry was extended to describe Kattegat. The transport of $^{137}$Cs was modelled using spherical horizontal coordinates with a horizontal resolution of 1/15° along the parallels and 1/30° along the meridians, and by using 20 sigma layers in vertical direction. Main rivers with seasonally varying discharge rates were included in model: Neva, Vistula, Daugava, Oder, Neman, Kemijoki, Torne-Alv, Narva, Dalalven and other smaller rivers. Total freshwater discharge rate was 484 km$^3$/year. The atmospheric forcing was obtained from ERA-Interim reanalysis data (http://apps.ecmwf.int/datasets/data/interim_ full_daily/). Air temperature, wind speed and direction, relative humidity, cloudiness and air pressure were interpolated from ERA-Interim data to the computational grid. Temperature, salinity, water velocity and surface elevation were prescribed along Kattegat from MyOcean reanalysis (http://www.myocean.eu/) for the North Sea. The sediment grain size was defined as 30 μm.

The simulation started on October 1st, 1985. It was assumed that initially only a homogeneous background concentration of $^{137}$Cs in water (15 Bq/m$^3$) exists. After one year spinup the concentration of $^{137}$Cs in the surface layer after Chernobyl accident was prescribed according to Fig. 2 and calculations were performed for the period October 31, 1986 to January 1st, 1991.

### *A.4 University of Seville (USEV)*

The USEV has applied a 2D depth-averaged model forced by the annual mean winds (southwest wind, 6 m/s). Ice cover and water density differences are not considered. A steady mean circulation is obtained, which is used to simulate the transport of radionuclides. The model spatial resolution is 2 minutes of arc, both in longitude and latitude. Bathymetry was obtained from GEODAS database.

The dispersion model includes three phases: water, suspended matter in the water column and bed sediments. An advection/diffusion equation is solved to simulate the transport of radionuclides in the water column. Interactions between the dissolved phase and solid phases (suspended matter and bed sediments) are described through a dynamic approach. Thus, uptake/release of radionuclides is considered to be described by a reversible reaction. This reaction is described by kinetic rates $k_1$ and $k_2$ as in previous works (Periáñez, 2008; 2012; Periáñez et al., 2013, among many others).

A constant and uniform suspended matter concentration over the domain, 4.5 mg/l, has been considered. Particle settling velocity is described by Stoke's law as usual. The model has been calibrated using the $^{137}$Cs inventories over the whole Baltic in water and sediments estimated from measurements. A standard value, determined for Cs from experiments (Nyffeler et al., 1984) has been used for $k_2$. The forward rate, $k_1$, can be determined from $k_2$ and the equilibrium distribution coefficient, $k_d$, as explained in the above mentioned references. Good results are obtained with $k_d = 3$ m$^3$/kg, which is close to the recommended value by the IAEA (2004): 4 m$^3$/kg. In addition, the sediment mixing depth (distance until which the dissolved phase interacts with the sediment) has been set to 2 cm. Finally, a correction factor that takes into account that part of the sediment particle surface may be hidden by other sediment particles is introduced ($\varphi = 0.001$). A detailed formulation of the model may be seen in the above mentioned references. In summary, three parameters are optimized: the equilibrium distribution coefficient, sediment mixing depth and sediment correction factor.

Once that the temporal evolution of $^{137}$Cs inventories in the Baltic in the water column and sediments are adequately reproduced by the model, mean $^{137}$Cs concentrations in several subbasins have been extracted from the model, without any extra tuning, and compared with field data.

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