A one dimensional dispersion model for radionuclides in the marine environment applied to the Chernobyl fallout over the northern Baltic Sea

by

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Abstract

As a result of the Chernobyl fallout the Baltic Sea was the most affected marine ecosystem. The occurrence of 'hot spots' at the water surface was characteristic for the contamination. A one dimensional vertical dispersion model was used to explain the distribution of the radionuclides cesium 137 and cesium 134 in the water column of the central Bothnian Sea for the first six months after the contamination. In addition to the physical dispersion processes, specific chemical characteristics of the radionuclides were taken into consideration. During the time of siumulation, 5 per cent of the radionuclides cesium 137 and cesium 134 were trapped in the sediment, and 50 per cent of the plutonium 239/240 activity concentration was deposited, however, it's activity concentration measured in the water column was close to the limit of detection.

Introduction

A contribution to monitoring the environmental radioactivity is supplied by numerical dispersion models. Box models were used to calculate radiation doses (Evans, 1985; Boelskifte, 1986). Numerical models, using the Lagrangian method, supplied prognoses of dispersion and environmental impact studies (Prandle, 1984; Mller-Navarra & Mittelstaedt, 1988; Pohlmann et al., 1988). But specific characteristics of the examined radionuclides were not taken into consideration

in theses models. For example, in most cases cesium 137 is found mainly in solution, whereas more than 90 per cent of the radionuclide plutonium 239/240 is adsorbed in suspension and sediment in shelf sea areas. Models using the Lagrangian method assumed the conservativity of the radionuclide. With these models not the dispersion of the released radionuclide was studied but therefore, the behaviour of a conservative model tracer was examined.

The nuclear power plant accident at Chernobyl demonstrated the weaknesses of existing dispersion models. During the time of the accident, sediment traps were in operation in the Black Sea, the Mediterranian Sea and the North Sea (Kempe & Nies, 1987; Fowler et al., 1987; Buessler et al., 1987). The evaluation of the trapped material demonstrated a rapid vertical transport of the deposited radionuclides. Within a short time, radionuclides were detected in the sediment. Their transport was not explained by turbulent mixing or in connection with an additional vertical sinking velocity for marine particles (McCave, 1976). In less than ten days, 10 per cent of the activity concentration of dissolved radionuclides such as cesium 134 and cesium 137 were found in water dephs greater 100 m.

The intention of this paper is to present a one dimensional vertical model for the dispersion of different non-conservative radionuclides in the marine environment. The model was applied to explain the vertical dispersion of the Chernobyl fallout in the northern Baltic Sea, i.e. the Bothnian Sea. At first independent of an advection field, the dispersion of a radionuclide, released into the marine environment, was explained under consideration of it's specific chemical characteristics (fig. 1). The released radionuclide is partitioned into five phases. Phase C1 represented the dissolved fraction of the radionuclide. Suspended particles in the water column adsorb a fraction (phase C2) of the dissolved radionuclide. During the relaxation time t, an equilibrium between the dissolved and adsorbed phase appears. Adsorption and the aggregation to large particles (phase C3) results in a rapid vertical transport. In addition to the segregation of these marine aggregates, a continuous exchange between the adsorbed and dissolved phase occurs. Alternating effects at the boundaries to the atmosphere and bottom are performing as sinks and sources for the inventory of the water column. The fraction of the radionuclide leaving the water column at this boundaries is represented by the phases C4 and C5.

In addition to the physical dispersion processes turbulent exchange (Kv, Kv1, Kv2) and sinking of suspended particles and marine aggregates (w1, w2), alternating processes between the phases Ci were taken into account using the exchange coefficients K1 to K16. Considered were adsorption (K1, K6, K7), desorption (K2, K5, K8), aggregation (K4), segregation (K3), accumulation (K9, K10, K11), deposition (K12, K13, K14) and finally resuspension (K15, K16). Sources and sinks for the whole system were represented by tracer production Q and radioactive decay l.

Fig. 1: Concept of the vertical dispersion model. The released radionuclide is divided by the alternating effects K1 to K16 into the five phases C1to C5. Each one of the phases is transported by the different physical dispersion processes Kv, Kv1, Kv2, w1 and w2. Sinks and sources are established by tracer production Q, Q1 and Q2 and radioactive decay l.

The Dispersion Model

Equations of the Model

To study the dispersion of a non conservative substance, the general equation of transport was extended by additional processes. They represent sinks and sources for the activity inventory of the water column. Underlying the alternating effects K1 to K16, the released radionuclide will be partitioned into five phases, which are influenced by different processes of transportation. The Eulerian description of local changes of the activity concentration of each phase Ci with time follows from fig. 1:

phase C1:

$$C1t = (Kv.C1z)z + C2.K2 + C3.K5 + C4.K8 + C5.K12 + Q1 - C1.(K1 + K6 + K7 + K9 + l1) \quad (1)$$

phase C2:

$$C2t = (Kv.C2z)z - w1.C2z + C1.K1 + C3.K3 + C4.K15 + C5.K13 + Q2 - C2.(K2 + K4 + K10 + l1) \quad (2)$$

phase C3:

$$C3t = (Kv.C3z)z - w2.C3z + C1.K6 + C2.K4 + C4.K16 + C5.K16 - C3.(K3 + K5 + K11 + l1) \quad (3)$$

phase C4:

$$C4t = (Kv1.C4z)z + w1.C2z + w2.C3z + C1.K7 - C4.(K8 + K15 + K16 + l1) \quad (4)$$

phase C5:

$$C5t = (Kv2.C5z)z + C1.K9 + C2.K10 + C3.K11 - C5.(K12 + K13 + K14 + l1) \quad (5)$$

It is not possible to solve the system of linear equations in an entirely analytical manner (Marsal, 1976). Ci is a function of place and time describing the activity concentration. The exchange coefficients K1 to K16 depend upon the amount of suspended material, density of sediment, time, depth and the characteristics of the examined radionuclide.

Single Processes

Parameterization of the vertical turbulent exchange Kv using a value of 10-4 m2.s-1 was shown to be appropriate (Kielmann, 1981; Mller-Navarra & Mittelstaedt, 1988). Measurements of field experiments using the passive tracer Rhodamin (Kullenberg, 1968) or determination of the vertical distribution of radionuclides from nuclear weapon tests (Rooth & stlund, 1972) supported this value.

The sinking of particles was described by an additional convective term wi.Ciz in equation (2) to (4). In principle, one has to distinguish between particle sizes smaller and larger than 32 mm. The first group represents something like a 'background noise' (McCave, 1976). The sinking velocities w1 of these particles are very low. The second group consists of large particles such as fecal pellets and macro/mirco flakes which are produced by the agglomeration of organic material (Fowler & Small, 1986; Alldredge & Gotschalk, 1988). The sinking velocities w2 of these particles range from less than some metres to more than 100 metres per day. The size distribution of the aerosols coming from Chernobyl

and transporting the radionuclides were in the range of the 'background noise' (SSK, 1987). The deposited radionuclide could only be transported to the sediment in the observed time range by the aggregation to large particles and thus sinking with high velocities w2.

Suspended material adsorbs dissolved elements. They can be dissolved in deeper, non-contaminated layers. The different activity concentration of the dissolved and adsorbed phase was decribed by the distribution coefficient Kd (IAEA, 1985; SSK, 1988). The time dependence until the moment of equilibrium was expressed by a relaxation time t shown in tab. 1 (Ries, 1980). The activity concentration of each phase Ci during the time of equilibrium was calculated knowing the mass m, the volume V and the total activity concentration C in each layer by:

$$C1 = (V.C)/(V+m.Kd) \quad (6)$$

$$C2 = (m.C.Kd)/(V+m.Kd) \quad (7)$$

In the equilibrium condition the transport coefficients Ki and Kj can be calculated (Ries, 1980). Using the Eulerian description, they have to be divided by the actual masses and
volumes. The coefficients of adsorption/desorption were determined from equation (8)

Table 1

Radiological characteristics

Element	Kd-factor		Relaxation time t	
	Suspension/Sediment	Half life l (dm3.kg-1)	Physical (d)	(d)
Cesium 137	3000	0.1	3	11000
Cesium 134	3000	0.1	3	840
Plutonium 239/240	10000	0.1 (*)	3 (*)	8900000

(*) assumed

and (9). Due to the time and depths dependence of the suspension, the coefficients Ki and Kj depend also upon these parameters.

$$Kj = (1/t.V.m)/(m.V+m2.Kd) \quad (8)$$

$$Ki = (1/t.V.m.Kd)/(V2+V.m.Kd) \quad (9)$$

At the ocean/atmosphere boundary there is a continuous exchange of momentum, energy and gas. On a global scale 180 million tons of aerosol per year was assumed to be produced at the sea surface (Grassl, 1987). These aerosols accumulate elements - 'bubble scavenging'. In laboratory experiments, factors of enrichment for actinides were found in the range of 30 to 600 (Walker et al, 1986).

In the sediment deposited particles are transported vertically by the process of bioturbation. This process can be described by Fickian's Law (Erlenkeuser & Balzer, 1988). The turbulent exchange coefficient Kv has to be substituted by a coefficient of bio-diffusion Kv1(Schink & Guinasso, 1977). The contribution of resuspension (K15, K16) cannot be neglected when studying the activity concentration of the water column. After the discharges of radioactive effluents from the reprocessing plant at Sellafield Works were reduced, the Irish seabed still acted as a source for radionuclides. Plutonium, which was adosrbed onto particle surfaces and deposited in the vicinity of Sellafield Works, was whirled up by tides or occasionally by storms and was transported out of the Irish Sea (Burton & Yarnold, 1988).

Parameterisation

The parameters K1, K2, K7 and K8 were caluclated from equations (8) and (9). Assuming an average suspension, all parameters were chosen as constant for all radionuclides. The same was valid for K7 and K8. In order to estimate the parameters K3 and K4, it was assumed that 10 per cent of suspension in the surface layer was aggregated. Owing to that, 10 per cent of the adsorbed phase C2 was bound into phase C3. If all aggregates were segregated, the whole phase C3 changes to phase C2. First, it was assumed that the parameters K5 and K6 were in the same range as the parameters K1 and K2. However, it has to be stated that these might be lower owing to the reduced free particle surface of marine aggregates. A constant enrichment factor of 100 for all elements under consideration was assumed in order to be able to determine the parameters K9 to K14. No value can be given for K15 and K16 at this moment. In tab. 2 the calculated parameters K1 to K16 as they were used in the following applications of the model for cesium 137 and plutonium 239/240 are represented.

Table 2

Exchange parameters K1 to K16 (s-1)

Parameter	Cesium	Plutonium
K1	1.1.10-4	1.1.10-4
K2	3.1.10-6	9.6.10-7
K3	1.1.10-5	1.1.10-5
K4	3.1.10-7	9.6.10-8
K5	3.1.10-6	9.6.10-7
K6	1.1.10-4	1.1.10-4
K7	3.9.10-6	3.9.10-6
K8	1.6.10-7	4.2.10-10
K9	1.1.10-12	1.1.10-12
K10	1.1.10-12	1.1.10-12
K11	1.1.10-12	1.1.10-12
K12	9.9.10-13	9.9.10-13
K13	9.9.10-13	9.9.10-13
K14	9.9.10-13	9.9.10-13
K15	0.0	0.0

K16 0.0 0.0

The Numerical Method

The Monte Carlo Method was chosen as the numerical method to solve the system of equation (1) to (5). This method yields the best results compared to other numerical methods, i.e. finite differences (Maier-Reimer, 1975).
To simulate the dispersion of a non-conservative element, it was necessary to associate the Monte Carlo particles with specific characteristics. They act as a transport vehicle for the deposited radionuclides, and each one represents a fraction of the dissolved or adsorbed activity concentration. The activity concentration in each layer resulted from the summation of the simulation particles or the activity concentration of each simulation particle in this special layer. After the summation, the alternating effects between the phases Ci and the radioactive decay l were determined.

Application

Assumptions

According to Walin (1972) the Bothnian Sea is a system which is mainly driven by meteorological forces. These are responsible for energy input and circulation. In front of the coast the system behaves in a baroclinic and in the central part in a barotropic way. This leads to the assumption that the vertical transport in the central part is more important than the horizontal transport.

To verify the results of the simulations, sufficient data of radiological measurements were available. They gave evidence about the activity deposition and the horizontal and vertical transport of the radionuclides which were depositied on the water surface. The contamination of the Baltic Sea owing to the reactor accident in Chernobyl was determined from the content of the radinuclides cesium 137 and cesium 134. The activity ratio at that time was in the order of 2:1. The highest deposition occurred in the northern part of the Baltic Sea, the Bothnian Sea, and the activity concentration increased by a factor of 40. The areal deposition of cesium 137 laid around of 35 kBq.m2 (Nies & Wedekind, 1987).

Geographical and hydrographical characteristics of the Bothnian Sea prevented a rapid dilution of the contaminated water. Fig. 2 and fig. 3 represent the activity concentration of the radionuclide cesium 137 in the surface layer of the Baltic Sea six months and fifteen months after the accident. The high initial activity concentration decreased by a factor of 0.5 from October 1986 to August 1987, but the highest activity concentration was still to be found as before in the Bothnian Sea.

Fig. 2: The cesium 137 activity concentration of the surface layer (Bq.m3) in October 1986, six months after the reactor accident in Chernobyl. The main deposition occurred in the northern Baltic Sea, the Bothnian Sea, and the activity concentration increased by a factor of 40.

Fig. 3: The cesium 137 activity concentration of the surface layer (Bq.m3) in August 1987, fifteen months after the reactor accident in Chernobyl. The highest surface activity concentration was still found in the Bothnian Sea.

Parts of the Model

Part 1

In a first application, the dispersion model (fig. 1) was simplified. Cesium 137 and cesium 134 were assumed to be completely dissolved. The radionuclides were transported by turbulent mixing only. All parameters Ki were zero and radioactive decay l was taken into account. A deposition of 35 kBq.m2 of cesium 137 and 17 kBq.m2 of cesium 134 compared to the surface deposition in the central part of the Bothnian Sea was assumed. Therefore, the whole inventory was dissolved into phase C1.

Fig. 4: Comparision between measurements of the cesium 137 (cesium 134) activity concentration (Bq.m3) on the 26th of September 1986 at station 63 (RV Gauss Cruise No. 91b) in the central part of the Bothnian Sea and results of the simulation after a simulation period of 179 days.

A comparison between measurements at a position in the central part of the Bothnian Sea and model data was undertaken (fig. 4), in order to verify the results of the simulation. Above 30 m depth the measurements agreed with the result of the simulation. Owing to the vertical turbulent exchange which cannot be constant as we asumed, the layering of the water column was not represented. The turbulent exchange has to be assumed to be dependent upon depth, if one wishes a better agreement below 30 m. Deeper than 90 m, the simulated activity concentration was too low which can be a result of a weak horizontal transport in the bottom layer representing a second source for radionuclides. Former surface water from the central Baltic Sea is entering the Bothnian Sea in deeper layers, driven by convective sinking.

As a result of this application, it was concluded that the vertical turbulent exchange coefficient Kv was of the correct magnitude and that the assumptions were sufficient to explain the distribution of a dissolved tracer in the water column. However, it is not possible to describe in this application the vertical distribution of a non-conservative tracer such as plutonium 239/240, and it is not possible to explain the high deposition of cesium 134 and 137 in the sediment as measured.

Part 2

The same parameters as in Part 1 were chosen. It was assumed that there was an equilibrium in the surface layer between the dissolved and adsorbed activity concentration after each model run covering a period of 24 hours (eq. 7, eq. 8). The transport of the adsorbed phase was followed by an additional vertical sinking velocity of 1.7.10-4, chosen from the range of the 'background noise'.

During the first days of the simulation, the surface activity concentration was very high. The adsorbed phase had a similarly high activity concentration (fig. 5). Eight days later, after the first particles reached the bottom (fig. 6), the acitivity concentration of the adsorbed phase decreased. A second simulation was carried out for the radionuclide plutonium 239/240 with a fictious surface deposition of 100 Bq.m2. 179 days later, 50 per cent of the surface deposition was trapped in the sediment, and less than 6 per cent of the cesium 137 deposition.

Fig. 5: Representation of the adsorbed phase C2 during a time of 179 days, each day summarized from bottom to surface (top one cesium 137, bottom one plutonium 239/240)

The assumed fictious deposition of plutonium 239/240 did not correspond to the real deposition by the Chernobyl fallout and might be a factor of 100 lower (Aarkrog, 1988). From this, a concentration resulted which was near to the limit of detection.

Fig. 6: Development of the activity deposition (Bq.m-2) (top one cesium 137, bottom one plutonium 239/240). After a simulation period of 179 days 50 per cent of the plutonium and 5 per cent of cesium 137 was bound into the sediment.
The adsorbed phase of the radionuclide plutonium 239/240 was observed as an intermediate concentration maximum and resulted in an early contamination of deeper layers. During the first days of the dispersion, the different transport mechanism of an absolutely dissolved tracer and a non-conservative tracer was obvious (fig. 7). After 179 days, the adsorption on suspended particles resulted in a slightly lower activity concentration (fig. 8) than obtained in Part 1.

Fig. 7: Vertical distribution of the cesium 137 (top one) and plutonium 239/240 (bottom one) activity concentration (Bq.m-3) during a period of 50 days.

Summarizing the results, it was obvious that Part 2 explained a contamination of the sediment by a dissolved radionuclide and an adsorbed radionuclide but not the high activity concentration which were found. The vertical distribution of the dissolved radionuclide cesium 137 in the water column was not explained.

Fig. 8: A comparison between measurements of the cesium 137 activity concentration on the 26th of September 1986 at station 63 (RV Gauss cruise 91b) in the central part of the Bothnian Sea and the results of the simulation after a simulation period of 179 days.

Part 3

In this part, alternating effects were taken into consideration to explain the high activity concentration in the sediment. Resuspension was neglected. For the parameters K1 and K14 the values from tab. 2 were chosen. To simulate the vertical flux of large marine particles the afore mentioned sinking velocity was increased by a factor of ten for a duration of sixty days paying attention to the developing plancton bloom in May 1986.

At the end of the simulation 7 per cent of the cesium 137 activity deposition was bound into the sediment (fig. 9). Although the activity deposition was high, the surface activity concentration from Part 3 corresponded with the measurements (fig. 10).

Fig. 9: Representation of the deposited phase C4 of cesium 137 during a simulation time of 179 days.

Fig. 10: A comparision between measurements of the cesium 137 activity concentration on the 26th of September 1986 at station 63 (RV Gauss cruise 91b) in the central part of the Bothnian Sea and the results of the simulation after a simulation period of 179 days. The alternating effects Ki were taken into consideration.

Discussion and Conlusion

The developed numerical dispersion model (fig. 1) was applied to the Chernobyl fallout in the Bothnian Sea. Due to the complexity of the model structure it was not possible to include in the calculations all processes which were mentioned afore. Therefore, the model was devided into different parts, also to demonstrate the different acting transport processes for the radionuclides cesium and plutonium. From Part 1 it was obvious that the assumed vertical turbulent exchange coefficient was in a reasonable order of magnitude. Although over a relative long period (i.e. 179 days) physically significant results were obtained in the simulation, it should be highlighted that the vertical structure of the water column might have an important influence on Kv. Especially, if the hydrographical situation was characterised by a permanent halocline or a seasonal thermocline, both are not unusual for the Baltic Sea. But in opposite to the central Baltic Sea the main gradients in temperature and salinity in the Bothnian Sea were weak which allowed the assumption of a constant Kv. It should be emphasized that the results of the simulations in Part 1 were in sufficient agreement with the measurements and a further discussion of the dependence of Kv upon the environmental conditions like temperature, salinity and wind field was done in other publications (e.g. KULLENBERG, 1968).

Part 2 and Part 3 attempted to explain the observed high cesium 137 contamination of the sediments within a few days and the dispersion of a non-conservative tracer like plutonium 239/240. It should be mentioned that for this first application the model's sensitivity to the exchange parameters Ki was not further tested. Only one model run was done. In figs. 4, 8 and 10 always the total activity concentration in the water column, i.e. the sum of the phases C1, C2 and C3, was compared with the measurements. The observed activity concentration in the Baltic Sea represented the total activity concentration in the water column. Therefore, the single activity concentration in the different phases of the model results were not examined. The assumption of constant coefficients seemed to be a weakness of the simulation results regarding high biological activity in the surface layer, input of suspended material at the surface via fresh water supply from river run-off, low concentrations of suspended material in the intermediate layer and a higher or increasing concentration near the seafloor. Also the dependence of the calculated parameters on the physical and chemical properties of the environment was excluded (e.g. LI, 1984). Another question was whether these factors were independent of the total activity concentration or not. However, summarizing the results from the calculations, one can conclude that the distribution of an almost dissolved tracer such as Cs 137 in the water column and the high contamination in the sediment observed after the Chernobyl fallout, was explainable by the consideration of the alternating effects K1 - K14 and their calculated order of magnitude. It was possible to explain the dispersion of a

non-conservative tracer by taking into account the specific characteristics of the radionuclide. In further investigations, a more exact determination of the parameters K1 - K16, a more realistic presentation of the vertical turbulent exchange coefficient Kv and an examination of the model's sensitivity to these parameters will be necessary. This paper should be considered as a first draft of a model structure for it's application in aeras of low horizontal mixing, slow dilution and fast vertical transport resulting out of biological activity.

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