

Long-term box modelling of ^{137}Cs in the Mediterranean Sea

J.A. Sanchez-Cabeza*, M. Ortega, J. Merino, P. Masqué

Institut de Ciència i Tecnologia Ambiental, Departament de Física, Universitat Autònoma de Barcelona, 08193 Bellaterra, Spain

Received 19 January 2001; accepted 5 December 2001

Abstract

Environmental radiotracers can be used to understand the underlying processes of the environment where they are found. Amongst others, ^{137}Cs , which is a soluble element in sea water, is transported with water masses in the oceans. A numerical model, in which the Mediterranean Sea was divided into 40 boxes (10 regions of 4 layers each), was developed to better understand the distribution and behaviour of ^{137}Cs in the Mediterranean Sea. It was run with a realistic source term, including global fallout, the Chernobyl accident, the nuclear industry and river runoff, and was used to predict ^{137}Cs concentrations during the period 1954–1994. ^{137}Cs surface water predicted concentrations ranged from 0 to 36 $\text{Bq}\cdot\text{m}^{-3}$. In most boxes the most prominent feature was the existence of two ^{137}Cs predicted concentration maxima, corresponding to maximum air concentrations due to nuclear weapons fallout (1963) and the Chernobyl accident (1986). Maximum concentrations were predicted in surface waters in all regions, generally decreasing as mean water depth increased. Largest spatial concentration gradients were predicted during, and shortly after, the global fallout maxima. After the global fallout maximum was reached, predicted concentrations decreased, first rapidly and later on more slowly, reaching relatively low levels immediately before the Chernobyl accident. The maximum ^{137}Cs predicted concentrations due to the Chernobyl accident in surface waters of each box showed widely different values because the deposition pattern was highly irregular. Levels reached values similar to those predicted immediately before the Chernobyl accident more rapidly than in the case of the global fallout maxima. In order to perform model validation, results were compared with annual mean values in each box and, in general, were found to be in good agreement. The main achievements of the model and its limitations are discussed. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Mediterranean Sea; Model; Radioactivity; ^{137}Cs ; Sea water

1. Introduction

Environmental radiotracers can be defined as radioactive elements that may be used as tools to understand underlying properties of the environment in which they are present. Various marine processes, covering different time scales, can be studied using radiotracers, depending on the phenomena involved and the obser-

vational techniques used. ^{137}Cs is, essentially, a soluble element in seawater (Coughtrey and Thorne, 1983) and has a radioactive half-life of 30.17 years. Thus, it is a useful radiotracer to study water mass transport in the oceans (Prandle, 1984; Nielsen, 1995; Sanchez-Cabeza et al., 1995). In this sense, the Mediterranean Sea has often been described as a small scale oceanographic laboratory (Margalef, 1985) and offers a unique opportunity to test the suitability of using ^{137}Cs as a long-term radiotracer of water mass transport on a large scale.

The aim of this work was to explain past and present (until 1994) levels of ^{137}Cs by using a numer-

* Corresponding author. Tel.: +34-93-581-1915; fax: +34-93-581-2155.

E-mail address: JoanAlbert.Sanchez@uab.es
(J.A. Sanchez-Cabeza).

ical model. To do this, a 40-box model was developed and run with realistic input functions under steady-state conditions. Results were compared with ^{137}Cs concentrations in Mediterranean Sea waters during the period 1954–1994 reported in the scientific literature.

2. Materials and methods

2.1. Model

The Mediterranean Sea was divided into 10 regions (Fig. 1) and its water column was split into four layers, taking into consideration the different water types present in each region (Table 1). Each box was

carefully defined taking into account the following aspects: topography, main water fluxes, input sources and data availability (used for model validation purposes). Excellent revisions of water circulation in the Mediterranean Sea can be found in Malanotte-Rizzoli and Hecht (1988), Özsoy et al. (1993), Millot (1999), Send et al. (1999) and Lascaratos et al. (1999).

The transfer rate between boxes (Table 2) was obtained from previous box models (Bethoux, 1980; Bethoux and Gentili, 1996). Specific studies about circulation in the Eastern (Pinardi et al., 1996; Robinson et al., 1992; Roether et al., 1996) and the Western Mediterranean Sea (Alberola et al., 1995; La Violette, 1995; Margalef, 1985) were also used when further information was needed. In the case of

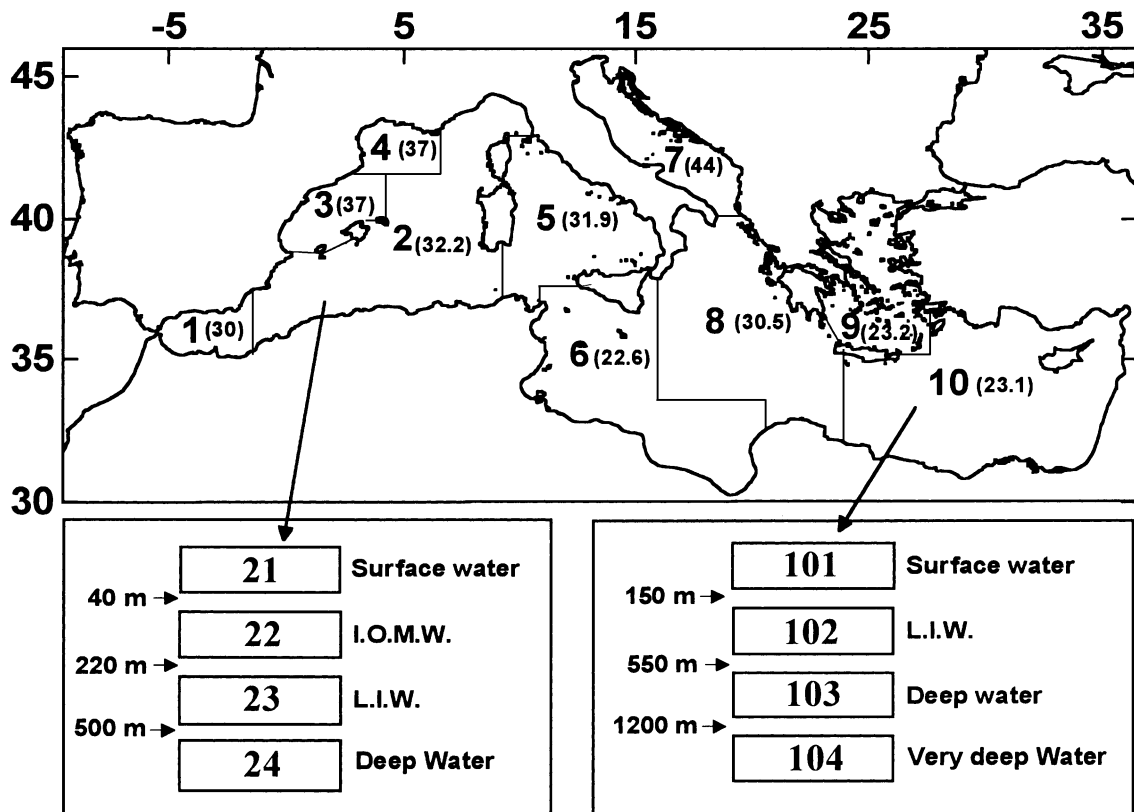


Fig. 1. Map of the Mediterranean Sea showing the considered regions and mean rainfall ($\text{cm}\cdot\text{year}^{-1}\cdot\text{cm}^{-2}$) in brackets. Each region is vertically divided into several boxes, depending on the water depth and water characteristics. The two subsets show the boxes considered in specific regions. The boxes considered correspond to the following regions: 1: Alboran Sea, 2: Central Occidental basin, 3: Catalan Sea, 4: Gulf of Lions, 5: Thyrrenian Sea, 6: Lybian Sea, 7: Adriatic Sea, 8: Ionian Sea, 9: Aegean Sea, 10: Levantine basin. The acronyms used in this figure are: IOMW, Intermediate Occidental Mediterranean Waters; LIW, Levantine Intermediate Water.

Table 1
Dimensions of boxes considered in the model of the Mediterranean Sea

Box	Volume (10 ¹³ m ³)	Width (m)	Box	Volume (10 ¹³ m ³)	Width (m)
11	4.0	50	61	2.0	50
12	8.8	170	62	6.8	170
13	0.16	330	63	13	330
14	0.26	550	64	28	700
21	1.9	40	71	0.97	70
22	8.3	180	72	1.5	150
23	15	330	73	3.3	330
24	87	1950	74	1.9	200
31	0.29	40	81	7.1	150
32	1.3	180	82	17	350
33	1.7	330	83	28	650
34	2.9	550	84	180	2100
41	0.22	40	91	1.2	70
42	0.95	180	92	2.5	150
43	1.7	330	93	5.5	330
44	7.0	1325	94	1.6	950
51	0.72	40	101	6.8	150
52	3.2	180	102	16	350
53	5.8	330	103	30	650
54	34	1950	104	48	1050

vertical transport, exchange rates (Table 2) include both convective and diffusive transport. In order to ensure mass conservation, fluxes were slightly adjusted when necessary. The water mass interchange between the Black Sea and the Aegean Sea is very small, and was ignored. As the input of Atlantic waters through the Gibraltar strait is fundamental to developing an understanding of water circulation in the Mediterranean Sea, a relatively small box (box 1, Alboran Sea) was considered. In order to avoid the definition of an Atlantic box, ¹³⁷Cs levels in surface Atlantic waters were considered to be equal to surface Alboran Sea waters.

The transfer of radiotracers between boxes was described by the following advection-diffusion equation:

$$\frac{\partial c}{\partial t} + \vec{\nabla}c \cdot \vec{v} = \vec{\nabla}_h(\epsilon \vec{\nabla}_h c) + \frac{\partial}{\partial z} \left(k_z \frac{\partial c}{\partial z} \right) + Q - \lambda c$$

where *c* = radionuclide concentration; \vec{v} = water velocity; $\vec{\nabla}_h = \frac{\partial}{\partial x} \vec{i} + \frac{\partial}{\partial y} \vec{j}$; ϵ = horizontal dispersion coefficient; k_z = vertical diffusion coefficient; *Q* = sources or sinks of radionuclide; λ = radioactive decay con-

stant, and the diffusion term is divided in the horizontal and vertical components.

Total diffusion can be considered as an addition of explicit and numerical diffusion. No explicit horizontal diffusion was considered since there was considerable numerical diffusion. The numerical diffusion coefficient was estimated as *ux* (Prandle, 1984) where *u* is the advection velocity and *x* is the distance between the centre of the two boxes, in contact, considered. For example, if the horizontal velocity was 0.1 m·s⁻¹ and *x* = 100 km, the diffusion coefficient was 10⁴ m²·s⁻¹, similar to that found in the literature (Flos, 1985). On the other hand, explicit vertical diffusion

Table 2
Exchange rates between boxes considered in the model of the Mediterranean Sea

Box		Exchange rate (10 ⁵ m ³ s ⁻¹)	Box		Exchange rate (10 ⁵ m ³ s ⁻¹)	Box		Exchange rate (10 ⁵ m ³ s ⁻¹)
From	To		From	To		From	To	
11	21	4.67	92	82	0.72	94	93	0.95
21	31	0.59	102	82	11.2	102	101	0.95
21	41	4.41	102	92	1.20	103	102	1.59
21	51	7.25	23	13	16.1	104	103	6.72
21	61	11.8	53	23	2.03	22	23	14.4
31	21	2.30	63	83	10.0	23	24	2.36
41	31	2.30	73	83	0.04	41	42	2.07
51	21	8.40	83	63	15.8	42	43	1.17
51	61	0.60	93	103	0.58	43	44	1.17
61	21	0.04	103	93	3.66	51	52	2.36
61	81	23.8	24	14	0.23	52	53	0.73
71	81	2.21	24	54	1.30	62	63	0.96
81	61	10.0	44	24	1.17	63	64	3.00
81	71	3.48	74	84	1.54	71	72	1.16
81	101	16.5	84	104	3.72	72	73	1.58
91	81	1.81	12	11	2.23	73	74	3.12
91	101	0.81	13	12	16.3	81	82	0.73
101	81	2.39	14	13	0.23	82	83	4.28
101	91	2.85	22	21	8.52	83	84	3.18
12	22	13.6	24	23	2.00	91	92	1.05
22	42	11.8	52	51	4.25	92	93	1.53
22	52	0.004	54	53	1.30	93	94	0.95
32	22	12.7	62	61	1.76	101	102	12.7
42	32	12.7	63	62	6.76	102	103	0.95
52	22	8.12	64	63	3.00	103	104	3.00
62	22	0.34	74	73	1.58	^a	11	2.52
62	52	10.7	82	81	0.95	^a	12	13.8
62	82	10.7	84	83	1.00	12	^a	16.3
82	62	17.7	92	91	0.95			
82	72	0.42	93	92	0.95			

^a Atlantic ocean.

was needed because vertical velocities were low and the vertical distance between boxes was also small. The vertical diffusion used was $k_z = 1.25 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$, similar to that used by other authors in analogous conditions (Schlitzer, 1988; Roether et al., 1994).

The numerical resolution of the differential equation was carried out with a FORTRAN program using the Runge–Kutta 4th order system. The time step was made variable. A full simulation needed a computation time of less than 1 min with a personal computer provided with a 200-MHz Pentium processor. The authors are well aware that a full sensitivity analysis of the model would be desirable, but this will be the object of a further refinement of the model in the future.

2.2. Source term

The ^{137}Cs sources considered in the model were global fallout from nuclear weapons, the Chernobyl accident, low-level liquid wastes from the nuclear industry and river runoff. As no long-term atmospheric ^{137}Cs sequences exist in Europe, its global deposition was estimated from ^{90}Sr measurements, as its ratio has been shown to be almost constant in

global fallout (UNSCEAR, 1982). During the 1958–1980 period, ^{90}Sr atmospheric concentrations were obtained from the UNSCEAR report (1982). During the period 1954–1958, a linear extrapolation was carried out, assuming that no deposition occurred before the first thermonuclear bomb test in 1954. During 1980–1993, ^{90}Sr atmospheric concentrations were obtained from measurements carried out at Risø, in Denmark (Aarkrog et al., 1997). In order to have a more reliable estimation of deposition throughout the Mediterranean Sea, ^{90}Sr concentrations were normalised to ^{137}Cs concentrations in rain observed in the Gulf of Lions area (Thein et al., 1980). The ^{137}Cs input due to global fallout is shown in Fig. 2. As described later, it should be noticed that the treatment of the deposition from the Chernobyl accident is based on estimations from coastal stations, as it changed the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio.

The ^{137}Cs input to surface waters can occur via dry and wet deposition. The importance of dry deposition was assessed using the expression $D = v_d S$ (Tveten, 1994), where D is the dry deposition ($\text{Bq} \cdot \text{m}^{-2}$), v_d is the dry deposition velocity ($\text{cm} \cdot \text{s}^{-1}$) and S is the ^{137}Cs air concentration ($\text{Bq} \cdot \text{m}^{-3}$). Using the recommended value for v_d , namely, $0.09 \pm 0.06 \text{ cm} \cdot \text{s}^{-1}$ (Tveten, 1994), and the ^{137}Cs air concentration in Denmark

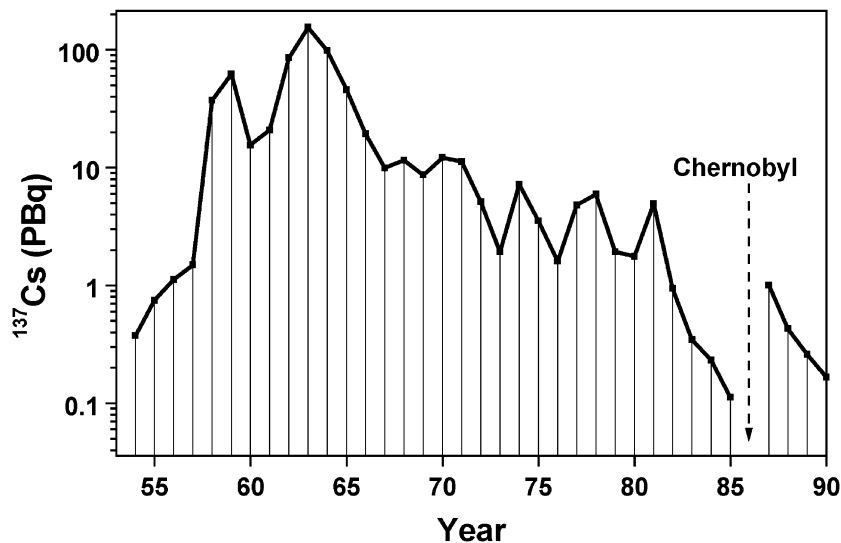


Fig. 2. Annual deposition of ^{137}Cs in the Northern Hemisphere. Because of the large variability of Chernobyl fallout, no estimations for this period are included in this figure.

Table 3
¹³⁷Cs input due to the Chernobyl accident (kBq·m⁻²)

Box	Deposition (kBq·m ⁻²)
11	0.06
21	0.5
31	0.06
41	1.4
51	6
61	0.5
71	8.5
81	0.7
91	4.1
101	0.5

during the period 1958–1980, we estimated that the ¹³⁷Cs dry deposition was less than 8% of the total deposition at that location (Aarkrog et al., 1992). Therefore, only wet deposition was considered in this model. For long-term modelling purposes, wet deposition can be considered as proportional to rainfall (Mitchell et al., 1990). Bethoux (1977) and Cruzado (1979) have estimated rainfall rates over the Mediterranean Sea. Thus, the ¹³⁷Cs deposition was further normalised by the ratio between the estimated rainfall in each region and that in the Gulf of Lions.

In 1986 the Chernobyl accident was responsible for a major input of, amongst other radionuclides, ¹³⁷Cs to Mediterranean surface waters (Table 3). In this case, the deposition pattern was patchy, depending on the trajectories of the contaminated air plumes and rainfall during their passage (Papucci et al., 1996). Therefore, ¹³⁷Cs deposition was estimated from local studies found in the literature: Florou (1996) in the Aegean Sea, Buffoni and Cappelletti (1997) in the Thyrrenian Sea, Holm et al. (1988) in the Gulf of Lions, and Molero et al. (1999) in the Catalan Sea. In the other boxes, ¹³⁷Cs deposition was

estimated from air measurements carried out at coastal laboratories (UNSCEAR, 1988).

Another important source of Chernobyl ¹³⁷Cs to the Mediterranean Sea is the contaminated waters of the Black Sea. This input is well described by the function $V=25.45e^{-0.99t}$, where V (TBq) is the total ¹³⁷Cs input, and t (years) is the time after the Chernobyl accident (Egorov et al., 1994). The ¹³⁷Cs input due to the nuclear industry (Table 4) has been recently reviewed during the Marina-Med project (Cigna et al., 1994). Finally, the ¹³⁷Cs input from river runoff (Table 4) was estimated as described by Fukai et al. (1981).

2.3. Observations

The model (Tracermed I) was designed to predict ¹³⁷Cs annual mean concentrations in each box. In order to validate the model, the literature was searched for existing data and organized in boxes corresponding to the model. More than 300 papers of potential interest were identified and data were extracted in a Mediterranean radiotracers database. These data are of a varied quality and have been produced by laboratories of a very different nature. Regretably, much of the bibliography makes no reference to Data Quality issues and, therefore, a quality flag could not be directly used by us. Instead, a preliminary judgement of the data quality was based on the published uncertainty, as our experience shows that uncertainties range from 2% to 10%. Values out of this range have been excluded from the validation. The list of publications used in the validation is as follows: Ballestra (1980); Fukai et al. (1980, 1981); Ballestra et al. (1981); Triulzi et al. (1989); Franic and Bauman (1993); Molero (1992); Cigna et al. (1994); Sanchez-Cabeza et al. (1995); Molero et al. (1995);

Table 4
¹³⁷Cs input due to the nuclear industry and rivers (GBq)

Box	¹³⁷ Cs (GBq)										
	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990
31	57	45	67	65	70	70	32	32	57	16	10
41	3817	3878	5578	1669	2126	2419	2534	2449	1963	1139	1680
51		45	67	65	70	70.5	32	32	57	16	10
71		0.9	1.0	2.0	1.0	1.0	1.4	8.2	0.5	0.4	0.1

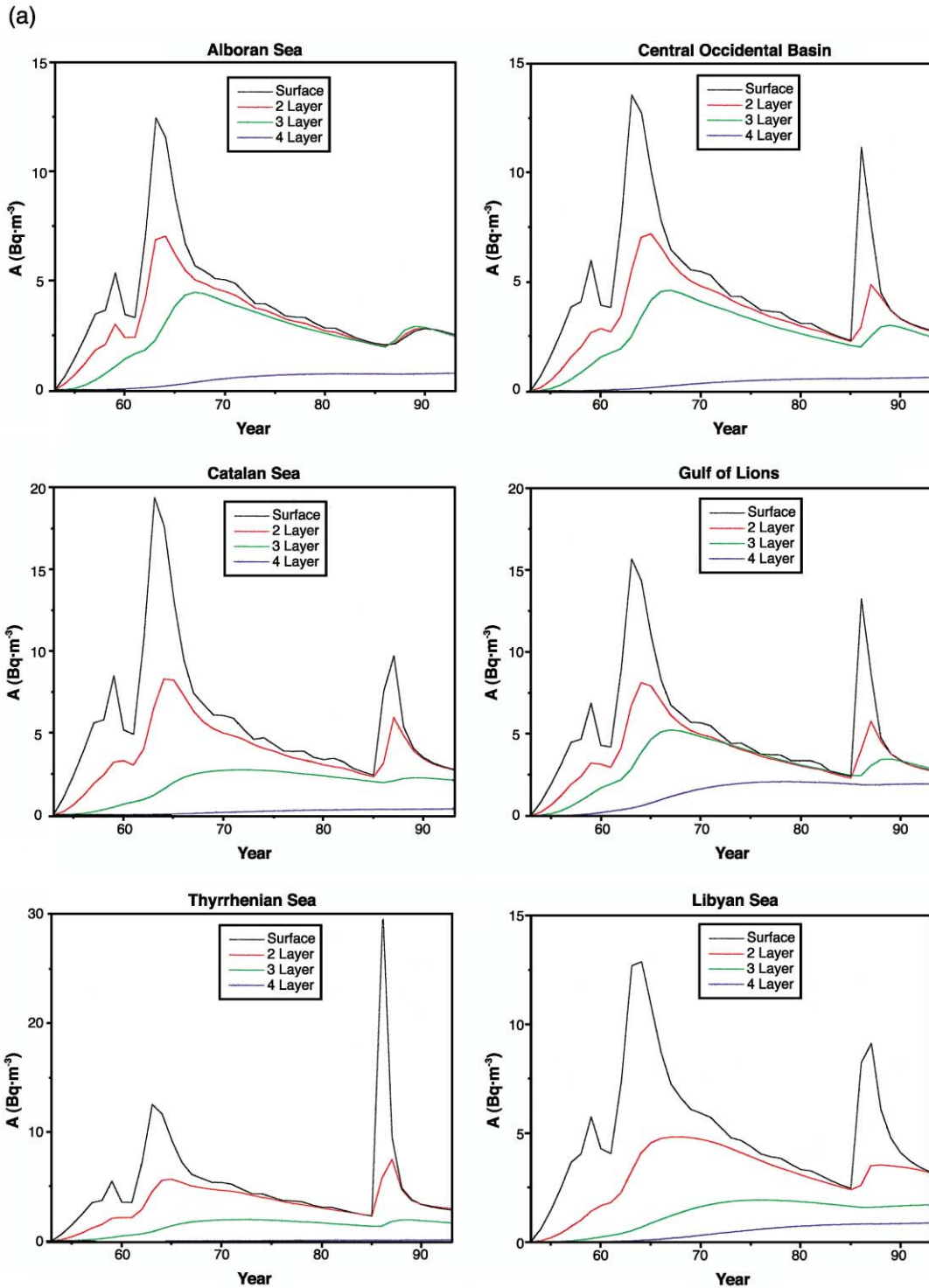


Fig. 3. Modelled time evolution of ^{137}Cs in all boxes of the Mediterranean Sea. Notice that vertical scales are not equal for all plots.

(b)

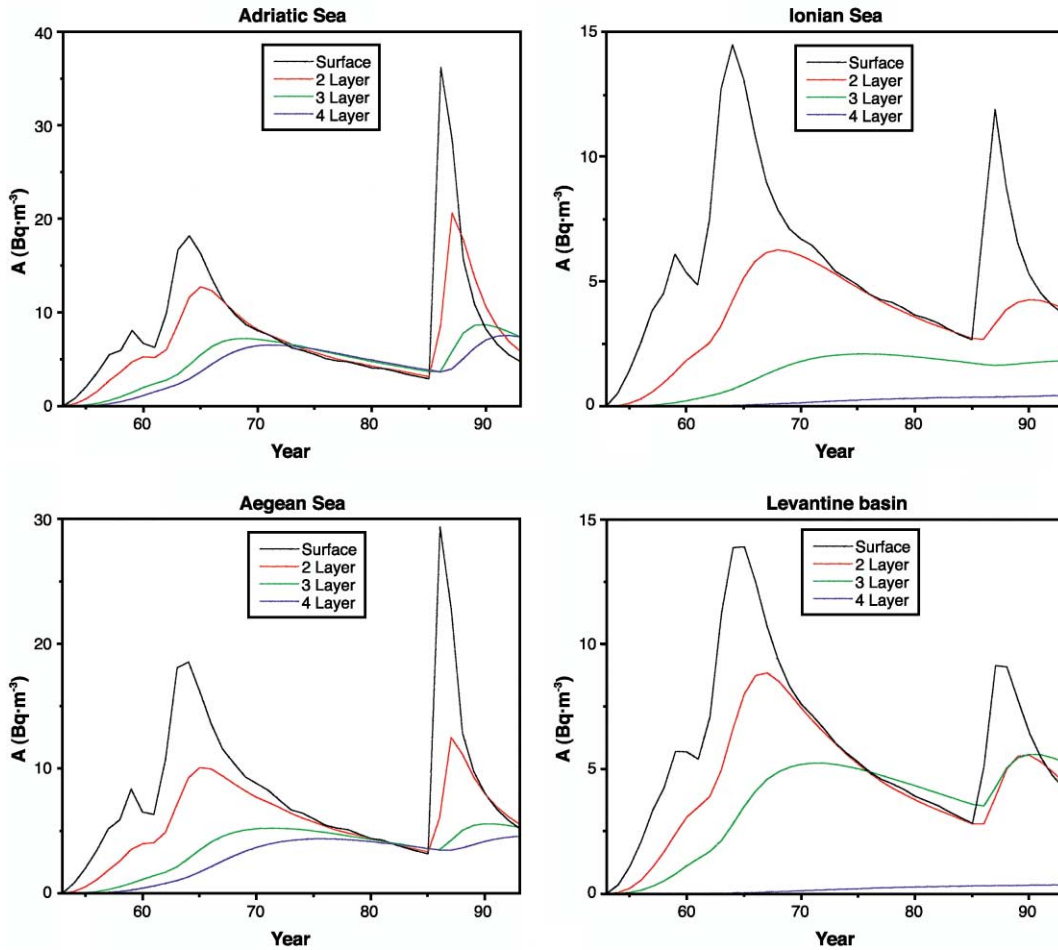


Fig. 3 (continued).

Arnaud and Rozet (1996); Florou (1996); Merino et al. (1997); ENEA (1997).

Accepted data were grouped by boxes and, depending on the number of values, were statistically analysed using the following criteria:

- 1 data point: the value was accepted and the interval of confidence ($p < 0.05$) was set to two times the data uncertainty.
- 2 data points: following the criteria given by IAEA (1998), an uncertainty weighed mean and uncertainty are used as mean value and confidence interval.

- 3 or more data points: the sub-data set was searched for outliers, which were then discarded. In all cases, remaining data followed a normal distribution (Kolmogorov–Smirnov nonparametric test) and the mean and standard error of the mean were computed.

3. Results

The evolution of ^{137}Cs levels in all boxes, during the period 1954–1994, was predicted by running the model. Results are presented in Fig. 3.

3.1. Temporal evolution in surface waters

^{137}Cs surface water predicted concentrations showed large variations during the studied period, as they ranged from 0 to $36 \text{ Bq}\cdot\text{m}^{-3}$. In all boxes, except in the Alboran Sea, the most prominent feature was the existence of two ^{137}Cs predicted concentration maxima, corresponding to the maximum air concentrations due to nuclear weapons fallout (1963) and the Chernobyl accident (1986). A secondary maximum, previous to 1963, was due to the temporal pattern of the global fallout deposition during the 1950s.

The predicted maximum concentration in the 1960s ranged, across the Mediterranean Sea, from $12 \text{ Bq}\cdot\text{m}^{-3}$ in the Alboran Sea, to $19 \text{ Bq}\cdot\text{m}^{-3}$ in the Catalan Sea. This maximum did not occur in all cases during 1963, but was delayed until 1964 in several eastern regions. Although deposition was rather homogeneous over the Mediterranean Sea, western basin surface waters were transported to the eastern basin where mixing with previously contaminated waters took place, thus delaying the observation of the maximum activity.

After the maximum was reached, predicted levels decreased, first rapidly and later on more slowly, reaching minimum pre-Chernobyl (i.e., immediately before the Chernobyl accident) predicted concentrations, which ranged from 3.2 to $2.2 \text{ Bq}\cdot\text{m}^{-3}$. The rapid fall-off reflected the decrease in air concentrations after the cessation of nuclear detonations in the atmosphere, whilst the later slower decrease was attributed to the slow transport and vertical mixing processes of surface waters in the Mediterranean Sea.

The maximum ^{137}Cs predicted concentrations due to the Chernobyl accident, which ranged from 2.6 to $36 \text{ Bq}\cdot\text{m}^{-3}$, showed largely different peak values because the deposition pattern was highly irregular. The maximum concentrations, 36 and $29 \text{ Bq}\cdot\text{m}^{-3}$, were predicted to occur in the Adriatic Sea and the Aegean Sea, respectively, where higher deposition was observed in the nearby terrestrial areas (UNSCEAR, 1988).

The time trends predicted after the Chernobyl accident were different from those observed after the global fallout maximum. First, the predicted peaks were sharp, which was due to the fact that the contamination could be considered as a pulsed input. Second, the levels rapidly returned to near pre-Chernobyl predicted concentrations as, although predicted

maxima were relatively high, the total deposition was smaller than that due to fallout, and mixing with noncontaminated waters in the basin rapidly diluted its effect. However, somewhat elevated levels were still predicted for 1993.

It should be noted that no Chernobyl maximum was predicted in the Alboran Sea, because of the small deposition observed in this area. In fact, levels some years after the accident were still slightly increasing because of the presence of contaminated waters coming from eastern regions.

3.2. Temporal evolution in the water column

Because of the atmospheric origin of the contamination, maximum concentrations were predicted in surface waters in all regions, generally decreasing as mean water depth increased. Largest spatial concentration gradients were predicted during, and shortly after, the global fallout maxima. Because ^{137}Cs entered underlying waters essentially by mixing with surface waters, fallout maxima were usually delayed in deeper waters. This delay was longer as mean water depth increased. For example, in the Central–Occidental basin, global fallout maxima occurred during 1963 in surface waters, during 1965 in the second layer (Mixed Atlantic Waters), and during 1966–1967 in the third layer (Levantine Intermediate Waters).

Before the Chernobyl accident, predicted concentrations in the second layer progressively approached those in surface waters and were, in all cases, almost identical just before the accident. Predicted concentrations in the third layer showed a maximum much later than surface waters. The relative importance of this maximum was variable, and it was almost undetectable in some regions such as the Thyrrenian Sea, the Catalan Sea and the Libyan Sea, where an almost stable predicted concentration was reached. In the fourth layer (Very Deep Mediterranean Waters), levels were in general very low and were steadily increasing until the present (1994). Only in the Aegean Sea, Adriatic Sea and the Gulf of Lions, where deep-water formation takes place (Millot, 1999; Lascaratos et al., 1999), were predicted concentrations slightly higher and they appeared to have reached a stable level.

After the Chernobyl accident, new vertical concentration gradients were predicted. Although the accident also affected the second layer waters, pre-

Chernobyl conditions were rapidly attained and predicted concentrations in the first two water layers were again equal by the end of the modeled period in most regions. Some impact in third layer waters was also predicted for most regions, although little effect was perceptible in the Thyrrenian Sea, the Libyan Sea, the Ionian Sea and the Catalan Sea. It is interesting to point out the prediction that third layer waters in the Levantine basin (Deep waters) should present more activity than the rest of the water layers, by the end of the modeled period, due to the entrance of deep water formed in the Aegean Sea and mixing with contaminated Levantine Intermediate Waters (second layer). The influence of the accident in the deepest layers was only detectable in the Aegean Sea and the Adriatic Sea, where its impact was relatively large.

4. Discussion

4.1. Model validation

The model was validated by comparison with annual mean concentrations of ^{137}Cs in Mediterranean Sea waters. Not unexpectedly, the number of available data points for model validation was limited. In general, existing data are dispersed over the whole basin and, except in the Adriatic Sea, no long-term series exist. This is an important limitation for the validation of any model using radiotracer data in the Mediterranean Sea.

The ratio between model predictions and observed ^{137}Cs water concentrations was determined for each box and followed a log-normal distribution (Fig. 4). The mean value was 0.95 ± 0.06 . Therefore, it was concluded that the model predictions were, in general, in keeping with observations.

4.2. Surface waters

Mean annual ^{137}Cs concentrations measured in surface waters are presented, together with model predictions, in Fig. 5. First, predicted values were in good agreement for the pre-Chernobyl period, as for example in the Adriatic Sea. However, predictions shortly after the Chernobyl accident were not so accurate. The reason for this distinct behaviour rests

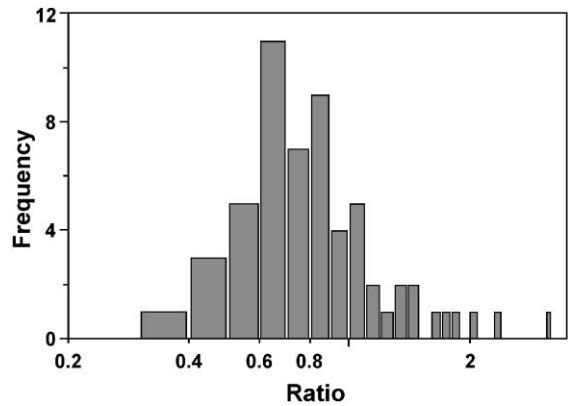


Fig. 4. Frequency distribution of the ratio between predicted and observed ^{137}Cs concentrations in Mediterranean Sea waters.

on the input function: whilst global fallout was deposited over the Mediterranean Sea rather homogeneously, Chernobyl deposition was not. The type of model presented here is especially well suited to describing systems in which temporal and spatial gradients are not large. In the contrary case, a higher-resolution model is needed. In fact, when the Chernobyl signal decreased, and levels were more homogeneous, the accuracy of predictions improved.

The model underestimated some of the concentrations found in the Western basin. This was due to the fact that the Central–Occidental basin compartment was too large, encompassing northern and southern coasts. This is also the case for the Ionian Sea. As most data were obtained in the northern parts, where rainfall (and therefore global fallout) was heavier, mean predictions in these and adjacent regions were underestimated compared to observations. This problem should be corrected in future versions of the model, which should consider the use of smaller boxes to study this basin. However, it should be borne in mind that any increase in the model resolution also reduces the number of experimental points per box and, therefore, makes model validation more difficult.

4.3. Vertical profiles

Measured and predicted vertical profiles are presented in Fig. 6. These included both the Western and Eastern basins and covered the time period from 1976 to 1994, although not in the same region. Selected

(a)

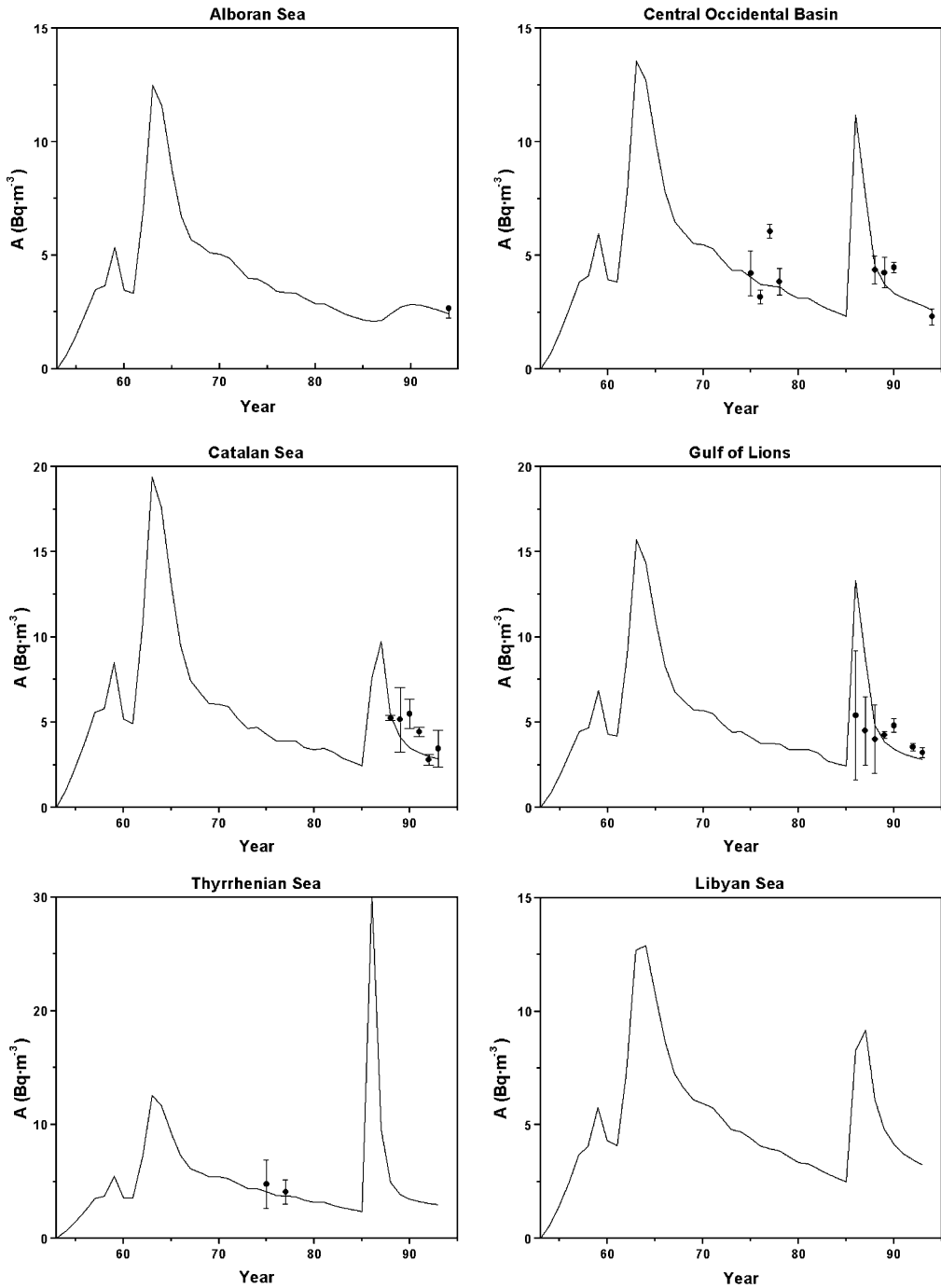


Fig. 5. Mean annual ^{137}Cs concentrations observed in Mediterranean Sea surface waters and model predictions. Error bars represent 1σ statistics of measured values.

(b)

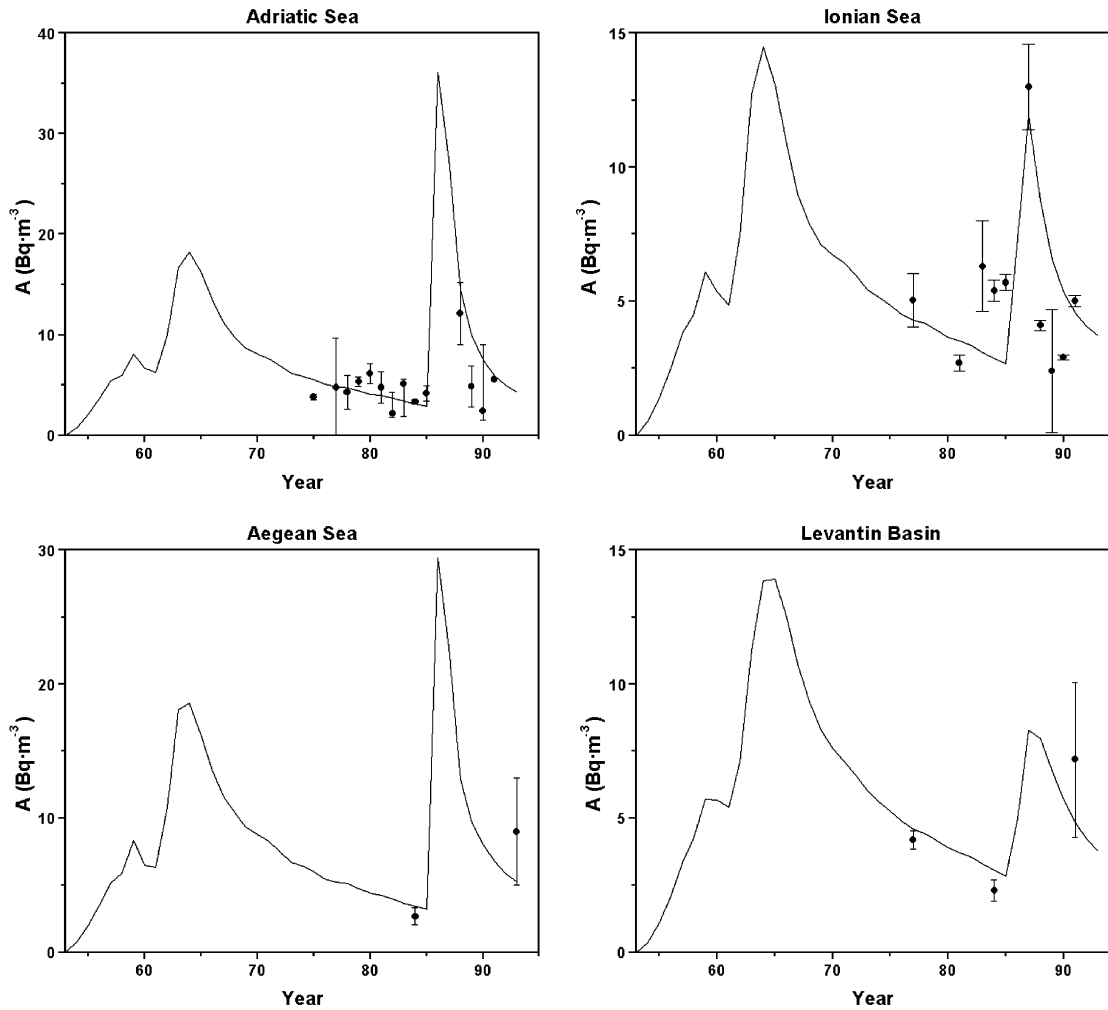


Fig. 5 (continued).

sites depended exclusively on availability of observed values. In general, predicted and observed levels decreased from surface to bottom waters, where levels were very low indeed. Consequently, observations in these waters showed greater uncertainties, making comparison more difficult. As discussed previously, apart from the 1963 and 1986 peaks, predicted levels in the first and second water layers were similar. This prediction was confirmed by the experimental values. However, some deviations were observed. This was clearly the case for the vertical profiles in the Thyrrenian Sea during 1977 (Fukai et al., 1980) and the

Catalan Sea during 1991 (Molero et al., 1995): although the profile shapes were correct, the model underestimated the concentrations. As both regions were adjacent to the Central–Occidental basin, it was concluded that predictions were underestimated because of the low resolution of the model in this region. The divergence observed in the Gulf of Lions profile is possibly due to the incomplete modelling of deep-water formation processes in this area.

In any case, taking into account the abovementioned difficulties, it should be concluded that model predictions of water column levels were, on average,

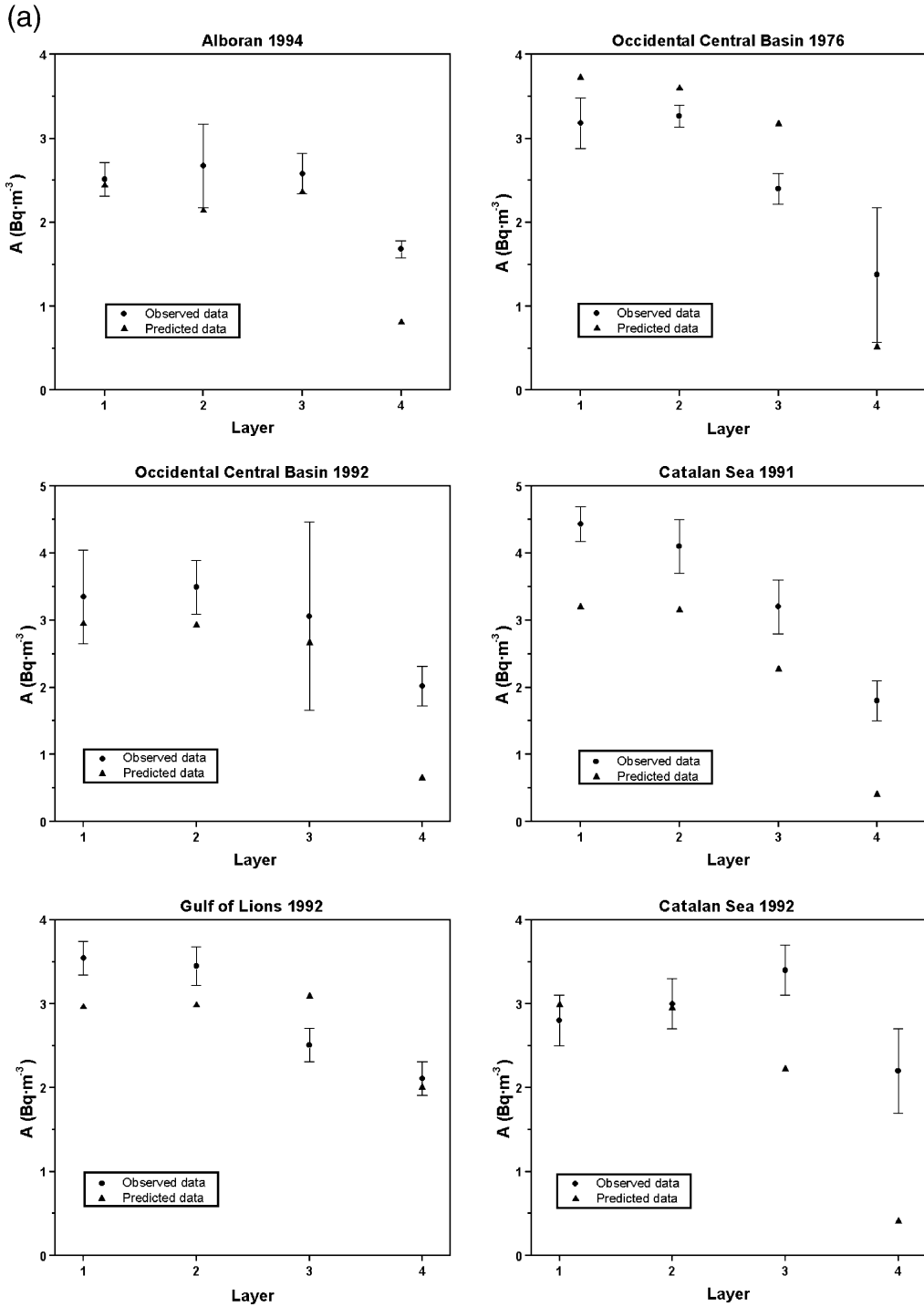


Fig. 6. Measured and predicted ¹³⁷Cs vertical profiles in the Mediterranean Sea. Error bars represent 1σ statistics of measured values.

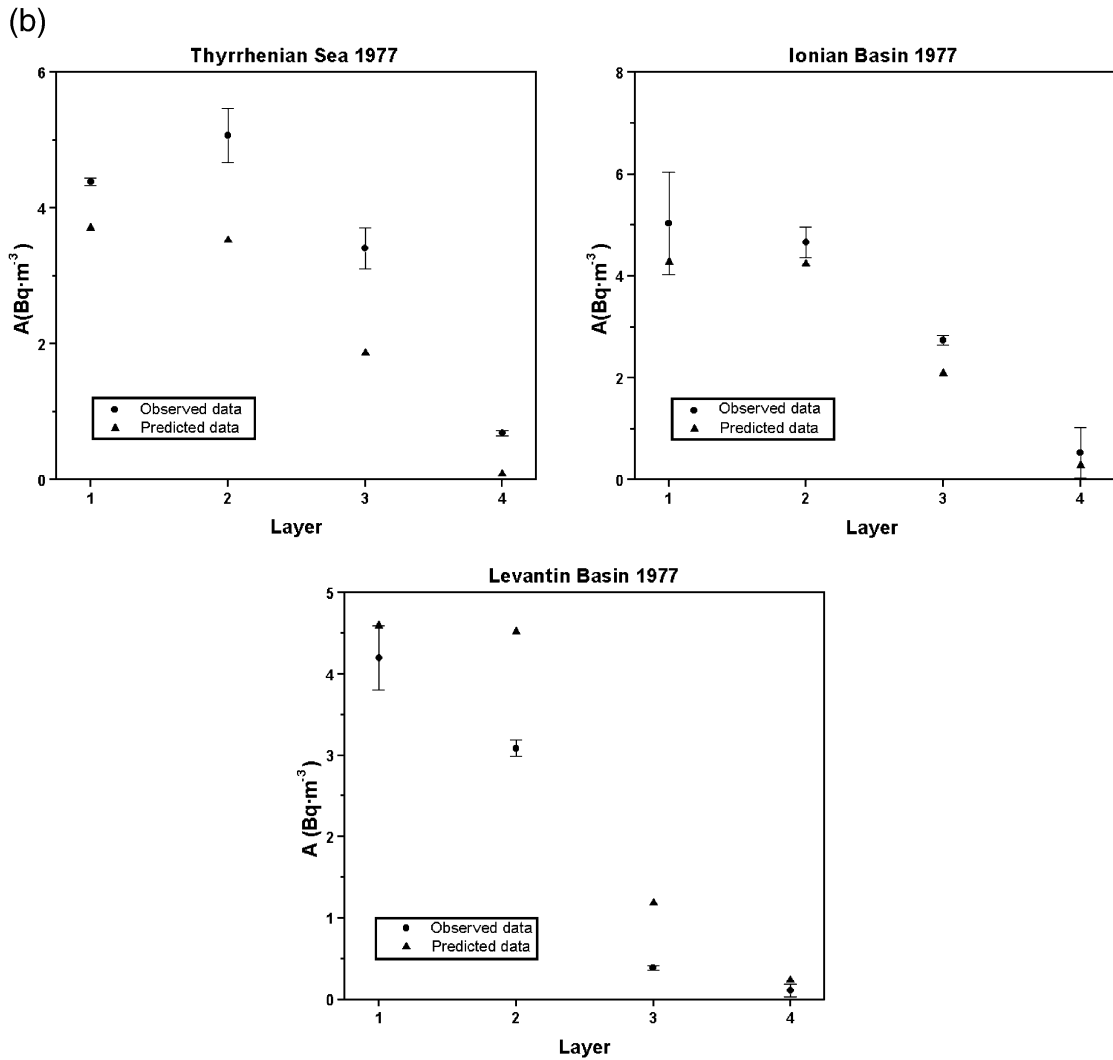


Fig. 6 (continued).

satisfactory, although they should be improved in the future versions of the model.

5. Conclusions

Predicted ^{137}Cs surface water concentrations showed wide variations, ranging from 0 to $36 \text{ Bq}\cdot\text{m}^{-3}$. In all boxes, except in the Alboran Sea, the most prominent feature was the existence of two ^{137}Cs predicted concentration maxima, corresponding to the maximum air concentrations due to nuclear weapons fallout (1963)

and the Chernobyl accident (1986). After the first maximum was reached, levels decreased, first rapidly and later on more slowly. The maximum ^{137}Cs predicted concentrations due to the Chernobyl accident showed different peak values, because the deposition pattern was highly irregular, showing maximum predicted concentrations in the Adriatic Sea and the Aegean Sea, where higher deposition was observed in the nearby terrestrial areas.

Because of the atmospheric origin of the ^{137}Cs contamination, maximum concentrations were predicted in surface waters in all regions, generally

decreasing as mean water depth increased. Largest temporal concentration gradients were predicted during, and shortly after, the global fallout maxima. Because ^{137}Cs entered underlying waters essentially by mixing with surface waters, fallout maxima were usually delayed in deeper waters.

Model results were compared with observed mean values calculated from data available in each box, if any. Not unexpectedly, the number of available data points for model validation was limited. This is an important limitation for the validation of any model using radiotracer data. The ratio between model predictions and observed ^{137}Cs water concentrations was determined for each box and followed a log-normal distribution. The mean value was 0.95 ± 0.06 , and it was concluded that, in general, the model predictions were in keeping with observations but could be improved.

In surface waters, predicted values were in good agreement for the pre-Chernobyl period. However, predictions shortly after the Chernobyl accident were not so accurate because this type of model is not well suited to describe systems with sharp temporal gradients. The model underestimated some of the concentrations found in the Western basin, because the Central–Occidental basin compartment was too large, encompassing northern and southern coasts, what should be solved in the future. In vertical profiles, predicted and observed levels decreased from surface to bottom waters. It was finally concluded that the model predictions were, on average, satisfactory.

Future work will advance in two directions: first, the model spatial resolution will be increased in order to better describe circulation in the basin; second, once the model be validated, it will be used to test existing physical models in the Mediterranean Sea by using fluxes derived from each model run under specific physical conditions. Validation may be better achieved in higher resolution models by the use of well-known stationary parameters such as salinity. This more advanced model will be thoroughly described and a full sensitivity analysis carried out.

Acknowledgements

This research was undertaken in the framework of the Mediterranean Targeted Project II-MATER (MTP

II-MATER) project. We acknowledge the support of the European Commission's Marine Science and Technology (MAST) Programme under contract MAS3-CT96-0051. The authors are thankful to Dr. Ll. Pujol, who helped us in the early stages of the project.

References

- Aarkrog, A., Botter-Jensen, L., Chen, Q.J., Dahlgaard, H., Hansen, H., Holm, E., Lauridsen, B., Nielsen, S.P., Strandberg, M., Sogaard-Hansen, J., 1992. Environmental Radioactivity in Denmark in 1990 and 1991. Riso-R-621(EN), Risø National Laboratory, Denmark.
- Aarkrog, A., Chen, Q.J., Clausen, J., Christensen, G.C., Dahlgaard, H., Ellis, K., Hansen, H., Holm, E., Joensen, H.P., Nielsen, S.P., Strandberg, M., 1997. Environmental Radioactivity in the North Atlantic Region Including the Faroe Islands and Greenland: 1992 and 1993. Risø National Laboratory, Denmark.
- Alberola, C., Millot, C., Font, J., 1995. On the seasonal and meso-scale variabilities of the Northern Current during the PRIMO-0 experiment in the western Mediterranean Sea. *Oceanol. Acta* 18, 163–192.
- Arnaud, M., Rozet, M., 1996. Radionuclides contents in water column during the Euromarge'95 cruise. Euromarge-NB Final Report, vol. IIIB, contribution 58. University of Barcelona, Spain.
- Ballestra, S., 1980. Radioactivité artificielle et environnement marine. Etude relative aux transuraniens ^{238}Pu , $^{239} + ^{240}\text{Pu}$, ^{241}Pu et ^{241}Am en Méditerranée. PhD thesis, Université de Nice.
- Ballestra, S., Thein, M., Fukai, R., 1981. Particulate transuranic elements in the Eastern Mediterranean Sea. V Journées Étud. Pollutions, Cagliari. CIESM, pp. 991–996.
- Bethoux, J.P., 1977. Contribution à l'étude thermique de la mer Méditerranée. Rapport n° 20. Laboratoire de Physique et Chimie Marines. Université Pierre et Marie Curie, Paris.
- Bethoux, J.P., 1980. Mean water fluxes across sections in the Mediterranean Sea, evaluated on the basis of water and salt budgets and of observed salinities. *Oceanol. Acta* 3, 79–88.
- Bethoux, J.P., Gentili, B., 1996. The Mediterranean Sea, coastal and deep-sea signatures of climatic and environmental changes. *J. Mar. Res.* 7, 383–394.
- Buffoni, G., Cappelletti, A., 1997. On the accumulation–dispersion processes of the tracer ^{137}Cs in the Italian seas. *J. Environ. Radioact.* 37/2, 155–173.
- Cigna, A., Delfanti, R., Serro, R., 1994. The Radiological Exposure of the Population of the European Community to Radioactivity in the Mediterranean Sea, Marina-Med Project Comm. European Communities, Bruxelles EUR 15564 EN.
- Coughtrey, P.J., Thorne, M.C., 1983. Radionuclide distribution and Transport in Terrestrial and Aquatic Ecosystems: A Critical Review of Data, vol. I. Balkema, Rotterdam.
- Cruzado, A., 1979. Climatology and hydrology of the Mediterranean Region. Personal Communication.
- Egorov, V.N., Polikarpov, G.G., Stokozov, N.A., Kulebakina, L.G., Lazorenko, G.E., 1994. Distribution of artificial radionuclides in water, bottom sediments and hydrobionts of the Black Sea fol-

- lowing the Chernobyl NPP accident and assessment of Cs-137 input to the seas of the Mediterranean basin through the Bosphorus. The radiological exposure of the population of the European Community to radioactivity in the Mediterranean Sea, Marina-Med project, RADIATION PROTECTION 70, EUR 15564 EN. European Commission, Luxembourg, pp. 363–391.
- ENEA, 1997. Mediterranean Environmental Data Bank, <http://192.107.66.21/www/massel.html>.
- Florou, H., 1996. ¹³⁷Cs in abiotic component and biota from the Aegean and Ionian sea — Greece. *Chem. Ecol.* 12, 253–258.
- Flos, J., 1985. The driving machine. In: Margalef, R. (Ed.), *Western Mediterranean*. Pergamon, Oxford, pp. 60–100.
- Franic, Z., Bauman, A., 1993. Radioactive contamination of the Adriatic Sea by ⁹⁰Sr and ¹³⁷Cs. *Health Phys.* 64, 162–169.
- Fukai, R., Ballestra, S., Vas, D., 1980. Distribution of cesium-137 in the Mediterranean Sea. *Management of Environment*. Wiley Eastern, New Delhi, pp. 352–360.
- Fukai, R., Ballestra, S., Thein, M., Guion, J., 1981. Input of transuranic elements through rivers into the Mediterranean Sea. Impacts of Radionuclide Releases into the Marine Environment. IAEA, Vienna, pp. 3–14.
- Holm, E., Fukai, R., Whitehead, N.E., 1988. Radiocesium and transuranium elements in the mediterranean sea: sources, inventories and environmental levels. *Conferencia Internacional sobre Radioactividad Ambiental en el Area del Mediterraneo*. Sociedad Nuclear Española, Madrid, pp. 601–617.
- IAEA, 1998. Evaluating the Reliability of Predictions Made Using Environmental Transfer Models IAEA, Vienna.
- Lascaratos, A., Roether, W., Nittis, K., Klein, B., 1999. Recent changes in deep water formation and spreading in the eastern Mediterranean Sea: a review. *Prog. Oceanogr.* 44, 5–36.
- La Violette, P., 1995. Seasonal and interannual variability of the Western Mediterranean Sea. *Coastal and Estuarine Studies* American Geophysical Union, Washington, DC.
- Malanotte-Rizzoli, P., Hecht, A., 1988. Large-scale properties of the Eastern Mediterranean: a review. *Oceanol. Acta* 11, 323–335.
- Margalef, R., 1985. *Western Mediterranean*. Pergamon, Oxford.
- Merino, J., Sanchez-Cabeza, J.A., Bruach, J.M., Masque, P., Pujol, L.I., 1997. Artificial radionuclides in a high resolution water column profile from the Catalan Sea (the Northwestern Mediterranean). *Radioprot.-Colloq.* 32, C2.85–C2.91.
- Millot, C., 1999. Circulation in the Western Mediterranean Sea. *J. Mar. Res.* 20, 423–442.
- Mitchell, P.I., Sanchez-Cabeza, J.A., Ryan, T.P., McGarry, A.T., Vidal-Quadras, A., 1990. Preliminary estimates of cumulative caesium and plutonium deposition in the Irish terrestrial environment. *J. Radioanal. Nucl. Chem.* 138, 241–256.
- Molero, J., 1992. Comportamiento y distribución de los radionúclidos de vida larga en ecosistemas marinos. Estudio relativo a radiocésio y a los transuránidos plutonio y americio en el entorno ambiental de la costa mediterránea española. PhD thesis, Universitat Autònoma de Barcelona.
- Molero, J., Sanchez-Cabeza, J.A., Merino, J., Pujol, L.I., Mitchell, P.I., Vidal-Quadras, A., 1995. Vertical distribution of radiocesium, plutonium and americium in the Catalan Sea (Northwestern Mediterranean). *J. Environ. Radioact.* 26, 205–216.
- Molero, J., Sanchez-Cabeza, J.A., Merino, J., 1999. Impact of ¹³⁴Cs and ¹³⁷Cs from the chernobyl reactor accident on the Spanish Mediterranean marine environment. *J. Environ. Radioact.* 43, 357–370.
- Nielsen, S.P., 1995. A box model for North-East Atlantic coastal waters compared with radioactive tracers. *J. Mar. Res.* 6, 545–560.
- Özsoy, E., Hecht, A., Ünlüata, Ü., Brenner, S., Sur, H.I., Bishop, J., Latif, M.A., Rozentraub, Z., Oguz, T., 1993. A synthesis of the Levantine Basin circulation and hydrography. *Deep-Sea Res. II* 40 (6), 1075–1119.
- Papucci, C., Charmasson, S., Delfanti, R., Gascó, C., Mitchell, P.I., Sanchez-Cabeza, J.A., 1996. Time evolution and levels of man-made radioactivity in the Mediterranean Sea. In: Guéguéniat, P., Germain, P., Métivier, H. (Eds.), *Radionuclides in the Oceans: Inputs and Inventories*. Les Éditions de Physique, France, pp. 178–197.
- Pinardi, N., Baretta, J., de Mey, P., Haines, K., Bassini, S., Lascaratos, A., Roether, W., Brasseur, P., Krestenitis, Y., Crise, S., Pierini, S., Allen, I., Williams, R., Vested, J., Le Traon, P.Y., 1996. I Mediterranean Eddy Resolving Modeling and Interdisciplinary Studies-II. Istituto per lo Studio Delle Metodologie Geofisiche Ambientali, S.T.A.R., Pisa, pp. 5–96.
- Prandle, D., 1984. A modelling study of the mixing of Cs-137 in the seas of the European continental shelf. *Philos. Trans. R. Soc. London, Ser. A* 310, 407–436.
- Robinson, A.R., Malanotte-Rizzoli, P., Hecht, A., Michelato, A., Roether, W., Theocharis, A., Ünlüata, Ü., Pinardi, N., Artegiani, A., Bergamasco, A., Bishop, J., Brenner, S., Christianidis, S., Gacic, M., Georgopoulos, D., Golnaraghi, M., Hausmann, M., Junghaus, H.-G., Lascaratos, A., Latif, M.A., Leslie, W.G., Lozano, C.J., Oguz, T., Özsoy, E., Papageorgiou, E., Paschini, E., Rozentraub, Z., Sansone, E., Scarazzato, P., Schlitzer, R., Spezie, G.C., Tziperman, E., Zodiatis, G., Athanassiadou, L., Gerges, M., Osman, M., 1992. General circulation of the Eastern Mediterranean. *Earth-Sci. Rev.* 32, 285–309.
- Roether, W., Roussenov, V.M., Well, R., 1994. A tracer study of the thermohaline circulation of the eastern mediterranean. In: Malanotte-Rizzoli, P., Robinson, A.R. (Eds.), *Ocean Processes in Climate Dynamics: Global and Mediterranean Examples*. Kluwer, Dordrecht, pp. 371–394.
- Roether, W., Manca, B.B., Klein, B., Bregant, D., Georgopoulos, D., Beitzel, V., Kovacevic, V., Luchetta, A., 1996. Recent changes in Eastern Mediterranean deep waters. *Science* 271, 333–335.
- Sanchez-Cabeza, J.A., Molero, J., Merino, J., Pujol, L.I., Mitchell, P.I., 1995. Cs-137 as a tracer of the Catalan current. *Oceanol. Acta* 18, 221–226.
- Schlitzer, R., 1988. Modelling the nutrient and carbon cycles of the North Atlantic: circulation, mixing coefficients and heat fluxes. *J. Geophys. Res.* 93, 699–721.
- Send, U., Font, J., Krahnemann, G., Millot, C., Rhein, M., Tintoré, J., 1999. Recent advances in observing the physical oceanography of the western Mediterranean Sea. *Prog. Oceanogr.* 44 (1–3), 37–64.
- Thein, M., Ballestra, S., Yamato, A., Fukai, R., 1980. Delivery of transuranic elements by rain to the Mediterranean Sea. *Geochim. Cosmochim. Acta* 44, 1091–1097.

- Triulzi, C., Tassi Pelati, L., Albertazzi, S., Morani, A., 1989. Presenza et distribuzione di ^{137}Cs e di radioattività naturel nel are Adratico (1979–1984). *Bolletino do Oceanologia Teorica ed Applicata*, numero speciale de Simposio ASCOP, pp. 177–191.
- Tveten, U., 1994. Atmospheric dispersion, deposition and redistribution of radioactive materials in the environment. In: Holm, E. (Ed.), *Radioecology—Lectures in Environmental Radioactivity*. World Scientific, Singapore, pp. 41–61.
- UNSCEAR, 1982. *Ionizing Radiation: Sources and Biological Effects* United Nations, New York.
- UNSCEAR, 1988. *Sources, Effects and Risk of Ionizing Radiation* United Nations, New York.