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^{137}Cs baseline levels in the Mediterranean and Black Sea: A cross-basin survey of the CIESM Mediterranean Mussel Watch programme

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Abstract

The common mussel *Mytilus galloprovincialis* was selected as unique biomonitor species to implement a regional monitoring programme, the CIESM Mediterranean Mussel Watch (MMW), in the Mediterranean and Black Seas. As of today, and upon standardization of the methodological approach, the MMW Network has been able to quantify ^{137}Cs levels in mussels from 60 coastal stations and to produce the first distribution map of this artificial radionuclide at the scale of the entire Mediterranean and Black Seas. While measured ^{137}Cs levels were found to be very low (usually <1 Bq kg^{-1} wet wt) ^{137}Cs activity concentrations in the Black Sea and North Aegean Sea were up to two orders of magnitude higher than those in the western Mediterranean Basin. Such effects, far from representing a threat to human populations or the environment, reflect a persistent signature of the Chernobyl fallout in this area.

Keywords: Bivalves; Radionuclides; Monitoring; Mussel watch; Mediterranean; Black Sea

1. Introduction

It is now widely accepted that establishing environmental quality threshold values for coastal management

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purposes requires more information on baseline levels than those currently available (e.g., UNEP, 1996; CEC, 2006; Warnau and Acuña, 2007). The demand for contaminant monitoring is especially critical for enclosed or semi-enclosed basins such as the Mediterranean and Black Seas that are characterized by extremely long water residence time (e.g., Özsoy, 1999).

Direct contaminant measurements in seawater are often technically challenging and costly. In addition, although such information are important for controlling direct and indirect waste releases, sustainable coastal zone management rather relies on the assessment of the bioavailable fraction of contaminants (e.g., Phillips, 1980; Rainbow, 1990; Thébault et al., 2005). To this end the use of mussels and oysters that are widely distributed in the coastal zone, sedentary, easy to collect and to identify, resistant to variation of environmental conditions and able to bioconcentrate efficiently several contaminants within their tissues (e.g. Cossa et al., 1980; Phillips, 1980; Cossa, 1988; Rainbow, 1990), has proven very effective. Since the early 1970s these bivalves have shown their value as "sentinel species" in biomonitoring programmes to assess levels and trends of contaminants in the coastal zone worldwide (e.g., Goldberg et al., 1983; Claisse, 1989; UNEP, 1992; Jernelov, 1996).

In 1998, a coordinated research programme was called by the International Mediterranean Commission (CIESM) during its 31st Congress in Athens. The resulting GIRMED project (Global Inventory of Radioactivity in the MEDiterranean Sea) aimed to assess the consequences of the Chernobyl fallout in the whole Mediterranean and Black Seas. However, despite the efforts deployed to collect large datasets, difficulties related to the diversity of indicators considered and methodological approaches did slow down the progress of a reliable and coherent inventory.

To respond to this challenge, CIESM implemented in 2002 a regional "Mediterranean Mussel Watch (MMW)" programme to record the levels and trends of radioactivity in the marine environment, using the common Mediterranean mussel *Mytilus galloprovincialis* as a biomonitor species (CIESM, 2002). This filled a major gap: until then, despite an increasingly pressing demand for a comprehensive monitoring of radioactivity and the existence of several ongoing national projects (e.g. France, Italy, Spain), no large-scale monitoring programme for radionuclides was coordinated at the Mediterranean basin scale.

The main objective of the CIESM MMW programme is to use a single common strategy throughout the network of participating Mediterranean marine stations and research institutes affiliated to CIESM, and to promote the production of adequately large and reliable datasets. This was set during the course of a specifically dedicated workshop that brought together 22 experts from 18 Mediterranean countries, who consensually agreed on the strategic objectives of the programme and on the common methodologies to be applied for field sampling and data validation, intercalibration, and treatment (see CIESM, 2002).

Currently, 21 marine environment laboratories from 16 Mediterranean countries are actively cooperating in this international, dynamic network. So far, 60 locations have been investigated along the coastline of the Mediterranean and Black Seas, using a single, standardized common methodology. Although incomplete (southern coasts are still poorly documented) the MMW network cooperation allowed the production of the very first peri-Mediterranean distribution map of intercomparable ^{137}Cs levels in mussels.

2. Materials and methods

The methodologies followed in the present study are fully described in the report of the expert workshop that was dedicated to this programme and held prior to its initiation (CIESM, 2002).

Briefly, the common Mediterranean mussel *Mytilus galloprovincialis* was selected as unique biomonitor species. Generally, sampling sites were selected among those already monitored within the activities of pre-existing national/international projects or, when set up *ex novo*, on the basis of a preliminary zoning of the littoral area. Subsequently, at least one sampling location per sector was selected in order to contemporarily ensure the geographic coverage of the monitored area and the sustainability of the network.

On a general basis, each sampling consisted in the collection of 200–300 mussels (corresponding to a wet wt of 3–5 kg). The organisms were then immediately rinsed onboard with seawater from the sampling station and transported to the home laboratory in a refrigerated container. Upon arrival at the laboratory, the organisms were cleansed from any external material and their byssus, opened using a micro-wave or steam oven, and then dried, freeze-dried or calcinated prior to radioanalytical measurements. For each sample, the condition index was calculated.

Trace level measurements of ^{137}Cs were performed by direct, low-background high-resolution Ge gamma spectrometry as a routine technique.

3. Results and discussion

In order to guarantee data comparability, all partners initially participated in an international intercalibration exercise, using a specifically-produced reference material (IAEA-437; *M. galloprovincialis* flesh). The intercalibration was organized jointly by the French Institute of Radioprotection and Nuclear Safety (IRSN) and the Marine Environment Laboratories of the International Atomic Energy Agency (IAEA-MEL) and coordinated by IAEA-MEL. The results of the intercomparison exercise were published as an IAEA Report (Pham and Sanchez-Cabeza, 2007). Recommended values for ^{137}Cs have been made available to all MMW participants.

The results provided by the different MMW partners for the 2004–2006 period, complemented with data from the IFREMER-coordinated EC Project MYTIMED (http://mytilos.tvt.fr/), allowed drawing a first comprehensive distribution map of ^{137}Cs activity concentrations in mussels *M. galloprovincialis* at the Mediterranean scale (Fig. 1).

On a general basis, ^{137}Cs activity concentrations were low, i.e. between 0.01 Bq kg^{-1} and 1 Bq kg^{-1} wet wt (see Table 1), and approached standard instrumental detection limits for this radionuclide (Pham et al., 2007). These values are in good agreement overall with previous, punctual measurements carried out by others in the region (e.g., Bologa et al., 1995; Papucci et al., 1996; Florou et al., 2000; Sanchez-Cabeza and Molero, 2000; Topcuoglu et al., 2001; Vassiliki-Angelique and Florou, 2006).

Although available data for the southern Mediterranean shore are still sparse, ^{137}Cs levels in mussels from the Black Sea and northern Aegean Sea were found to be up to two orders of magnitude higher than those from the western Mediterranean basin. Furthermore, we found a strong inverse relationship between the concentration of ^{137}Cs measured in mussels and the distance of their sampling locations from the Chernobyl Nuclear Power Plant (Fig. 2). The regression is clearly driven by the data obtained within a perimeter of about 2000 km, comprising the stations in the Black Sea and North Aegean. Obviously, the role played by the drainage basins of the Danube, Dniepr and Don rivers in conveying the Chernobyl radioactive fallout to the Black Sea is such that the effects of this major nuclear accident are still evident. As a matter of fact, these rivers are known to influence ^{137}Cs concentrations from the Black Sea all the way to the northern Aegean Sea (Delfanti et al., 2006) due to their large fluvial inputs that feed a net, positive outflow of Black Sea-originating waters to the Aegean Sea through the Dardanelles strait (Unluata et al., 1990; Kourafalou, 2007).

By way of comparison, ^{137}Cs values measured in other regions of the word reach up to 0.2 Bq kg^{-1} dry wt in mussels from the Pacific Coast of Japan (Ishikawa et al., 2004) and 0.4 Bq kg^{-1} dry wt in specimens from the US West Coast (Valette-Silver and Lauenstein, 1995). While 35 times higher (on a dry wt basis), the most elevated ^{137}Cs values measured in our survey still remain well below radioactive levels for which restoration measures would be recommended (IAEA, 2004, and references therein). For example, even the highest ^{137}Cs concentrations found in the present study (i.e., 1.5 Bq kg^{-1} wet wt; Fig. 1) are 400 times lower than those authorized by the European Union in seafood intended for human consumption (i.e., 600 Bq kg^{-1} wet wt; AAE, 1999). Nevertheless the precautionary approach would dictate that the evolution of ^{137}Cs and/or other radionuclides such as U and Pu isotopes be

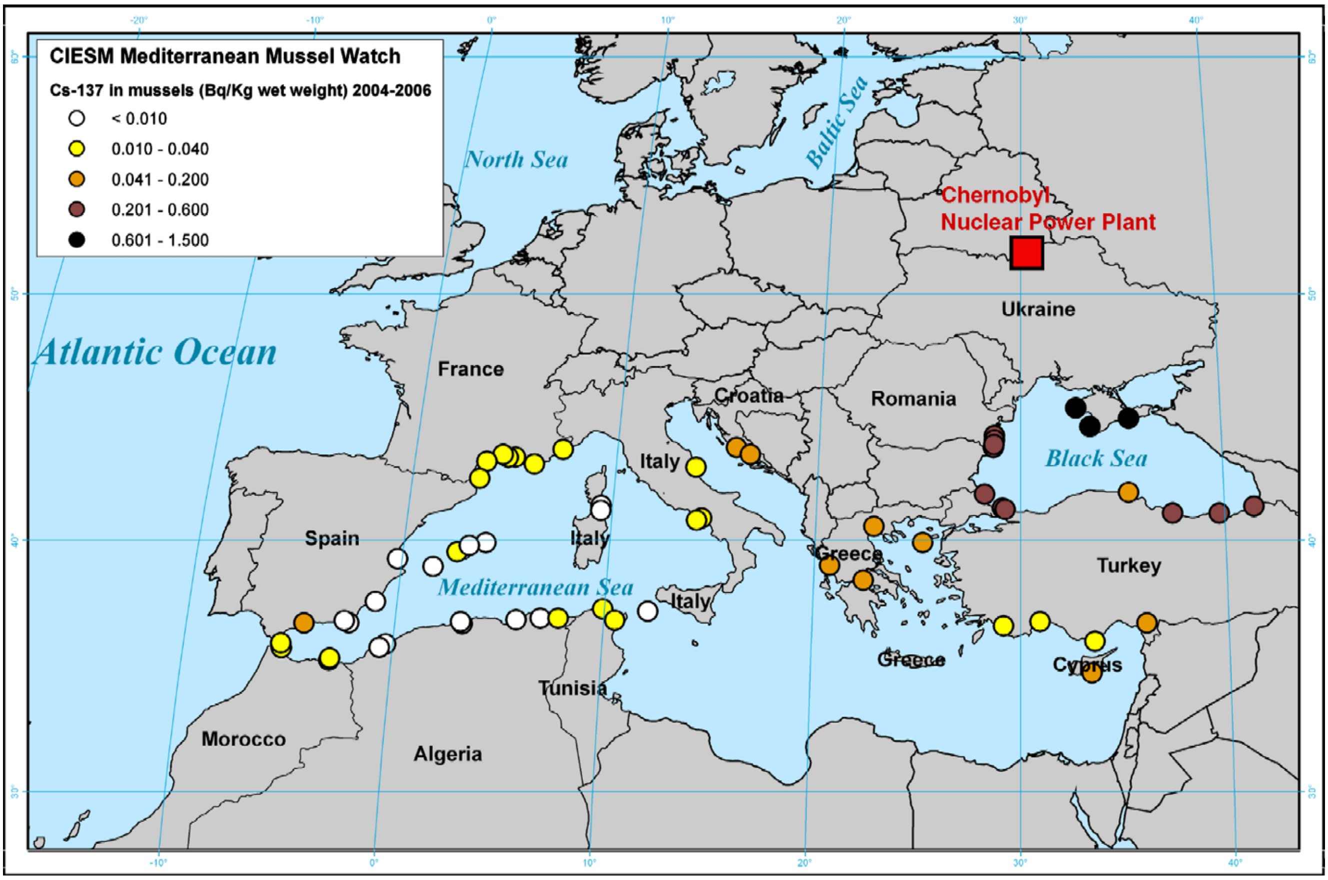

Fig. 1. Distribution of ^{137}Cs concentrations (Bq kg^{-1} wet wt) in mussels *Mytilus galloprovincialis* collected in 2004–2006.

Table 1
^{137}Cs concentrations in mussels *Mytilus galloprovincialis* collected in 2004–2006

Country	Station	^{137}Cs (Bq kg^{-1} wet wt)	Longitude	Latitude
Algeria	Arzew	<0.01	−0.32511	35.86343
Algeria	Bouharoun	<0.01	3.24168	36.67626
Algeria	El kala	<0.01	6.95715	36.89710
Algeria	Oran	0.007 ± 0.003	−0.61097	35.72707
Algeria	Alger	0.010 ± 0.005	3.15457	36.76072
Algeria	Djijel	0.010 ± 0.004	5.80333	36.82798
Algeria	Annaba	0.011 ± 0.004	7.82058	36.89942
Croatia	Strobec	0.08 ± 0.03	15.90029	43.73756
Croatia	Vranjic	0.07 ± 0.03	16.61151	43.46136
Cyprus	Zygi, fish farm	0.11 ± 0.06	33.40166	34.70800
France	Nice	0.02 ± 0.01	7.28937	43.66719
France	Toulon	0.02 ± 0.01	5.93691	43.07646
France	Carteau	0.02 ± 0.01	4.95754	43.35628
France	Faraman	0.03 ± 0.01	4.61554	43.34074
France	Saintes maries	0.02 ± 0.01	4.33572	43.46510
France	Sete	0.02 ± 0.01	3.57399	43.18528
France	Banyuls	0.02 ± 0.01	3.32526	42.50128
France	Bonifacio	0.01 ± 0.01	9.43465	41.41309
Greece	Preveza	0.20 ± 0.01	20.75056	39.01070
Greece	Lemnos	0.12 ± 0.05	25.24913	39.91612
Greece	Thessaloniki	0.10 ± 0.03	22.84312	40.56909
Greece	Galaxeidi	0.15 ± 0.04	22.40572	38.40915
Italy	Maddalena	0.010 ± 0.004	9.43968	41.20349
Italy	Banco di Pantelleria	0.008 ± 0.003	12.09727	37.16510
Italy	Napoli	<0.05	14.24972	40.82611
Italy	Bagnoli	<0.05	14.16389	40.81416
Italy	S. Benedetto del Tronto	<0.05	13.95472	42.95306
Morocco	M'diq	0.020 ± 0.012	−5.29239	35.72221
Morocco	Nador	0.040 ± 0.015	−2.95248	35.21534
Morocco/Spain	Ceuta	0.016 ± 0.004	−5.33330	35.90378
Morocco/Spain	Mellila	0.026 ± 0.004	−2.92492	35.30912
Romania	Constantza	0.28 ± 0.04	28.70866	44.22925
Romania	Eforie	0.27 ± 0.03	28.70894	44.04105
Romania	Vama veche	0.32 ± 0.03	28.65987	43.73691
Romania	Mangalia	0.28 ± 0.04	28.68211	43.85896
Spain	Cartagena-El Portus	0.010 ± 0.005	−1.04092	37.56756
Spain	Cabo de Gata	0.010 ± 0.005	−2.20269	36.71504
Spain	Cala Trebeluja-Menorca	0.010 ± 0.005	3.98231	39.91805
Spain	Palma de Mallorca	0.020 ± 0.005	2.63228	39.54290
Spain	Alcudia	0.010 ± 0.005	3.19141	39.80098
Spain	Valencia	0.010 ± 0.005	−0.22257	39.27232
Spain	Santa Eulalia-Ibiza	0.010 ± 0.005	1.54890	38.96663
Spain	Almeria	0.010 ± 0.004	−2.43079	36.80213
Spain	Málaga-El Candado	0.077 ± 0.008	−4.34607	36.70655
Tunisia	Bizerte	0.012 ± 0.003	9.91907	37.26940
Tunisia	Korbous	0.012 ± 0.005	10.53598	36.80728
Turkey	Igneada	0.32 ± 0.07	28.23704	41.85873
Turkey	Kilyos	0.25 ± 0.07	29.12371	41.30091
Turkey	R.feneri	0.24 ± 0.04	29.25281	41.24976
Turkey	Sinop	0.16 ± 0.12	35.31911	41.93896
Turkey	Unye	0.22 ± 0.13	37.44878	41.10645
Turkey	Yomra	0.23 ± 0.04	39.73335	41.10645
Turkey	Rize	0.23 ± 0.03	41.45645	41.37750
Turkey	Antalya	0.03 ± 0.01	30.94450	36.75677
Turkey	Botas	0.06 ± 0.02	36.06653	36.70341
Turkey	Akkuyu	0.030 ± 0.014	33.56729	35.96795
Turkey	Fethiye bay	0.020 ± 0.011	29.19216	36.58535
Ukraine	Cape tarkanhut	0.7 ± 0.2	32.77635	45.34573
Ukraine	Cara-dag	1.0 ± 0.2	35.41115	44.92717
Ukraine	Streletskay bay	1.5 ± 0.3	33.47149	44.58735

Values are expressed in Bq kg^{-1} wet wt ± 1σ propagated analytical uncertainty.

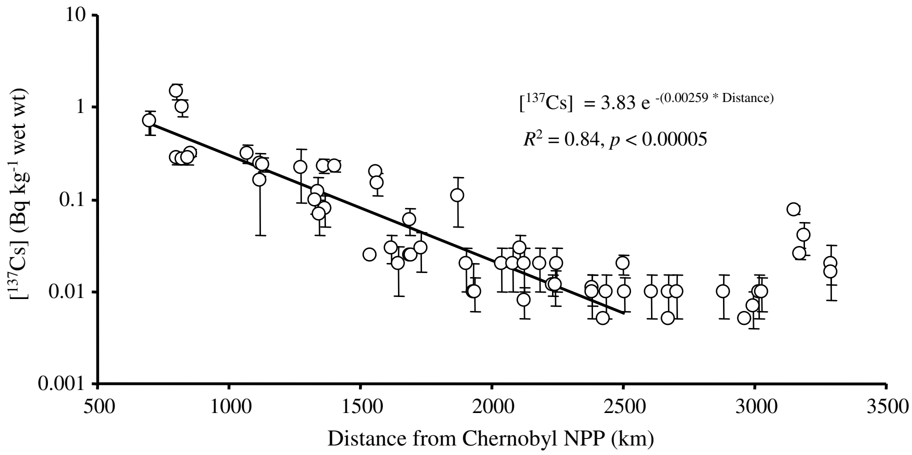

Fig. 2. Relationship between ^{137}Cs concentrations in mussels *Mytilus galloprovincialis* (Bq kg^{-1} wet wt) and distance from the Chernobyl Nuclear Power Plant (NPP) (km). Error bars indicate 1σ propagated analytical uncertainty. Nonlinear regression analysis has been carried out using Statistica© 6.0 software and taking into account data collected in a perimeter of 2500 km around the Chernobyl NPP.

followed closely in particular sectors of the Mediterranean and Black Seas, a view that is reinforced by most recent geo-political developments.

One remarks that seafood is a 3 billion euro a year industry in Europe with shellfish accounting for 790 million euro of this business (IFREMER, 2002). There is a real risk that these organisms may become unsuitable for human consumption due to their well-known ability to accumulate high levels of pollutants. In this respect, the CIESM scientific network has the unique potential of adopting common strategies to generate intercomparable data at the regional scale. Thereby, the levels of key contaminants can be reliably biomonitored in coastal areas, where most fishery and farming activities are carried out. Thus, following the successful completion of MMW Phase I, CIESM is now set to launch the second phase of its programme with the main objectives of: (1) broadening the network by the association of six additional Mediterranean countries, (2) extending the monitoring activities to other radionuclides and selected key trace- and emerging-contaminants, and (3) making all the data produced on a common basis fully available on-line through the MMW database now under construction (Rodriguez y Baena and Thébault, 2007).

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