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A Mediterranean Mussel Watch for radionuclides : what is the best information that can be gained?

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Bivalve molluscs and particularly mussels of the genus *Mytilus* have been used worldwide to document or "monitor" the existing levels and spatial and temporal trends in concentrations of a wide variety of marine contaminants. Many of these bioindicator, or "sentinel", monitoring exercises using mussels have often met with mixed success and resulted in merely identifying hot spots of contamination that were either already known or suspected beforehand (Goldberg *et al*., 1978, 1983; Farrington *et al*., 1983). Much of the ambiguity that may have arisen was partially caused by a poor understanding of the behaviour of the contaminant in the mussels, and as a result proper interpretation of the environmental data was difficult to achieve. One of the problems is that mussels often reflect only the bioavailable fraction of a contaminant, and in this respect, do not give a true picture of the existing ambient levels in the surrounding waters. Another is that for various physiological reasons mussels are poor sentinels for environmental contamination of specific heavy metals. Radionuclides like the long-lived transuranic elements are no exception because they too behave as heavy metals, although there is far less information on their physiological behaviour in mussels than exists for the conventional heavy metal contaminants. For this reason, it is wise to review some of the main environmental and physiological factors which may play a role in the bioaccumulation and retention of radionuclides that are measured in Mussel Watch activities.

A second important point to make is that in order to know what kind of useful information will be gained from this exercise, the project must have a well-defined goal, i.e., what is the specific purpose of gathering these data? A rather large perspective is needed, for ultimately one must understand the complex relationship between contaminant loading in the ambient environment, the mussel's response to it, and the actual entity or target being protected (White, 1984). This strategy has been underscored by an earlier Mussel Watch workshop (NAS, 1980) which stated that implicit in proposing a biomonitoring programme is having a conceptual model of the physical, biological and social systems affected. Furthermore, it would be impossible to construct any rational monitoring system without such a concept, and its development should be a primary goal of the proposed Mediterranean programme. Such a conceptual, holistic model might take the form of that shown in Fig. 1.

As radionuclide monitoring will be the first stage of this effort, what can be said about the application of this model to establishing criteria for monitoring radionuclides in mussels from around the Mediterranean? First of all, the radionuclide loads for the key nuclides (e.g., ^{137}Cs and $^{239+240}$Pu) are fairly well-known for coastal waters in many areas of the western Mediterranean. Most of the radionuclides entering the Mediterranean originate from atmospheric fallout with rel-

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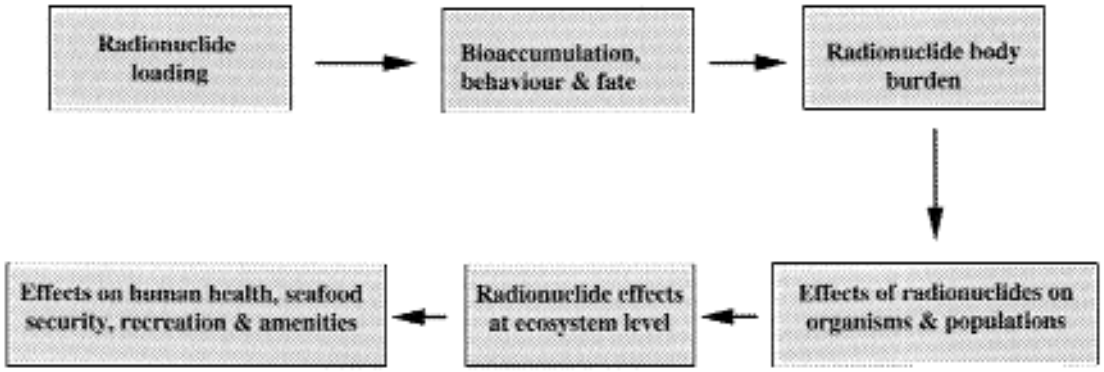


Fig. 1. Six box conceptual model for setting goals and priorities for monitoring radionuclides using sentinel organisms like marine mussels.

atively very small amounts coming down rivers like the Rhône, Ebro and Po. Hence, outside the proximity of those minor, localized source terms, most artificial radioactivity accumulated by Mediterranean mussels will be fallout-derived including minor amounts of ^{137}Cs from the Chernobyl accident which now are barely measurable in marine environmental samples. With respect to the second box in Figure 1, information on bioaccumulation, elimination and fate of artificial radionuclides in Mediterranean mussels (either *M. galloprovincialis* or *M. edulis*) is somewhat limited and restricted to only a handful of published papers (e.g., Keckes *et al*., 1967; Fowler *et al*., 1975, 1981; Guary and Fowler, 1977, 1978, 1981; Whitehead *et al*., 1988; Nolan and Dahlgaard, 1991; Fisher *et al*., 1996). Nevertheless, transfer rate information in this literature is important to take on board when establishing sampling strategies for the various radionuclides of interest, and for interpreting field data obtained from mussels living under the different environmental conditions found around the Mediterranean basin.

Obtaining data on existing radionuclide levels or "body burdens"in mussels (third box) has been the central focus of previous Mussel Watch efforts (Goldberg *et al*., 1978, 1983; Farrington *et al*., 1983; Vallette-Silver and Lauenstein, 1995), and it will likely remain so in the proposed project for the Mediterranean. However, in the case of radionuclides, focusing attention on the remaining boxes and protecting those entities as a primary goal of this project is open to debate. Most Mussel Watch programmes rightly have made pleas to research the relationships between contaminant body burdens, reproduction and survival (NAS, 1980), and some have included examining sub-lethal effects such as scope for growth measurements in mussels (Martin and Severeid, 1984). However, the focus there has been on toxic contaminants known to cause effects at the organism level such as POPs and certain toxic heavy metals. The case for artificial radionuclides is different in that wherever examined in the marine environment, no effects of ionizing radiation or radionuclide toxicity at extant ambient concentrations, or impacts of radionuclides, have been found in organisms or ecosystems (IAEA, 1976, 1988). Thus, based on both historical and scientific perspective, GESAMP (2001) have recently concluded that the "impacts" of authorized releases of radionuclides "*on human health and the environment at global and regional levels are generally of minor significance*". This is why according to GPA/LBA criteria for food security, public health and safety, marine resources and ecosystem health, the impact of radionuclides vis à vis all other types of contaminants is considered low or negligible (*ibid*.). Given these conclusions drawn from a broad spectrum of scientific study, it seems clear that the only realistic goal for a Mediterranean Mussel Watch focused on radionuclides will be to obtain radionuclide body burden levels on a spatial and perhaps temporal scale for use as baseline information in the event of any major accidents or deliberate releases in the future.

It also should be kept in mind that far more information is available on radionuclide levels in Mediterranean water and sediments than for concentrations in mussels. Hence, to date, most estimates of spatial and temporal trends of long-lived fallout radionuclides in the Mediterranean have come from broadbased, decadal monitoring and research studies of changes of concentrations and resultant inventories in the water from coastal and open sea areas (e.g., see Fowler *et al*.,

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2000). Such studies clearly show that fallout-derived radionuclide levels are decreasing in the Mediterranean as elsewhere. In the case of mussels, and likely for other sentinel organisms as well, comparable systematic biomonitoring data for radionuclides do not exist for the Mediterranean outside of "snapshot" measurements made in certain locations. For example, $^{239+240}$Pu analysis of mussels from Marseilles made some 30 years ago showed a concentration of approximately 40 mBq/kg dry in soft parts (Murray and Fukai, 1978). This concentration lies about midway in the range of values (1.2-88.4 mBq/kg dry) reported for $^{239+240}$Pu mussels from the US coast collected in 1990 (Vallette-Silver and Lauenstein, 1995). As fallout levels in the Mediterranean have fallen considerably since 1973, it is likely that present day plutonium levels in mussels will be somewhat lower. The same Mediterranean study showed that mussel shell contained roughly three times higher concentrations than soft parts. The increased concentrations of $^{239+240}$Pu in a shell sample coupled with the knowledge that residual plutonium is strongly retained in shell with little loss over time (Fowler *et al*., 1975), suggests that mussel shell may be a better integrator of past and present plutonium levels in the water than soft parts of the animal.

As mentioned above, in any monitoring programme involving the use of mussels as sentinels, it is important to keep in mind certain factors which can affect the levels of the radionuclides measured. As often stated, one chief disadvantage of using mussels is the rapidity with which they purge themselves of contaminants. However, for certain radionuclides this may not be the case as attested to by biological half lives for long-lived radionuclides like plutonium ranging from six months to two years (Fowler *et al*., 1975; Guary and Fowler, 1981). Nevertheless, a priori knowledge of elimination half-lives is important when establishing the sampling frequencies for a given radionuclide. Such transfer rates are often governed by temperature, salinity, the physical-chemical species of the radionuclide, and the amount of time and the manner in which the mussels have been exposed to the radionuclide. Thus, given the wide variety of environmental conditions under which mussels live in the Mediterranean coastal environment, some knowledge of the behaviour of the radionuclide in mussels will greatly assist in the interpretation of any observed variations in radionuclide body burden over time and space. Where radiotracer technology and appropriate experimental facilities are not available, carefully designed mussel transplant experiments at specific locations may furnish similar rate data. In any event, implementing a Mussel Watch exercise in which only information on body burden levels is obtained would be extremely short-sighted, and wherever possible ancillary data on radionuclide concentrations in the surrounding waters and sediments should be acquired, particularly in areas where measurements have not been made previously.

Another obstacle to obtaining basin-wide data for the Mediterranean is the fact that mussels do not exist or are poorly represented along many coasts on the eastern and southern shores. This limitation may be partially overcome by the use of non-endemic mussels transplanted to those locations (provided they can survive there), or by selecting a closely-related bivalve species although both these alternatives also carry some disadvantages.

It is hopefully evident from the facts presented above that an initial Mediterranean Mussel Watch for radionuclides can only have a limited goal, i.e. to obtain a basin-wide snapshot of current levels of long-lived radionuclides in mussels where they exist. Further temporal measurements may have some value only if sampling frequencies are properly determined, and this will have to be based on the targeted radionuclides. Such an effort should also depend on a thorough cost-benefit analysis with the real benefits of further basin-wide artificial radionuclide monitoring being carefully weighed.

While contaminant radionuclide monitoring is the primary goal of the initial stages of the proposed project, some additional effort should be made to establish levels of the natural radionuclide ^{210}Po in the mussel samples. This is important for two reasons. First, ^{210}Po is considered to be the most important contribution to the radiation dose received by humans via seafood consumption (Aarkrog *et al*., 1997) and information on extant levels in seafood from many areas of the Mediterranean is lacking. Second, elevated levels of ^{210}Po can enter the coastal waters through industrial, mining and agrochemical activities. Thus, enhanced ^{210}Po concentrations in mussels can provide a strong signal for various types of land-based activities which contaminate coastal areas. Therefore, with respect to technological enhancement of natural radionuclides, ^{210}Po could be considered as a marine contaminant.

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In conclusion, we should keep in mind the limited goals and scope of a Mediterranean Mussel Watch for radionuclides and use it to assemble a qualified group of analysts and environmental scientists who can devise a truly integrated monitoring network which uses a sentinel organism to gain more insight into existing levels of key radionuclides around the Mediterranean basin. When this initial task is accomplished, it would then be most profitable for the Mussel Watch project to turn its attention to an examination of the more high priority pollutants such as POPs, various heavy metals, and anti-foulants.

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