TOTAL BETA RADIOACTIVITY AND GAMMA SPECTROMETRICAL ANALYSES OF BLACK SEA WATER, FLORA AND FAUNA SAMPLES BETWEEN 1982-1983

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ABSTRACT

Results are presented for total beta radioactivity and gamma radionuclides in samples of water, algae, molluscs and fishes collected from the Black Sea during the period 1982-1983.

RESUME

Les données concernant la radioactivité bêta totale et les analyses par spectrométrie gamma d'échantillons d'eau de mer et de certains organismes (macrophytes, mollusques, poissons), de la mer Noire, pour les années 1982-1983 sont présentées dans ce travail.

The monitoring of natural and artificial radioactivity, both in the Romanian sector of the Black Sea and in the Danube river, was frequently performed during the last two decades (BOLOGA,1982). Generally, sediment, water and biota samples were analyzed by different procedures. Because of the interest in knowing the present levels of marine radioactivity resulting from atmospheric fallout and radionuclide transport by the Danube, total beta radioactivity and gamma spectrometrical analyses of environmental samples were carried out between 1982-1983.

MATERIAL AND METHODS

Sea water samples were collected nearshore along the Romanian Black Sea coast between Constantza $(44^{\circ}10^{\circ}N)$ and Jupiter $(43^{\circ}50^{\circ}N)$, and offshore the Danube Delta, at Constantza, Tuzla, Cape Caliacra and the prebosporic sector (up

to 30 nautical miles offshore). Biological samples were collected along the Romanian coast (up to 2 m depth) at Mamaia, Constantza, Agigea, Eforie Sud and Jupiter.

The sea water samples were evaporated to obtain salts. The marine organisms - macrophytes (whole thallus), molluscs (shell) and fishes (whole body) - were washed, weighed (fresh weight), ashed at 480°C and reweighed (ash).

Total beta radioactivity was assayed in aliquots of salt or ash for 1,000 s at least 5 times each using a low background Nuclear Enterprises beta counter with an efficiency of 15%.

Gamma spectrometrical analyses were performed by means of a Quanta-8100 analyzer with a Ge(Li) detector of 30 cm³. The counting time for each sample was about 60,000 s. The background of the instrument housed in a low background room was 0.9 cps in the 50-1,500 keV energy range. The spectra were processed by means of PDP-11/04 computer using modified SPECTRAN-III calculation programs (SIMA,1978).

In total 114 samples were analyzed for total beta radioactivity and 29 samples for gamma radionuclides. The activity due to 40 K was not subtracted (as it is in the reference data) from the measured values.

RESULTS AND DISCUSSION

Total beta radioactivity in sea water from the Romanian Black Sea coast is due mainly to 40 K and other natural radionuclides such as 226 Ra, 228 Ac, 232 Th, etc. In shallow water total concentration ranged between 24 - 68 pCi 1⁻¹ for 1982 and between 28 - 68 for 1983. In comparison on the Romanian shelf 48 pCi 1⁻¹ were measured in 1964 and 42 pCi 1⁻¹ in 1967 (GEORGESCU and SKOLKA,1970). During the period 1977-1978 values ranged between 6 - 29 pCi 1⁻¹ (GEORGESCU and SIRBU,1979). To a distance of 30 nautical miles offshore, the values ranged between 26 - 39 pCi 1⁻¹ in 1982 and between 28 - 57 in 1983.

Along the western coast of the Black Sea (off the Danube Delta, Cape Caliacra and the Bosporus) the values ranged between $12 - 41 \text{ pCi l}^{-1}$ during 1982. Off the Danube Delta, the value of 13 pCi l⁻¹ was situated between those levels which have been measured in Danubian fresh water

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 $(3 - 6 \text{ pCi } 1^{-1}, \text{ IORGULESCU } \text{et al.,1976} \text{ and } 0 - 22 \text{ pCi } 1^{-1},$ GEORGESCU and SIRBU,1979) and those for marine water (20 - 70 pCi 1^{-1}). Somewhat higher values of 42 and 37 pCi 1^{-1} were obtained off Cape Caliacra, and in the prebosporic sector, respectively. These data for both nearshore and offshore water are significantly lower than for example, the 4^{O} K radioactivity (287 pCi 1^{-1}) reported for the Nor-thern Irish Sea (WOODHEAD,1970), or that (331 pCi 1^{-1}) generally indicated for sea water (BURTON,1975).

Total beta radioactivity of the marine macroalgae <u>Enteromorpha linza</u> and <u>Ceramium elegans</u> during different seasons ranged between 12 - 70 and 16 - 78 pCi g⁻¹ ash, respectively, in 1982, and between 59 - 297 and 24 - 173pCi g⁻¹ ash, respectively in 1983. The differences appear to be due to variable calcination factors, not to the sampling site or season. For example, these values are very similar for both algae when expressed on fresh weight $(1 - 2 \text{ pCi g}^{-1})$. These values are similar to those previously obtained for the same species in 1978, i.e. 73 and 120 pCi g⁻¹ ash, respectively (BOLOGA and CHIOSILA, personal communication).

By gamma spectrometrical analyses of the principal marine macrophytes along the Romanian Black Sea coast (1983), the presence of one cosmogenic radionuclide (7 Be), three natural radionuclides (40 K, 226 Ra, 228 Ac) and eight fission products (54 Mn, 60 Co, 95 Nb, 95 Zr, 106 Ru, 125 Sb, 137 Cs, 144 Ce) was detected. Among the species analyzed, the highest 40 K content was found in <u>Cystoseira barbata</u>. Furthermore among these species, <u>Bryopsis plumosa</u> displayed relatively high levels of 226 Ra (and daughters) and 228 Ac (and daughters).

Total beta measurements of the marine molluscs (<u>Myti-lus galloprovincialis</u>, <u>Mya arenaria</u> and <u>Rapana thomasiana</u>) made in 1982 indicated that levels approached those of the natural background.

Total beta radioactivity in eight species of marine fishes sampled in 1983 ranged between <19 to 157 pCi g⁻¹ ash.

Gamma spectrometrical analyses of these fish species allowed determining the concentrations of 7_{Be} , ${}^{40}\text{K}$, ${}^{54}\text{Mn}$, ${}^{60}\text{Co}$, 106_{Ru} , 125_{Sb} , 137_{Cs} , 144_{Ce} , ${}^{226}\text{Ra}$ and ${}^{228}\text{Ac}$. For

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example the 40 K content ranged between 29 - 331 pCi g⁻¹ ash or between 1 - 20 pCi g⁻¹ fresh weight. The highest values were found in <u>Odontogadus</u> merlangus and <u>Sprattus</u> sprattus. The levels of the other radionuclides were very low.

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Discussion

S. DANALI-COTSAKI: Which methodology do you use for measuring gross beta activity in sea water? What is the percentage of salt in your samples? When you count gross beta activity in fish, do you also take into account the bones, or only the edible part? How do you explain the big concentration differences (11 - \sim 1000) in your algae samples? Is it a question of the season or only of the species?

A. BOLOGA: The sea water samples have been evaporated to salt (mean salinity of the Romanian Black Sea coastal waters is $\sim 15\%$) and aliquots of the salt have been used for measuring total beta radioactivity. The data on the total beta radioactivity of some marine fishes are based on whole body measurements (bones and edible parts). A big difference between the results concerning the total beta readioactivity among the analyzed marine algae is observed only for Bryopsis plumosa as compared to other species. Like other previous measurements of ours have shown, it seems to be a question of the species rather than season.

I. GEORGESCU: The macrophytes Cystoseira barbata and Enteromorpha linza are two species which have proved useful as bioindicators of artificial radionuclides in the Black Sea. There have been several previous studies using these species which were carried out by the Politechnical Institute at Bucharest.

A. BOLOGA: Experimental studies recently carried out at our Institute in Constanta have also confirmed the usefulness of these two species as indicators of 137Cs and 84Sr (Cystoseira) and 59Fe and 65Zn (Enteromorpha).