Application of the marine organisms as a “biological monitor” to evaluation of the background levels of radioactivity in the coastal environment

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Abstract. A study was carried out for investigating the availability of marine organisms such as a common species of brown and green algae (Sargassum thunbergii and Bryopsis sp.), and mollusks including some species of gastropods of the family of Buccinum and squids (Todarodes pacificus, Thysanoteuthis rhombus, etc.) for exploring in the marine environment the background levels of 99Tc, radium and its progenies, and 108mAg, respectively. The geographically wide distribution of the organisms above mentioned made it possible to compare the background levels of radioactivity from coast to coast in Japan. In addition, the comparison of the specific activity of 108mAg in the viscera of the mollusks among the species inhabiting in the layers at different depths allowed drawing a general trend of vertical distribution for the nuclide. All the organisms except gastropods studied here were known to have one year’s span of life. The burden of radionuclides in the organisms, therefore, resulted from the uptake not in the past years, but that in the most recent year during which they lived. They are expected to be used as a monitor extremely sensitive to the counterpart radionuclides. The application of biological monitors to the environmental monitoring programme would be useful especially for tracing the behaviour of long-lived radionuclides such as 99Tc (half-life: 2.111 × 10^5 y) and 108mAg (half-life: 418.21 y) along with naturally occurring radionuclides, which might be accumulated in the marine ecosystem.

1. Introduction

Although the radioactive contamination in the environment has lowered to the level at which humankind don’t receive any significant dose, it is still of importance to provide detailed information on the contaminant radionuclides in a monitoring programme, because of a growing concern of the public for the dietary safety and the environmental quality. The general public recognize the implication of food web through which hazardous substances are transferred to higher nutrition levels, eventually to humankind, and accumulated from the surrounding environment up to a great extent, especially in the aquatic ecosystem. It would be necessary, therefore, to organize the monitoring programme so as to obtain a full scope of the radiological situation in the environment of interest, and to give a full account of the results of monitoring in order to promote understandings of the public.

Marine organisms sometimes show a high affinity specifically to a chemical element or a radionuclide and accumulate it to the levels high enough to be readily detected by the ordinary measurement techniques. It would be possible to apply such organisms as a “biological monitor” to the environmental surveillance programme not only for just monitoring the releases of radionuclides but also for tracing the temporal and spatial changes of their distribution in the marine environment, although the level of radioactivity released under control are generally too low to detect directly in seawater. In addition, the “biological monitor” may provide information on the possible or maximum extent of contamination in marine products, even if the organisms are not used for human consumption.

The present study reports the results of an investigation on the concentrations of radionuclides such as 99Tc, 108mAg, and radium and its progenies in the marine organisms which were suggested to be suitable for a biological monitor by a preliminary screening study. The marine organisms used here were a common species of brown algae (Sargassum thunbergii), green algae (Bryopsis sp.), and mollusks including gastropods of the family of Buccinum, squids and so on. The former two artificial radionuclides mentioned above presumably originate from the worldwide fallout and still remain in the environment, although three decades have passed since the atmospheric releases from nuclear test explosions ceased. All the radionuclides of interest have extremely long half lives and are important from the radioecological viewpoint whether it may be artificial or naturally occurring, particularly with respect to establishment of the nuclear fuel cycle or radiation protection of the environment. The
measurements of stable elements were also carried out in order to determine the elemental composition, especially for the elements classified into the same group on the periodic table as the radionuclides studied. Thus, the background levels of radionuclides were determined for the coastal environment in Japan by using the data as indices.

2. Sampling and Analytical Methods

2.1. $^{99}$Tc in Brown Algae

It is well known that brown algae take up $^{99}$Tc analogously as iodine and accumulate it up to 1,500 times as high as the concentration in the surrounding water for Fucus serratus for example [1]. The species of brown algae such as Fucus vesiculosus and Ascophyllum nodosum representative both in the Irish Sea and in the North Sea are, therefore, often used in the monitoring programme in the marine environment for the liquid releases of radioactive effluents from the nuclear facilities [2]. The adjacent seas to Japan teem with algae and produce a variety of seaweed products which are consumed as food stuff and are the important source of iodine intake by the Japanese, although these two species are not always common in the North-west Pacific Ocean. In the present study, we used the species, Sargassum thunbergii, as a $^{99}$Tc monitor, taking into account of the results of the preliminary study on the comparison of the concentrations of the nuclide among several species of brown algae [3]. The species, Sargassum thunbergii, is not used for human consumption, but is likely to have advantageous features for the monitoring use; that is, Sargassum thunbergii is widely distributed along the coast of Japan and grows on rocks readily accessible in the intertidal zone.

As much as approximately ten kilograms of the algae was collected at each sampling site and thoroughly washed with seawater after transporting to the laboratory. The sample was divided into five sub-samples, weighed in a drying vessel, forced-air dried at 110°C and incinerated at 450 °C to ash in an electric muffle furnace. Each sub-sample reduced to ash was dissolved with 4M HNO$_3$ and $^{99}$Tc was extracted by tri-n-octylamine, followed by stripping with 5M NaOH and successively by back-extraction with 2-butanone as reported before [4, 5]. After being separated and purified, $^{99}$Tc was co-precipitated with 10 mg of Cu$^{2+}$ ion. The precipitate was filtered on filter paper and mounted on a stainless steel plate. The radioactivity of the nuclide was measured with a low-background beta-ray spectrometer (Fuji Electric Inc. Model PICOBETA PBS-1), which was composed of two types of detectors, namely GM gas-flow counter as a gate detector and plastic scintillation counter for the energy analysis designed to be operated in the coincidence mode, a beta-ray energy analyzing module for logarithmically converting the energy of beta-rays detected, and a pulse-height analyzer. Technetium-99 radioactivity was thus determined by averaging all the values obtained for the sub-samples. In addition to Sargassum thunbergii, $^{99}$Tc concentrations were determined for comparison for Ascophyllum nodosum which was obtained through an import trader in the form of dried meal produced in Norway and Canada.

2.2. Radium and its Progenies in Green Algae

Green algae may be of less economical importance as marine resources than brown algae, because of less numbers of edible species and less biomass, however, they are still a major member of organisms constituting an ecosystem in the intertidal zone. It would be necessary, therefore, to be fully aware of the role of green algae playing in the mass flow in the coastal environment. A preliminary screening study showed that a species of green algae, Bryopsis maxima, had a unique elemental composition, especially for alkali earth elements; the concentrations of Sr and Ba were 13,100 µg/g-dry and 4,630 µg/g-dry, respectively, whereas the respective average concentrations were 545 ± 491 µg/g-dry and 5.19 ± 4.89 mg/g-dry for 50 species of marine algae collected in Japan [6]. The present study examined the availability of green algae of the family of Bryopsidaceae as a biological monitor for radium classified into alkali earth metal same as Sr and Ba. The amount of several hundreds grams to some kilograms of algae was collected for Bryopsis maxima at the sampling points located in Ibaraki and Chiba, and for Bryopsis corticulans in Hiroshima. Samples were thoroughly washed with seawater in the laboratory, weighed, forced-air dried at 110 °C, incinerated to ash at 450 °C in an electric furnace. The sample
reduced to ash was put into a plastic cylindrical vessel having an inside diameter of 5 cm and 5 cm height after being pulverized in a alumina mortar and served for gamma spectrometry analysis using a Ge semiconductor detector (EG&G ORTEC Model GMX-30200) coupled with a PHA (EG&G ORTEC Model MCA-7700). The counting efficiency of the measuring device was determined by counting a set of the standard samples which had the same geometry as the samples and were tagged with known radioactivity $^{133}$Ba and $^{152}$Eu. Radioactivity of $^{226}$Ra was determined by calculating the peak area corresponding to 186.101 keV after taking into consideration of the background counts and also evaluating the contribution of $^{235}$U (185.7121 keV, 57.25%) by checking the presence of photo-peaks corresponding to 143.7642 keV (10.968%), 163.3582 keV (5.084%) and so on. On the other hand, radioactivity of $^{228}$Ra was derived indirectly from $^{228}$Ac radioactivity, which would reach the radioactive equilibrium to $^{228}$Ra within a few days.

2.3. $^{108m}$Ag in Mollusks

Some species of gastropods belonging to the family of Buccinidae, that is, B. isaotakii, B. striatissimum, B. tenuissimum, B. tsubai and Neptuna constricta, were collected from 2000 to 2003 at four fishing ports located rather in the northern part of Japan, where they were landed. They are commercially distributed usually within a local market and consumed as a local specialty. A total of some tens kilogram of each species was used for the present study. The soft part of an individual organism was separated from outside shell by cracking with a plastic hammer after weighing and heating in a microwave oven, and dissection into two parts, i.e. edible part including ventral muscular foot and proboscis, and viscera including digestive gland, gonad, gill and other organs. Each of edible part and viscera from all individuals was mixed and lumped together; thus, approximately 1 to 4 kilogram of composite sample of edible part and viscera was obtained for each specimen.

Squids were also collected at eight fishing ports and through three fish processing companies located mainly in the northern part of Japan. The fish processing companies usually deal not only inshore squids but also those caught in the pelagic fishery or imported from the countries mainly facing the Pacific Ocean. In the present study, one of the most commercially important species, Todarodes pacificus (Japanese common flying squid) was collected from all the points mentioned above another three species of the same family, Ommastrephidae; Ommastrephes bartramii (neon flying squid), Nototodarus sloani (Wellington flying squid) and Illex argentinus (Argentine shortfin squid) were obtained from the fish processing companies. In addition, Thysanoteuthis rhombus (diamondback squid) and Sepia esculenta (golden cuttlefish) were obtained at the ports facing the Japan Sea. The amount of some tens kilogram was used for the present study for each species. Individual organisms were dissected into three parts, i.e. mantle, arms including eyes and funnel and viscera including liver and other organs.

The samples were reduced to ash and prepared for gamma spectrometry analysis in the same procedures as algae samples as mentioned above.

Following the gamma spectrometry analysis, the samples were served for an analysis of stable silver. The amount of one gram of ash sample was taken in a tall beaker and completely decomposed with nitric acid and hydrogen peroxide on a hot plate. Nitrates dried up in the beaker were dissolved with nitric acid and hot water and transferred into a volumetric flask for adjusting the volume to 100 ml as well as the concentration of HNO$_3$ to 1N. Then, the concentration of silver in the solution was determined by using ICP-AES (Shimazu ICPS-7500).

3. Results and Discussion

3.1. $^{99}$Tc in Brown Algae

An example of beta-ray spectrum obtained for Ascophyllum nodosum by 50,000 seconds’ counting was represented in Fig. 1. Technetium-99 is a pure beta particle emitter and generally needs a beta-ray counting method after chemical separation of its fraction from the matrix of samples for determining its radioactivity, as long as a method of radioactivity measurement is adopted. It would be difficult,
however, to thoroughly get rid of possible radioactive contaminants such as U, Th, their progenies and other elements which could form anionic species such as Ru through chemical separation. It is advantageous to use the present technique for determining $^{99}$Tc radioactivity, not only because it enables to easily identify the presence of the nuclide on the spectrum by examining the energy of the end point in the higher side of the peak, which corresponds to the maximum energy of beta particle, i.e. 293.7 keV, but also because it enables to find out possible radioactive contaminants as shown in Fig. 1. The detection limit of $^{99}$Tc radioactivity was estimated to be approximately 13 mBq, when the measurement was carried out by a 600,000 seconds’ run at the precision level of 5%. Chemical yield through the procedures above mentioned was estimated to be approximately 70% by a test run using $^{95m}$Tc as a yield monitor and overall error was 20% [5].

![Beta ray spectrum obtained by 50,000 seconds’ counting for Ascophyllum nodosum.](image)

The result of measurements was represented in terms of geographical distribution of $^{99}$Tc concentrations in *Sargassum thunbergii* in Japan as shown in Fig. 2. The values given here may be underestimated probably by a factor of 0.7 because the chemical yield was not taken into account at all in the present study. It could be concluded that $^{99}$Tc originated principally from the worldwide fallout judging from the wide distribution of the nuclide and that the background level of $^{99}$Tc in *Sargassum thunbergii* would amount to some tens mBq/kg-wet at the utmost. There was not so great difference in the levels of the nuclide, in addition, between algae from the Japan Sea side and those from the area facing the Pacific Ocean, although the levels might be significantly higher partly in the northern area along the coast of the Japan Sea. Remarkably high concentration of the nuclide amounting to 1,500 mBq/kg-wet was observed in 1989 in Ibaraki Prefecture nearly in the central part of Japan Islands, where a total of twenty-one nuclear facilities and institutions including a nuclear spent fuel reprocessing plant as well as a power plant were located. Radioactive wastes released in liquid effluents from the nuclear facilities presumably contributed to a rise of the level of the nuclide in the adjacent coastal environment. The level of $^{99}$Tc in algae rapidly decreased thereafter to the level comparable to those observed in other locations in a few years.

The measurement of $^{99}$Tc was also carried out for the species of brown algae other than *Sargassum thunbergii*, including not only domestic species but also those of foreign growth. Two typical species taxonomically belonging to the family of *Laminariaceae* grown in Japan were served for the measurement; one was commercially available *Laminaria sp.*, which was one of the most important
algae for human consumption, and the other was *Eisenia bicyclis*, which played an ecologically important role as a member of algae forming seaweed beds in the coastal environment. The presence of $^{99}$Tc was not observed in *Laminaria sp.*, while relatively high concentration of $^{99}$Tc (503 mBq/kg-wet) was observed in the sample of *Eisenia bicyclis*, which was taken in 1987 at the same point in Ibaraki Prefecture as mentioned above. The presence of the nuclide was not observed any more, however, in the samples taken at the same time at the points some tens kilometers apart in the north. When it is taken into account that the concentration in *Sargassum thunbergii* collected simultaneously with *Eisenia bicyclis* was 276 mBq/kg-wet, the latter species might be an alternative as a biological monitor, although the former showed more advantageous features. The concentrations of $^{99}$Tc were 290 Bq/kg-dry and 3 Bq/kg-dry (58 Bq/kg-wet and 0.6 Bq/kg-wet supposing the ratio of dry weight to fresh weight was 20 %) respectively for two samples of brown algae, *Ascophyllum nodosum* offered by an import trader; one was imported from Norway and the other was from Canada. It is well known that a number of marine organisms including brown algae are collected and served for radioactivity measurements in the monitoring programme in the marine environment adjacent the nuclear facilities in England, and that high concentrations of $^{99}$Tc were often observed in *Ascophyllum nodosum* as well as in *Fucus vesiculosus*; e.g. 320 Bq/kg-wet in *A. nodosum* taken at Cape Wrath in Scotland in 2001[2]. Roughly speaking, the level of $^{99}$Tc was likely to be lowered by the oceanographic diffusion process by a factor 5 in the coast of Norway and 500 times in the coast of Canada. The levels of $^{99}$Tc in the coastal environment in the Northwest Atlantic, which did not have so heavy burden of radioactivity, still might be 5 times as high as those in the coast of Japan.

![Fig.2. Geographical distribution of $^{99}$Tc in Sargassum thunbergii along the coast of Japan.](image)

3.2. Radium and its Progenies in Green Algae

The biomass of green algae of the family of *Bryopsidaceae* is generally not large, although they are commonly distributed along the coast of Japan. An ordinary gamma spectrometry analysis using a Ge semiconductor detector would need the amount of some tens grams of samples reduced to ash at least in a routine programme. It may be not so easy to collect the corresponding fresh amount of algae of this family. When the difficulty in sample collection is taken into account, this species might not be suitable for a 'biological monitor', however, it should be noted that the algae accumulates alkali earth elements, especially elements of large mass number as mentioned above. Attention should be directed, therefore,
Radioactivity of $^{226}$Ra was determined not from activity of $^{214}$Bi but directly by calculating the area of the peak corresponding to 186.21 keV from itself. It could be concluded that the peak did not include any contribution from gamma-ray (185.71 keV, $I_{\gamma}$: 57.25%) from $^{235}$U, judging from the fact that none of the co-existing peaks expected for the nuclide (143.76 keV, $I_{\gamma}$: 10.97%, 163.36 keV, $I_{\gamma}$: 5.1%, etc.) were present on the spectrum. It is likely, in addition, that the presence of $^{238}$U was not significant in the sample, if any, because the presence of $^{234}$Th (63.29 keV, $I_{\gamma}$: 4.85%), decay product of $^{238}$U was not observed on the spectrum. Radioactivity of $^{228}$Ra was derived, on the other hand, from the activity of $^{228}$Ac at a radioactive equilibrium.

Radioactive concentrations of $^{226}$Ra in the species of Bryopsis ranged from 40 Bq/kg-wet to 92 Bq/kg-wet, whereas those of $^{228}$Ra varied more widely from location to location, ranging from 28 Bq/kg-wet to 211 Bq/kg-wet. The site dependence of $^{228}$Ra concentration in algae would reflect the extent of the burden of terrestrial radioactivity in the adjacent coastal environment, while $^{226}$Ra concentration was rather uniform in the marine environment owing to its long half-life (1,600y); namely, the inventory of $^{226}$Ra in the marine environment was large enough not to fluctuate by an additional input from the terrestrial environment. The result of the present study would agree with a well-known fact that the terrestrial radiation exposure is higher in the west part of Japan than in the east. The species of Bryopsis would be useful as a biological monitor for monitoring the effects of terrestrial radioactivity, which could be readily qualified by comparing the ratio of $^{228}$Ra to $^{226}$Ra in the algae as an index as shown in Fig. 4.

Special attention should be directed to the properties of the species of Bryopsis as a ‘hyperaccumulator’, from the radiation protection point of view. The hypothetical intake of the algae as foodstuff would result in radiation exposure to humankind, although the algae are not used for the purpose. The estimates of annual doses to an adult resulting from 5 grams of daily intake of the algae of the concentrations given in the present study would range from 0.06 mSv to 0.29 mSv altogether for $^{226}$Ra and $^{228}$Ra. The intake of algae could be one of the most critical pathways delivering radiation dose due
to naturally occurring radionuclides as well as to artificial radionuclides such as $^{90}$Sr. On the other hand, dose rates to the algae from radium and its progenies were estimated in the range from 1.1 nGy/h to 4.6 nGy/hr, supposing the internal dose rate per unit concentration given for leaves and needles of trees in the terrestrial environment given in Ref. [7] could be also used for the algae of the present study. The values were remarkably higher than those given for leaves and needles of trees. Care also should be taken, therefore, to radiation dose to algae themselves.

![Fig. 4. Site dependence of $^{228}$Ra/$^{226}$Ra ratio in algae, Bryopsis sp.](image)

3.3. $^{108m}$Ag in Mollusks

An example of gamma spectrum obtained for *Buccinum* by 200,000 second’s counting was represented in Fig. 5. The gamma spectrum for viscera of mollusks was usually characterized by prominent photo-peaks corresponding to 46.5391 keV, 63.292 keV, 92.381 keV, 92.802 keV, 185.7155 keV, which were emitted from $^{210}$Pb, $^{234}$Th and $^{235}$U. In addition to those from naturally occurring radionuclides, gamma rays at 433.937 keV, 614.276 keV and 722.907 keV from $^{108m}$Ag were observed for most of all the mollusks studied and that at 661.660 keV from $^{137}$Cs was also observed for squids. Radioactivity of $^{108m}$Ag and $^{137}$Cs was determined from the peak corresponding to 722.0907 keV and 661.660 keV, respectively. The least measurable levels of radioactivity concentration corresponded to approximately 15 mBq/kg-wet both for $^{108m}$Ag and $^{137}$Cs, when the threshold level was chosen three times of the fluctuation of the background continuum of spectrum. The $^{108m}$Ag concentrations observed in gastropods in the present study were slightly higher than the values of 10 to 30 mBq/kg-wet reported for oysters (*Crassostrea gigas*) from the early 1980s to the early 1990s on the northeast coast of Japan [8], whereas the $^{108m}$Ag concentrations in squids were comparable with those reported [9]. It would be worth notice the fact that the level of $^{137}$Cs was less than the detection limit in edible parts of snails, or much lower, if any, than in those of squids. Species of *Buccinidae* are commonly distributed in the coastal sea around the Japan Islands and inhabit in mud of the bottom at the depth of some hundreds to one thousand meters, where the $^{137}$Cs burden would be smaller than in the environment of surface water of the sea. These species are classified into carnivores from the standpoint of feeding habit and fed on bivalve mollusks, other gastropods and echinoderms as well as dead organisms which deposits onto the bottom. It is likely that the lower levels of $^{137}$Cs in species of *Buccinidae* than in squids reflected the smaller burden of $^{137}$Cs in the bottom environment than in the surface waters where squids lived their
entire lives, although some extent of the $^{137}\text{Cs}$ transfer occurred with deposition of dead organisms as a feed. On the other hand, the biological availability of silver was considered to be much higher than that of cesium, judging from the similarity of the vertical profile of silver concentration in the sea to that of nutrient elements such as phosphorus \cite{10}. Vigorous biological activity would deplete the level of $^{108}\text{mAg}$ released in the surface of the sea by incorporating into the existing silver cycling in the ecosystem and resulted in a rapid downward transfer. Thus, the level of $^{108mAg}$ would become comparable in the environment between the surface waters and the bottom of the sea.

The presence of stable silver was observed only in the viscera of mollusks in which radioactive silver was detected. A comprehensive study \cite{9} on radioactive and stable silver concentration in the liver of various species of squids showed that concentrations from 85.4 to 310 µg/g-ash were obtained for $Todarodes$ $pacificus$ caught off the coasts of Japan from 1991 to 1993, whereas the values from 250 to 411 µg/g-ash and from 58.8 to 188 µg/g-ash were observed respectively for $Ommastrephes$ $bartrami$ caught in the north-west Pacific from 1983 to 1992 and other species caught in the south Pacific from 1979 to 1993. The stable silver concentrations ranging from 25.0 to 94.8 µg/g-ash for $Todarodes$ $pacificus$ and from 9.5 to 10.4 µg/g-ash for pelagic squids in the present study were slightly lower than those reported. The values of $^{108mAg}$ specific activity were, however, comparable or a little bit higher than those reported.

A geographical distribution of the concentrations of $^{108mAg}$ in the viscera of mollusks along with its specific activity was represented in Fig.6. A relatively high value of $^{108mAg}$ radioactivity observed both for diamondback squid and $Buccinum$ $tenuissimum$ resulted from a high extent of accumulation of silver existing in its dwelling environment. Generally speaking, the $^{108mAg}$ specific activity fluctuated not so widely but within a rather narrow range corresponding to an order of magnitude at most. It is likely that there were no great differences in the burden of $^{108mAg}$ in the environment between the coasts facing to the Pacific Ocean and those to the Sea of Japan, because of similar values of $^{108mAg}$ specific activity between snails caught in the respective region of the sea. The comparable values of $^{108mAg}$ specific activity between the bottom-dwelling snails and the mid-water swimming squids suggested rather uniform distribution of $^{108mAg}$ from the surface to the bottom of the sea in comparison with the stratified distribution of $^{137}\text{Cs}$. It could be concluded that the $^{108mAg}$ specific activity in the viscera of mollusks was equal to that in seawater, so far as any discrimination was not caused in the silver cycling in the environment or in the silver metabolisms in organisms between radioactive and stable silver, for
instance. If a round value of 50 Bq/g is given to the $^{108m}\text{Ag}$ specific activity in the environment of the Sea of Japan, the inventory of $^{108m}\text{Ag}$ per unit area could be estimated to be 19 MBq/km$^2$, when the concentration of 0.28 ppb [11] for silver in seawater and the depth of 1,350 m for the mean depth of the Sea of Japan [12] both being taken into account. A study on the inventory of the fallout radionuclides in the sea showed that the total amount of $^{137}\text{Cs}$ in a water column was estimated to be comparable to the cumulative deposition of the radionuclides observed in the cities on the latitudes corresponding to the sampling points for the Pacific and to be less by approximately 30% than the terrestrial deposition for the Sea of Japan; that is, the total amount of $^{137}\text{Cs}$ in a water column were estimated as 127 mCi/km$^2$ and the respective terrestrial deposition were 147 mCi/km$^2$ from 1945 to 1977 for the Pacific, whereas $^{137}\text{Cs}$ in a water column were 130 mCi/km$^2$ and those of the terrestrial fallout were 173 mCi/km$^2$ for the Sea of Japan [13]. The cumulative radioactivity of $^{137}\text{Cs}$ due to the terrestrial deposition was calculated as 1.9 and 2.6 GBq/km$^2$ as of 1990 after correction of physical decay in two cities facing the Sea of Japan. If the amount corresponding to 70% of the total terrestrial fallout $^{137}\text{Cs}$ still remains in the Sea of Japan, the inventory of $^{137}\text{Cs}$ per unit area of the sea would be approximately 1.5 GBq/km$^2$. Roughly speaking, the inventory of $^{108m}\text{Ag}$ would, therefore, correspond to 1% of that of $^{137}\text{Cs}$ in the environment in the Sea of Japan and the value of the ratio would increase due with the course of time.

Fig. 6. Geographical distribution of $^{108m}\text{Ag}$ (mBq/kg-wet) and its specific activity (Bq/g, values in parentheses) in the viscera of various species of mollusks.

$\text{Tp}$: Todarodes pacificus, $\text{Ob}$: Ommastrephes bartramii, $\text{Ns}$: Nototodarus sloani, $\text{Ia}$: Illex argentinus, $\text{Tr}$: Thysanoteuthis rhombus, $\text{Se}$: Sepia esculenta, $\text{Bi}$: Buccinum isaotakii, $\text{Bs}$: Buccinum striatissimum, $\text{Bts}$: Buccinum tsubai, $\text{Bte}$: Buccinum tenuissimum, $\text{Nc}$: Neptunea constricta

Silver radioisotopes, particularly $^{110m}\text{Ag}$, are regarded as one of the most important radionuclides in the monitoring program to be carried out near the nuclear power plants, because of a high yield in the neutron activation process ($\sigma_\gamma$ of $^{109}\text{Ag}$: 4.72 b) as well as its relatively long half-life time (249.79 d). The element of silver is composed of two isotopes of mass number, i.e. $^{107}\text{Ag}$ and $^{109}\text{Ag}$, the natural abundance being 51.8397 and 48.4647%, respectively. When the neutron activation process produces unit radioactivity of $^{110m}\text{Ag}$ in a nuclear power plant, radioactivity of $^{108m}\text{Ag}$ to be simultaneously yielded can be estimated to be $1.3 \times 10^4$ times that of $^{110m}\text{Ag}$, if the cross section of $^{107}\text{Ag}$ ($\sigma_\gamma$: 0.338 b) being taken into account. On the other hand, the isomers of mass number of 107 would be yielded by
fission of $^{235}$U by neutrons at thermal energies, although the fission yield is fairly small (0.146 %) [14]. The corresponding amount of $^{108m}$Ag to the yield of the isomers may be produced in the fuels of the nuclear power plants. Not only nuclear power plants, but spent fuel reprocessing plants are likely, therefore, to be the source of release of the radionuclide into the environment. It would be meaningful to trace the existence of $^{108m}$Ag in the monitoring program as a measure for judging whether radionuclides released from the plant are accumulated in the environment or not, even if it does not respond the release from the plant so sensitively as other short-lived radionuclides. Mollusks can be used as a ‘biological monitor’ for this purpose and gastropods are possibly more advantageous than others, squids for example, because they provide information on the environmental quality specifically to the area of interest.

REFERENCES


