Thesis for Master Degree

Distribution of ^{210}Po and ^{210}Pb in the Pelagic Ecosystem around Jeju Island and in the Oysters and Mussels in Korean Coast

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Content

List of Tables

List of Figures

List of Abbreviation

$^{209}N_1$: Background subtraction of the alpha spectrum for detector geometry for ^{209}Po in the 1st count.

$^{209}N_2$: Background subtraction of the alpha spectrum for detector geometry for ^{209}Po in the 2nd count.

$^{210}A^{m}_{Po}$: Activity concentration of emitted ^{210}Po from the 2nd plating time to the 2nd counting time.

$^{210}A_{Pb}$: Activity concentration of emitted ^{210}Pb to ^{210}Po after ion exchange (i.e., the ion exchange column).

$^{210}A'_{Pb}$: Loss in activity concentration of ^{210}Pb from the sampling time to before ion exchange.

$^{210}A_{Pb-insitu}$: Activity concentration of ^{210}Pb at the sampling time (=^{210}Pb).

$^{210}A_{Po}$: Activity concentration of ^{210}Po at the 1st plating time before correction.

$^{210}A_{Po-ingrowth}$: The correction to ingrowth from ^{210}Pb to ^{210}Po.

$^{210}A_{Po-insitu}$: Activity concentration of ^{210}Po at the sampling time (=^{210}Po).

$^{210}N_1$: Background subtraction of the alpha spectrum for detector geometry for ^{210}Po during the 1st count.

$^{210}N_2$: Background subtraction of the alpha spectrum for detector geometry for ^{210}Po during the 2nd count.

A_{spike}: The first-added ^{209}Po as a yield tracer for ^{210}Po.

A^{2}_{spike}: The second-added ^{209}Po as a yield tracer for ^{210}Pb.

AED: The annual effective dose from foods.

CE_{Po210}: ^{210}Po internal dose coefficient change Bq to Sv.

CF: Concentration factor.

Chl-a: Chlorophyll-a.

C_{Po210}: Result of ^{210}Po activity concentration in this study.

I_{Po210}: Amount of ingestion to food per year per capita.

K_d: Distribution coefficient.

ηC: Recovery of ^{210}Pb.

SPM: Suspended particle matter.

λ_{Bi}: Proportionality constant of ^{210}Bi.

λ_{Pb}: Proportionality constant of ^{210}Pb.

λ_{Po}: Proportionality constant of ^{210}Po.

λ_{Pot}: Proportionality constant of ^{209}Po.

T_1: Time from the 1st mid-plating to the 1st counting mid-time.

T_2: The time from the 1st spike to the 1st plating mid-time.

T_3: The time from sampling to the 1st plating mid time.

T_4: The time from the 2nd mid-plating to the 2nd counting mid time.

T_5: The time from the 2nd spike to the 2nd plating mid time.

T_6: The time from the 2nd plating mid-time to ion-exchange column time.

T_7: The time from ion exchange/ion-exchange column to sampling time.

제주 주변 해역의 표영 생태계와 한국 연안의 굴과 홍합에서 ^{210}Po과 ^{210}Pb의 분포

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요 약

해양생물체에 많은 영향을 주는 자연방사성 동위원소인 ^{210}Pb과 ^{210}Po는 ^{238}U의 딸 원자인 가스형태의 ^{222}Rn으로 인해 해양생태계로 들어오게 된다. 각 분류군별로 서식지, 먹이를 먹는 방식, 먹이종류 등 여러 원인으로 인해, 각각의 해양생물은 상이한 ^{210}Pb과 ^{210}Po 체내 농도를 갖는다. 그로 인해 해수 대비 생물에 농축하는 방사성 동위원소 또한 다양한 범위를 보인다. 식품 섭취를 통한 ^{210}Po의 유효선량 중 많은 부분이 수산물에 의한 것으로 알려져 있으며, 그 중 연체동물은 ^{210}Po을 높은 농도로 축적한다는 연구 결과들이 있다.

제주 해역의 해양 영양단계(식물플랑크톤-동물플랑크톤-멸치-고등어)에 따른 ^{210}Po과 ^{210}Pb의 축적정도를 알아보았다. 제주 해역 표층해수의 총 ^{2210}Po과 ^{210}Pb의 농도는 0.83±0.004 와 1.27±0.03 $mBq{\cdot}kg^{-1}$이며, 해수 중 용존성 물질의 ^{2210}Po과 ^{210}Pb의 농도는 0.75±.06 과 1.22±0.09 $mBq{\cdot}kg^{-1}$이다. 식물플랑크톤의 ^{210}Po과 ^{210}Pb의 해수 대비 농축계수는 각각 1.5×10^5와 2.6×10^4로 ^{210}Po이 약 5배 더 높았다. 동물플랑크톤의 ^{210}Po농축계수는 식물플랑크톤과 비슷한 반면에 ^{210}Pb 농축계수는 약 5배 낮아 동물플랑크톤은 배설물을 통한 ^{210}Pb의 배출이 식물플랑크톤보다 상대적으로 더 빠르게 이루어지는 것을 보여주었다. ^{210}Po농축계수는 멸치가 플랑크톤에 비하여 수 배 더 높은 값을 보였

다. 반면에, 고등어의 근육은 멸치에 비해 ^{210}Po농축계수가 약 100배 이상 낮아 ^{210}Po은 상위 영양단계로 갈수록 낮아지는 것을 보였다. 멸치와 고등어의 내장 부위는 근육에서 보다 ^{210}Po농축계수가 8 - 38 배 높아 ^{210}Po 농축은 내장 부위에서 높게 이루어지는 것을 보였다. 상위 영양단계로의 ^{210}Pb 농축은 식물플랑크톤-동물플랑크톤-멸치로 가면서 각 영양단계마다 약 5배 감소하는 경향을 보였다. 이후 멸치에서 고등어로 전이되는 과정에서는 고등어의 근육과 내장부위의 ^{210}Pb농축계수가 멸치보다 30 - 70%로 이전 영양단계의 농축 보다 더 적은 감소가 이루어졌다.

한국은 굴과 홍합의 생산량과 자급률이 높다. 따라서 한국에서 생산되는 굴과 홍합의 ^{210}Po과 ^{210}Pb의 농도분포를 파악하고, 이들의 섭취로 인한 연간 유효선량을 추정해 보았다. 굴과 홍합의 ^{210}Po농도는 각각 41.3 - 206과 42.9 - 46.7 Bq·(kg·ww)$^{-1}$로 나타났다. 굴의 ^{210}Po농도는 서해안에서 상대적으로 높은 농도를 보였으며, 동일 해역 표층해수 중의 부유 물질 농도와 매우 밀접한 양의 1차 상관관계(R^2=0.89)를 보였다. 굴과 홍합의 ^{210}Pb농도는 각각 2.7 - 8.2와 2.0 - 4.2 Bq·(kg·ww)$^{-1}$로 나타났다. 동일 해역 표층해수 중의 부유 물질 농도와 비교적 밀접한 양의 1차 상관관계(R^2=0.74)를 보였다. 굴과 홍합 가식부의 ^{210}Po농도와 한국 성인의 굴과 홍합의 평균 섭취량으로부터 추정된 ^{210}Po 연간 유효선량은 각각 21 - 104와 5.01 - 5.46 μSv·y^{-1}였다. 한국에서 굴 섭취로 의한 ^{210}Po의 연간 유효선량은 다른 나라에 비해 비교적 높았으나, 홍합은 다른 나라에 비해 낮은 값을 보였다. 한국에서 굴과 홍합의 섭취로 인한 ^{210}Po의 연간 유효선량은 평균 76±42 μSv·yr^{-1}로 음식물 섭취로 인한 ^{210}Po의 연간 총 유효선량의 약 28±16%, 총 수산물 섭취 중 약 35±19%를 차지하는 것으로 나타났다.

KEYWORDS: ^{210}Po; ^{210}Pb; Bio Concentration Factor 생물농축; Annual Effective Dose 연간유효선량.

Chapter 1. Introduction

Polonium was discovered by Pierre and Marie Curie during their study on the radioactivity of uranium and thorium in 1898 (Figgins, 1961). They called it ‘Radium F’, later renaming it polonium after Marie Curie’s native land of Poland (Latin: Polonia). ^{210}Po emits a high-energy alpha (5.3 MeV) particle among the ^{238}U decay series, and this radioactivity amounts to 1.66 TBq· $^{-1}$, causing the substance to have about the main effective dose in marine biota (Argonne National Laboratory Environmental Science Division (ANL), 2007; Cherry & Shannoh, 1974). According to the hazard function (HF) model, intake of 1 MBq·(kg·bw)$^{-1}$ of ^{210}Po per day would shorten the life span to 28 days due to the total damage to the kidney and other organs with accompanying severe loss of lymphocytes, white blood cells, red blood cells and hemoglobin (Scott, 2007).

^{210}Pb and ^{210}Po flux in the earth consist of a variety of types. Their presence can be brought about naturally (resuspension of soil, sea salt spray, volcanic activity, and so forth) or by artificial fossil fuel burning (via tetraethyl lead combustion, dispersion of phosphate fertilizers and gypsum byproducts, etc). ^{210}Pb and ^{210}Po of the highest concentrations are generally natural in source; relevant in the ^{238}U decay series are daughter atoms of ^{222}Rn, which is emitted from ^{238}U in the crust of the earth as 99% of its total type. An inert gas of ^{222}Rn (half-life: 3.8 days) becomes ^{210}Pb (half-life: 22.2 years), ^{210}Bi (half-life: 5.0 days) and ^{210}Po (half-life: 138.4 days), through to the short half-life daughter atoms (Fig. 1). ^{210}Pb of the metal atom is dispersed (both on land and on the surface of the ocean) via precipitation and attached aerosol. The in-flow of radionuclides on the land was re-suspended due to the transpiration and dust; the in-flow in the ocean settled as per Stokes' law, by attaching to a suspended particle (Karali et al., 1996; Pietrzak-Fil & Skowronska-smolak, 1995; Preiss et al., 1996).

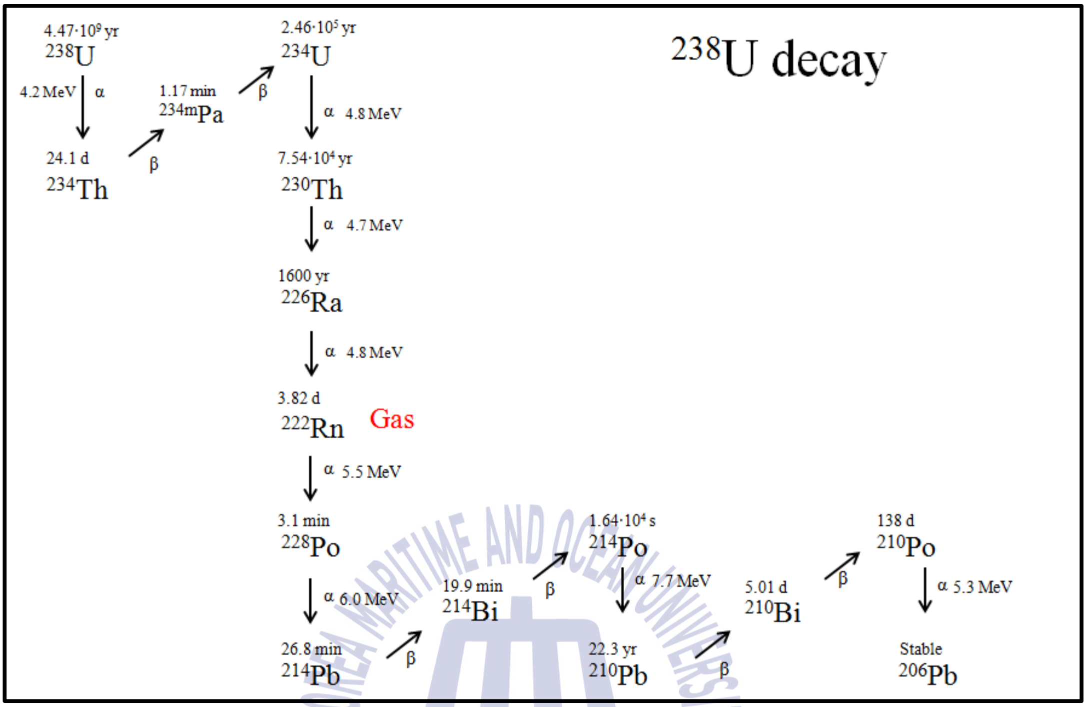

Fig. 1 The ^{238}U decay chain

^{210}Pb is present in biota in high concentrations, specially ^{210}Bi and ^{210}Po, which are the ^{210}Pb daughter atoms and have a high activity concentration in the marine biota (Holtzman, 1996). Among the natural radionuclides, ^{210}Po is the one that most effectively accumulates in marine biota, replacing sulfur, selenium, and tellurium as the same 16 periods with ^{210}Po in a periodic table of the elements (Cherry & Heyraud, 1981). ^{210}Pb and ^{210}Po, though showing high accumulations in marine biota, also show variations along a range that is determined by the surrounding environment and the taxonomic group of the biota. Activity concentrations of ^{210}Pb and ^{210}Po in marine biota differs from site to site according to whether a site has or does not have industrial sewage, whether the amount of suspended matter due to water current is different, and also the amount of ^{238}U (Fowler, 2011; Samad et al., 2010; Štrok & Smodiš, 2011). Activity concentrations of ^{210}Pb and ^{210}Po change due to the type of marine biota; each taxonomic group has a different habitat, feeding behavior, and feeding type. Mollusks in particular have high radioactive concentrations of ^{210}Po, from 1.7 times to up to a maximum of 40 times of that in other marine biota (Aközcan & Ugur, 2013; Alam & Mohamed, 2011a; Heyraud & Cherry, 1979). Most mollusks are suspension feeders, and take up both directly suspended particulate matter in seawater and food organisms such as phytoplankton, which contain heavy metals and other toxic substances including in situ algae-produced bio-toxins (Anderson et al., 2002). In many cases, heavy metals in oysters and mussels have been used to monitor concentrations of dissolved heavy metals in seawater (Goldberg et al., 1983; Rainbow, 1995). High concentrations of radionuclides can also be monitored using oysters and mussels (Connan et al., 2007; Rožmarić et al., 2012). Feeding type and source of food also lead to organisms having higher or lower concentrations of radionuclides. Planktivorous fish are found to have higher radioactive concentrations of ^{210}Po than are the carnivorous fish of upper trophic level (Aközcan & Ugur, 2013; Cherry et al., 1989; Lazorenko et al., 2002).

Instance in which suspended particles (floating particles) in ^{210}Po is more present in a given organism as compared to those who show dissolved ^{210}Po on testing more likely reflect the ingestion of heavy metals (Bacon et al., 1976). Concentration factors change according to the radioactivity concentration in seawater. Therefore, concentration factors of ^{210}Pb and ^{210}Po are determined according to what sea is tested. Previous research about the concentration factor (CF) of ^{210}Pb and ^{210}Po following the food chain of the marine biota in the pelagic and bottom ecosystem in the southern Atlantic revealed a higher range of CF for ^{210}Po plankton (1×10^4 - 1×10^6) than for fish (1×10^3 - 7×10^5). Specifically, planktivorous sardines (7×10^5) showed higher ^{210}Po CFs than did carnivorous tuna (5×10^3) (Carvalho 2011; Carvalho et al. 2011). The CF of ^{210}Pb and ^{210}Po following trophic levels of the phytoplankton (8×10^4), zooplankton (5×10^5), fish (4×10^5) was researched in Korea in the Donghae (Suh et al., 1995). Currently, most research is examining each CF without following the food chain (Alam & Mohamed, 2011a; Aoun et al., 2015; Lubna et al., 2011).

The CFs of ^{210}Pb and ^{210}Po in the marine-ecosystem food chain is not frequently researched in Korea. The predominant types of fish in the South-North area around Jeju Island include anchovies (*Engraulis japonicus*) and mackerels (*Scomber japonicas*), which comprised 28% and 18% of the total fish among the 51 species across four seasons for which measurements were available (Korea institute of Ocean Science & Technology (KIOST), 2005). Diatom and copepod composed a high share of the plankton in the study area; copepods showed a high share in the Kuroshio warm current, including study area (Kim et al., 2013; KIOST, 2005). The process of zooplankton consuming phytoplankton helps conduct the primary food source toward the high-trophic level biota (Kim et al., 2013). A shoal of anchovy has spread around the Korean peninsula recently (March 2016). These anchovies prepared for winter in South area around the Korean peninsula; this location is where they take in phytoplankton and zooplankton, and they then selective feed on copepod in the summer (Kim et al., 2013; National Fisheries Research and Development Institute (NFRDI), 2010). Mackerel moved along the Tsushima warm current and the East China Sea warm current due to the seasonal migration. Following the Tsushima warm current, the presence of mackerel spread to the east sea of Korea; the main spawning season was April to May (NFRDI, 2010). Mackerel also are predators of anchovy; in a previous study, of the mackerel caught in the South Sea of Korea, 37% had anchovy in their digestive systems, and anchovy is generally considered a large part of mackerel's diet (Yoon et al., 2008). Chapter 3 shows the measured activity concentration of ^{210}Pb and ^{210}Po in each biota and analyzes the transfer of ^{210}Pb and ^{210}Po according to the trophic level of the phytoplankton, zooplankton, anchovy and mackerel in the sea area around Jeju Island.

According to the concentration factor and activity concentration in marine biota, radionuclides are accumulation in people through seafood intake (Cherry & Shannon, 1974; Skwarzec & Falkowski, 1988). The annual effective dose is commonly divided into medical exposure and natural exposure of radiation. The annual effective dose through food intake (effective radionuclides due to the decay of the radionuclides uranium and thorium) is 0.34 $mSv \cdot yr^{-1}$. The effective dose of ^{210}Po at 0.28 $mSv \cdot yr^{-1}$ occupies about 84% of the total annual effective dose. Eighty-seven percent of this annual effective dose from food is from seafood, at 0.25 $mSv \cdot yr^{-1}$ (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000; International Atomic Energy Agency (IAEA), 2011). Previous research in different countries has confirmed that ingestion of ^{210}Po is high due to consumption of seafood (Lee et al., 2009; Ota et al., 2009; Pietrzak-Flis et al., 1997; Sivakumar, 2014).

Oysters and mussels (as mollusks) have high concentrations of ^{210}Pb and ^{210}Po, with various ranges of activity concentrations for ^{210}Po. Consequently, the annual effective dose of ^{210}Po can also easily change following consumption of oysters and mussels (Aközcan, 2013; Connan et al., 2007; Khan et al., 2014; Lee & Wang, 2013; Rani et al, 2014; Rožmarić et al., 2012; Štrok & Smodiš, 2011). The production rates of oysters and mussels in Korea are very high. The Food and Agriculture Organization of the United Nations (FAO) Aquaculture Production Dataset (Fish Stat, 2002- 2012) reported that Korea produces more than 3 times the average world oyster production and 1.5 times more than the average mussel production. However, the level of activity of ^{210}Po and the effective dose from the digestion of oysters and mussels in Korea has not been researched in spite of the high levels of production and consumption. Chapter 4 discusses research in which the distribution of ^{210}Po was investigated in the tissues of oysters and mussel collected in a major production area in Korea, along with the annual effective dose of ^{210}Po from the intake of oysters and mussels.

Chapter 2. Materials and Methods

2.1 Sampling

Sampling for the determination of ^{210}Pb and ^{210}Po levels in oysters, mussels, and fish from the sea areas around Korea was carried out in 2013 and 2014. Seawater, plankton, anchovy and mackerel were collected in and around Jeju Island (Fig. 2).

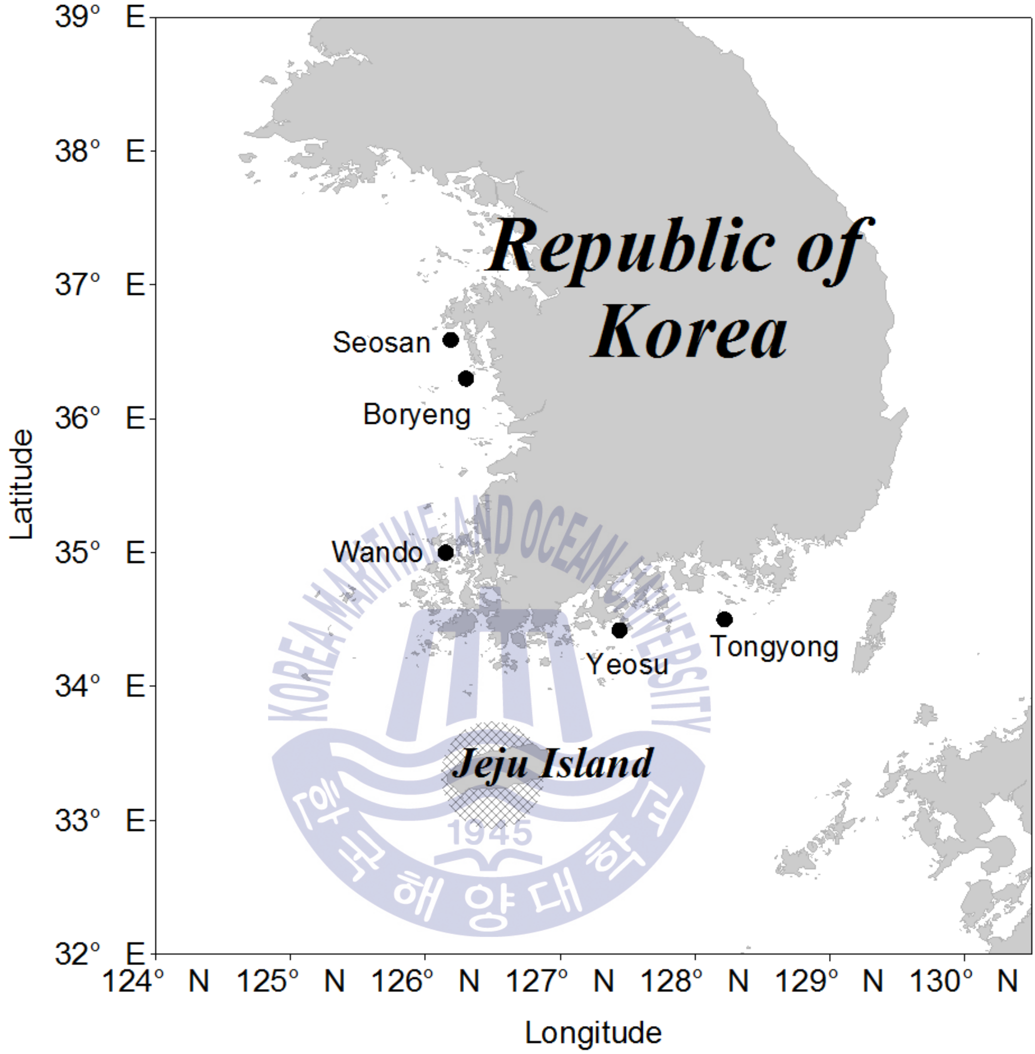

Fig. 2 Study sampling locations (shown with cross lines and dots)

2.1.1 Seawater sampling procedure

Seawater sampling was carried out using a pump in May 2014 in and around Jeju Island, 2 km away from the harbor of Ham-Duk. Approximately 20 kg of seawater was filtered through a 0.45 μm cartridge filter. Unfiltered total seawater, filtered seawater, and total seawater were analyzed separately. All seawater samples were acidified with 50 ml guaranteed-pure (GR) concentrated HCl to 6 mol·L^{-1}, followed by addition of stable Pb and ^{209}Po (A_{spike}) as a yield tracer. For seawater in which there was known to be a specific concentration of suspended particulate matter (SPM), the SPM content was determined using an 0.4 μm filter, and the non-filtered water was packed into a 4 L sterilization water bottle after separation from the filtered 1 L of seawater.

Manganese was used to extract polonium (^{209}Po and ^{210}Po) from seawater samples by adding saturated KMnO4 and 0.4 mol·L^{-1} $MnCl_2$ (ratio 1:2) to the acidified seawater sample. After equilibration for several hours, the solution was brought to pH 8 - 9 with $NH_4(OH)$, and precipitates isolated by centrifugation.

2.1.2 Plankton collection

Samples were collected using a marine phytoplankton net with a mesh size of 20 μm to 300 μm and a marine zooplankton net with a mesh size of 300 μm. Some of the tiny zooplankton was possibly mixed in the phytoplankton because size was used to separate the two. The phytoplankton and zooplankton were towed in surface waters to the sea water sampling site, which was located in close proximity to their collection.

2.1.3 Fishes collection and sampling procedure

Fish samples were collected from the local market (Hanlim-Hang) with assistance from the Korea Fisheries Resources Agency. Two species were selected for analysis according to their feeding behavior and trophic level: one of them was the planktivore fish Japanese Anchovy (*Engraulis japonicus*) and the other the carnivore fish chub Mackerel (*Scomber japonicus*). Mackerel was generally dissected for analysis of radionuclides in the muscles, skin, and internal tissues (liver and whole internal tissue). Anchovy was divided into head, muscle, and internal tissues. These preparations were performed to determine the concentration of ^{210}Po and ^{210}Pb in each part of the body.

2.1.4 Oyster and mussel collection and sampling procedure

Oysters (*Crassostrea gigas*) (average shell length of 10 cm) were collected from 5 hanging culture farms (Seosan, Boryeong, Wando, Yeosu, Tongyeong) and mussels (*Mytilis coruscus*) were collected at 2 sites (Yeosu, Tongyeong) from November 26 - 29, 2013. Soft tissues were separated from the collected oysters, which were rinsed using distilled water to remove sale, then frozen at -15°C. A freeze-dryer was used to lyophilize the frozen samples for at least 36 h, and dried power was produced with a grinder.

2.1.5 After sampling

All samples were transferred at a temperature of – °C without seawater samples. To reduce errors, each sample (minimum weight, 10 kg) was divided into 3 bundles after being ground and shaken. Stable Pb and ^{209}Po (A_{spike}) served as an added yield tracer in all samples. After preconditioning, the count for ^{210}Po sampling was performed as soon as possible because of its short half-life. Wet or dried samples were added in the Teflon beaker.

2.2 ^{210}Po analysis

2.2.1 Digestion of samples

Samples were placed in the 250 ml Teflon beaker, spiked with an aliquot of ^{209}Po as a yield tracer (A_{spike}), and digested with a concentrated HNO_3 solution (Nitric acid 65%, Merck KGaA Darmstadt, Germany) and H_2O_2 (Hydrogen peroxide, Junsei chemical Co. Ltd, Japan). The mixture of samples and acids were evaporated near dryness at 80°C to 100°C. The remaining residue was digested using concentrated HF (Suprapur Hydro fluoric acid 40%, EMD Millipore corporation, USA) to concentrated HNO_3. Samples were heated at 90°C for 3 hours. The digested solution was evaporated to near dryness at 80°C. To change the remaining HNO_3 to HCl, concentrated HCl (Hydrochloric acid fuming 37%, Merck KGaA Darmstadt, Germany) was added in the beaker and evaporated repeatedly (about 10 times) to dry it completely. Last, the remaining residue was diluted with 100 ml 0.5 mol·L^{-1} HCl solution. Triplicate samples were used to reduce errors.

2.2.2 Preparation of the silver planchet

The silver planchet (99.9% Ag, Φ 24.1 mm × 0.15 mm, Aldrich) was washed with acetone for 1 hour, and acetone was then removed using dust-free tissue. A thin layer of enamel paint was applied to one side of the planchet; it was then punched to make a hole for hanging thread (Fig. 3).

2.2.3 Spontaneous plating

One-half gram of ascorbic acid was added into the sample (which was diluted with 0.5 mol·L^{-1} HCl). The magnetic stirrer speed was adjusted to 220 rpm; the researchers subsequently waited for 30 minutes for the ascorbic acid to dissolve and combine with Fe^{3+}. We then placed the silver planchet in the beaker for 15 hours (Lee et al., 2014). ^{209}Po and ^{210}Po were spontaneously plated on the silver planchet (Fig. 3). After plating, the planchet was rinsed with distilled water, labeled, and put in a well-sealed plastic bag.

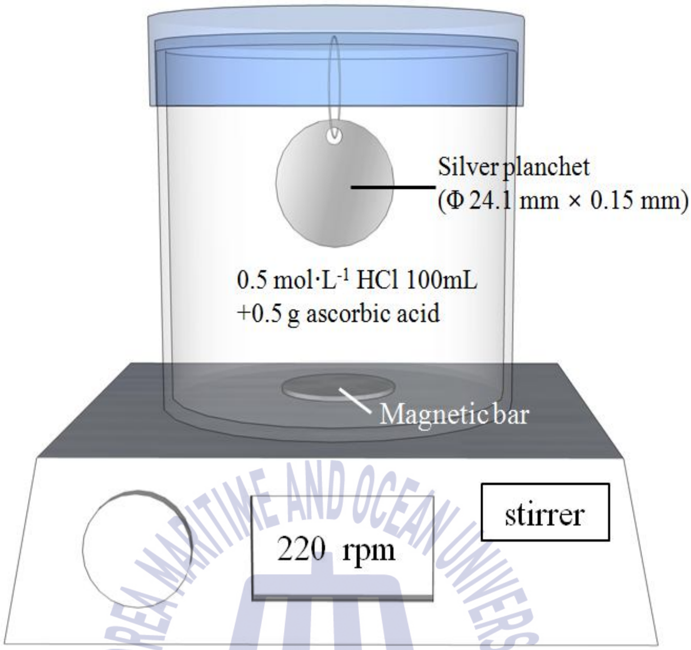

Fig. 3 Spontaneous plating set for ^{210}Po onto an Ag planchet.

2.2.4 Alpha counting

The silver planchet was counted to over a 1000 count using an alpha spectrometer (Canberra series 35 MCA, equipped with an A450-18AM PIPS detector with an active surface area of 450 mm^2). The full width at the half maximum of detection is < 20 KeV. The counter baseline was estimated for a more accurate result, with the baseline considered as 1 counter per hours above 3 MeV. The activity concentration of ^{210}Po was corrected from those of ^{210}Pb measured after one and a half years. The following formula was used for the ^{210}Po calculations [1-4].

$$^{210}A_{Po}(dpm) = \frac{^{210}N_1}{^{209}N_1} \times E^{\lambda_{Po} T_1} \times E^{-\lambda_{Pot} T_2} \times A_{spike} \quad [1]$$

$$^{210}A_{Po-ingrowth} = {}^{210}A_{Pb-insitu} \times \quad [2]$$

$$\left[\frac{\lambda_{Bi} \times \lambda_{Po} \times E^{-\lambda_{Pb} T_3}}{(\lambda_{Bi} - \lambda_{Pb})(\lambda_{Po} - \lambda_{Pb})} + \frac{\lambda_{Bi} \times \lambda_{Po} \times E^{-\lambda_{Bi} T_3}}{(\lambda_{Pb} - \lambda_{Bi})(\lambda_{Po} - \lambda_{Bi})} + \frac{\lambda_{Bi} \times \lambda_{Po} \times E^{-\lambda_{Po} T_3}}{(\lambda_{Pb} - \lambda_{Po})(\lambda_{Bi} - \lambda_{Po})}\right]$$

$$^{210}A_{Po-insitu}(dpm) = ({}^{210}A_{Po} - {}^{210}A_{Po-ingrowth}) \times E^{-\lambda_{Pb} \times T_3} \quad [3]$$

$$^{210}A_{Po-insitu} = (Bq \cdot kg^{-1}) = {}^{210}A_{Po-insitu}(dpm) \times (sample)(kg) - 1 \times 10^{3} \cdot 60^{-1} \quad [4]$$

$^{210}A_{Po}$ represents the activity concentration of ^{210}Po in the plating time before correction; Accurate measurement of ^{210}Po is required for correction of the ^{210}Po activity concentration, which grows in the ^{210}Pb during the interval between sampling time and plating time ($^{210}A_{Po-ingrowth}$). The activity of $^{210}A_{Po-insitu}$, after the correction to ingrown Pb, excludes $^{210}A_{Po-ingrowth}$ at $^{210}A_{Po}$ and then allows the correction of the activity concentration of ^{210}Po, which is emitted during the period from sampling time to plating time. Using formula [4] allows the correction of weight and units for activity concentrations of ^{210}Po in the samples (GEOTRACES Standards and Intercalibration (S&I), 2014).

2.3 ^{210}Pb analysis

2.3.1 Ion-exchange column

The residue of samples after spontaneous plating was evaporated almost to the point of burning. The remaining residue (ascorbic acid) was digested using concentrated HNO_3. After digestion, samples were evaporated repeatedly (about 3 to 4 times). At the end of evaporation, the HNO_3 medium was changed to 9 $mol \cdot L^{-1}$ HCl.

The remanent polonium ion was removed for use in the AG® 1-X8 ion-exchange column (100 - 200 mash chloride form, Bio-Rad Laboratories, Inc. USA). The ionic passage through the ion-exchange column was collected in a 25 ml PE bottle. The samples with added ^{209}Po (A^{2}_{spike}) tracer were kept during 6 months. To measure the recovery of the tracer, 1-ml of the passage through the ion-exchange column samples was saved.

2.3.2 Recovery of Pb

Stable Pb for recovery was measured in saved 1 ml samples after passage through the ion-exchange column by inductively coupled plasma mass spectrometry (ICP-MS) (X serise ICP-MS, Thermo Fisher Scientific Ico., USA). All samples were diluted with 1 mol·L^{-1} HNO_3 because ICP-MS cannot handle high-concentration materials. The Pb_{input}/Pb_{output} ratio is known that measures stable Pb by ICP-MS.

2.3.3 ^{210}Pb analysis

Recovery of ^{210}Pb (ηC) is measured in remanent stable Pb by ICP-MS. In the samples that were kept for 6 months, ^{210}Po was analyzed in the same way it was ^{210}Po analyze was analyzed to same method of 2.2 ^{210}Po analyze. ^{210}Pb was calculated using the following formula [5-9] (S&I, 2014).

$$^{210}A^{m}_{Po}(dpm) = \frac{^{210}N_2}{^{209}N_2} \times E^{\lambda_{Po}T_4} \times E^{-\lambda_{Pot}T_5} \times A^{2}_{spike} \quad [5]$$

$$^{210}\mathrm{A}_{\mathrm{Pb}}(\mathrm{dpm}) = \frac{^{210}\mathrm{A}^{\mathrm{m}}_{\mathrm{Po}}}{(1 - E^{-\lambda_{\mathrm{Po}} T_6})} \quad [6]$$

$$^{210}\mathrm{A}'_{\mathrm{Pb}}(\mathrm{dpm}) = \frac{^{210}\mathrm{A}_{\mathrm{Pb}}}{\eta \mathrm{C}} \quad [7]$$

$$^{210}\mathrm{A}_{\mathrm{Pb-insitu}}(\mathrm{dpm}) = {}^{210}\mathrm{A}'_{\mathrm{Pb}} \times E^{\lambda_{\mathrm{Pb}} T_7} \quad [8]$$

$^{210}\mathrm{A}^{\mathrm{m}}_{\mathrm{Po}}$ was known to express the ^{210}Po emitted between the plating time and counting time. $^{210}\mathrm{A}_{\mathrm{Pb}}$ represents the emitted ^{210}Pb to ^{210}Po ratio after the ion-exchange column based on $^{210}\mathrm{A}^{\mathrm{m}}_{\mathrm{Po}}$. $^{210}\mathrm{A}'_{\mathrm{Pb}}$ represents how much ^{210}Pb was lost by the samples before the ion-exchange column, and is then corrected to the activity concentration of ^{210}Pb at the sampling time ($^{210}\mathrm{A}_{\mathrm{Pb-insitu}}$). Formula [9] is used to correct the sample weight and result uit.

2.4 Q/A and Q/C

The reference material IAEA-414 (Radionuclide in mixed fish from Irish Sea and North Sea) was determined simultaneously for Q/A and Q/C of this study. Each sample was tested in triplicate. The certified value of the reference material is 2.1 ± 0.4 Bq·(kg·dw)$^{-1}$. The measured activity concentration of the reference material in this study was within the certified value (Table 1).

Table 1 Analytical accuracy of ^{210}Po using the reference material, IAEA-414

(radionuclides in mixed fish from the Irish Sea and North Sea)

(unit: Bq·(kg·dw)$^{-1}$)

Median (95% Confidence interval)	This study	Remark
2.1 (1.8 - 2.5)	2.2 ± 0.1	n=3

Chapter 3. Concentration factor of ^{210}Pb and ^{210}Po with the trophic level of phytoplankton, zooplankton, anchovy and mackerel in the coastal water of Jeju Island, Korea

3.1 The distribution of the activity concentration of ^{210}Po and ^{210}Pb in the coastal water and in plankton

The mean ^{210}Po concentrations measured in total phases and dissolved phases of seawater were 0.83 ± 0.004 and 0.75 ± 0.06 mBq·kg^{-1}, respectively, while the mean ^{210}Pb concentrations measured in total phases and dissolved phases of seawater were 1.27 ± 0.03 and 1.22 ± 0.09 mBq·kg^{-1}, respectively, in Jeju Island at the collection date (May 2014) (Table 2). This study, which measured the activity concentration of ^{210}Po and ^{210}Pb in the surface water of the coastal water around Jeju Island, was included in previous research that measured the activity concentration of ^{210}Po during the total phase (0.65 – 05 mBq·kg^{-1}) and dissolved phase (0.26 – 48 mBq·kg^{-1}) and of ^{210}Pb during the total phase (0.9 – 35 mBq·kg^{-1}) and the dissolved phase (0.3 – 38 mBq·kg^{-1}) in the surface water of the coastal water around the Korean peninsula (Hong et al., 2008; Hong et al., 1999; Kim & Kim, 2014; Kim & Yang, 2004). The activity concentration of ^{210}Po for the total and dissolved phases in the East China Sea and the Yellow Sea were close to those found in the coastal water of Jeju Island (0.65 –)5 and 0.26 –)3 $mBq \cdot kg^{-1}$, respectively), while the total-phase and dissolved-phase concentrations of ^{210}Pb were 0.9 – 35 and 1.13 – 38 $mBq \cdot kg^{-1}$, respectively. These previous results include the results of this study (Nozaki et al., 1991).

Table 2 The activity concentrations and distribution coefficients of ^{210}Po and ^{210}Pb in the coastal surface water around Jeju Island

(unit : $mBq \cdot kg^{-1}$)

Seawater	Total	Dissolved	Particulate	K_d
^{210}Po (± STD)	0.83 ± 0.004	0.75 ± 0.06	0.08 ± 0.06	$1.9 \times 10^5 \pm 2.6 \times 10^5$
^{210}Pb (± STD)	1.27 ± 0.03	1.22 ± 0.09	0.05 ± 0.06	$6.6 \times 10^5 \pm 3.1 \times 10^4$
$^{210}Po/^{210}Pb$ (± STD)	0.66 ± 0.02	0.62 ± 0.07	1.79 ± 2.79	

^{210}Po is relatively more scarce than ^{210}Pb, as the $^{210}Po/^{210}Pb$ ratio is 0.66 in the total phase and 0.62 in the dissolved phase. It is shown that removal of ^{210}Po is faster in the coastal water than ^{210}Pb in the coastal water, as ^{210}Po has a high distribution coefficient about the inorganic particles in marine ecosystems (Bacon et al., 1988; Sarin et al., 1994). The measured $^{210}Po/^{210}Pb$ ratio around Jeju Island included the Kuroshio current flow from of the southern side of the East China Sea (0.41-0.70) (Nozaki et al., 1991; Stewart et al., 2010; Tateda et al., 2003). The distribution coefficient (*Kd*) of ^{210}Po and ^{210}Pb in the seawater around Jeju Island was calculated by activity concentration of ^{210}Po (^{210}Pb) in the total and dissolved phases in the seawater of the study area and from the concentration of SPM following formula [10] (IAEA, 2004).

$$K_d = \frac{[(A^{T}_{Po(Pb)} - A^{D}_{Po(Pb)})/SPM]}{A^{D}_{Po(Pb)}} \quad [10]$$

where $A^{T}_{Po(Pb)}$ means activity concentration of ^{210}Po (^{210}Pb) during the total phase in the seawater around Jeju Island, and $A^{D}_{Po(Pb)}$ is the activity concentration of ^{210}Po (^{210}Pb) in the dissolved phase. SPM concentration in the study area was 0.56 $mg{\cdot}kg^{-1}$. Formula [10] gives a *Kd* for ^{210}Po and ^{210}Pb of 1.9×10^{5} and 6.6×10^{5}; the distribution coefficient of ^{210}Pb is 3 times higher than that for ^{210}Po (Table 2). The *Kd* of ^{210}Po and ^{210}Pb in seawater in the open ocean was 2.0×10^{7} and 1.0×10^{7}, and for the marginal sea was 2.0×10^{7} and 1.0×10^{5} (IAEA, 2004). The *Kd* of ^{210}Po calculated in this study was 100 times lower than the IAEA reported value, while the *Kd* of ^{210}Pb is similar to that of the IAEA reported value. Finally, the *Kd* of ^{210}Po, which was studied during May and July around the Straits of Korea, showed a lower range than the reported value at 2.5×10^5 to 4.1×10^6 (Hong et al., 2008).

The activity concentrations of ^{210}Po and ^{210}Pb in the phytoplankton (their primary producers in the marine ecosystem) are 116 ± 24 and 31 ± 6.6 $Bq\cdot(kg\cdot ww)^{-1}$, and in and zooplankton (their primary consumers) were 107 ± 2.1 and 6.89 ± 0.93 $Bq\cdot(kg\cdot ww)^{-1}$ (Table 3). Formula [11] was used to calculate the concentration factor (CF) of ^{210}Po and ^{210}Pb from the activity concentration of ^{210}Po and ^{210}Pb in the seawater (IAEA, 2004).

$$CF = \frac{A_{Po(Pb)}^{sample}}{A_{Po(Pb)}^{D}} \qquad [11]$$

In this formula, $A_{Po(Pb)}^{sample}$ represents the activity concentration of $^{210}Po(^{210}Pb)$ in each sample, and $A_{Po(Pb)}^{D}$ is the activity concentration of $^{210}Po(^{210}Pb)$ in the dissolved phase of seawater. Concentration factors (CF) of ^{210}Po and ^{210}Pb were 1.5×10^5 and 2.6×10^4 in the phytoplankton and 1.4×10^5 and 5.7×10^3 in the zooplankton (Table 5). Previous research indicated a concentration change of ^{210}Po and ^{210}Pb with differences in plankton size; ^{210}Po did not change dependent on the difference in plankton size, but ^{210}Pb concentrations changed in that those of size 60 - 200 μm size showed a higher concentration relative to plankton that were of a size greater than 200 μm (Strady et al., 2015). The activity concentration of ^{210}Po in the phytoplankton measured in the bay of Jin-Hea was 99.5 - 139 $Bq\cdot(kg\cdot ww)^{-1}$, with a CF of 1×10^5. The CF of ^{210}Po to phytoplankton in the bay of Jin-Hea and around Jeju Island were not significantly different from one another (Kim & Yang, 2004).

Table 3 The activity concentrations and concentration ratios of ^{210}Po and ^{210}Pb in the trophic levels of phytoplankton, zooplankton, anchovy and mackerel around Jeju Island

(unit: Bq·(kg·ww)$^{-1}$)

Common name *(Species name)*	Part	^{210}Po (± STD)	^{210}Pb (± STD)	$^{210}Po/^{210}Pb$ (± STD)
Phytoplankton		116 ± 24	31.4 ± 6.6	3.7 ± 1.1
Zooplankton		107 ± 2.1	6.89 ± 0.93	15.5 ± 2.1
Anchovy (*Engraulis japonius*)	Whole body	264 ± 1	1.41 ± 0.52	187 ± 69
	Head	236 ± 19	1.36 ± 0.14	173 ± 22
	Muscle	115 ± 10	0.58 ± 0.18	198 ± 64
	Internal organs	968 ± 160	2.40 ± 0.24	403 ± 78
Mackerel (*Scomber japonicus*)	Muscle	0.8 ± 0.03	0.21 ± 0.05	3.7 ± 0.9
	Skin	2.9 ± 0.6	0.73 ± 0.13	3.9 ± 1.1
	Internal organs	30.1 ± 6.5	1.8 ± 0.5	16.5 ± 5.8
	Liver	66 ± 22	2.46 ± 0.35	27 ± 10

3.2 The distribution of the activity concentration of ^{210}Po and ^{210}Pb in the anchovy and mackerel

According to National Fisheries Research & Development Institute, the anchovy (*Engraulis japonicus*) caught in the area around the north-west coast of Jeju Island was more than one year of age due to its 10 cm body size; the current may have driven it to the South Sea of Korea. Activity concentrations of ^{210}Po and ^{210}Pb in the anchovy (*Engraulis japonicus*) captured around Jeju Island were 264 ± 1 and 1.41 ± 0.52 Bq·(kg·ww)$^{-1}$, with the ^{210}Po value 187 times higher than the ^{210}Pb value (Table 3). Previous studies of activity concentrations of ^{210}Po in the anchovy have shown a diverse range from 24.9 to 281 Bq·(kg·ww)$^{-1}$ with those of ^{210}Pb in the anchovy ranging from 0.18 to 28.1 Bq·(kg·ww)$^{-1}$ (Aközcan, 2013; Çatal et al., 2012; Khan & Wesley, 2012; Lazorenko et al., 2002; Štrok & Smodiš., 2011). The activity concentration of ^{210}Po and ^{210}Pb in each part of the anchovy living around Jeju Island has a different concentration. ^{210}Po and ^{210}Pb concentrations in muscle were 115 ± 10 and 0.58 ± 0.18 Bq·(kg·ww)$^{-1}$, respectively; in the head, they were 236 ± 19 and 1.36 ± 0.14 Bq·(kg·ww)$^{-1}$, and in the internal organs 968 ± 160 and 2.40 ± 0.24 Bq·(kg·ww)$^{-1}$. The activity concentration of ^{210}Po in the anchovy is 2 times higher than in plankton. Concentration of ^{210}Po in internal organs was shown to be high－ times higher than in muscle. ^{210}Po bio-magnification is evident along the trophic level of planktivorous anchovy. In contrast, the concentration of ^{210}Pb in the whole body and head were 5 times lower than in plankton. Concentration of ^{210}Pb in the internal organs is 3 times lower than plankton. It was shown that ^{210}Pb was lost at trophic levels.

Previous research has shown that the activity concentration of ^{210}Po in mackerel varies from 3.6 to 30.2 Bq·(kg·ww)$^{-1}$, as does the ^{210}Pb range (from 0.56 to 9.5

Bq·(kg·ww)$^{-1}$); these variations depend on the sea area where these values are measured (Aközcan & Ugur, 2013; Aoun et al., 2015; Khan & Wesley, 2012). According to the National Fisheries Research & Development Institute, mackerel (*Scomber japonicas*) caught around the north-west coast of Jeju Island was over 2 years old, as its size was 30 cm long, 330 g average weight. Its appearance in the South Sea of Korea may reflect spawning migration. ^{210}Po and ^{210}Pb were analyzed in the mackerel of the upper trophic level (a higher trophic level than for the anchovy), and the mackerel as divided into muscle, skin, internal organs, and liver. The liver showed the highest concentration of ^{210}Po and ^{210}Pb, at 66.2 ± 21.9 and 2.46 ± 0.35 Bq·(kg·ww)$^{-1}$, respectively. The muscles had the lowest concentration of ^{210}Po and ^{210}Pb, at 0.80 ± 0.03 and 0.21 ± 0.05 Bq·(kg·ww)$^{-1}$, respectively, when compared to the other parts of the mackerel (Table 3). The activity concentrations of ^{210}Po and ^{210}Pb in the internal organs were 30.1 ± 6.5 and 1.82 ± 0.51 Bq·(kg·ww)$^{-1}$, respectively, 50% to 70% lower than that in the liver. The activity concentrations of ^{210}Po and ^{210}Pb in the internal organs were 2.9 ± 0.6 and 0.73 ± 0.13 Bq·(kg·ww)$^{-1}$, respectively, 3.5 times higher than those of muscle. The activity concentration of ^{210}Po in mackerel muscle is 330 times lower than anchovy among the mackerel feed animal; the concentration of ^{210}Pb is 6.7 times lower than anchovy. In the anchovy, which is at a lower trophic level than the mackerel, the concentrations of both radionuclides were higher than in the mackerel liver, and also higher than they were for the highest ^{210}Po concentration in the internal organs of the mackerel.

Range of the ^{210}Po and ^{210}Pb in marine biota can change by taxonomic group, inhabited environment, and feed (Aközcan & Ugur, 2013; Alam & Mohamed, 2011b; Heyraud & Cherry, 1979). Fishes were classified by feed kind in the same habitat. The ranges of activity concentrations of ^{210}Po and ^{210}Pb in planktivorous fishes (measured in the muscles) was 23.1 – 0 and 0.6 –)9 Bq·(kg·ww)$^{-1}$, respectively. The ranges of activity concentration of ^{210}Po and ^{210}Pb in carnivorous fishes were 0.8 – 16 and 0.2 – .6 Bq·(kg·ww)$^{-1}$ (both measured in muscle; Table 4) (Štrok & Smodiš, 2011; Suriyanarayanan et al., 2010; Musthafa & Krishnamoorthy, 2012; This study). Previous studies showed that planktivorous fishes have higher activity concentrations of ^{210}Po and ^{210}Pb than do carnivorous fishes. In a culture experiment of fish using different feed kinds (plankton and other kinds of feed), fishes that fed on plankton had higher activity concentrations of ^{210}Po than did those who consumed the other feed (Cherry et al., 1989).

Table 4 Comparison of the activity concentrations and concentration ratios of ^{210}Po and ^{210}Pb in the planktivorous and carnivorous fish with those in other studies.

(*: muscle) (unit: Bq·(kg·ww)$^{-1}$)

Site	common name	Species name	Type*	^{210}Po (± STD)	^{210}Pb (± STD)	$^{210}Po/^{210}Pb$ (± STD)
Lebanese coastal	Marbled spinefoot	*Siganus Rivulatus*	P	140 ± 10	99 ± 4	1.4 ± 0.1
	Common pandora	*Pagellus Erythrinus*	C	4.6 ± 0.8	1.6 ± 0.1	2.9 ± 0.5
East coast of India	Mozambique tilapia	*Oreochromis mossambicus*	P	81.6 ± 6.7	2.0 ± 0.6	4.1 ± 1.3
	Fringescale sardinella	*Sarcinella fumbriata*	C	41.3 ± 3.1	0.7 ± 0.4	57.4 ± 2.9
Slovenian coast	European pilchard	*Sardina pilchardus*	P	23.1 ± 0.9	1.1 ± 0.1	21.6 ± 2.8
	Flathead grey mullet	*Mugil cephalus*	C	8.2 ± 0.5	0.3 ± 0.1	32.8 ± 17
South coast of India	Indian oil sardine*	*Sardinella longiceps*	P	190 ± 5	13 ± 1	14.6 ± 1.2
	Indian Salmon*	*Eleutheronema tetradactylum*	C	116 ± 7	1.0 ± 0.5	116 ± 58
Jeju Island of Korea	Japanese anchovy*	*Engraulis japonicus*	P	115 ± 10	0.6 ± 0.2	198 ± 64
	Chub mackerel*	*Scomber japonicus*	C	0.8 ± 0.03	0.2 ± 0.1	3.7 ± 0.9

*Feeding type: P: Planktivore, C: Carnivore

(Lebanese coastal : Aoun et al., 2015; East coast of India: Musthafa and Krishnamoorthy, 2011; Slovenian coast: Strok and Smodis, 2011; South coast of India: Suriyanarayanan et al., 2010; Jeju island of Korea: This study)

3.3 Transferral of ^{210}Po and ^{210}Pb through the trophic levels

CFs of ^{210}Po at the trophic levels while tracking the phytoplankton, zooplankton, anchovy and mackerel around Jeju Island were highest in phytoplankton, at 1.5×10^5. Zooplankton has a lower CF than did phytoplankton at 1.4×10^5. The CF of ^{210}Po in the anchovy was 3.5×10^5, several times higher than in phytoplankton. This results in the anchovy having higher concentrations of ^{210}Po than does plankton (Table 5; Fig. 4). The CF of ^{210}Po in anchovy muscle was similar to that of phytoplankton at 1.5×10^5, but 8 to 9 times higher than that of phytoplankton in the internal organs at 1.3×10^6; ^{210}Po is highly concentrated in the internal organs due to the take feed. The CF of ^{210}Po of carnivorous mackerel on the upper trophic level (at a higher level than anchovy) was ten times to more than hundreds times lower than that of anchovy as 1.1×10^3 and 4.0×10^4 in the muscles and internal organs. The finding that planktivorous fish have higher concentrations of ^{210}Po than do carnivorous fish (as was shown by previously completed research) is important but not surprising, as anchovy (a planktivorous fish) has a high concentration of ^{210}Po in this study (Fowler 2011; Suriyanarayanan et al. 2010).

CF of ^{210}Pb to phytoplankton was 5 times lower than ^{210}Po with 2.6×10^4. CF of ^{210}Pb to zooplankton was 25 times lower than ^{210}Po with 5.7×10^3. Zooplankton is low concentration of ^{210}Po than ^{210}Pb than phytoplankton. In previous study on $^{210}Po/^{210}Pb$ ration in the excrement of the zooplankton (2.2 ± 0.3) was higher than $^{210}Po/^{210}Pb$ ratio of the zooplankton through the feed (6), relatively. ^{210}Pb excreted better than ^{210}Po via excrescence of the zooplankton (Beasley et al., 1978). Zooplankton have lower concentrations ^{210}Pb than of ^{210}Po. Also, upon culture experiment of measurement residues for ^{210}Po and ^{210}Pb in euphausiids (*Meganyctiphanes norvegica*) after their feeding, it appeared that ^{210}Po in the body was 44% of the total intake of ^{210}Po and passed ^{210}Po by excreta was 26% of total intake ^{210}Po, whereas ^{210}Pb in body was 3.5% of total intake ^{210}Pb and passed ^{210}Pb by excreta was 84% of total intake ^{210}Po. It is shown that ^{210}Pb passed to a greater extent than did ^{210}Po through the body (Stewart et al., 2005). CF of ^{210}Pb in the anchovy of the planktivorous fish was 5 times and 7 times lower than zooplankton as 2×10^3 and 4.8×10^2 in the whole body and muscle, as the excretion of ^{210}Pb in the anchovy is better. The CF of the ^{210}Pb to mackerel of the carnivore fishes was half that of anchovy－ 7×10^2 in the muscles and similar to anchovy with 1.5×10^3 in the internal organs (Table 5).

Table 5 The concentration factors of ^{210}Po and ^{210}Pb in the trophic levels of phytoplankton, zooplankton, anchovy and mackerel around Jeju Island

Common name (*Species name*)	Part	CF ^{210}Po (± STD)	CF ^{210}Pb (± STD)
Phytoplankton		$1.5\times10^{5} \pm 3.5\times10^{4}$	$2.6\times10^{4} \pm 5.8\times10^{3}$
Zooplankton		$1.4\times10^{5} \pm 1.2\times10^{4}$	$5.7\times10^{3} \pm 8.8\times10^{2}$
Anchovy (*Engraulis japonius*)	Whole body	$3.5\times10^{5} \pm 2.8\times10^{4}$	$1.2\times10^{3} \pm 4.3\times10^{2}$
	Head	$3.1\times10^{5} \pm 3.5\times10^{4}$	$1.1\times10^{3} \pm 1.4\times10^{2}$
	Muscle	$1.5\times10^{5} \pm 1.8\times10^{4}$	$4.8\times10^{2} \pm 1.5\times10^{2}$
	Internal organs	$1.3\times10^{6} \pm 2.4\times10^{5}$	$2.0\times10^{3} \pm 2.5\times10^{2}$
Mackerel (*Scomber japonicus*)	Muscle	$1.1\times10^{3} \pm 9.5\times10^{1}$	$1.7\times10^{2} \pm 4.4\times10^{1}$
	Skin	$3.8\times10^{3} \pm 8.8\times10^{2}$	$6.0\times10^{2} \pm 1.1\times10^{2}$
	Internal organs	$4.0\times10^{4} \pm 9.1\times10^{3}$	$1.5\times10^{3} \pm 4.3\times10^{2}$

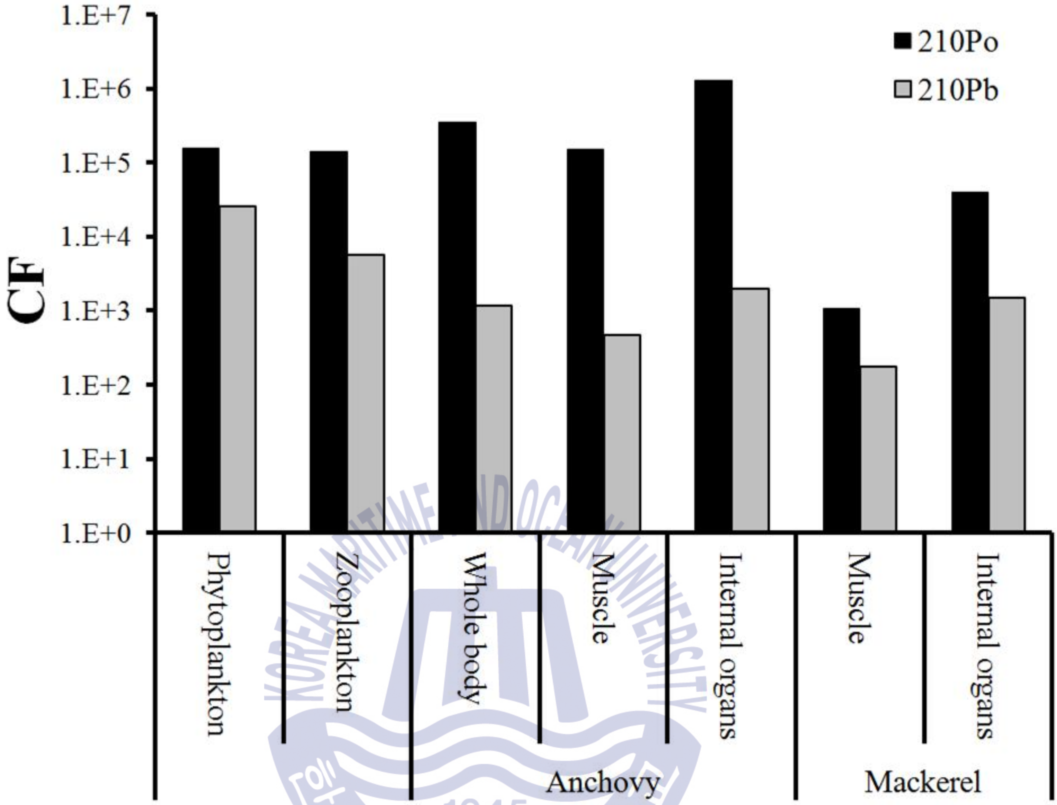

Fig. 4 The concentration factors of ^{210}Po and ^{210}Pb in phytoplankton, zooplankton, anchovy and mackerel around Jeju Island

Chapter 4. Annual effective dose of ^{210}Po from oysters and mussels

4.1 The activity concentration of ^{210}Po in oysters and mussels

Table 6 shows the measured ^{210}Po labels in the edible soft tissue of samples collected from 5 regions along the Korean coast. Results for the specific activity of ^{210}Po in oysters ranged from 41.3 to 206 Bq·(kg·ww)$^{-1}$. The overall range for the specific activity with oysters caught along the western coast of Korea (yellow sea) (activity concentration of ^{210}Po: 158 ± 14 to 206 ± 10 Bq·(kg·ww)$^{-1}$), such as Seosan, was greater (and broader) than for the southern coast of Korea (activity concentration of ^{210}Po: 41.3 ± 3.7 to 55.8 ± 21.2 Bq·(kg·ww)$^{-1}$). Oysters captured along the coast of Korea were compared with the activities of ^{210}Po determined in other countries (Table 7). Tongyoung and Yeosu oysters showed activity concentrations 2 times higher than those observed in France and Taiwan, where the range was from 14.3 to 25.9 Bq·(kg·ww)$^{-1}$ (Connan et al., 2007; Lee & Wang, 2013), and oysters (*Crassostrea madrasensis*) inhabiting the east coast of India were a different species compared to the oysters (*Crassostrea gigas*) prevalent in the bodies of water around Korea; however, their activity values were similar to those for oysters captured along the south coast of Korea (Satheeshkumar., 2016). The Slovenian oyster (*Ostrea edulis*) is another species with a similar range (56.7 to 124 Bq·(kg·ww)$^{-1}$) (Štrok & Smodiš, 2011). However, the activity concentrations of ^{210}Po in the oysters of the west sea of Korea were considerably higher than those shown in previous studies, where the observed range was from 110 ± 18 to 206 ± 10 Bq·(kg·ww)$^{-1}$.

The activity of ^{210}Po in the edible soft tissue of mussels in the Tongyoung and Yeosu was 46.7 ± 0.7 to 42.9 ± 3.2 Bq·(kg·ww)$^{-1}$. The radioactivity concentration of ^{210}Po in the mussels caught along the southern coast were similar to those in oysters (Table 6). The ^{210}Po activity observed in the mussels caught in 5 other countries (Croatia; India; Slovenia; Turkey; France) across different species showed a distribution across a very wide range of 22.1 to 776 Bq·(kg·ww)$^{-1}$ (Aközcan, 2013; Connan et al., 2007; Khan et al., 2014; Rani et al., 2014; Rožmarić et al., 2012; Štrok & Smodiš, 2011). The activity of ^{210}Po in the tissue of mussels in Turkey showed the highest range of values, of 332 to 776 Bq·(kg·ww)$^{-1}$, which was 10 times higher than the radioactivity concentration of ^{210}Po from mussels caught along the Southern coast (Aközcan, 2013). The mean activity of ^{210}Po in the mussel of Korea as 44.8 ± 2.7 Bq·(kg·ww)$^{-1}$ was about twice as high as that observed in Croatia and India, which respectively showed radioactivity concentrations of has 22.1 and 31 Bq·(kg·ww)$^{-1}$ (Table 7) (Khan et al., 2014; Rožmarić et al., 2012).

Table 6 Activity concentrations of ^{210}Po and ^{210}Pb in the soft tissues of the oysters and the mussels collected in Korean coast

(n=3) (unit: Bq·(kg·ww)$^{-1}$)

Site	Oyster		Mussel	
	^{210}Po (± SD)	^{210}Pb (± SD)	^{210}Po (± SD)	^{210}Pb (± SD)
Seosan	206 ± 10	4.4 ± 0.7		
Boryeong	110 ± 18	3.4 ± 0.2		
Wando	158 ± 14	8.2 ± 7.1		
Yeosu	55.8 ± 21.2	2.7 ± 0.2	42.9 ± 3.2	2.0 ± 0.5
Tongyeong	41.3 ± 3.7	3.6 ± 0.4	46.7 ± 0.7	4.2 ± 0.3

Table 7 Comparison of ^{210}Po activity concentrations in the oysters and the mussels of other countries with those of this study

(unit: Bq·(kg·ww)$^{-1}$)

	Country	Species	^{210}Po
Oysters	France	*Crassostrea gigas*	14.3 to 34
	Slovenia	*Ostrea edulis*	56.7 to 124.6
	Taiwan	*Crassostrea gigas*	25.9
	India	*Crassostrea madrasensis*	45.2
	Korea	*Crassostrea gigas*	41.3 to 206
Mussel	France	*Mytilus edulis*	156.2 to 275
	Slovenia	*Mytilus galloprovincialis*	78.7
	Croatia	*Mytilus galloprovincialis*	22.1 to 207
	Turkey	*Mytilus galloprovincialis*	332 to 776
	India	*Perna indica*	31 to 186
		Perna viridis	36 to 212
		Perna perna	320
	Korea	*Mytilis coruscus*	42.9 to 46.7

(Oyster: France [Connan et al., 2007]; Slovenia [Štrok & Smodiš, 2011]; Taiwan [Lee & Wang, 2013] India [Satheeshkumar., 2016] Korea [This study] Mussel: France [Connan et al., 2007] Slovenia [Štrok & Smodiš, 2011] Croatia [Rožmarić et al., 2012] Turkey [Aközcan, 2013] India [Khan et al., 2014; Rani et al., 2014] Korea [This study])

The activity concentration of ^{210}Po in marine biota can change by their habitat, feeding type, physiological process, and size (Alam & Mohamed, 2011b). The oyster filter feeder appeared to indicate that different activity concentrations existed of ^{210}Po in the bodies of the oysters according to the features of the suspended matter. In previous studies, oysters selectively fed with a preference for phytoplankton or random phytoplankton and inorganic matter (Ward et al., 1998; Nasr, 1984). The correlation between suspended particle matter (SPM) and chlorophyll-a (Chl-a), as an indirect indicator of plankton in the sea water around the site where the oysters were caught, was examined to determine the reason of the different activity concentration of ^{210}Po. The concentration of SPM and Chl-a was used during 11 years (from 2003 to 2013) in a national marine environmental monitoring system made by the Environmental Management Corporation (Marine Environment Information System (MEIS), 2003-2013) (Table 8). The concentration of ^{210}Po in oysters at each site from 2003 to 2013 maintained a trend similar to that of SPM of each November and of the average for the year (Fig. 5).

The activity concentration in oysters was shown to have a high positive correlation (R^2=0.89) with the SPM measured in November for the captured oysters. Further a high positive correlation also existed (R^2=0.76) with the SPM of the annual average of 11 years (Fig. 6). The reported in vivo half-life of the metallic elements in the oyster (*Crassostrea gigas*) was 23 to 60 days (Ozaki &Panietz, 1981). Consequently, it is interesting to note that the activity concentration of ^{210}Po, mainly reflecting radionuclide sorption onto suspended matter, was similar for the concentration of suspended matter at each site in November. The concentration of Chl-a, which was related with a growing oyster population as an indirect indicator of plankton in November (R^2=0.62), showed a low negative correlation with the activity concentration of ^{210}Po. Meanwhile, to gain knowledge about the characteristics of suspended matter in the seawater, we compared the concentration of SPM to the value of Chl-a per SPM (Chl-a·SPM^{-1}). SPM had a negative correlation with Chl-a·SPM^{-1} (Fig. 7). It is indicated that the total concentration of the SPM contributed very little organic suspended matter to the re-suspended matter. Calvalho et al. (2011) reported that the activity concentration of ^{210}Po in the edible part of the oyster was correlated with the concentration of SPM; this activity concentration for ^{210}Po showed a stronger correlation with inorganic suspended matter than with Chl-a (an indirect indicator of plankton). The distribution coefficient (*Kd*) of ^{210}Po and ^{210}Pb in suspended matter, in contrast to seawater (2×10^{7}), was higher than those in phytoplankton (7×10^{4}) and zooplankton (3×10^{4}) (IAEA, 2004). Consequently, the oysters that lived where there was a high content level for re-suspended matter among total suspended matter might have had a relatively high activity concentration of ^{210}Po.

Table 8 Activity concentrations of ^{210}Po in the soft tissue of oysters and concentrations of suspended particulate matter in the surface water of sampling areas (MEIS, 2003-2013)

	^{210}Po	SPM (㎎·L^{-1})		Chl-a (㎍·L^{-1})	
	(Bq·(kg·ww)$^{-1}$)	November	Year average	November	Year average
Tongyeong	41.3 ± 3.7	6.9 ± 3.3	8.2 ± 3.1	2.7 ± 1.3	4.1 ± 0.8
Yeosu	55.8 ± 21.2	8.5 ± 5.0	10 ± 4	2.4 ± 1.3	4.0 ± 1.5
Wando	158 ± 14	18 ± 21	22 ± 20	2.4 ± 2.2	2.5 ± 1.5
Boryeong	110 ± 18	10 ± 7	12 ± 4	1.8 ± 0.9	9.1 ± 6.7
Seosan	206 ± 10	17 ± 14	18 ± 5	1.2 ± 0.7	2.4 ± 1.1

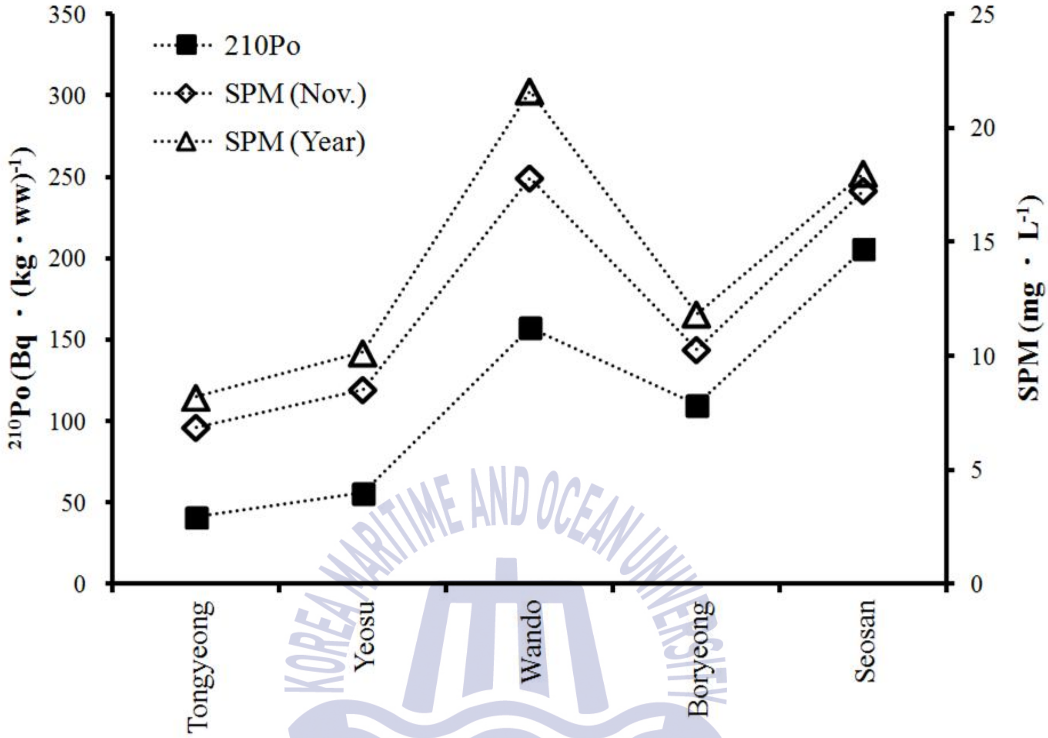

Fig. 5 Each site of activity concentrations of ^{210}Po in the soft tissues of oysters and concentrations of suspended particulate matter (SPM) in the surface water of sampling areas observed in November and across four seasons from 2003 to 2013 (MEIS, 2003-2013)

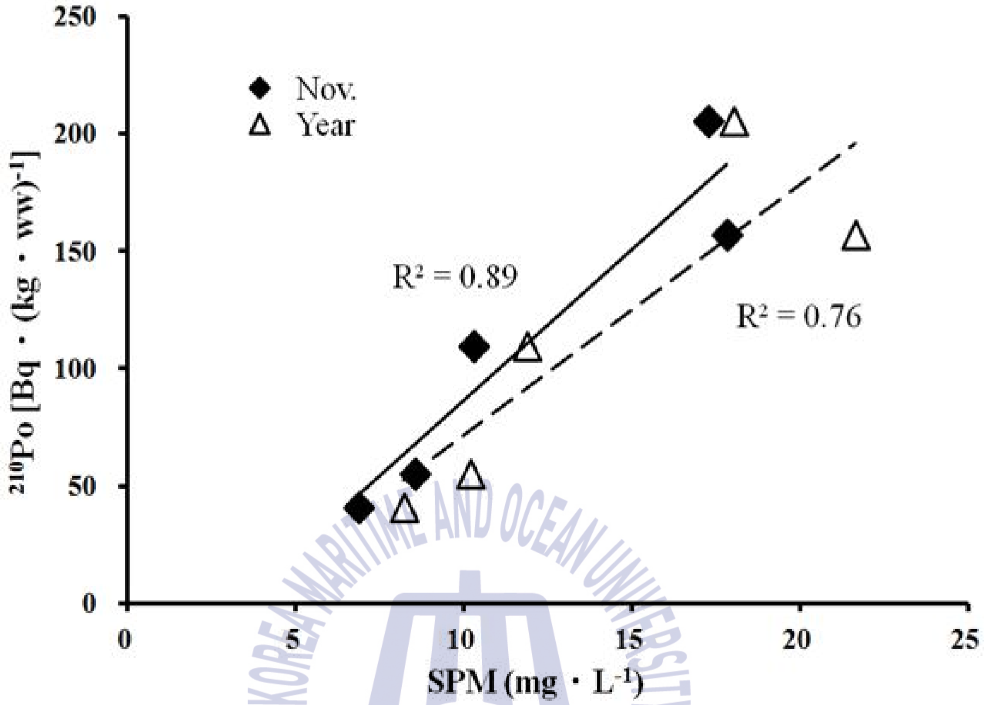

Fig. 6 Correlation between the activity concentrations of ^{210}Po in the soft tissues of oyster and the concentrations of suspended particulate matter (SPM) in the surface water of sampling areas observed in November and across four seasons from 2003 to 2013 (MEIS, 2003-2013)

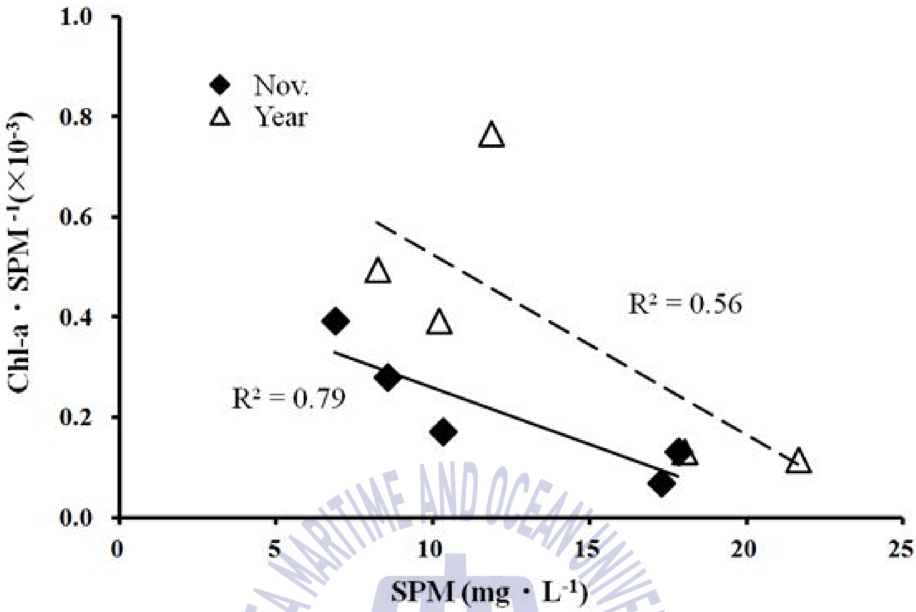

Fig. 7 Correlation between the concentrations of suspended particulate matter (SPM) and the ratios of the concentrations of chlorophyll-a (Chl-a) to the SPM (Chl-a·SPM ($\times10^{-3}$)) in the surface water of sampling areas observed in November and across 4 seasons from 2003 to 2013 (MEIS, 2003-2013)

4.2 Assessment of effective dose

The intake of ^{210}Po for humans caused by ingestion of oysters collected in Korea was calculated. Each isotope has a different change efficiency Becquerel (Bq) to Sievert (Sv) in organ. ^{210}Po change efficiency via ingestion was 1.2×10^{-6} $Sv\cdot Bq^{-1}$ (IAEA, 2011). In Korea, the adult annual intake of oysters and mussels were 507 and 117 $g\cdot yr^{-1}$; this estimate is based on primitive data in 2013 from the Korea Centers for Disease Control and Prevention. The annual effective dose via ingestion of ^{210}Po in oysters and mussels from the coast of Korea was calculated by the following formula [12].

$$AED\ (Sv\cdot yr^{-1}) = CE \times I \times C \qquad [12]$$

In [12], AED is the annual effective dose from foods; CE_{Po210} is ^{210}Po internal dose co-efficiency change Bq to Sv (1.2×10^{-6} $Sv\cdot Bq^{-1}$) (IAEA, 2011); I_{Po210} is amount of ingestion to food per year per capita ($kg\cdot yr^{-1}$) (primitive data in 2013 from the Korea Centers for Disease Control and Prevention); C_{Po210} is the result of ^{210}Po activity concentration in this study ($Bq\cdot kg^{-1}$) (Table 6). The annual effective dose of ^{210}Po via oysters and mussels was compared with that of previous studies, and was shown to have a wide range in other countries Annual effective dose of ^{210}Po via oyster and mussel in other countries was shown to a wide range (Table 9). The annual effective dose of ^{210}Po via oysters in Korea (21 - 104 $\mu Sv\cdot y^{-1}$) can be compared with those of Taiwan (41 $\mu Sv\cdot yr^{-1}$) (Lee & Wang, 2013). However, the annual effective dose of ^{210}Po via oysters in Korea was over 10 times higher than those of France (10 - 24 $\mu Sv\cdot yr^{-1}$) and India (12.7 $\mu Sv\cdot yr^{-1}$) (Connan et al., 2007; Khan et al., 2014; Rani et al., 2014).

Table 9 Comparison of annual effective doses of ^{210}Po from the ingestion of mussel and oysters in other countries with this study.

(unit: $\mu Sv \cdot yr^{-1}$)

	Country	Species	Annual effective dose
Oysters	France	*Crassostrea gigas*	10 - 24
	Taiwan	*Crassostrea gigas*	41
	India	*Crassostrea madrasensis*	12.7
	Korea	*Crassostrea gigas*	21 - 104
Mussel	France	*Mytilus edulis*	50
	Slovenia	*Mytilus galloprovincialis*	8.5
	Croatia	*Mytilus galloprovincialis*	53 - 497
	Turkey	*Mytilus galloprovincialis*	1,992 - 4,332
	India	*Perna indica*	5.1 - 30.5
		Perna viridis	6.1 - 34.9
		Perna perna	1728
	Korea	*Mytili scoruscus*	5.01 - 5.46

(Oyster: France [Connan et al., 2007]; Slovenia [Štrok & Smodiš, 2011]; Taiwan [Lee & Wang, 2013] India [Khan et al., 2014] Korea [This study] Mussel: France [Connan et al., 2007] Slovenia [Štrok & Smodiš, 2011] Croatia [Rožmarić et al., 2012] Turkey [Aközcan, 2013] India [Khan et al., 2014; Rani et al., 2014] Korea [This study])

The compared annual effective dose of ^{210}Po via mussels ranged from 5.01 to 5.46 $\mu Sv \cdot y^{-1}$ compared with those of previous studies showing a range from 5.1 to 4,332 $\mu Sv \cdot y^{-1}$; the annual effective dose of ^{210}Po via mussels in the Korea was relatively low (Aközcan, 2013; Connan et al., 2007; Štrok & Smodiš, 2011; Rožmarić et al., 2012; Khan et al., 2014; Rani et al., 2014; This study). The annual effective dose of ^{210}Po via mussels along the southern coast of India and Slovenia also was within a low range of 5.1 - 8.5 $\mu Sv \cdot y^{-1}$, similar to those of Korea (Štrok & Smodiš, 2011; Khan et al., 2014); however, in Turkey the range from 1,192 to 4,332 $\mu Sv \cdot y^{-1}$ was a hundred times higher than that of Korea (Aközcan, 2013). That resulted from the high activity concentration of ^{210}Po in the mussels in these seas.

The reported annual effective dose of ^{210}Po in Korean adults through the intake of all food was 269 $\mu Sv \cdot y^{-1}$; 80% of this was attributable to seafood intake (Lee et al., 2009). Korean adults were exposed to an annual effective dose of ^{210}Po of 76 ± 42 $\mu Sv \cdot yr^{-1}$ from only the consumption of oysters and mussels, which is based on annual average ingestion of oysters and mussels in Korea. This amount represented 28±16% an annual effective dose of ^{210}Po taken in via all food, and 35±19% according to annual effective dose of ^{210}Po from the consumption of seafood. The average annual effective dose of ^{210}Po in Korean adults from the intake of oysters and mussels was very high when one considers that oysters and mussels consisted of 3% of the total amount of seafood consumed by Korean adults. Meanwhile, the recommended CF of ^{210}Po contrasted with seawater for mollusks and crustaceans is 2.0×10^4, and for fish and seaweed is 2.0×10^3 (IAEA, 2004). However, the activity concentrations of ^{210}Po in oysters was different by the site, and the average annual effective dose of ^{210}Po for Korean adults via seafood intake can vary according to the site of capture and the type of seafood. Given this, the average annual effective dose of ^{210}Po via seafood intake was rated accurately due to the high concentration of ^{210}Po in seafood.

Chapter 5. Conclusions

The activity concentrations of ^{210}Po and ^{210}Pb within the trophic levels of phytoplankton, zooplankton, anchovy and mackerel in the coastal waters of Jeju Island were determined, and their accumulation along the trophic levels were studied in May 2014. The activity concentrations of ^{210}Po in the corrected total phase and dissolved phase of seawater were measured as 0.83 ± 0.004 and 0.75 ± 0.06 $mBq \cdot kg^{-1}$, and for ^{210}Pb were measured to 1.27 ± 0.03 and 1.22 ± 0.09 $mBq \cdot kg^{-1}$. The concentration of ^{210}Pb was 1.5 times higher than ^{210}Po. The CFs of ^{210}Po and ^{210}Pb contrasted with seawater to phytoplankton were 1.5×10^5 and 2.6×10^4, respectively; the CF of ^{210}Po was 5 times higher. The CF of ^{210}Po compared to zooplankton was similar to the CF of ^{210}Po to phytoplankton, whereas the CF of ^{210}Pb compared to zooplankton was 5 times lower. The CF of ^{210}Po to anchovy was over 10 times higher than for zooplankton. The CF of ^{210}Po compared to upper-trophic level mackerel was one hundred times lower than for the anchovy. The CF of ^{210}Po to anchovy and mackerel internal organs is 8 times to 38 times higher than those of muscle, and ^{210}Po is highly concentrated in the internal organs.

In the phytoplankton-zooplankton-anchovy trophic levels, the CF of ^{210}Pb was decreased by five times along the trophic level in order, and in anchovy-mackerel was decreased by 30% to 70%. (Fig. 8). The concentration factor ratio between ^{210}Po and ^{210}Pb ($CF(^{210}Po)/CF(^{210}Pb)$) increased 5 to 12 times under the trophic level step of phytoplankton-zooplankton-anchovy, but decreased over 10 times under the trophic level step of anchovy to mackerel (Fig. 9).

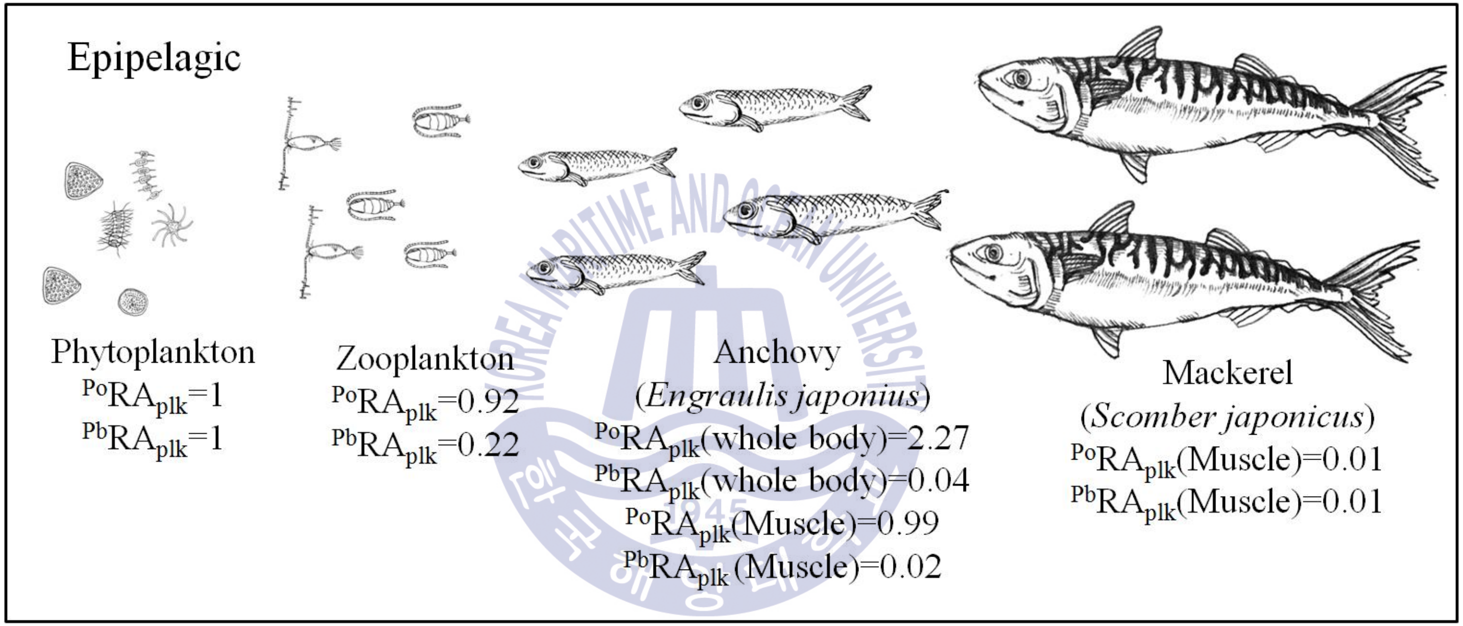

Fig. 8 The relative concentration factors in zooplankton, anchovy and mackerel to the concentration factors of ^{210}Po and ^{210}Pb in phytoplankton around Jeju Island.

Relative concentration factor: $^{Po(Pb)}RA_{plk}$=(CF of $^{210}Po(Pb)$ in marine biota)(CF of $^{210}Po(Pb)$ in phytoplankton)$^{-1}$

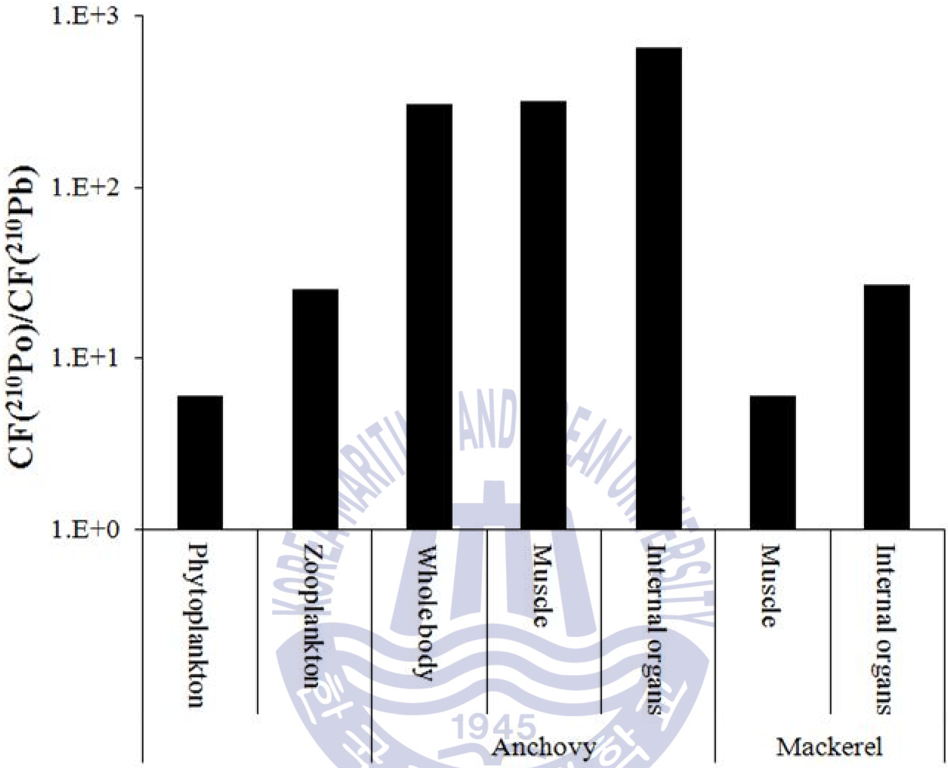

Fig. 9 The ratios of ^{210}Po and ^{210}Pb concentration factors in phytoplankton, zooplankton, anchovy and mackerel around Jeju Island

The activity concentrations of ^{210}Po in the soft tissues of oysters and mussels collected along the Korean coast were 41.3 ± 3.7 to 206 ± 10 Bq·(kg·ww)$^{-1}$ and 42.9 ± 3.2 to 46.7 ± 0.7 Bq·(kg·ww)$^{-1}$, respectively. The activity concentrations of ^{210}Po in oysters were different according to the site where they were captured, as oysters from the southwest coast of Korea have a higher activity concentration of ^{210}Po than do oysters caught along the west coast. The activity concentration of ^{210}Po in oysters is positively correlated with concentrations of SPM. Specifically, the correlation between concentration of ^{210}Po and the annual mean concentration of SPM (R^2=0.76) was lower than the correlation between concentration of ^{210}Po and the concentration of SPM (R^2=0.89) in November when corrected for the oysters in the present study. These findings indicate that the concentration of ^{210}Po in the edible part of oyster was affected by/due to the inorganic suspended matter.

The annual effective dose of ^{210}Po via oyster and mussel intake was in the range of 5.01 to 104 μSv·yr^{-1}. The annual effective dose and radioactivity concentrations from the Korean coast for oysters (21 - 104 μSv·yr^{-1}) were higher than those in other countries excepting Taiwan and France. The annual effective dose via mussels was 5.01 - 5.46 μSv·yr^{-1}, 10 to 20 times lower than the annual effective dose via oysters in this study. When comparisons are drawn between the annual effective dose via mussels in Korea and in other countries, the annual effective dose of ^{210}Po via mussels in the South coast of India and Slovenia is in a low range from 5.1 - 8.5 μSv·yr^{-1}, similar with those of Korea. The average annual effective dose of ^{210}Po in the Korean adult via intake of oysters and mussels was 76 ± 42 μ Sv·yr^{-1} was 28 ± 16% according to annual effective dose of ^{210}Po about the Korean adult via intake of all food, and 35 ± 19% according to the annual effective dose of ^{210}Po in Korean adults via. The average annual effective dose of ^{210}Po in Korean adults that was attributable to lobster and oyster was very high, allowing for oyster and mussel consumption in Korea that comprises 3% of Korea's annual food intake.

Finally, more data are needed on natural radionuclides in seafood from around the Korean peninsula to fully assess annual the effective dose of natural radionuclides in terms of its impact on good. In this study it appears unlikely that the pelagic biota around Jeju Island are exposed to a radiation concentration different from that affecting animals in coastal or deep-sea environments.

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