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Radon 222 in and over shallow seas

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Abstract. The formulae for the concentration of ^{222}Rn in a shallow sea and the flux density of ^{222}Rn from the sea surface were derived assuming that the seawater was well mixed vertically. The concentrations of ^{222}Rn and the flux densities of ^{222}Rn estimated using the formulae agree approximately with those measured. In addition, in the case of $-z_b$ (the depth of water) = 50 m and $-z_0$ (the thickness of a stagnant water film) = 45 μm, the flux densities of ^{222}Rn were estimated to be 0.085 mBq m^{-2} s^{-1} for K_{sw}^{Rn} (the eddy diffusivity of well mixed isothermal layer) = 0.0022 m^2 s^{-1} and 0.12 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.02 m^2 s^{-1}, and in the case of $-z_b$ = 100 m and $-z_0$ = 45 μm, those were estimated to be 0.041 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.0022 m^2 s^{-1} and 0.076 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.02 m^2 s^{-1}. Furthermore, the contributions of ^{222}Rn from the sea surface to the atmospheric concentrations of ^{222}Rn over the sea were 1.1 % (1.7 %) of that measured in the Seto Inland Sea and 3.4 % of that measured around Andros Island.

1. Introduction

Information on ^{222}Rn at sea is considerably less than that on land. As far as we know, reports have been published on the atmospheric concentration of ^{222}Rn over the oceans (Broecker, 1965; Mochizuki, 1978, 1982; Bonsang and Lambert, 1984; Rangarajan et al., 1984/85; Smethie et al., 1985; Hansen, et al., 1990; Miyake et al., 1990; Balkanski et al., 1992; Tanji et al., 1992; Hussain et al.,1998), on the concentration of ^{222}Rn in seawater (Broecker, 1965; Broecker et al. 1967; Broecker and Peng, 1971, 1974; Peng et al., 1974, 1979; Dueñas et al., 1983; Smethie et al., 1985; Kawabata et al., 2003; Hwang et al., 2005; Saito et al., 2012), and on the flux density of ^{222}Rn from the sea surface (Hoang and Servant, 1972; Wilkening and Clements, 1975; Dueñas et al., 1983; Schery and Huang, 2004; Zahorowski et al., 2013).

In this report, first we derive the formulae for the concentration of ^{222}Rn in a shallow sea and the flux density of ^{222}Rn from the sea surface. Next, we compare the concentrations of ^{222}Rn and the flux densities of ^{222}Rn estimated using the formulae with those measured by other investigators. Finally, we estimate the atmospheric concentrations of ^{222}Rn over the sea only due to the flux densities of ^{222}Rn from the sea surface, and evaluate the contributions of the estimates to the atmospheric concentrations of ^{222}Rn measured at a position (34°38'N, 134°19'E) in the Seto Inland Sea and at Frazer's Hog Cay on the edge of the Great Bahama Bank.

2. Theory

2.1. A steady state mixing model

If the seawater between the sea-air and sediment-water interfaces is assumed to be well mixed vertically in a shallow sea, a sketch of the concentration profile of ^{222}Rn in the sea is shown in Figure 1.

According to Broecker (1971), under the assumption of horizontal homogeneity, the flux density of ^{222}Rn from the sea surface is represented by

$$F_{Rn,sw,0} = -\frac{D_{sw,0}^{Rn}}{0 - z_0}(P\alpha - C_{Rn,sw,0}) . \quad (1)$$

Considering the atmospheric burden of ^{222}Rn is very small ($C_{Rn,sw,0} \gg P\alpha$) and rewriting Equation (1), we obtain

$$F_{Rn,sw,0} = -\frac{D_{sw,0}^{Rn}}{z_0} C_{Rn,sw,0} , \quad (2)$$

where $F_{Rn,sw,0}$ (atoms m^{-2}) is the flux density of ^{222}Rn from the sea surface, $D_{sw,0}^{Rn}$ (m^2 s^{-1}) the molecular diffusivity of ^{222}Rn in seawater, $-z_0$ (m, $z_0 < 0$) the thickness of a "stagnant" water film, $C_{Rn,sw,0}$ (atoms m^{-3})

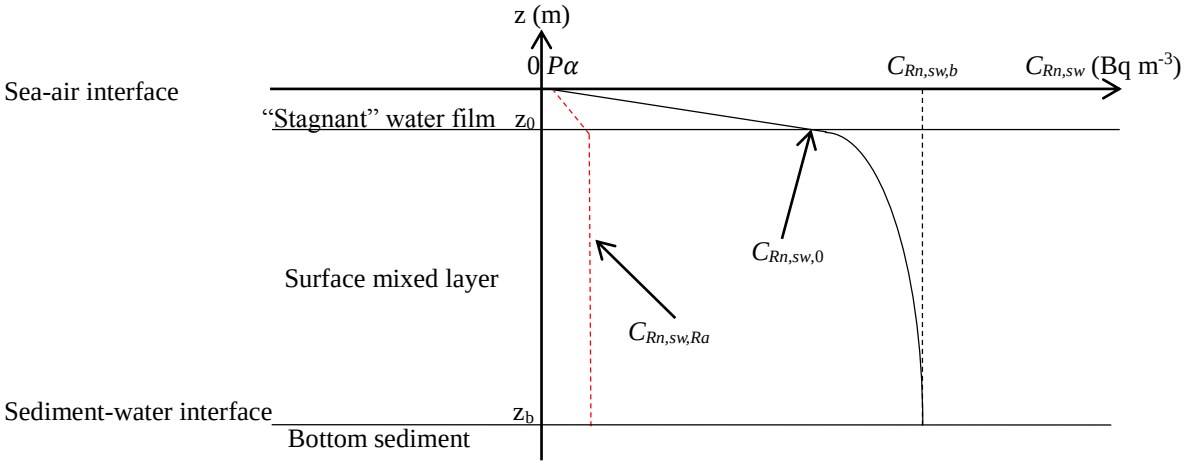

Figure 1. Concentration profile of ^{222}Rn in the sea.

the concentration of ^{222}Rn in seawater at the base of the stagnant water film, P (atm) the partial pressure of ^{222}Rn in the atmosphere near the sea surface, and α (atoms m^{-3} atm^{-1}) the solubility of ^{222}Rn in seawater.

Assuming steady state, one-dimensional diffusion of ^{222}Rn in the well mixed isothermal layer below the base of the stagnant film is represented by Fick's second law:

$$K_{sw}^{Rn}\frac{\partial^2 C_{Rn,sw}}{\partial z^2} = -\lambda\left(C_{Rn,sw,Ra} - C_{Rn,sw}\right), \tag{3}$$

where K_{sw}^{Rn} (m^2 s^{-1}) is the eddy diffusivity of well mixed isothermal layer, z (m) the depth from the sea-air interface and taken as negative in the downward direction, $C_{Rn,sw}$ (atoms m^{-3}) the concentration of ^{222}Rn in seawater at depth z, $C_{Rn,sw,Ra}$ (atoms m^{-3}) the concentration of ^{222}Rn, which is in radioactive equilibrium with ^{226}Ra, in seawater at depth z, λ (s^{-1}) the decay constant of ^{222}Rn.

The amount of ^{222}Rn produced in sediment per unit time is given by $\varrho_{bs}A_{Ra,bs}(1-w_{bs})/100$ (atoms m^{-3} s^{-1}). Radon 222 passes through the pore water of sediment only by molecular diffusion. Hence, assuming steady state, one-dimensional diffusion of ^{222}Rn in the pore water of sediment is given by Fick's second law:

$$D_{sw,0}^{Rn}\frac{\partial^2 C_{Rn,sw}}{\partial x^2} = -\left\{\frac{\delta_{bs}\varrho_{bs}A_{Ra,bs}(1-w_{bs})}{100n_{bs}} - \lambda C_{Rn,sw}\right\} \tag{4}$$

where δ_{bs} is the ^{222} Rn escape-to-production ratio of sediment, which is the ratio of the amount of ^{222}Rn that escapes into the seawater-filled pore space relative to the amount produced in the sediment at equilibrium state, ϱ_{bs} (kg m^{-3}) the bulk density, $A_{Ra,bs}$ (Bq kg^{-1} of dry sediment) the concentration of ^{226}Ra in solid material, w_{bs} (%) the water content (calculated as the percentage of seawater weight to the weight of the wet sediment), n_{bs} the porosity, x the depth from the sediment-water interface and taken as negative in the downward direction.

The continuity of the concentration requires that the concentration of ^{222}Rn at the base of stagnant film is equal to that at the top of the mixed layer. Since the stagnant film is at the most 1 mm thick (Bolin, 1960; Schindler et al., 1972; Broecker and Peng, 1974), $-z$ is in fact equal to the distance below the sea surface. Considering this and solving Equations (3) and (4), we have

$$C_{Rn,sw} = C_{Rn,sw,Ra} - \left[\frac{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\sqrt{\lambda D_{sw,0}^{Rn}}\,\frac{\delta_{bs}\varrho_{bs}A_{Ra,bs}(1-w_{bs})}{100\lambda}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}\right.$$
$$\left.+\frac{\left\{\left(\frac{D_{sw,0}^{Rn}}{z_0}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\right\}C_{Rn,sw,Ra}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}\right]\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z\right)$$

$$+\left[\frac{\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\sqrt{\lambda D_{sw,0}^{Rn}}\,\frac{\delta_{bs}\varrho_{bs}A_{Ra,bs}(1-w_{bs})}{100\lambda}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}})\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}\right.$$

$$\left.-\frac{\left\{\left(\frac{D_{sw,0}^{Rn}}{z_0}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\right\}C_{Rn,sw,Ra}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}\right]\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z\right). \quad (5)$$

Substituting Equation (5) into Equation (2), we have

$$F_{Rn,sw,0}=\frac{D_{sw,0}^{Rn}}{z_0}\left[\frac{2\sqrt{\lambda K_{sw}^{Rn}}\sqrt{\lambda D_{sw,0}^{Rn}}\,\frac{\delta_{bs}\varrho_{bs}A_{Ra,bs}(1-w_{bs})}{100\lambda}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}\right.$$

$$\left.-\frac{\sqrt{\lambda K_{sw}^{Rn}}\left\{\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)-\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+2n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right\}}{\left(\frac{D_{sw,0}^{Rn}}{z_0}+\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}-n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)+\left(\frac{D_{sw,0}^{Rn}}{z_0}-\sqrt{\lambda K_{sw}^{Rn}}\right)\left(\sqrt{\lambda K_{sw}^{Rn}}+n_{bs}\sqrt{\lambda D_{sw,0}^{Rn}}\right)\exp\left(-\sqrt{\frac{\lambda}{K_{sw}^{Rn}}}z_b\right)}C_{Rn,sw,Ra}\right], \quad (6)$$

where $-z_b$ (m, z_b<0) is the depth of the sediment-water interface, namely the depth of water.

Considering measurements carried out at our observation station in the Seto Inland Sea which is a large shallow inland sea in Japan (see Section 3), we make the following seven points about our calculations using Equations (5) and (6).

1. The concentrations of ^{226}Ra in surface seawater measured in the Seto Inland Sea (Okubo, 1981) are almost the same as those in the Pacific Ocean (Peng et al., 1979; Okubo et al., 1979; Ku et al., 1980; Nozaki et al., 1990), in the Sea of Japan (Harada and Tsunogai, 1986) and in Tokyo Bay (Nakano-Ohta and Sato, 2006) which is as shallow as the Seto Inland Sea. Hence, we take $C_{Rn,sw,Ra}$ to be 1.7 Bq m^{-3} which is the average concentration of ^{226}Ra in seawater obtained in the area located about 27 km southeast from the station (Okubo, 1981) (see Figure 2).
2. Sediment is made up of solid and liquid material. Since the water content of sediment at the station and its surrounding region ranges from 20 to 70 % (Comprehensive Water Quality Study of the Seto Inland Sea, 2015a), we take w_{bs}, ϱ_{bs} and n_{bs} to be 44.9 %, 1513 kg m^{-3} and 0.694, respectively (Nakagawa et al., 2004; Bennett and Lambert 1971).
3. The Seto Inland Sea sides of the Chugoku and Shikoku regions consist mainly of granite. Hence, we assume that the sediment of the Seto Inland Sea consists of the decomposed granite. Miyake et al. (1974) reported that the average concentration of ^{238}U (which we consider to be in radioactive equilibrium with ^{226}Ra) in granitic rocks was 25.9 Bq kg^{-1} at the surrounding region of the station. From this, we take $A_{Ra,bs}$ to be 26 Bq kg^{-1} of dry sediment.
4. The ^{222}Rn escape-to-production ratio of decomposed granite*[1)] is found to be about 0.4 (Barretto et al., 1974). However, soil water also contains ^{222}Rn that escapes from solid. Therefore, we take δ_{bs} to be 0.5. Broecker et al. (1968) also took this value.
5. Since the seawater temperature of the station and its surrounding region is uniform to the sea floor in autumn and winter (The marine Meteorological Society, 2013; Comprehensive Water Quality Study of the Seto Inland Sea, 2015b), the surface mixed layer is considered to extend to the sediment-water interface. Hence, to investigate the effect of the eddy diffusivity of well mixed isothermal layer, we use values of 0.0022 and 0.02

*[1)]The ratio of the amount of ^{222}Rn that escapes into the air-filled pore space relative to the amount produced in the decomposed granite at equilibrium state.

Table 1. Flux density of ^{222}Rn and the concentration of ^{222}Rn in surface seawater.

Flux density of ^{222}Rn (mBq m^{-2} s^{-1})	Concentration of ^{222}Rn in surface seawater (Bq m^{-3})	Reference
- - - - -	8.1	Broecker (1965). Samples are taken in water 0.7-6 m deep on the Bahama Banks.
3.2 (1500atoms m^{-2} s^{-1})	- - - - -	Wilkening and Clements (1975). Samples are taken 4 m offshore of Cocoanat Island in water 3.3 m deep in Hiro Bay.
0.14 (67 atoms m^{-2} s^{-1})	33	Dueñas et al. (1983). Samples are taken 2 m offshore in water 2.8 m deep in Málaga Bay.
- - - - -	140 (8.6 dpm ℓ$^{-1}$)	Hwang et al. (2005). Samples are taken in water 1-3 m deep in Bangdu Bay on Jeju Island.
- - - - -	20 (near the sea surface) 26 (near the sea floor)	Saito et al. (2012). Samples are taken in water 10-25 m deep in Hiuchi Nada in November.

m^2 s^{-1} for BOMEX composite ^{222}Rn profile and composite ^{222}Rn profile at ocean station Papa, respectively (Peng et al., 1974).

6. To investigate the effect of the water depths, we use values of 3 m (see Table 1), 11 m (see Section 3) and 20 m (see Section 3).
7. The film thickness $-z_0$ can hardly be more than 1 mm (Bolin, 1960), and the thickness ranging from 10 to 300 μm was observed (Broecker and Peng, 1971, 1974; Schindler et al., 1972; Peng et al., 1974,1979; Smethie et al., 1985). Therefore, in order to investigate the effect of the film thickness, we use values of 10, 35*[2)], 45*[3)], 100, 200 and 300 μm.

2.2. Atmospheric concentration of ^{222}Rn

The depth of marine atmospheric mixed layer is about 900 m, and its diurnal variation is not larger than that of atmospheric mixed layer on land (Hus, 1979; Kuribayashi et al., 2011). Hence, we make the following three points about the marine atmospheric mixed layer in order to estimate the atmospheric concentration of ^{222}Rn over the sea only due to the flux density of ^{222}Rn from the sea surface.

1. Since the marine atmospheric mixed layer was observed during a period of more than 80 % of a year over the East China Sea (Kuribayashi et al., 2011), we assume that it is always present over the sea.
2. We assume that the background concentration of ^{222}Rn in marine atmospheric mixed layer is constant.
3. Assuming that atmospheric conditions always remain neutral stability, we select case NNN (normal turbulence conditions) in Figure 1 in Jacobi and André (1963) as the turbulent diffusivity of marine atmospheric mixed layer. Hence, we use the profile of ^{222}Rn estimated for the NNN case in Figure 2 in Jacobi and André (1963) as the concentration of ^{222}Rn except for the background in marine atmospheric mixed layer.

3. Study site and Experiment

Atmospheric concentrations of ^{222}Rn and its short-lived progeny were measured on board Marinus II maru*[4)] (our observation station) on 15 and 16 December 1998, at the position (34^{O}38'N, 134^{O}19'E) located about 4.5 km south from Hinase Islands in the Seto Inland Sea. The location is identified by the letter OS in Figure 2. The Seto Inland Sea is about 450 km from east to west and 15-55 km from north to south, and occupies a total surface area of 19,700 km^2. It is bordered by the main lands of Honshu, Shikoku, Kyushu, Kii-suido Channel and Bungo-suido Channel to the Pacific Ocean, and Kanmon-kaikyo Strait to the Sea of Japan. It has a mean depth of 31 m and is

*[2)] The film thickness calculated using a transfer velocity of 7×10^{-5} m s^{-1} corresponding to a residence time for CO_2 in the atmosphere of about five years (Bolin, 1960).

*[3)] The average of film thickness obtained by Broecker and Peng (1974), Peng et al. (1979) and Smethie et al. (1985).

*[4)] The Marinus II Maru is a small craft (Length: 11.25 m, Capacity: 31 passengers) and belongs to Okayama University.

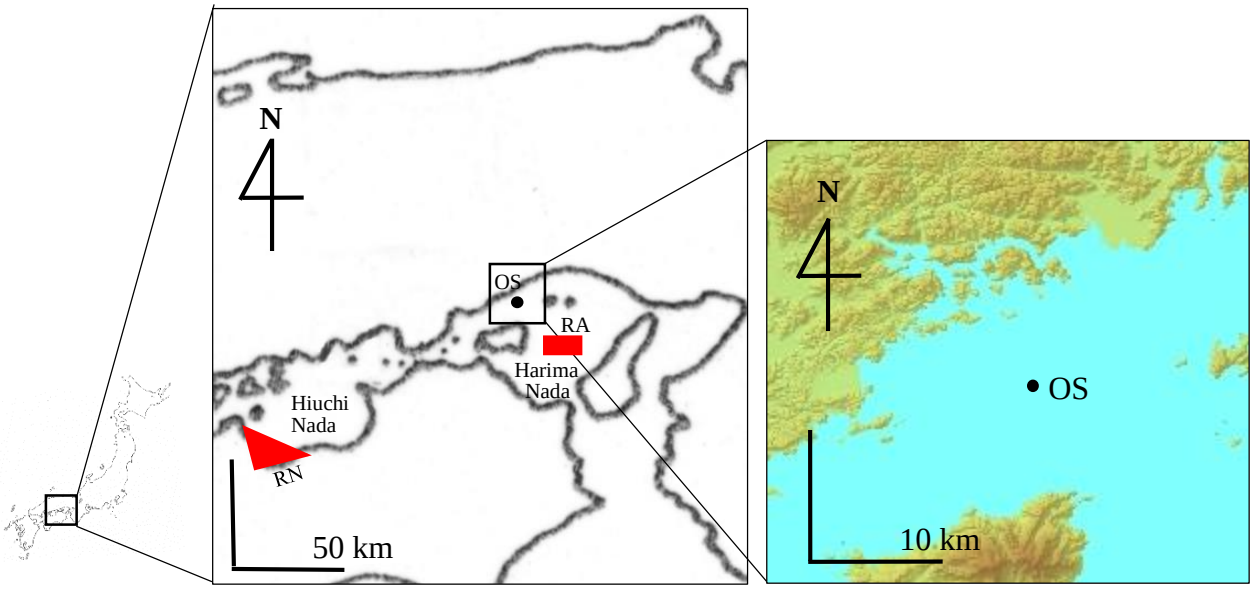

Figure 2. Map showing the location of our observation station (● OS), one of the areas where the concentrations of ^{226}Ra in seawater were measured by Okubo (1981) (▬RA) and the area where the concentrations of ^{222}Rn in seawater were measured by Saito et al. (2012) (◣RN).

dotted with more than 700 islands (The Marine Meteorological Society, 2013). The observation station has a water depth of about 11 m.

We assume that, although atmospheric conditions are different between the station and Nagoya University, the atmospheric concentration of ^{222}Rn except for the background at the station is due to ^{222}Rn exhaled from the section within 80 km radius (mainly within 40 km radius) from the station (Sakashita et al., 1996). Nearly half of the section is covered by the sea, the average water depth of which is about 20 m.

The atmospheric concentration of ^{222}Rn was measured with a ^{222}Rn monitor (Pylon Electronics Inc., Model PMT-TEL with Model AB-5). Air was drawn through a vessel having desiccant (phosphorus pentoxide) to remove humidity, a Whatman® glass microfiber filter (Grade GF/F) to remove all activity except for gaseous radon, and a chamber for measurement at a flow rate of 1.3 ℓ min^{-1}. When the radon-bearing air moves through the chamber, it undergoes decay to progeny. Alpha particles emitted from the nuclei of ^{222}Rn and its short-lived progeny in the chamber were counted with a ZnS (Ag) scintillation detector.

The atmospheric concentration of short-lived ^{222}Rn progeny (the equilibrium-equivalent concentration of ^{222}Rn, EC_{Rn}), which will hereafter be designated as the atmospheric concentration of ^{222}Rn progeny (EC_{Rn}), was measured with a WL monitor (Tracerlab Instruments, Model WLM-200-plus). Air was drawn through a membrane filter of 0.8μm pore size and 47 mm diameter (Advantec Toyo Co. Ltd.) at a flow rate of 1.5 ℓ min^{-1}. Alpha particles emitted from the nuclei of short-lived ^{222}Rn progeny on the filter were counted with a silicon surface barrier detector set in front of the filter.

These Instruments were installed on the small craft by the author, Dr. Eiji Yunoki (Okayama Prefectural Institute for Environmental Science and Public Health), and Dr. Munetoshi Kanayama (Okayama University) (see Figure 3). When the instruments were installed, they were protected from sea sprays.

4. Results and discussion

Figure 4 shows the concentration profiles of ^{222}Rn in seawater estimated using Equation (5) and 7 points in Section 2.1. The concentration profiles of ^{222}Rn in Figures 4 (a), (b) and (c) were estimated using K_{sw}^{Rn} = 0.0022 m^2 s^{-1}, and those in Figures 4 (d), (e) and (f) were done using K_{sw}^{Rn} = 0.02 m^2 s^{-1}. In the case of $-z_b$= 3 m, the

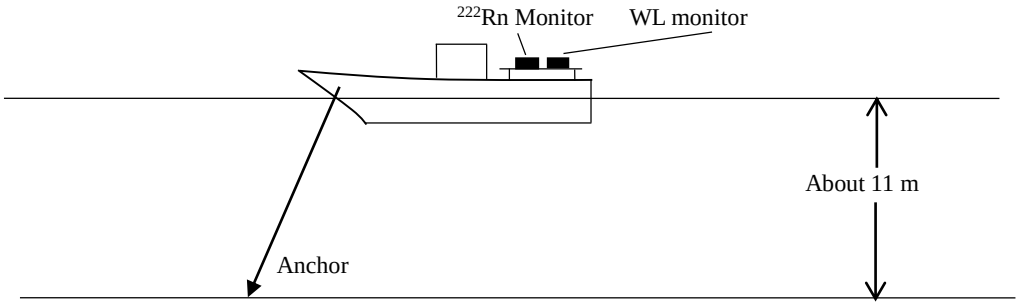

Figure 3. A sketch of our observation station.

concentrations of ^{222}Rn estimated using K_{sw}^{Rn} = 0.0022 m^2 s^{-1} for each $-z_0$ were almost the same as those estimated using K_{sw}^{Rn} = 0.02 m^2 s^{-1} for the same $-z_0$, and they were almost constant with z. In the case of $-z_b$= 11 m, the concentrations of ^{222}Rn estimated using K_{sw}^{Rn} = 0.02 m^2 s^{-1} for each $-z_0$ were almost constant with z, but those estimated using K_{sw}^{Rn} = 0.0022 m^2 s^{-1} increased with an increase in –z. In addition, we note that, in the case of $-z_b$= 11 m, the concentrations of ^{222}Rn near the sea surface estimated using K_{sw}^{Rn} = 0.02 m^2 s^{-1} for each $-z_0$ were higher than those estimated using K_{sw}^{Rn} = 0.0022 m^2 s^{-1} for the same $-z_0$. The case of $-z_b$= 20 m had the same tendency as the case of $-z_b$= 11 m.

Taking $-z_0$ = 100 μm*[5)] and $\delta_{bs}\varrho_{bs}A_{Ra,bs}(1-w_{bs})/100 = 2.9\times10^3$ Bq m^{-3} (four times as large as that assumed by Broecker (1965) in considering the following discharge of coastal groundwater), the concentration of ^{222}Rn in surface seawater is found to be almost the same as the average concentration of ^{222}Rn in seawater measured on the Bahama Banks (Broecker, 1965) (see Table 1), and is also found to be within the range of our estimates shown in Figures 4 (a) and (d). Furthermore, the concentrations of ^{222}Rn in seawater at a position 2 m offshore of the coast of Málaga Bay (Dueñas et al., 1983) (see Table 1) are also found to be within the range of our estimates shown in Figures 4 (a) and (d). The concentrations of ^{222}Rn in seawaters in Bangdu Bay on Jeju Island (Hwang et al., 2005) (see Table 1) were higher than those measured by Broecker (1965) and Dueñas et al. (1983) and those estimated here (see Figure 4). As pointed out by Hwang et al. (2005), this may be due to a relatively large discharge of coastal groundwater in which the concentrations of ^{222}Rn were two to three-fold higher than those in seawater. Contrary to this, in Hiuchi Nada in the Seto Inland Sea (about 130 km southwest from the observation station, see Figure 2), the concentrations of ^{222}Rn in seawaters, which were elevated by mixing with groundwater and/or river water, are within the range of our estimates. Considering that the concentrations of ^{222}Rn in groundwater and in river water are higher than that in seawater and considering the difference in the concentration of ^{226}Ra in sediment and the ^{222}Rn escape-to-production ratio of sediment, our estimates might be reasonable.

Taking $A_{Ra,bs}$ = 26 Bq kg^{-1} (0.7 pg g^{-1}, in the volcanic ash soil of Mauna Loa (Wilkening, 1974)), $D_{sw,0}^{Rn} = 1.43\times10^{-9}$ m^2 s^{-1} for a seawater temperature of 26.5 ^{O}C (near Hiro Bay (Peng et al., 1979)), $-z_0$ = 45 μm and K_{sw}^{Rn} = 0.02 m^2 s^{-1} *[6)], and selecting points 1, 2 and 4 in Section 2.1 as the other conditions, the concentration of ^{222}Rn in surface seawater and the flux density of ^{222}Rn from the sea surface in Hiro Bay are estimated to be 16 Bq m^{-3} (7.5×10^6 atoms m^{-3}) from Equation (5) and 0.50 mBq m^{-2} s^{-1} (240 atoms m^{-2} s^{-1}) from Equation (6), respectively. The estimated flux density of ^{222}Rn is about one sixth of that measured by Wilkening and Clements (1975) (see Table 1). Even if $-z_0$ varies from 10 to 300 μm, the estimated values range from 0.26 ($-z_0$= 300μm) to 0.58 mBq m^{-2} s^{-1} ($-z_0$= 10μm). The difference between the estimated and measured values may be due to a relatively large

*[5)]This value was assumed by Broecker (1965) and is considered to be reasonable in the Atlantic and Pacific oceans (Peng et al., 1979).

*[6)]For $-z_b$ = 3 m, the concentration of ^{222}Rn in seawater in the case of K_{sw}^{Rn} = 0.02 m^2 s^{-1} is almost the same as that in the case of K_{sw}^{Rn} = 0.0022 m^2 s^{-1}.

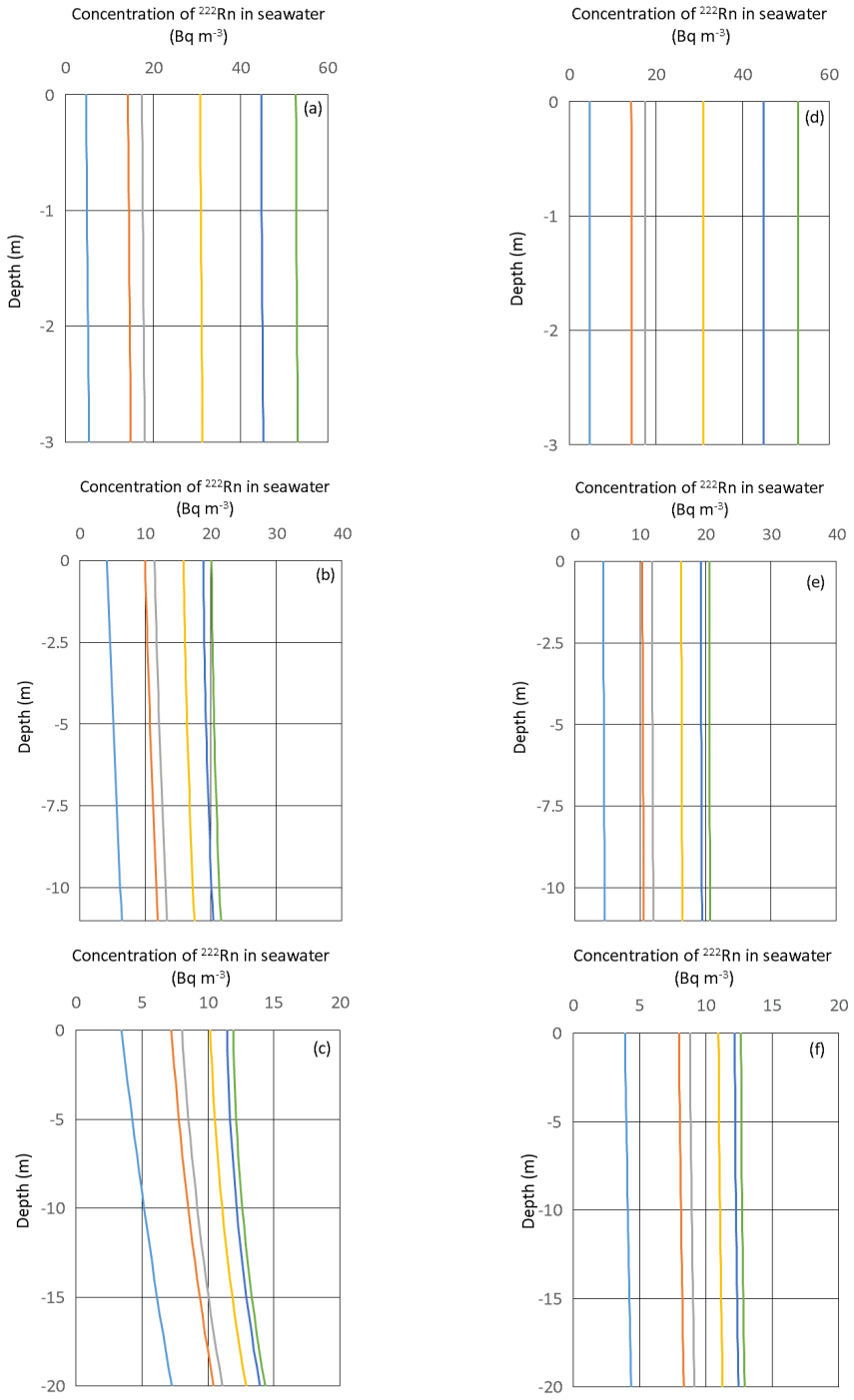

Figure 4. Vertical profiles of ^{222}Rn. ——: 10 μm, ——: 35 μm, ——: 45 μm, ——: 100 μm, ——: 200 μm, ——: 300 μm. (a) K_{sw}^{Rn}=0.0022 m^2 s^{-1}, $-z_b$=3 m, (b) K_{sw}^{Rn}=0.0022 m^2 s^{-1}, $-z_b$=11 m, (c) K_{sw}^{Rn}=0.0022 m^2 s^{-1}, $-z_b$=20 m, (d) K_{sw}^{Rn}=0.02 m^2 s^{-1}, $-z_b$=3 m, (e) K_{sw}^{Rn}=0.02 m^2 s^{-1}, $-z_b$=11 m, (f) K_{sw}^{Rn}=0.02 m^2 s^{-1}, $-z_b$=20 m.

discharge of coastal groundwater like Bangdu Bay. To our deep regret, Wilkening and Clements (1975) lacking the measured concentrations of ^{222}Rn in seawater, we can not discuss this any further.

In Málaga Bay, although the concentrations of ^{222}Rn in seawater were high, the flux densities of ^{222}Rn from the sea surface were almost the same as those from the open ocean surface (Wilkening and Clements, 1975). The film thickness $-z_0$ was estimated to be 270 μm using the data two-thirds of which were obtained in the case of a

wind velocity of less than 3.1 m s^{-1}. Considering that a film thickness of 295 μm was obtained in the case of a wind velocity of less than 3 m s^{-1} (Schindler et al., 1972; Broecker and Peng, 1974), the concentrations of ^{222}Rn and the flux densities of ^{222}Rn obtained in Málaga Bay may be reasonable.

Large shallow seas are almost coupled with continental shelves (e.g., the Chukchi, East Siberian, Laptev, Kara and Barents shelves and so on by which one-third of the total area of the Arctic Ocean is underlain, the northwest European shelf including the North Sea (having an average water depth of 90 m) and the Baltic Sea (having an average water depth of 55 m), the Gulf of Carpentaria (having a maximum water depth of 70 m), Hudson Bay (having an average water depth of about 100 m), the Gulf of Thailand (having an average water depth of 45 m), the Java Sea (having an average water depth of 46 m), the East China Sea shelf (the part where the depth of water is less than 100 m), and the South China Sea shelf (the part where the depth of water is less than 100 m)). The continental shelf zones comprise 7.6 % of the surface area of the world oceans (Sverdrup et al., 1942). Using Equation (6), the five points except for points 6 and 7 in Section 2.1 and $-z_0$ = 45 μm*3), the flux densities of ^{222}Rn in the case of $-z_b$ = 50 m were estimated to be 0.085 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.0022 m^2 s^{-1} and 0.12 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.02 m^2 s^{-1} and those in the case of $-z_b$ = 100 m were estimated to be 0.041 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.0022 m^2 s^{-1} and 0.076 mBq m^{-2} s^{-1} for K_{sw}^{Rn} = 0.02 m^2 s^{-1}. When we take the average flux density of ^{222}Rn for $-z_b$ = 50 m to be 0.10 mBq m^{-2} s^{-1} and that for $-z_b$ = 100 m to be 0.059 mBq m^{-2} s^{-1}, it is found that, except for the South China Sea shelf and part of the East China Sea shelf, the flux densities of ^{222}Rn from large shallow seas estimated by Schery and Huang (2004) are smaller than our estimates. It may also be important for defining the background concentrations of ^{222}Rn in marine atmospheric mixed layer to know accurately the flux densities of ^{222}Rn from large shallow seas.

Let us estimate the concentrations of ^{222}Rn in marine surface air due to the flux density of ^{222}Rn from the sea surface. As mentioned in Section 3, the atmospheric concentration of ^{222}Rn except for the background at our observation station is assumed to be due to ^{222}Rn exhaled from the section within 80 km radius (mainly within 40 km radius) from the station, and nearly half of the section is covered by the sea, the average water depth of which is about 20 m. Furthermore, in the case of $-z_b$ = 20 m, the flux densities of ^{222}Rn estimated using Equation (6) varied from 0.042 mBq m^{-2} s^{-1} (K_{sw}^{Rn} = 0.0022 m^2 s^{-1}, $-z_0$= 300 μm) to 0.41 mBq m^{-2} s^{-1} (K_{sw}^{Rn} = 0.02 m^2 s^{-1}, $-z_0$ = 10 μm). The estimates range from 0.6 % to 6.0 % of that measured on land near the station (6.81 mBq m^{-2} s^{-1}, Kataoka et al. (2001)). Taking these facts into consideration, the estimated contributions of nearly half of the section to the atmospheric concentration of ^{222}Rn at the station (about 3 m above the sea) varied from 0.004 to 0.044 Bq m^{-3}. Table 2 gives the atmospheric concentrations of ^{222}Rn and ^{222}Rn progeny (EC_{Rn}) and the ratios of the atmospheric concentration of ^{222}Rn progeny (EC_{Rn}) to the atmospheric concentration of ^{222}Rn (the ^{222}Rn progeny (EC_{Rn})/ ^{222}Rn ratios) measured at the observation station. In addition, Figure 5 shows time variation of the atmospheric concentrations of ^{222}Rn and ^{222}Rn progeny (EC_{Rn}) and the ^{222}Rn progeny (EC_{Rn})/ ^{222}Rn ratio obtained at a very small part of Okayama City, Uchio located about 12 km north of the coast of the Seto Inland Sea and about 4 km west of the lake made from a part of the creek of the Inland Sea. The atmospheric concentrations of ^{222}Rn and ^{222}Rn progeny (EC_{Rn}) and the ^{222}Rn progeny (EC_{Rn})/ ^{222}Rn ratios measured at the station were almost the same as those measured in the daytime at Uchio. The atmospheric concentrations of ^{222}Rn measured at the station were also almost the same as those obtained in the daytime at a very small part of Kasaoka City, Kabutonishi Town which is located about 3 km north of the coast of the Inland Sea and about 1 km north-north east of a cove (New Kasaoka Port) (Kataoka et al., 2012). Therefore, the largest contribution (0.044 Bq m^{-3}) is 1.1 % of the smaller value of the atmospheric ^{222}Rn concentrations measured at the station (4.06 Bq m^{-3}) and 1.7 % of the smallest value measured on land (Uchio) near the station during the daytime (2.5 Bq m^{-3}, see Figure 5(a)).

Similarly, taking the average water depth of the Great Bahama Bank to be 13 m, at a position (corresponding to our observation station) located about 60 km west from the southern tip of Andros Island, the contribution of

Table 2 Concentrations of ^{222}Rn and its progeny, and their ratio on the Inland Sea.

Date	Time (JST)	Concentration of ^{222}Rn (Bq m^{-3})	Time (JST)	Concentration of ^{222}Rn progeny (EC_{Rn}) (Bq m^{-3})	^{222}Rn progeny (EC_{Rn})/^{222}Rn ratio
15 December 1998	1349-1449	4.06±0.27	1400-1500	2.8±0.4	0.69±0.11
	1449-1549	4.18±0.27	1500-1600	2.5±0.3	0.60±0.08
16 December 1998	- - - - -	- - - - -	1100-1200	5.8±0.8	- - - - -
	1151-1251	5.25±0.32	1200-1300	4.8±0.5	0.91±0.11

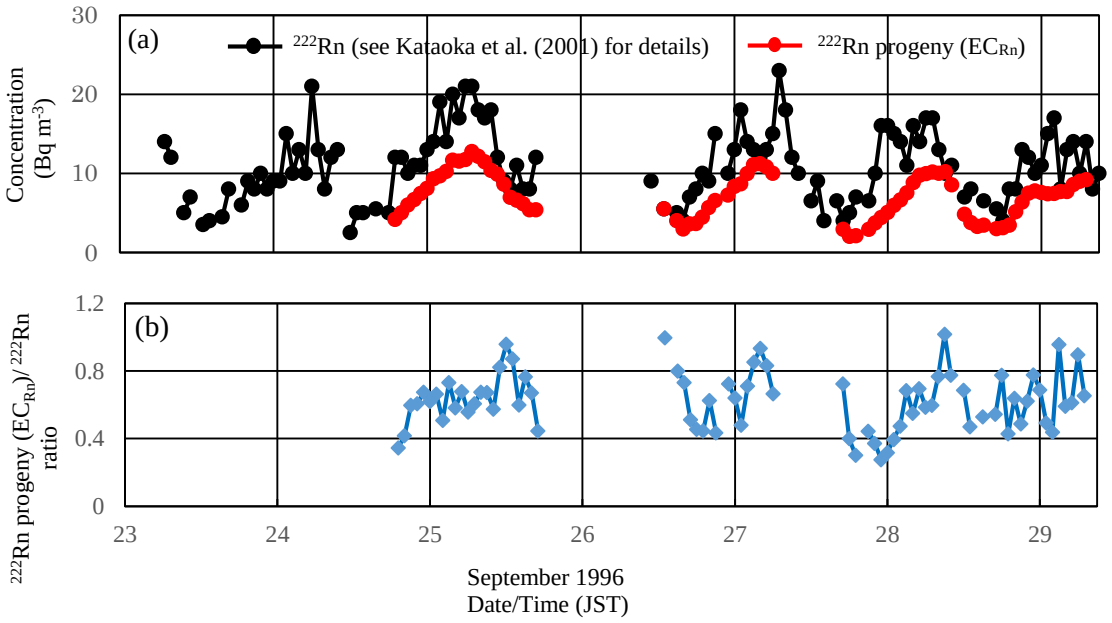

Figure 5. Time variation of (a) the concentrations of ^{222}Rn and ^{222}Rn progeny (EC_{Rn}) and (b) the ^{222}Rn progeny (EC_{Rn})/ ^{222}Rn ratio.

^{222}Rn exhaled from the sea surface of the section within 80 km radius from the position to the atmospheric concentration of ^{222}Rn over the sea is about 3.4 % of that measured at Frazer's Hog Cay on the eastern tip of Chub Cay, in the southern Berry Islands of Bahamas (Broecker, 1965).

5. Conclusions

The concentrations of ^{222}Rn in shallow seas and the flux densities of ^{222}Rn from the sea surface estimated assuming that the seawater was well mixed vertically agree approximately with those measured. However, it is noted that, in order to represent advection by groundwater and river water, three-dimensional (3D) flow and dispersion model should be used instead of one-dimensional (1D) flow and dispersion model.

Except for the South China Sea shelf and part of the East China Sea shelf, the flux densities of ^{222}Rn from large shallow seas estimated by Schery and Huang (2004) were smaller than those estimated in this paper. Furthermore, roughly estimated, the contributions of ^{222}Rn from the sea surface to the atmospheric concentrations of ^{222}Rn over the sea were 1.1 % (1.7 %) of that measured in the Seto Inland Sea and 3.4 % of that measured around Andros Island. Hence, we believe from these results that the flux densities of ^{222}Rn from shallow seas are necessary to be re-estimated and that those in the source term of ^{222}Rn used for atmospheric modeling are also necessary to be modified.

To our deep regret, Broecker (1965) lacked the measured flux density of ^{222}Rn from the sea surface. As far as we know, regarding shallow seas, there are no reports containing a set of measured data on the atmospheric

concentration of ^{222}Rn over the sea, the flux density of ^{222}Rn from the sea surface, the concentration of ^{222}Rn in seawater, the concentration of ^{222}Rn in sediment pore water and the ^{226}Ra concentration in sediment, the ^{222}Rn escape-to-production ratio of sediment, and the concentrations of ^{222}Rn in groundwater and in river water. Hence, it is highly desirable to accumulate such datasets in shallow seas.

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