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Water-sediment distribution and behaviour of Polonium (^{210}Po) in a shallow coastal area with high concentration of dissolved organic matters in water, North Vietnam

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Abstract: The behaviour and water-sediment distribution of particle-reactive Polonium-210 in the marine environment with high concentration of total suspended particulates (TSP) and dissolved organic matters (DOC) along the coast of the Tonkin Gulf (North Vietnam) were investigated. It was revealed that the water-sediment distribution coefficient, $K_d(s)$, of ^{210}Po varied from 2.39×10^3 to 1.9×10^4 (L kg^{-1}) and from 7×10^3 to 2.5×10^5 (L kg^{-1}), respectively, in the rainy and dry season. This implies that in the aquatic environment ^{210}Po tends to be of high affinity to suspended particulates. The ^{210}Po $K_d(s)$ was positively correlated with salinity in both rainy and dry seasons. With DOC the $K_d(s)$ was positively correlated in the rainy season, but in the dry season the relationship tended to be reverse. This behaviour of ^{210}Po in the coastal region was explained by the variation of pH of seawater and by the complexation of the isotope with DOC. The $K_d(s)$ found in this study was in an order lower compared to that reported by Malaysian researchers for the Thailand Gulf. The most important source of ^{210}Po was suggested to be from in-situ generation by the decay of its grand-parent ^{210}Pb which mainly derived from the atmospheric fall-out.

Keywords: Polonium-210, water-sediment distribution coefficient, coastal region, Tonkin Gulf, Vietnam

I. INTRODUCTION

The distribution and behaviour of the natural-series radionuclide polonium-210 (^{210}Po , $T_{1/2} = 138$ days) in the marine environment has been a focus of study for nearly 50 years. Initially, the primary interest in this alpha-emitter originated from the relatively high levels of ^{210}Po measured in some marine biota and the enhanced natural radiation dose they receive from it [1-3]. Additionally, it was noted that ^{210}Po levels in the sea may become enhanced due to direct anthropogenic inputs from the atmosphere and land runoff, e.g. coal combustion and certain mining, industrial, shipping and agricultural activities related to phosphorus fertilizer

application, which could result in increased public and occupational radiation exposure [4-7]. In other scientific fields, owing to its unique physical and chemical properties, ^{210}Po and its grand-parent lead-210 (^{210}Pb , $T_{1/2} = 22.3$ years) were shown to be useful biogeochemical tracers of particle scavenging processes in the sea as well as a tool for delineating specific marine food chain processes [8-10]. Recently the disequilibria between ^{210}Po and ^{210}Pb in seawater and suspended particles have been studied by oceanographers as a possible analogue for assessing particulate organic carbon (POC) flux through the water column [11-15].

Lead-210 is produced throughout the water column by the decay of dissolved Ra-226 ($T_{1/2} = 1,600$ years) and its successive short-lived daughters. This part of ^{210}Pb is called as supported. Besides, ^{210}Pb is also introduced into the sea surface from decay of atmospheric Rn-222 ($T_{1/2}=3.8$ days) and it is called as unsupported or excess lead-210. By beta decays, ^{210}Pb produces ^{210}Po and in certain conditions between the two radio-nuclides would be existed secular equilibrium. Virtually all ^{210}Po in seawater is produced in-situ by the decay of ^{210}Pb [16]. In the oceans, both ^{210}Pb and ^{210}Po may be recycled or settle along with solid particles and eventually be deposited in the marine sediments. Detail on the various pathways through which these nuclides may proceed in the atmospheric or marine environment was described by Du et al. [17]. It was shown that the ^{210}Po and ^{210}Pb mobility in the water column is accomplished by an aggregation of colloidal material and adsorption onto particulate matter [18], therefore they are called particle-reactive, i.e. non-conservative nuclides. On the other hand, ^{210}Po has more tendencies to a biological removal and appears to be more actively involved in the biogeochemical cycle compared to ^{210}Pb [19]. The removal of ^{210}Pb and ^{210}Po from the sea surface into the deep sea is mostly by the scavenging of particulate matters suspended in the water column and adsorption *via* particulate-water interface interaction [20]. In estuaries with high concentrations of dissolved organic matter (DOM), the abundance of these particle-reactive nuclides in the water column tends to be relatively high [21, 22] due to the complexation of metal ions with DOM. Thus, in organic-rich waters, the residence times and ultimate fate of these nuclides are strongly influenced by complexation with DOM. Studies on the geochemical processes of ^{210}Po in open sea and oligotrophic aquatic environment have been conducted by numerous oceanographers as well as

environmentalists and they were summarized by Fowler [23]. However, the water-suspended matters/sediment distribution of ^{210}Po in shallow coastal region with high DOM, turbid, low salinity water is still very limited [24, 25]. It appears that to adequately answer such questions about radiation doses as well as to better understand how ^{210}Po can be best used to measure specific marine biogeochemical processes, an in-depth knowledge of the behaviour of this radionuclide in a variety of marine species (water, suspended matter, sediment, biota) and food chains is necessary.

In this study, ^{210}Po concentration was quantified along the coastal line of the Tonkin Gulf in the Bien Dong (Eastern Sea) with the aim to (i) determine the background level of the nuclide in seawater and sediment in a region having active agricultural, aqua-cultural, traffic and mining as well as coal burning activities; (ii) assess the effect of dissolved organic carbon (DOC) content and pH/salinity in water column on the water-sediment distribution coefficient ($K_d(s)$) of ^{210}Po ; (iii) evaluate important sources of ^{210}Po generation in the marine environment of the study area.

II. MATERIALS AND METHOD

A. Study area

Fig 1 depicts a map of the study area along with sampling locations. The study area was the coastal line along the Tonkin Gulf of the Eastern Sea (North Vietnam) that prolongs from the Thai Binh estuary ($20^\circ 17.33'\text{N}$, $106^\circ 35.20'\text{E}$) to the Van Don Island District (Quang Ninh province, $21^\circ 09.21'\text{N}$, $107^\circ 22.02'\text{E}$). This covers the coastal areas of the Thai Binh, Hai Phong and Quang Ninh provinces that are located within the Red River delta (RRD). Thai Binh province is specialized mainly with rice production. Hai Phong is the biggest and most modern container terminal in the North of Vietnam with through-put capacity of 500,000 TEU's a year [26]. In Quang Ninh province coal mining is the main

activity with a production capacity of around 42 million ton a year (by the 2011) [27]. The coal mining technology applied in Quang Ninh is open excavation thus a huge amount of soil in piles left behind that could threaten the habitat of the Ha Long Bay and the adjacent Bai Tu Long Bay also. This is particularly true in the

Nui Beo (Hon Gai area). With the coal abundance, several coal power plants, e.g. Uong Bi with 710 MWe and Cam Pha (260MWe) as well as cement factories with total capacity of 2.3 million ton a year are currently operated in the locality.



Fig. 1. Sampling sites along the coast from Tra Ly estuary (Thai Binh province) to Van Don Island (Quang Ninh province)

The RRD covers an area of around 1.5 million ha, out of which 47% is used for agriculture and aqua-culture, and 90% of the agricultural area (670,000 ha) is used for annual crops, e.g. rice, corn, sweet potato, etc. The agricultural production in the RRD is much affected by the Red River and Thai Binh River systems as the source of fresh water for irrigation. The discharge of the Red River and the Thai Binh River into the sea, in average, is about $3,300 \text{ m}^3 \text{ s}^{-1}$ and $1,060 \text{ m}^3 \text{ s}^{-1}$, respectively [28, 29]. In rice farming, Vietnamese farmers are used to apply inorganic (N-P-K) fertilizer at a rate of 373.3 kg ha^{-1} of which 30% is P_2O_5 [30].

The RRD is located within a typical wet and hot subtropical climate determined by monsoons. In winter or dry season (November through February), the weather is quite cold

with little rain, and summer or rainy season (April through July) is hot, sunny and rainy. The average annual rainfall is approximately 1,600 mm. The highest rainy season occurs from May to October. In summer, rainfall accounts for 80–85% of total annual precipitation. Average temperatures range from 12°C in December and January, the coolest months, to more than 37°C in July, the hottest month. Mean humidity is greater than 80% throughout the year [29].

B. Sample collection

Table I shows the locations of the sampling campaigns. The first campaign was conducted by the end of the rainy season (Oct–Nov, 2014) and the second one was by the end of the dry season (March, 2015). Table I indicates also the characteristics of each sampling site.

All water and sediment samples were taken from a distance of around 1 km from the seashore at the highest tide with a water column of 3.0-3.5 m, except for the Cat Ba location where water column at high tide was deeper than 5 m. Water samples of 10 L were taken at a depth of 0.5 m from the surface into HDPE cans. Surface sediment (from surface to a depth of 5-7 cm) with an amount of around 0.5 kg (wet weight) was taken using a stainless steel Ponar grab [31] then transferred into dark-wide mouth glass bottles with caps.

Table I. Sampling locations and its characteristics

| Sample ID | Sampling location | Location description |
|-----------|---|---|
| S1 | Thai Binh (201° 17.33'N, 106° 35.20'E) | Aqua-culture area where clams are raised. The location is under the direct impact of fresh water discharge from the Thai Binh River system through Tra Ly, Diem Dien, Van Uc estuaries. Water is turbid. The area is exposed to the air during neap tide. At high tide the water column was as high as 3.0-3.5 m. |
| S2 | Ca Ba I (20° 51.30'N 106° 58.58'E), and | Aqua-culture area where oysters and fishes are raised in cages. The location is under indirect impact from the Bach Dang estuary. Water is relatively clear. During the neap the depth of water column was 2-3 m and during the high tide it was deeper than 5 m. |
| S3 | Cat Ba II (20° 48.15'N 106° 56.57'E) | Aqua-culture area specialized with oyster raising |
| S4 | Tuan Chau (Ha Long Bay) (20° 55.39' N 106° 59.22' E) | A tourism area with intensive boat traffic. Coal mining is carried out deep inland (30-35 km from the seashore). Several streams from the mountainous area discharge fresh water into the Bay. |
| S5 | Cai Lan port (Ha Long Bay) (20° 58.32' N 107° 05.05' E) | A deep-water port. There is a cement factory |
| S6 | Hon Gai (Ha Long Bay) (20° 55.13' N 107° 07.06' E) | Residential and administrative area |
| S7 | Ha Tu (Ha Long Bay) (20° 58.28' N 107° 13.27' E) | A coal mining area |
| S8 | Quang Hanh (Bai Tu Long Bay) (20° 59.43' N 107° 15.33' E) | A coal mining area |
| S9 | Cam Pha (Bai Tu Long Bay) (20° 59.35' N 107° 19.16'E) | The location is under heavy impact of the coal mining activity from the Nui Beo Mine. |
| S10 | Van Don (Bai Tu Long Bay) (21° 02.59'N 107° 21.52'E) | Aqua-cultural area with fish in cage raising |

The samples were transported to the laboratory in Hanoi city for further treatment and measurement for the ^{210}Po activity as well as total dissolved organic carbon (DOC) content. Salinity of seawater was measured directly in the location using an appropriate probe (TOA, Japan).

C. Sample treatment

In the laboratory, water samples, were first vacuum filtered to remove suspended particulates using Millipore filters of 0.45 meshes. An aliquot of the filtrate was taken to quantify the content of total dissolved organic matters (DOC) by wet oxidation on a TOC analyzer (Shimadzu, Japan). Eight mBq of ^{209}Po (chloride solution) as an internal chemical recovery standard was added to the filtrate, thoroughly mixed and then allowed the content to reach the isotopic equilibrium for one day. Afterwards, 10 ml of saturated solution of FeCl_3 as a carrier was added to the filtrate and then $\text{Fe}(\text{OH})_3$ was precipitated by gently adding NH_4OH 1M under continuous stirring. The suspension was allowed to settle overnight then the supernatant was siphoned out. The $\text{Fe}(\text{OH})_3$ with adsorbed constituents, e.g. polonium and other heavy metal ions, was centrifuged. The precipitate was dissolved in 10 ml HCl 1M containing some mg of ascorbic acid to reduce Fe^{3+} , if any, present in the chloride solution. Both ^{209}Po and ^{210}Po from the chloride solution will then spontaneously deposit onto silver plates of 25 mm diameter which directly contacted the solution, at 80 °C for 4 hours to form polonium sources.

A procedure for sediment treatment to get polonium source was described elsewhere [32]. Briefly, it was as follows.

Sediment samples were, first, wet sieved through 1 mm mesh sieve. The suspension was allowed to settle for one day then the supernatant was decanted. The sediment was allowed to dry at room temperature followed by drying at 70 °C overnight. For each analysis

around 2 g of the dried sediment was taken and 8-10mBq of ^{209}Po (PoCl_2 solution) as an internal chemical recovery standard was added. The sample was digested by a mixture of HCl: HNO_3 : HF: H_2O_2 on a hot sand plate till clear solution was obtained. The solution was evaporated to almost dryness then dissolved into 10 ml of HCl 5M. Polonium in the solution was thrice purified by liquid extraction with a mixture of diethyl dithiocarbamate (DDTC) 0.1% in CHCl_3 (10 ml each time). Polonium in the organic phase was twice back-extracted in 5 ml of concentrate HNO_3 and acid phases were combined and evaporated to almost dryness. Note that, do not rise temperature during the evaporation over 80 °C as polonium is an easily volatile substance. Residue of HNO_3 was decomposed by H_2O_2 and then the solid content obtained was allowed to cool down to room temperature followed by dissolving into 10 ml HCl 1 M at 80 °C. If the solution was brown then some mg of ascorbic acid needs to be added to reduce the Fe^{3+} . Polonium sources were prepared by its deposition onto silver plates, similarly to those described above for water samples.

D. Radioactivity measurement and data processing

Activity of ^{209}Po and ^{210}Po was counted on an alpha spectrometer (Alpha-analyst, Canberra, USA) equipped with a PIPS (Passivated Ion-Implanted Planar Silicon) detector. The active area of the detector is 450 mm^2 and its resolution is 18 keV. The activity of ^{209}Po and ^{210}Po was quantified based on the area of the energetic peaks of 4.88 MeV and 5.30 MeV, respectively. The data were processed using Genie-2000 for α -analyst software supplied by the supplier. The activity of ^{210}Po in the samples was corrected to the date of sampling.

The accuracy of the procedure was checked using a certified reference material CRM-4357 (marine sediment) from the NIST

and the deviation from the certified value for ^{210}Po was not exceeded 5-7%.

Water-sediment distribution coefficient $K_d(s)$ is defined as the ratio of specific concentration activity in sediment to those in water [33]. So, $K_d^{210\text{Po}}(s)$ for ^{210}Po is defined as:

$$K_d^{210\text{Po}}(s) = \frac{A_{\text{sed}}^{210\text{Po}}}{A_w^{210\text{Po}}} \quad (1)$$

Where $A_{\text{sed}}^{210\text{Po}}$ is the radio-activity concentration of ^{210}Po in sediment (Bq kg^{-1} dry weight), and $A_w^{210\text{Po}}$ is the radio-activity concentration of ^{210}Po in water (Bq L^{-1}).

The coefficient $K_d(s)$ was used widely to characterize the biogeochemical processes in the lake and marine environment [15, 34-36].

III. RESULTS AND DISCUSSIONS

Table II presents the data of the activity concentration of ^{210}Po in sediment and water samples, the content of DOC and salinity in seawater are also shown in the fourth and fifth

columns, respectively. The value of water-sediment distribution coefficient, $K_d(s)$, of ^{210}Po estimated based on Eq. (1) for each sampling site is presented in the last column of Table II. It should be noted that the content of total suspended matters (TSM) in seawater from the study area during the rainy season (Oct-Nov, 2014) was so high ranging from 0.5 g L^{-1} (in Van Don site) to 1.12 g L^{-1} (in Thai Binh site) with mean \pm stdev (0.6 ± 0.5) g L^{-1} . In contrast, the TSM content in seawater in the dry season was a half of those in the rainy season (0.3 ± 0.2) g L^{-1} (both data not show here). This implies the higher erosion rate from inland during the rainy season compared to those during the dry season. Pham Van Ninh and his co-workers studied the marine environment along the coast of Vietnam had reported that the river systems in the North Vietnam annually discharge into the Tonkin Gulf around 3.1 million ton suspended matter, out of which more than 70% was the contribution from the rainy season (May-October) [37].

Table II. Activity concentration of ^{210}Po in sediment and water along with DOC and salinity in seawater from the study area during the rainy and dry seasons; the $K_d^{\text{Po}-210}(s)$ derived for each sampling site based on Eq. (1) is presented in the last column

| Sampling site | ^{210}Po in sediment, Bq kg^{-1} DW | ^{210}Po in seawater, mBq L^{-1} | DOC in seawater, mg L^{-1} | Salinity, ppt | $K_d^{\text{Po}-210}(s)$, L kg^{-1} |
|---|---|--|-------------------------------------|----------------|---|
| <i>Rainy season (October-December 2014)</i> | | | | | |
| Thai Binh | 196 ± 12 | 8.5 ± 2.2 | 7.3 ± 0.5 | 14.4 ± 0.6 | $(2.3 \pm 1.2)10^4$ |
| Cat Ba I | 106 ± 9 | 15.4 ± 2.5 | 7.8 ± 0.4 | 22.3 ± 0.5 | $(7.0 \pm 1.3)10^3$ |
| Cat Ba II | 111 ± 17 | 7.4 ± 1.7 | 6.7 ± 0.2 | 22.2 ± 0.7 | $(1.5 \pm 0.4)10^4$ |
| Tuan Chau | 108 ± 17 | 56.8 ± 3.2 | 5.8 ± 0.2 | 15.5 ± 0.4 | $(1.9 \pm 0.3)10^3$ |
| Cai Lan port | 170 ± 11 | 22.4 ± 6.5 | 6.9 ± 0.3 | 18.6 ± 0.4 | $(7.6 \pm 0.6)10^3$ |
| Hon Gai | 122 ± 15 | 6.4 ± 2.7 | 2.6 ± 0.2 | 25.6 ± 0.3 | $(1.9 \pm 0.2)10^4$ |
| Ha Tu | 139 ± 13 | 15.4 ± 7.3 | 5.7 ± 0.6 | 24.7 ± 0.6 | $(9.0 \pm 0.1)10^3$ |
| Quang Hanh | 133 ± 11 | 9.5 ± 2.6 | 4.6 ± 0.2 | 24.5 ± 0.3 | $(1.4 \pm 0.3)10^4$ |
| Cam Pha | 122 ± 8 | 17.4 ± 6.6 | 6.8 ± 0.4 | 22.2 ± 0.3 | $(3.7 \pm 0.1)10^3$ |
| Van Don | 129 ± 15 | 7.0 ± 3.4 | 3.5 ± 0.5 | 26.4 ± 0.4 | $(1.9 \pm 0.5)10^3$ |
| <i>Rainy season (March-April 2015)</i> | | | | | |
| Thai Binh | 185 ± 15 | 9.7 ± 3.6 | 4.3 ± 0.2 | 15.9 ± 1.1 | $(1.9 \pm 1.0)10^4$ |
| Cat Ba I | 115 ± 21 | 14.6 ± 1.8 | 3.7 ± 0.3 | 15.6 ± 0.6 | $(7.0 \pm 0.6)10^3$ |
| Cat Ba II | 145 ± 11 | 1.9 ± 2.5 | 4.9 ± 0.2 | 15.7 ± 0.6 | $(7.6 \pm 0.3)10^4$ |
| Tuan Chau | 131 ± 14 | 0.5 ± 0.3 | 6.1 ± 0.2 | 26.3 ± 0.4 | $(2.5 \pm 0.2)10^5$ |
| Cai Lan port | 136 ± 22 | 0.6 ± 0.5 | 5.1 ± 0.2 | 24.3 ± 0.2 | $(2.3 \pm 0.2)10^5$ |

| | | | | | |
|------------|----------|----------|---------|----------|--------------------------|
| Hon Gai | 117 ± 19 | 0.7±0.5 | 4.9±0.2 | 25.6±0.4 | (1.6±0.4)10 ⁵ |
| Ha Tu | 158 ± 12 | 1.4±0.3 | 4.7±0.2 | 23.5±0.3 | (1.1±0.3)10 ⁵ |
| Quang Hanh | 153 ± 16 | 21.9±4.7 | 5.2±0.3 | 17.8±0.5 | (7.0±1.0)10 ³ |
| Cam Pha | 137 ± 16 | 0.7±0.6 | 6.0±0.1 | 28.5±0.2 | (1.9±0.3)10 ⁵ |
| Van Don | 142 ± 19 | 1.1±0.5 | 3.6±0.1 | 26.8±0.2 | (1.3±0.5)10 ⁵ |

Chung and Wu [38] found the ²¹⁰Po concentration in surface water in the open northern Eastern Sea (South China Sea) to be as high as 1.6-2.8 mBq L⁻¹. As seen from Table 2, the ²¹⁰Po concentration in water of the Tonkin Gulf during the dry season was mostly comparable with those found by Chung and Wu [38]. However, in the rainy season ²¹⁰Po in water of the Gulf was from 3 to almost 20 times higher than it was in the open sea (Table 2). This observation could be explained by the higher content of DOC in the coastal water compared to those in the open sea as it could be expected that during the rainy season there was more intense discharge of organic matters to the sea from inland. More detail explanation for the influence of the DOC on the mobilization of ²¹⁰Po in the aquatic environment will be given in the following discussion.

From Table II one can see that the $K_d(s)$ of ²¹⁰Po in the rainy season ranged from 1.9x10³ to 2.3x10⁴ (L kg⁻¹), i.e. it varied in within an order depending upon the sampling site. The highest $K_d(s)$ was found in the Thai Binh site and the lowest was in the Van Don site (Table 2). On the other hand, in the dry season the $K_d(s)$ ranged from 7x10³ to 2.5x10⁵(L kg⁻¹) that was in an order higher compared to those in the rainy season. High value of $K_d(s)$ for ²¹⁰Po in the aquatic environment reflects a fact that this isotope is capable of fast adsorption by the TSM in the water column and ultimately deposited into the bottom. For comparison, the $K_d(s)$ of ²¹⁰Po in the estuarine environment of Mersing River in Malaysia that discharges into the Thailand Gulf was found to be ranging from 5x10⁴ to 501x10⁶ L g⁻¹ depending on the sampling sites and seasons [24]. However, the authors of this study applied another formula (not follow eq.1)

to estimate the $K_d(s)$ viz. $K_d(s) = A_{sed}/A_w \cdot [TSM]$, where [TSM] is the content of total suspended matters in seawater samples, in gram. Unfortunately, in that study the [TSM] was not shown. Supposed the [TSM] in seawater of the Mersing River estuary to be 0.5 g L⁻¹, i.e. in the range for water in the Tonkin Gulf in the rainy season, then the $K_d(s)$ of ²¹⁰Po for the Malaysian case could be as high as from 10⁵ to 5x10⁹(L kg⁻¹) that are from 10² to 10⁴ times higher compared to the $K_d(s)$ of ²¹⁰Po in the Tonkin Gulf. On the other hand, IAEA recommended to use a $K_d(s)$ value of 2x10⁷ (L kg⁻¹) for ²¹⁰Po to simulate geochemical processes in coastal regions [33] with a note that this value was chosen to be equal to the $K_d(s)$ of periodically adjacent elements, but not yet determined experimentally. The difference of $K_d^{210Po}(s)$ found in the Tonkin Gulf from those in the Thailand Gulf as well as the IAEA recommended value could be explained only due to the higher content of DOC in water in the former place (see latter). It should be noted that the study of ²¹⁰Pb and ²¹⁰Po distribution between water and suspended matters in the estuarine environment with high TSM and DOC content is still very scarce, and the $K_d(s)$ found experimentally spreads over a wide range [24, 25].

In this study, the $K_d(s)$ determined for the rainy season was in an order lower than that for the dry season (Table 2). This could be explained by the reduction of the TSM content in seawater during the dry season. Numerous researchers [24, 25, 39-41] have also observed the negative correlation between $K_d(s)$ of ²¹⁰Po as well as its grandparent ²¹⁰Pb and the content of TSM in the open sea. They called this phenomenon as “particle concentration effect,

PCE". The PCE could be understood if the average ^{210}Po supply flux were constant over the year, so high content of suspended matters would dilute the concentration of the radioisotope in the solid phase leading to lowering the $K_d(s)$, according to the Eq.1.

It was well known that estuaries tend to be very dynamic systems with a high degree of temporal and spatial variability in factors such as pH, salinity, DOC and turbidity which could affect the $K_d(s)$ value. High levels of DOC can lead to relatively low lead (Pb) and plutonium

(Pu) $K_d(s)$ [42]. Particulate attached Pu may be released upon contact with low pH river water [43].

Fig 2 depicts the relationship between $K_d(s)$ of ^{210}Po and DOC content and salinity in the rainy season. Figure 3 presents the relationship of $K_d(s)$ of ^{210}Po vs. DOC in the dry season. The trend of ^{210}Po $K_d(s)$ variation with salinity in the dry season was very similar to those in the rainy season that the $K_d(s)$ positively correlated with salinity ($R^2=0.72$, Fig. 2), so this result was not shown in Fig. 3.

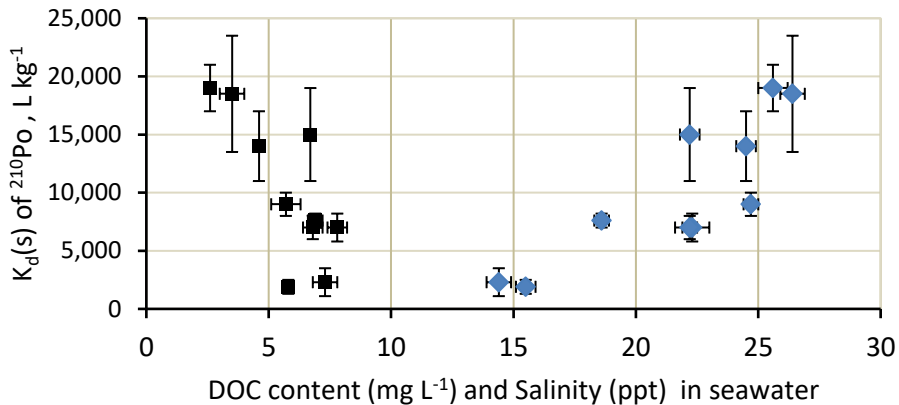


Fig. 2. Relationship between $K_d(s)$ of ^{210}Po and dissolved organic matter (DOC) content (black square points) and salinity (green diamond points) in seawater in the rainy season (Oct-Nov, 2014)

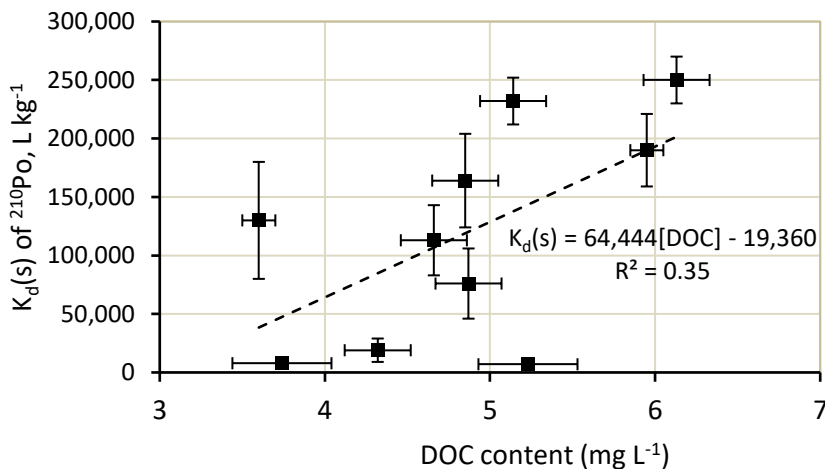


Fig. 3. Relationship between $K_d(s)$ of ^{210}Po and DOC in the dry season (Mar-Apr, 2015)

The positive correlation between $K_d(s)$ of ^{210}Po and salinity (Fig.2) indirectly reflects the influence of pH on the behaviour of the isotope in the marine environment. Salinity in

the coastal marine environment depends upon the amount of fresh water discharged from inland through rivers systems. Mixing of low pH fresh water with seawater should tend the

pH of brackish water to be lower compared to those in pure seawater, and as a consequence polonium attached on particulates could be re-dissolved like the case of plutonium that was observed by Hamilton-Taylor et al. [43]. Therefore, the lower salinity (more freshwater) the lower $K_d(s)$ of polonium would be expected in the brackish water environment.

From Fig. 2 one can see that the higher DOC contents the lower $K_d(s)$. This could be explained by a fact that ^{210}Po can form with DOC complexes which maintain in seawater as colloids not easily be separated by filtration. So, the higher DOC contents the more ^{210}Po in the colloidal component that made $K_d^{210\text{Po}}(s)$ to be lower, according to the Eq. (1) and the relationship $K_d(s)$ vs. DOC was negative (Fig 2). The negative trend of $K_d(s)$ vs. DOC was also observed for other particle-reactive isotopes like ^{210}Pb and $^{239+240}\text{Pu}$ [24, 42]. However, in this study the negative relationship between $K_d(s)$ and DOC was true only in the rainy season. In the dry season the trend of $K_d(s)$ vs. DOC relationship was reverse (Fig. 3) although the correlation was not so strong, $R^2=0.35$ (Fig. 3). The trend of positive correlation of $K_d(s)$ and DOC content

in seawater in the dry season could be explained as follows.

In the dry season, there was no intense discharge from inland that could make the ^{210}Po -DOC complexes-colloids to aggregate themselves and settle to the bottom and from there the colloidal aggregates do not much re-suspend. So the concentration of ^{210}Po in the water phase became lower compared to that in the rainy season. Additionally, in the dry season due to no intense fresh water discharges the pH in brackish water could be high enough that facilitates more ^{210}Po to attach on particulates leading to the increase of $K_d(s)$.

The question is that where does the ^{210}Po in the water column in the coastal region derive from, and how high the rate of its supply is?

It is generally thought that $^{210}\text{Pb}/^{210}\text{Po}$ enters a lake or sea *via* four major pathways: atmospheric deposition, tributaries, catchment runoff, and decay of the grand-grandparent isotope, ^{226}Ra , within the system [17, 44]. Figure 4 depicted one-box model of components and processes that was widely employed in oceanographic studies with the use of the disequilibria of the ^{238}U -natural radioactive chain [45-47].

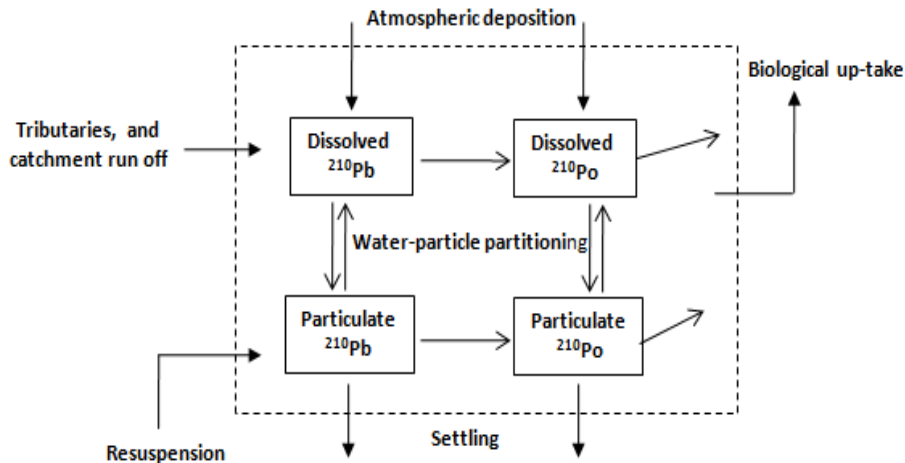


Fig. 4. Model components and processes employed in oceanographic studies with the use of the disequilibria of the ^{238}U -natural radioactive chain. The outer box shows the boundary separating the system and its environment. Solid arrows with solid ends represent fluxes between the system and the larger environment (atmospheric flux and settling). Solid arrows with sketched ends represent internal processes within the system. Arrows pointing from ^{210}Pb to ^{210}Po and arrows pointing from ^{210}Po to space represent isotopic decay

For lakes and oceans in different geographical locations and with variable hydrological conditions, the relative importance of input sources may vary. The flux input of ^{210}Pb and ^{210}Po from the atmospheric deposition was studied either by direct measurements, i.e. collection of wet and dry deposition [48-50] or by estimation from soil or sediment inventories [50-52]. For ^{210}Pb , a compilation in 1990 gave an average rate for direct deposition measurements of $1.02 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ and $0.91 \text{ dpm cm}^{-2} \text{ yr}^{-1}$ estimated based on the inventories [52] (dpm means disintegration per minute). From the eight studies summarized by Kim et al. [53], the average depositional flux of ^{210}Pb in North America within the period from 1977 to 1996 was $(1.0 \pm 0.3) \text{ dpm cm}^{-2} \text{ yr}^{-1}$. Xu et al. [54] studied sedimentation rate in some islands of the Xisha (Hoang Sa by Vietnamese) archipelago and found that the ^{210}Pb fallout flux in that area was of $126 \text{ Bq m}^{-2} \text{ yr}^{-1}$ or around $0.8 \text{ dpm cm}^{-2} \text{ yr}^{-1}$. The ^{210}Pb inventory in 8 sediment cores taken from the Ha Long Bay during 2011-2012 [55] showed the flux of ^{210}Pb deposition in that area to be very close ($0.85 \text{ dpm cm}^{-2} \text{ yr}^{-1}$) to those value found by the Chinese team for Sisha.

The magnitude of in-situ production of ^{210}Pb (decay of ^{226}Ra associated with sediment) can be estimated from sediment profiles through a comparison of supported and unsupported ^{210}Pb . Among the nine cores in depositional areas presented by Klump et al. [56], the ratio of unsupported to supported ^{210}Pb in surface sediments ranged from 5 to 91. Similarly, Evans et al. [57] studied sedimentation rates and depositional processes in the Lake Superior (North America) and found that supported and unsupported ^{210}Pb activities were $1.5\text{--}3.0 \text{ dpm g}^{-1}$ and $11\text{--}216 \text{ dpm g}^{-1}$ respectively, or ratios of excess to supported ^{210}Pb to vary from 7 to 70. On average, supported ^{210}Pb was only 3% of unsupported ^{210}Pb in the surface sediments as

estimated from the results of the two studies. Results of the Nguyen Quang Long et al. [55] study on the sedimentation rate for the Ha Long Bay also showed the contribution of supported to excess ^{210}Pb in the surface sediment to be within 2-3%. Clearly, ^{226}Ra in sediment originated from stream and catchment runoff containing soils not developed on granite rock base like those in the Red River's Delta, is not a significant source of ^{210}Pb relative to the atmospheric deposition; even the oceanic ^{226}Ra concentration (1.63 Bq m^{-3}) would result in a generation of only $6.3 \times 10^{-4} \text{ dpm cm}^{-2} \text{ yr}^{-1}$ of ^{210}Pb [58].

It is more difficult to evaluate the relative importance of all potential sources of ^{210}Po in the marine environment. It was observed that the general ratio of Po:Pb in atmospheric deposition is <0.5 in the northern zone of Canada [48]. Balistrieri et al. [58] observed a $^{210}\text{Po} : ^{210}\text{Pb}$ atmospheric flux ratio of 0.11 ± 0.09 in locations along the west coast of North America. A recent study reported a ratio of 0.1 for aerosols in the Chicago area [60]. Measured $^{210}\text{Po} : ^{210}\text{Pb}$ activity ratios in size-fractionated aerosols from the coast of Japan had a mean value of 0.21 ± 0.13 [61]. However, fallout $^{210}\text{Po} : ^{210}\text{Pb}$ ratio of 0.1 is commonly used for the oceanographic studies nowadays.

From the fallout ^{210}Pb activity of $126 \text{ dpm m}^{-2} \text{ yr}^{-1}$ estimated for the Bien Dong area [53, 54], a generation of $50.4 \text{ dpm m}^{-2} \text{ yr}^{-1}$ for ^{210}Po can be calculated assumed the equilibrium coefficient between ^{210}Pb and ^{210}Po in aquatic environment to be 0.4 [23]. This implies that, in Tonkin Gulf the major source of ^{210}Po in sediment would be in-situ decay of ^{210}Pb but not from the direct atmospheric deposition. The suggestion was proven by the measurement of Nozaki et al. [62] for the Yellow Sea and East China Sea. The authors of this study have found that the direct deposition of ^{210}Po from the atmosphere was as low as $0.01 \text{ dpm m}^{-2} \text{ yr}^{-1}$.

Other sources of ^{210}Po viz. from tributaries discharging particulates containing ^{226}Ra associated with phosphorus fertilizer and from sediments containing ashes released from the coal burning are not as well documented as for ^{210}Pb . However, as it was mentioned previously that the contribution of supported ^{210}Pb into the total ^{210}Pb activity concentration in sediment of the Tonkin Gulf was 2-3% [55] so it was obvious a fact that the portion of ^{210}Po produced from the in-situ decay of supported ^{210}Pb was much smaller compared to the portion of ^{210}Po originated from the decay of unsupported ^{210}Pb . This means that sediment from tributaries and from runoff containing ashes of coal burning were the minor sources of ^{210}Po in the Tonkin Gulf. Therefore, the major source of ^{210}Po to the Tonkin Gulf was in-situ production from decay of ^{210}Pb which was mainly derived from the atmospheric fallout.

IV. CONCLUSIONS

Results of this study confirmed that in the aquatic environment ^{210}Po is mostly attached onto particulates and then deposited to the bottom. The mobilization of ^{210}Po in the aquatic environment is much affected by pH and DOC content in water. The water-particulate/sediment distribution coefficient $K_d(s)$ of ^{210}Po in the environment of the Tonkin Gulf was found to be lower than that of the Thailand Gulf as well as that recommended by IAEA for the open sea and this was explained due to the higher DOC content in water of the former location. The most important source of ^{210}Po in the aquatic environment in the study area is suggested to be in-situ production from its grand-parent ^{210}Pb that mainly derived from the atmospheric fall out.

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