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²¹⁰Po and ²¹⁰Pb concentration in zooplankton of the Syrian coastal waters (eastern Mediterranean Sea)

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Abstract

²¹⁰Po and ²¹⁰Pb activity concentrations and their concentration ratio (CR) in marine zooplankton collected for the first time from the Syrian coastal waters (the eastern Mediterranean Sea) have been determined. The average activity concentrations of ²¹⁰Po and ²¹⁰Pb were 243±36 and 26.4±4.3 Bqkg⁻¹ dw. The ²¹⁰Po/²¹⁰Pb activity ratio was found to be more than one in all zooplankton samples with an average of 9.2 (8.4 to 10), which indicates that the accumulation of ²¹⁰Po in zooplankton does not come from the decay of the ²¹⁰Pb only, and reflects a preferential bioaccumulation of ²¹⁰Po over ²¹⁰Pb. In addition, the Concentration Ratio (CR) for ²¹⁰Po and ²¹⁰Pb reached 10⁴ and 10³, respectively. This study will contribute to the radioecological reference database for zooplankton in the Mediterranean Sea.

Keywords: Zooplankton; Syrian coast; Concentration ratio (CR); Polonium; ²¹⁰Po, Lead; ²¹⁰Pb.

Introduction

Marine organisms accumulate ²¹⁰Po, a naturally occurring radioactive alpha emitter, mainly in their soft tissues, from the water column and their own food. Therefore, ²¹⁰Po is considered as an important source of internal radiation dose to marine organisms (Aarkrog *et al.*, 1997; IAEA, 2004; Fowler, 2011). The beta-emitter ²¹⁰Pb has less affinity for soft tissues, but plays an indirect biological role, as a continuous source of ²¹⁰Po. Concentration of ²¹⁰Po and ²¹⁰Pb can be locally enhanced due to direct inputs from the atmosphere (via decay of their parent ²²²Rn gas) and land runoff via leaching (Al-Masri *et al.*, 2002; Kim *et al.*, 2005; Othman & Al-Masri, 2007).

In the ocean, zooplankton feed on phytoplankton and provide a food source to thousands of species both directly and indirectly. They play an important role in aquatic food webs, as the first link between the primary producers and consumers on higher trophic levels including fishes, which are very important for human diet (Bacha & Amara, 2009; Siokou-Frangou *et al.*, 2010).

Zooplankton have been shown to play a crucial role in the accumulation, transfer and biogeochemical cycling of number of organic compounds, trace elements and radionuclides, by feeding in surface waters and producing

sinking fecal pellets to the bottom. Their importance is a function of their largest biological surface area owing to their small size and large biomass (Altabet & Small, 1990; Reinfelder & Fisher 1991; Stewart and Fisher, 2003; Stewart *et al.*, 2008; Fowler, 2011; Turner, 2015).

Zooplankton has been extensively studied in many seas and oceans of the world from radioactivity point of view (Skwarzec & Bojanowski, 1988; Jeffrey *et al.*, 2004; Mohamed and Kuan, 2005; Carvalho, 2011; Meli *et al.*, 2013; Färber Lorda *et al.*, 2013; Faganeli *et al.*, 2016; Kim *et al.*, 2016; Cho & Kim, 2016). In the Mediterranean Sea, most research has concentrated on its western part where the activity concentration of ²¹⁰Po and ²¹⁰Pb in zooplankton ranged from 217 to 274 BqKg⁻¹ dw and from 24 to 25 Bq Kg⁻¹dw., respectively (Strady *et al.*, 2015). The ²¹⁰Po/²¹⁰Pb activity concentration ratio in zooplankton indicated that these organisms concentrate polonium-210 more than lead-210. However, in spite of this global and regional attention to zooplankton, there is no much data on zooplankton in the eastern Mediterranean Sea waters.

Over decade, the activity concentration and concentration factors (CFs) of ²¹⁰Po and ²¹⁰Pb in some marine organisms such as algae, mollusk, fish and jellyfish have been reported (Othman *et al.*, 1994; Al-Masri *et al.*, 2000; Al-Masri

et al., 2002; Al-Masri *et al.*, 2003; Ammar *et al.*, 2009; Mamish *et al.*, 2015). However, to our knowledge, there is no data reported for radioactivity in zooplankton for the Syrian coastal waters, and the main objective of this work was to acquire baseline data on ^{210}Po and ^{210}Pb activity concentrations for this area (Eastern Mediterranean Sea).

Materials and Methods

Sampling and samples preparation

Zooplankton and seawater samples (three samples from each type) were collected during five cruises using well-equipped small boat, between May 2014 and May 2015, from two Syrian coastal sites located approximately 1 km off the coast of Latakia city (Fig. 1). The first site (MR) is located opposite the sport city (N 35° 33' 19.95", E 35° 43' 49.07"), which is considered as one of the most important tourist areas on the Syrian coast, the water depth ranges from 17 to 19 m. The second site (MN) is located opposite the Al-Kaber Al-Shamali river estuary (N 35° 29' 44.32", E 35° 48' 0.44"), where river water carries urban, industrial and agricultural runoff waters and discharges directly into the sea. Water depth in this site ranges from 5 to 7 m.

Zooplankton was sampled by means of near-surface horizontal hauls at a depth of one meter from the surface, using a standard WP2 net (diameter 56 cm, mesh size 200 μm) (Suthers & Rissik, 2008). Each tow was continued for 30 minutes at a low speed of about 3 knots per hour, in order to keep the plankton net in the horizontal position at the required depth. Once the net was lifted out of the water, it was rinsed down gently from the outside with ambient seawater. Net was visually inspected to ensure all zooplankton were rinsed off the net cloth and concentrated into the sample bucket. The sample was transferred to a clean sample storage bottle. The zooplankton sam-

ples were refrigerated as soon as possible after collection.

At the laboratory, suspended matter and phytoplankton were removed from the samples by tweezers under a microscope, and then the samples were sieved with a same-sized mesh filter. The wet sample was weighted, then after oven drying for 3 days at 60°C, the zooplankton was homogenized in a mortar and dry weight was taken. The analysis of ^{210}Po and ^{210}Pb were performed on the dry weight samples.

Surface seawater samples were also collected at a depth of one meter from the surface, using a 10 liters Niskin-Type water sampler. At the laboratory, seawater samples were filtered through 45 μm pore size membrane filter and acidified by concentrated hydrochloric acid; pH value was determined to be less than 2 to reduce the adsorption of radionuclides on the inner surface of the storage containers and to prevent the growth of algae.

Radioactivity measurements

Activity concentrations of ^{210}Po and ^{210}Pb in zooplankton were determined using the spontaneous deposition of ^{210}Po onto a rotating silver disc technique (Saito and Cunha, 1997). The analyses were performed on 0.5 g aliquots of dried and homogenized zooplankton sample, and ^{208}Po tracer was added to each sample for radiochemical recovery. Each sample was digested using a combination of mineral acids (nitric and hydrochloric acid) for at least 24 h. When the solution was clear, the sample was gently evaporated to near dryness. The residue was dissolved in 100 ml of 0.5 mol l⁻¹ hydrochloric acid. The solution was heated to 80°C and ^{210}Po was spontaneously plated onto a rotating silver disc after reduction of iron with ascorbic acid. Alpha counting of ^{208}Po tracer and ^{210}Po was performed using an alpha spectrometer (Oasis, Oxford) with a passive ion-implanted silicon detector (active area 300 mm², background counts per day 3.6 and the mini-

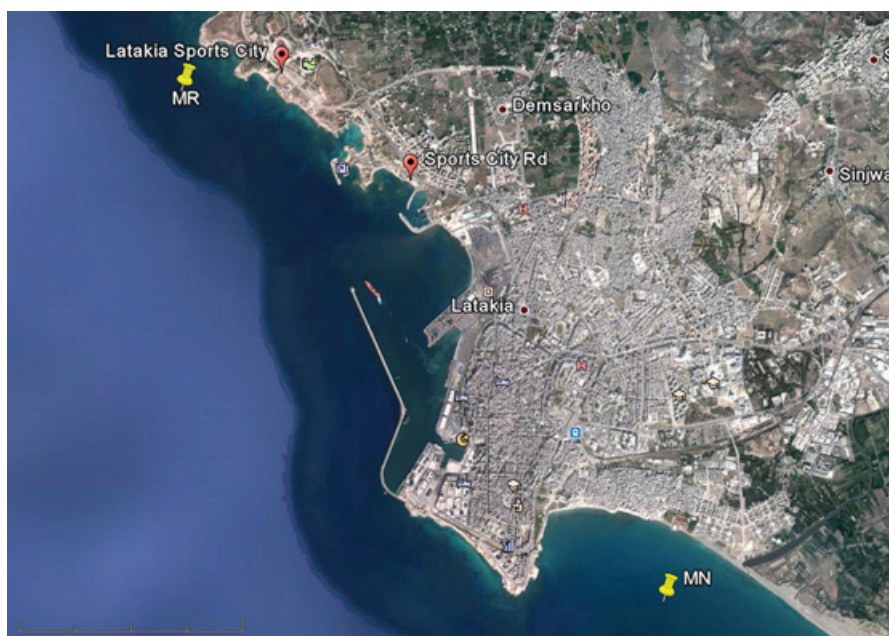


Fig. 1: Sampling Location.

mum depletion thickness 100 mm). The plating solutions were stored at least for 6 months, then evaporated and digested as mentioned for ^{210}Po , the plating and counting were then repeated to measure the ingrowths of new ^{210}Po from ^{210}Pb and to calculate the ^{210}Pb activity concentration in the original sample. For seawater samples, ^{210}Po and ^{210}Pb were co-precipitated with MnO_2 from 20 l of filtered seawater (La Rosa *et al.*, 2001). The precipitate was dissolved in 1.5 mol l⁻¹ of hydrochloric acid and processed as described for zooplankton samples. The lower limit of detection of the method used was 0.4 Bq kg⁻¹ dw.

Concentration ratio (CR)

Concentration Ratio ($\text{CR}_{\text{wo-media}}$) is the usual approach to predict the activity concentration of a radionuclide in the biota from the activity concentration in water (Tagami and Uchida, 2013). It was calculated from the ratio of the radionuclide concentration in zooplankton (Bq kg⁻¹ fresh weight) to that in water (Bq l⁻¹) following this equation (IAEA, 2014):

$$\text{CR}_{\text{wo-media}} =$$

$$\frac{\text{Activity concentration in zooplankton (Bq kg}^{-1}\text{, fresh weight)}}{\text{Activity concentration in (filtered) seawater (Bq l}^{-1}\text{)}}$$

Results and Discussion

The results presented here are given in activity concentrations in mBq l⁻¹ (\pm standard deviation) for seawater samples and Bq kg⁻¹ dw (\pm standard deviation) for zooplankton samples.

^{210}Po and ^{210}Pb in Seawater

Table 1 shows the activity concentrations of ^{210}Po , ^{210}Pb and $^{210}\text{Po}/^{210}\text{Pb}$ ratio in seawater. The average activity concentrations of ^{210}Po and ^{210}Pb in seawater at the two sites were 1.38 \pm 0.2 and 0.75 \pm 0.04 mBq l⁻¹, respectively for the first site (MR), and 1.54 \pm 0.3 and 0.85 \pm 0.05 mBq l⁻¹, respectively for the second site (MN). The average activity ratios of $^{210}\text{Po}/^{210}\text{Pb}$ for the first and second sites samples were 1.85 and 1.80, respectively. The

mean total activity concentrations of ^{210}Po and ^{210}Pb in seawater including all dates and sites were 1.46 \pm 0.2 and 0.8 \pm 0.1 mBq l⁻¹, and the mean $^{210}\text{Po}/^{210}\text{Pb}$ was 1.82. The activity concentration of ^{210}Po and ^{210}Pb in both sites were very close and this due to their spatial proximity. There is however a difference between MN and MR sites in all dates except August, but the differences stay within the error bars. May be it is due to the influence of the Al-Kabir Al-Shamali River, a coastal river where the greatest abundance of river water with agriculture, industrial and human contaminants are discharged into the sea during rainy seasons, while it is almost dry in summer and the river impact is negligible. However, these surface seawater results are within the world surface seawater values (1 mBq l⁻¹ for ^{210}Po , and between 0.1 and 0.4 mBq l⁻¹ for ^{210}Pb , Table 2). The average activity concentrations of ^{210}Po and ^{210}Pb of 44 surface seawater samples collected from the Syrian coastal waters ranged between 1.1 and 1.4 mBq l⁻¹ for ^{210}Po and between 0.5 and 0.7 mBq l⁻¹ for ^{210}Pb (Mamish *et al.*, 2015).

^{210}Po and ^{210}Pb in zooplankton

Table 3 shows the activity concentrations of ^{210}Po , ^{210}Pb and $^{210}\text{Po}/^{210}\text{Pb}$ ratios in the zooplankton. The average activities of ^{210}Po and ^{210}Pb in zooplankton at the two sites were 239 \pm 28 and 25.8 \pm 3.3 Bq kg⁻¹ dw for the first site (MR), and 247 \pm 36.6 and 27 \pm 4.4 Bq kg⁻¹ dw for the second site (MN), respectively. The average total activity concentrations for both sites were 243 \pm 36 and 26.4 \pm 4.3 Bq kg⁻¹ dw, respectively. As for seawater, there was still a relatively higher value in the MN site in comparison with MR, for all sampling times except August. The activity ratios ranged from 8.4 to 10, with average activity ratios for MR and MN at 9.3 and 9.2, respectively. The average including all dates was 9.2. This value agrees with the fact that ^{210}Po is more accumulated in zooplankton than ^{210}Pb , a well-known effect already described in the literature (Mohamed and Kuan, 2005; Carvalho, 2011; Strady *et al.*, 2015). Several authors (Carvalho, 2011; Fowler, 2011; Färber Lorda *et al.*, 2013; Strady *et al.*, 2015) explained the over unity of the $^{210}\text{Po}/^{210}\text{Pb}$ in zooplankton by the fact that ^{210}Po is bind to amino acids and proteins, and concentrated to high levels within the organisms through

Table 1. Activity concentration and activity concentration ratio of ^{210}Po and ^{210}Pb in filtered seawater (mBq l⁻¹).

| Sampling date (Season) | ^{210}Po | | ^{210}Pb | | $^{210}\text{Po}/^{210}\text{Pb}$ | |
|---------------------------|-------------------|---------------|-------------------|----------------|-----------------------------------|------|
| | MR | MN | MR | MN | MR | MN |
| 28/5/2014 (Spring) | 1.5 \pm 0.1 | 1.7 \pm 0.2 | 0.71 \pm 0.1 | 0.84 \pm 0.1 | 2.1 | 2.0 |
| 29/8/2014 Summer | 1.2 \pm 0.1 | 1.1 \pm 0.1 | 0.69 \pm 0.1 | 0.78 \pm 0.1 | 1.7 | 1.4 |
| 21/10/2014 Autumn | 1.3 \pm 0.1 | 1.8 \pm 0.3 | 0.80 \pm 0.1 | 0.92 \pm 0.1 | 1.6 | 2.0 |
| 10/3/2015 Spring | 1.3 \pm 0.1 | 1.4 \pm 0.1 | 0.76 \pm 0.1 | 0.82 \pm 0.1 | 1.7 | 1.7 |
| 15/5/2015 Spring | 1.6 \pm 0.1 | 1.7 \pm 0.2 | 0.79 \pm 0.1 | 0.91 \pm 0.1 | 2.0 | 1.9 |
| Average | 1.38 | 1.54 | 0.75 | 0.85 | 1.85 | 1.8 |
| Standard error | 0.15 | 0.26 | 0.04 | 0.07 | 0.22 | 0.26 |
| RSD % | 10.6 | 17 | 5.78 | 6.3 | 11.7 | 14 |

Table 2. ^{210}Po and ^{210}Pb in seawater of the seas and oceans of the world.

| Location | ^{210}Po mBq l ⁻¹ | ^{210}Pb mBq l ⁻¹ | $^{210}\text{Po}/^{210}\text{Pb}$ | Reference |
|---|--|--|-----------------------------------|---|
| North Atlantic – Portages coast | 1.7±0.08 | 2.27±0.16 | 0.75 | Carvalho, 2011 |
| NW Mediterranean Sea, Gulf of Lion, France | 3-5 | 11-12 | 0.3-0.5 | Strady <i>et al.</i> , 2015 |
| N. Adriatic Sea – Gulf of Trieste | 0.56-3.75 | 2.2 | | Faganeli <i>et al.</i> , 2016 |
| Central Adriatic Sea – Italy | 0.33±0.14 | 0.42±0.26 | 0.78 | Meli <i>et al.</i> , 2013 |
| Southern Baltic Sea | 0.49±0.11 | | | Skwarzec and Bojanowski, 1988 |
| SW Pacific Ocean | 0.47-1.58 | 0.81-3.19 | | Jeffrey <i>et al.</i> , 2004 |
| Korean Coastal Water | 0.75±0.06 | 1.22±0.09 | 0.62±0.07 | Cho and Kim, 2016; Kim <i>et al.</i> , 2016 |
| E. Mediterranean Sea - Syrian Coast, Lattakia | 1.1-1.8 | 0.7-0.9 | 1.4-2.1 | This study |

Table 3. Activity concentrations and activity concentration ratios of ^{210}Po and ^{210}Pb in zooplankton (Bq kg⁻¹ dw.).

| Sampling date | Drying factor % | | ^{210}Po | | ^{210}Pb | | $^{210}\text{Po}/^{210}\text{Pb}$ | |
|-----------------------|-----------------|------|-------------------|--------|-------------------|-------|-----------------------------------|-----|
| | MR | MN | MR | MN | MR | MN | MR | MN |
| 28/5/2014 | 0.09 | 0.08 | 233±28 | 247±31 | 25±5 | 27±8 | 9.3 | 9.1 |
| 29/8/2014 | 0.13 | 0.16 | 185±22 | 172±19 | 21±4 | 19±6 | 8.8 | 9.1 |
| 21/10/2014 | 0.08 | 0.13 | 272±43 | 287±46 | 32±5 | 34±6 | 8.5 | 8.4 |
| 10/3/2015 | 0.13 | 0.14 | 265±38 | 275±41 | 27±9 | 29±11 | 9.8 | 9.5 |
| 15/5/2015 | 0.15 | 0.18 | 240±29 | 254±57 | 24±5 | 26±7 | 10 | 9.8 |
| Average | | | 239 | 247 | 25.8 | 27 | 9.3 | 9.2 |
| Standard error | | | 28 | 36.6 | 3.3 | 4.4 | 0.5 | 0.4 |
| RSD % | | | 11.7 | 14.8 | 12.9 | 16.4 | 5.6 | 4.5 |

Table 4. CR values of ^{210}Po and ^{210}Pb in zooplankton.

| Sampling date | CR ^{210}Po | | CR ^{210}Pb | |
|----------------|----------------------|----------|----------------------|----------|
| | MR | MN | MR | MN |
| 28/05/2014 | 1.40E+04 | 1.31E+04 | 3.17E+03 | 2.89E+03 |
| 29/08/2014 | 1.85E+04 | 1.97E+04 | 3.96E+03 | 3.90E+03 |
| 21/10/2014 | 1.67E+04 | 2.07E+04 | 3.20E+03 | 4.80E+03 |
| 10/03/2015 | 2.87E+04 | 3.50E+04 | 4.62E+03 | 4.95E+03 |
| 15/5/2015 | 2.25E+04 | 2.69E+04 | 4.56E+03 | 5.14E+03 |
| Average | 2.01E+04 | 2.31E+04 | 3.89E+03 | 4.34E+03 |
| Standard error | 5.12E+03 | 7.40E+03 | 6.29E+02 | 8.40E+02 |
| RSD % | 2.55E+01 | 3.21E+01 | 1.61E+01 | 1.94E+01 |

various metabolic activities. In contrast, ^{210}Pb is not retained by the organism metabolism, and zooplankton released ^{210}Pb more quickly via fecal pellet excretion and do not accumulate it in their bodies as much as ^{210}Po .

Table 4 shows the activities of ^{210}Po and ^{210}Pb in zooplankton from different regions. It can be seen that the reported values are within the activity ranges reported in other areas in the world.

Concentration ratio (CR)

Table 5 shows the Concentration Ratio (CR) of ^{210}Po and ^{210}Pb in zooplankton, which reached 10^4 and 10^3 , respectively. However, the reported CR values are found to be within the reported values in other areas, as seen in Table 4.

Table 5. ^{210}Po and ^{210}Pb activity concentration and CR values for zooplankton in seas and oceans of the world.

| Location | ^{210}Po | | ^{210}Pb | | $^{210}\text{Po}/^{210}\text{Pb}$ | CR | | Reference |
|--|----------------------------|--------------------------------|--------------------------------|--------------------------------|-----------------------------------|-------------------------|-------------------------|---------------------------------|
| | Bq Kg ⁻¹ dw. | Bq Kg ⁻¹ wet wt. | Bq Kg ⁻¹ dry wt. | Bq Kg ⁻¹ wet wt. | | ^{210}Po | ^{210}Pb | |
| North Atlantic – Portages coast | | 38±14 | | 8.4±2.6 | 4.4 | | | Carvalho, 2011 |
| NW Mediterranean Sea, Gulf of Lion, France | 217-277 | | 24-25 | | 10-12 | | | Strady <i>et al.</i> , 2015 |
| N. Adriatic Sea – Gulf of Triest | 240-1800 | | 1122 | | | 1.1x10 ⁴ | | Faganeli <i>et al.</i> , 2016 |
| Central Adriatic Sea – Italy | 133.2±47 | 8.9±3.1 | 79.6±56 | 5.3±3.9 | 1.67 | 2.4x10 ⁴ | 1.3x10 ⁴ | Meli <i>et al.</i> , 2013 |
| Southern Baltic Sea | 143±107 | 20.9±14 | | | | 42x10 ³ | | Skwarzec and Bojanowski 1988 |
| SW Pacific Ocean | 565-1737 | | 47-551 | | | 1.3-3.3x10 ⁵ | 0.9-9.1x10 ⁴ | R. Jeffrey <i>et al.</i> , 2004 |
| Malaysia , South China Sea | 365±21 | | 94±7 | | 4 | | | Mohamed and Kuan, 2005 |
| Coastal Water of the Jeju Island, Korea | | 107±2 | | 7±1 | 0.62±0.07 | 1.4x10 ⁵ | 5.7x10 ³ | Cho and Kim, 2016 |
| Korean costal water | | 32-137 | | | | 4-18x10 ⁴ | | Kim <i>et al.</i> , 2016 |
| W Mediterranean Sea - Monaco | | | | | | 1-3x10 ⁴ | | Fowler, 2002 |
| N. Mediterranean coastal waters | 175-878 | | 7.5-486 | | | | | Färber <i>et al.</i> , 2013 |
| E Mediterranean Sea-Syrian Coast Lattakia | 172-287 | 21-45.7 | 19-34 | 2.3-4.7 | 8.4-10 | 2.2x10 ⁴ | 4.2x10 ³ | This study |

Conclusion

^{210}Po and ^{210}Pb activity concentrations have been determined in zooplankton of Lattakia coastal waters for the first time along the Syrian coast (the Eastern Mediterranean Sea). The activity concentrations of ^{210}Po and ^{210}Pb found in this study are within the worldwide reported values. Small variations of activity concentrations in zooplankton collected close to river estuary, may be due to the influence of river inputs. The activity ratio of $^{210}\text{Po}/^{210}\text{Pb}$ is more than one in zooplankton, ranging from 8.4 to 10. The concentration ratios (CR) as well as the activities in the surface waters are within the range of world values.

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