



Article The Atmospheric Input of Dissolvable Pb Based on the Radioactive ²¹⁰Pb Budget in the Equatorial Western Indian Ocean

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Abstract: To estimate the atmospheric deposition flux of ²¹⁰Pb in the equatorial western Indian Ocean, we determined the dissolved (<0.45 μ m) and particulate ²¹⁰Pb (>0.45 μ m) in the water column. In addition, we calculated the atmosphere-derived dissolvable Pb in seawater using the budget of ²¹⁰Pb. The dissolved ²¹⁰Pb and total ²¹⁰Pb were higher in the surface layer and, overall, showed a decreasing distribution with depth. In particular, radioactive ²¹⁰Pb activities in the surface-to-upper layer (<1000 m depth) were 1.5 to 2 times higher than those reported in the 1970s (in nearby regions), suggesting that there has been additional ²¹⁰Pb input in recent years. Based on the mass balance of the total ²¹⁰Pb budget in the water column, we estimated the atmospheric deposition flux of ²¹⁰Pb and the residence time of Pb for the first time in this region. The atmospheric deposition flux of ²¹⁰Pb was estimated to be 0.1–0.5 dpm cm⁻² yr⁻¹, and these values agreed with the general global estimations for the major oceans (0.1–0.7 dpm cm⁻² yr⁻¹). Considering the residence time of ²¹⁰Pb (29–41 years) in the water column (estimated from the ²¹⁰Pb inventory and ²³⁴Th-based Pb scavenging rate), the atmospheric input of seawater-dissolvable Pb was quantified to be 0.08–0.1 nmol cm⁻² yr⁻¹, which is about eight times higher than the estimated input in the early 1990s in the region. Therefore, these results imply that radioactive ²¹⁰Pb could be a useful tracer for quantifying Pb flux in seawater.

Keywords: ²¹⁰Pb; atmospheric input; seawater-dissolvable Pb; flux; Indian Ocean

1. Introduction

As a naturally occurring radionuclide from the ²³⁸U decay series, ²¹⁰Pb (half-life $(T_{1/2}) = 22.3$ years) is mostly derived from the decay of ²²²Rn in the atmosphere and is mainly generated via the in situ decay of ²²⁶Ra in deep water [1]. ²¹⁰Pb shows the highest concentration at the atmosphere–ocean interface layer due to dry deposition (in the form of particles) and wet deposition (directly supplied to the upper ocean via precipitation) [2,3]. ²¹⁰Pb has a half-life suitable for tracing the behavior of particulate matter and is used to estimate the biogeochemical cycle of chemical species due to its adsorption properties towards particles in an aquatic environment [4–9].

Although the sources of ²¹⁰Pb and Pb could be different, it has been suggested that they have similar behaviors and removal mechanisms in seawater [3]. Therefore, ²¹⁰Pb can be used to understand the observed behavior of Pb in the ocean. ²¹⁰Pb has been studied to trace the behavior of anthropogenic Pb in the marine environment using ratios of Pb/²¹⁰Pb [10–14]. Recently, seawater-dissolvable Pb was quantified using the scavenging rate of ²¹⁰Pb [14]. Despite these various studies, Pb is known to have a wide range of solubility in seawater (13–90%) [15]. Therefore, the determination of Pb solubility in seawater remains a challenging issue, and thus, Pb solubility-related studies in various environments and geographical locations are required.



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The Indian Ocean differs from the Pacific and Atlantic Oceans in that it accounts for 30% of the global oceans, but it has a range of only 25° N latitude [16]. Due to these geographic characteristics and its complex seafloor topography, this region has distinct and complex physical features, such as seasonal climate variations (monsoons) and various patterns of ocean currents and circulations [17]. The Indian Ocean is surrounded by rapidly developing countries such as India and South Africa. As a result of the increased hightemperature industrial activities, the late phase-out of leaded gasoline (in the late 1990s and mid-2000s), and the weak environmental regulations of these countries [18–22], Pb emissions in the Indian Ocean have increased over the past few decades. For example, a recent study reported that the Pb emissions from coal combustion have increased by almost 15 times in India [23]. In addition, the concentrations of Pb in seawater were extremely high in the Indonesian coastal region (range of 600–2900 nM) [24–26] and Indian Ocean coast near Kenya (35-340 nM) [27]. Moreover, wildfires from Australia and Indonesia may have transported atmospheric Pb into the ocean through an easterly wind [28]. The Indian Ocean is one of the areas with scarce Pb data and is the least explored compared to the Pacific and Atlantic oceans [23], although Pb inventories in this region have likely been increasing compared to the past.

Therefore, in this study, (i) we investigated the distributions of radioactive ²¹⁰Pb in the water column of the equatorial Indian Ocean and compared them to those reported in the past (before the 1980s) [29] to evaluate the changes in Pb inventories in this region, and (ii) we evaluated the atmospheric inputs of ²¹⁰Pb based on the mass balance of the ²¹⁰Pb in the water column of the Indian Ocean. Then, (iii) we also quantified the atmospheric seawater-dissolvable Pb inputs coupled with recently reported Pb concentrations (inventory) in our study area, since Pb solubility in the ocean is still controversial.

2. Materials and Methods

2.1. Sampling

To determine the dissolved and particulate ²¹⁰Pb in seawater, water samples from the equatorial western Indian Ocean were obtained during a research cruise in April 2018 (Figure 1). Seawater samples (8 L, n = 28) for ²¹⁰Pb analysis were collected in high-density polyethylene (HDPE) bottles from Niskin samplers from 3 stations. The samples were filtered (0.45 μ m, polycarbonate, Millipore), and the dissolved water samples were acidified with 6 N HCl (pH < 2) immediately after sampling to prevent ²¹⁰Pb from adsorbing onto the bottle; then, the filtered samples were stored in petri dishes at room temperature until analysis.

2.2. Analytical Procedure

In this study, the ²¹⁰Pb in seawater was measured using ²¹⁰Po, which was analyzed according to a previously published protocol [31]. Briefly, a ²⁰⁹Po spike (1 dpm g⁻¹), stable Pb (for monitoring the chemical yield) (1 dpm g⁻¹), and an Fe³⁺ (100 mg g⁻¹) carrier were added to all samples and stirred for 6 h. Ammonium hydroxide was used to adjust the pH to ~8 for the co-precipitation of ²¹⁰Po and Fe(OH)₃. The supernatants were removed, and then, the precipitates were digested with HNO₃ and HCl to remove any organic matter in the samples. The particulate (filtered) samples were digested with a solution of concentrated HNO₃ and HCl (1:1 v/v) and repeatedly heated until the sample was completely dissolved.

All the samples were dried down after rinsing with 0.5 M HCl, and then, 100 mL of 0.5 HCl and 0.5 g of ascorbic acid (to reduce Fe^{3+}) were added to the samples. Po was plated on a silver (Ag) planchet (Φ 24.1 mm and 0.15 mm thickness) (99.9% Ag, Aldrich, Burlington, MA, USA) coated with commercial nail polish on one side for 15 h with stirring. The ²¹⁰Po activities on the silver planchet were counted using alpha spectrometry (Alpha Analyst, Mirion Technology, (Former Canberra, Australia), Canada, USA). The measured counts were corrected for the background of the alpha spectrometry, the decay of ²¹⁰Po during counting, the recovery of the ²⁰⁹Po spike, the decay of ²¹⁰Pb from sampling to



plating, the recovery of the ²⁰⁹Po spike, the decay of ²¹⁰Pb from sampling to plating, and the reagent blank.

Figure 1. Sampling locations (square symbols) for ²¹⁰Pb (stations 13, 19, and 34) and dissolved Pb (1, 5, 13, 19, 24, 29, 34, and 38; data from [30]) together with all stations covered by the previous (**a**) dissolved ²¹⁰Pb and (**b**) total ²¹⁰Pb [29] (from GEOSECS data in 1970s) data (triangle symbols), and (**c**) Pb data from the Indian Ocean. (**a**) Sampling locations for dissolved ²¹⁰Pb and surface activities of dissolved ²¹⁰Pb (dpm 100 L⁻¹). (**b**) Sampling locations for total ²¹⁰Pb and surface activities of total ²¹⁰Pb. (**c**) Sampling locations for dissolved Pb, including various mean annual surface currents (colored arrows) and wind directions (black arrows) during the sampling period (April to May) in the western Indian Ocean.

After removing the silver plate on which the ²¹⁰Po was adsorbed, ²¹⁰Pb analysis was performed using the remaining solution. The samples were heated while adding a sufficient amount of conc. HNO₃ to the sample to decompose any ascorbic acid contained in the solution. The samples were dried after rinsing with 9 M HCl, and 5 mL of 9 M HCl was added to the samples. To separate ²¹⁰Pb from ²¹⁰Po, 50 mL of 9 M HCl was conditioned by passing it through a column (~2.5 cm length of quartz wool, 5–6 cm length of resin, and some glass wool) filled with an anion exchange resin (AG1-x8, 100-200 mesh, Bio-Rad Laboratories, Inc., Hercules, CA, USA), followed by passing the samples and washing them 4 times with 5 mL of 9 M HCl. The eluted samples were stored in vials, and then, a 209 Po tracer (1 dpm g^{-1}) was added to the samples and they were incubated for at least 6 months to generate ²¹⁰Po, a daughter nuclide of ²¹⁰Pb. The generated ²¹⁰Po was measured through the same process. The concentration of ²¹⁰Pb was calculated using the measured ²¹⁰Po concentration, incubation time, and recovery rate, which was calculated through the Pb concentration, measured using an inductively coupled plasma mass spectrometer (ICP-MS) (Element 2, Thermo Fisher Scientific, Waltham, MA, USA). To calculate the recovery of the Pb carrier added to the sample, the standardization of Pb was performed using a 500-fold dilution (0–50 ppb) of the Pb carrier in Milli-Q water. The Pb carrier for standardization and the diluted samples for the calculation of Pb recovery were measured using an ICP-MS. The recovery of Pb was calculated using a calibration curve between the measured sample and the result of the Pb carrier dilution (count s^{-1}). ²¹⁰Pb activity was calculated from the measured ²¹⁰Po activity, which revealed that the average chemical yield of stable Pb was $84.07 \pm 15.07 \%$ (n = 87).

3. Results and Discussion

3.1. Hydrological Properties

Potential temperature and salinity data were used to identify major water masses along the occupied transect (Figure 2). In this study region, the Indian Ocean possesses different water mass characteristics, such as temperature and salinity, and we observed various water masses in this study: ITW (Indonesian Throughflow Water), STUW (South Indian Subtropical Underwater), SICW (South Indian Central Water), ROSW (Red Sea Overflow Water), AAIW (Antarctic Intermediate Water), IDW (Indian Deep Water), and CDW (Circumpolar Deep Water). The ITW, STUW, SICW, SAMW, and RSOW are observed in the surface-to-upper intermediate layer (0–1000 m), the AAIW and IDW are observed in the intermediate layer (1000–2000 m), and the CDW is observed in the deep layer (2000–4000 m). The water masses observed in this study were defined according to previous similarly conducted studies for this study region, e.g., [32–36].



Figure 2. T–S diagram indicating the identified water masses in the equatorial western Indian Ocean and major water masses (**left figure**) and distributions of dissolved Pb by depth (data from Kim et al. [30]) in this study area (modified from recent work of Kim et al. [30]) (**right figure**). Isopycnals are shown as gray lines. Abbreviations: ITW (Indonesian Throughflow Water), STUW (South Indian Subtropical Underwater), SICW (South Indian Central Water), ROSW (Red Sea Overflow Water), AAIW (Antarctic Intermediate Water), IDW (Indian Ocean Deep water), and CDW (Circumpolar Deep Water).

3.2. Distribution of Radioactive ²¹⁰Pb in the Indian Ocean

The horizontal distributions of dissolved ²¹⁰Pb, particulate ²¹⁰Pb, and total ²¹⁰Pb (dissolved + particulate) in the surface layer (0–20 m) in this study are shown in Figure 1b,c. All the ²¹⁰Pb data are presented in Table 1.

Station	Depth (m)	Dissolved ²¹⁰ Pb (dpm 100 L ⁻¹)	Particulate ²¹⁰ Pb (dpm 100 L ⁻¹)	Total ²¹⁰ Pb (dpm 100 L ⁻¹)
5	.00° S, 60.	00° E (2018.04.11)		
	0	21.47 ± 0.79	0.97 ± 0.17	22.44 ± 0.81
	50	12.05 ± 0.44	0.31 ± 0.2	12.36 ± 0.48
	100	8.71 ± 0.45	0.44 ± 0.09	9.14 ± 0.46
	200	10.54 ± 0.6	0.6 ± 0.06	11.14 ± 0.6
.1 10	300	8.48 ± 0.46	1.20 ± 0.11	9.68 ± 0.47
st. 13	500	9.19 ± 0.43	0.6 ± 0.07	9.79 ± 0.44
	1000	14.46 ± 0.6	0.68 ± 0.08	15.14 ± 0.6
	2000	9.9 ± 0.49	0.87 ± 0.08	10.77 ± 0.49
	3000	10.96 ± 0.46	1.81 ± 0.12	12.77 ± 0.48
	4035	10.67 ± 0.46	0.99 ± 0.08	11.66 ± 0.46

Table 1. Activities of ²¹⁰Pb in the equatorial western Indian Ocean.

Station	Depth (m)	Dissolved ²¹⁰ Pb (dpm 100 L ⁻¹)	Particulate ²¹⁰ Pb (dpm 100 L ⁻¹)	Total ²¹⁰ Pb (dpm 100 L ⁻¹)
5	.27° S, 67.	90° E (2018.04.15)		
	0	13.94 ± 0.55	0.96 ± 0.11	14.9 ± 0.56
st. 19	50	10.85 ± 0.59	1.07 ± 0.25	11.92 ± 0.64
	100	8.39 ± 0.4	0.71 ± 0.12	9.10 ± 0.41
	200	8.76 ± 0.4	0.8 ± 0.09	9.56 ± 0.41
	300	8.96 ± 0.4	0.78 ± 0.09	9.73 ± 0.41
	500	9.46 ± 0.41	0.84 ± 0.12	10.3 ± 0.43
	1000	12.21 ± 0.54	0.88 ± 0.15	13.10 ± 0.56
	2000	14.48 ± 0.64	0.73 ± 0.07	15.21 ± 0.64
	3006	11.64 ± 0.49	1.70 ± 0.16	13.34 ± 0.52
20).00° S, 67	.05° E (2018.04.23)		
	0	19.25 ± 0.71	0.82 ± 0.07	20.07 ± 0.72
	50	20.36 ± 0.67	1.42 ± 0.11	21.78 ± 0.68
20.00	100	19.17 ± 0.72	0.88 ± 0.16	20.05 ± 0.73
	200	16.03 ± 0.58	0.79 ± 0.1	16.82 ± 0.59
st. 34	300	13.37 ± 0.55	0.82 ± 0.08	14.19 ± 0.55
	500	11.55 ± 0.48	1.02 ± 0.11	12.57 ± 0.49
	1000	13.87 ± 0.5	0.87 ± 0.09	14.74 ± 0.51
	1500	16.38 ± 0.72	1.06 ± 0.1	17.44 ± 0.72
	2040	11.64 ± 0.44	2.37 ± 0.12	14.01 ± 0.45

Table 1. Cont.

Dissolved ²¹⁰Pb and total ²¹⁰Pb showed slightly higher activities in the western part (60.00° E) (21.47 dpm 100 L⁻¹ and 22.44 dpm 100 L⁻¹, respectively) than stations in the eastern region (st. 19 and st. 34) (16.60 \pm 3.76 dpm 100 L⁻¹ and 17.49 \pm 3.65 dpm 100 L⁻¹, respectively). These results may be attributed to the more lithogenic materials and/or matter originating from land from the African continent and Mascarene Plateau (Figure 1) [37] along the South Equatorial Current (SEC, orange arrow in Figure 1a).

The vertical distributions of dissolved ²¹⁰Pb and total ²¹⁰Pb in the water column are shown in Figure 3. The vertical distributions of both dissolved ²¹⁰Pb and total ²¹⁰Pb in the Indian Ocean showed higher activities in the surface layer and lower activities in the deeper layer. The average activities of dissolved ²¹⁰Pb and total ²¹⁰Pb were 11.32 \pm 0.56 dpm 100 L⁻¹ and 13 \pm 1.21 dpm 100 L⁻¹, respectively. The activities of both dissolved ²¹⁰Pb and total ²¹⁰Pb and total ²¹⁰Pb and total ²¹⁰Pb and total ²¹⁰Pb mere 11.32 \pm 0.56 dpm 100 L⁻¹ and 13 \pm 1.21 dpm 100 L⁻¹, respectively. The activities of both dissolved ²¹⁰Pb and total ²¹⁰Pb mere 11.32 \pm 0.56 dpm 100 L⁻¹ and 13 \pm 1.21 dpm 100 L⁻¹, respectively. The activities of both dissolved ²¹⁰Pb and total ²¹⁰Pb and total ²¹⁰Pb mere 11.32 \pm 1.72 dpm 100 L⁻¹ and 15.1 \pm 1.79 dpm 100 L⁻¹, respectively).

In the surface layer, the dissolved and total ²¹⁰Pb activities were about 1.9 times higher than those reported in a previous study [29] (Geochemical Ocean Sections Study (GEOSECS) cruise data from the 1970s) (Figure 3). In the same study area, dissolved Pb metal concentrations were also about 2.3 times higher than those reported in previous studies [30,38].

The total and dissolved ²¹⁰Pb activities in the surface-to-intermediate layer (<1000 m) in this study were consistently 1.5 to 2 times higher than those in previous observations (in the 1970s GEOSECS data) of the equatorial western Indian Ocean. The ²¹⁰Pb activities in this study and previous measurements of ²¹⁰Pb activity data from various ocean/seawater samples are presented in Table 2. The total and dissolved ²¹⁰Pb activities (<1000 m) in this study were also similar to or relatively higher than those in the north Atlantic, some Pacific regions, and the Antarctic. These results may be due to the following reasons. First, ²¹⁰Pb may be introduced by the long-range transport of coastal water masses, such as from the Indonesian coastal region (600–2900 nM dissolved Pb) [24–26] and African coast (near Kenya) (35–340 nM dissolved Pb) [27] along the Indian Ocean subtropical gyre. Second, the increased Pb may come from modern anthropogenic sources (e.g., coal combustion, mining and smelting operations, etc.) from various industries in the surrounding countries. For example, Schaule and Patterson [39] suggested that the Pb increase is congruent with that

observed for ²¹⁰Pb concentrations in the same water sampled at the same times. Recently, Witt et al. [40] suggested that the ratios of stable Pb isotopes (^{206,207,208}Pb) are consistent with coal combustion and its increased importance as a source of Pb around the Indian Ocean. Moreover, Lee et al. [22] suggested that the Pb in the Chagos coral (located near the study area) reflects the predominance of India's industrial (gasoline and coal) Pb in this region. Third, Pb released into the atmosphere from wildfires in southeast Asia and Australia may have been transported by seasonal and easterly winds and deposited into the Indian Ocean. Das et al. [41] determined that the ratios of Pb isotopes (²⁰⁸Pb/²⁰⁷Pb and ²⁰⁶Pb/²⁰⁷Pb) increased during wildfire haze periods in Indonesia, and suggested that the suspension of the crustal material was the dominant emission source of total suspended particulate (TSP) matter. Fourth, it could be the result of the additional dissolution of particulate Pb from the artificial origin (i.e., especially fine particulate forms smaller than 50 µm, which are dissolved very efficiently), which is continuously introduced to the water column on the way to being exported to the deeper layer. Overall, the ²¹⁰Pb in the upper layer (<1000 m) in the water column of in this study region was consistently increased.



Figure 3. Vertical profiles of (**a**) dissolved ²¹⁰Pb and (**b**) total ²¹⁰Pb in the Indian Ocean. The GEOSECS data from the 1970s [29] obtained from nearby stations in our study area are shown for comparison (see Figure 1).

Table 2. Comparison of ²¹⁰Pb activities in seawater from various ocean/marginal sea regions.

Site	Data	Denth (m)	²¹⁰ Pb Activity (dpm 100 L ⁻¹)		Poforonco
Site	Date	Deptil (III)	Total	Dissolved	Reference
This study	2018.04	<1000	13.74 (9.10–22.44) (n = 28)	12.91 (8.39–21.47) (n = 28)	
This study	2018.04	>1000	13.60(10.77-17.44)(n = 28)	12.24 (9.90–16.38) (n = 28)	
In dian Oraan	1070-	<1000	9.51 (4.95–13.46) (n = 28)	9.35(4.50-15.70)(n = 36)	[20]
Indian Ocean	19705	>1000	11.16(5.51-14.51)(n = 70)	10.47 (4.50 - 13.70) (n = 82)	[29]
Nouth Design	2000 05	<1000	19.25(11.64 - 31.23)(n = 11)	18.96(11.40-31.10)(n = 11)	[40]
North Pacific	2009.05	>1000	26.26(23.89-28.62)(n=2)	25.95(23.60-28.30)(n = 2)	[42]
North Atlantia	2010 10	<1000	12.93 (6.56-24.10) (n = 57)	12.36(6.18-23.37)(n = 64)	[42]
North Atlantic	2010.10	>1000	11.38 (5.10–17.72) (n = 25)	10.56 (1.63 - 22.30) (n = 40)	[43]
South Antarctic	2007.07	<1000	4.12(0.72-12.20)(n = 170)	3.03(0.24-10.28)(n = 161)	[44]
Plaak Coa	1000 06	<1000	4.22 (0.89-22.4) (n = 49)	2.53 (0.50 - 21.88) (n = 50)	[45]
DIACK Sea	1900.00	>1000	3.96(3.40-5.09)(n=7)	1.52(1.15-2.97)(n=7)	[40]
Pacific Ocean marginal					
sea	2013.10	<350	3.09 (1.49–6.95) (n = 27)	2.01 (0.90–2.99) (n = 27)	[46]
(ECS: East China Sea)					
East Pacific Zonal	2012 11	<1000	11.90 (5.51–40.20) (n = 63)	11.57 (5.35–39.80) (n = 63)	[47]
Transect (EPZT)	2013.11	>1000	16.92 (5.12 - 26.30) (n = 64)	15.45 (1.87–25.20) (n = 64)	[1/]
Fast Sea /Sea of Japan	2015.05	<1000	9.66 (5.20–16.40) (n = 16)		[14]
Lust sea, sea of jupan	2013.03	>1000	5.21 (3.80–7.20) (n = 13)		

3.3. ²¹⁰Pb Budget

The budget of ²¹⁰Pb in the Indian Ocean is estimated using the box of a steady-state scavenging model (0–300 m). At steady state ($\partial A/\partial t = 0$), by neglecting advection and diffusion, the rate of change of ²¹⁰Pb activity can be expressed as following Equation (1):

$$\frac{\partial A_{210Pb}}{\partial t} = \lambda_{210Pb} (A_{226Ra} - A_{210Pb}) + F_{Atm} - (k_{210Pb} A_{210Pb}) = 0$$
(1)

where A is the inventory of each radionuclide (dpm cm^{-2}) in the 0–300 m depth water column, and λ , F_{Atm} , and k represent the decay constant of ²¹⁰Pb (0.0311 yr⁻¹), the atmospheric depositional flux of 210 Pb (dpm cm⁻² yr⁻¹), and the first-order scavenging rate constant (yr⁻¹), respectively. The atmospheric input of ²¹⁰Pb should be balanced with the in situ-production from mother nuclei, ²²⁶Ra (F_{Ingrowth}), the in situ decay of ²¹⁰Pb (F_{decay}), and the settling of flux to the deeper layer (Fexport) (Figure 4). We estimated FIngrowth by multiplying the ²²⁶Ra inventory and the decay constant of ²¹⁰Pb, and F_{decay} by multiplying the ²¹⁰Pb inventory and the decay constant of ²¹⁰Pb. The inventory of ²²⁶Ra in the south Indian Ocean was taken from recently published results [48]. Fexport was calculated by multiplying the 210 Pb inventory and the first-order scavenging rate constant (yr⁻¹). The first-order scavenging rate constant was obtained from previous published results (k_{210Pb} ; 0.02-0.07 yr⁻¹ in 0–300 m of water column), which was calculated using the ²³⁴Th-based export flux of Pb [37] in the same station (also using the same sample) in this study. The unknown constant F_{Atm} was calculated by assuming a steady state. Each calculated term was schematized as a box model in Figure 4. The F_{Atm} (atmospheric depositional flux of ²¹⁰Pb) was calculated to be 0.10 ± 0.05 – 0.50 ± 0.16 dpm cm⁻² yr⁻¹. The atmospheric depositional flux of ²¹⁰Pb estimated in this study was comparable to those in the major oceans, such as the North Pacific (0.22–0.30 dpm cm⁻² yr⁻¹ [49]), the Equatorial Pacific $(0.11-0.51 \text{ dpm cm}^{-2} \text{ yr}^{-1} \text{ [50]})$, the North Atlantic $(0.4-0.69 \text{ dpm cm}^{-2} \text{ yr}^{-1} \text{ [51,52]})$, the northeasternmost part of the Indian Ocean (~0.4 dpm cm⁻² yr⁻¹ [53]), and the Arabian Sea (0.73 dpm cm⁻² yr⁻¹ [54]) near the northern Indian Ocean.



Figure 4. A schematic box model accounting for the ingrowth, decay, export, and atmospheric flux of 210 Pb (dpm cm⁻² yr⁻¹) in the equatorial western Indian Ocean.

From the ²¹⁰Pb budget, the residence time of total ²¹⁰Pb in the water column was calculated using the following Equation (2):

$$\tau = \frac{1}{k_{210Pb}} = \frac{A_{210Pb}}{A_{226Ra}\lambda_{210Pb} - A_{210Pb}\lambda_{210Pb} + F_{Atm}}$$
(2)

Here, τ is the residence time of the total ²¹⁰Pb, and it was estimated to be in the range of 28.7–40.9 (average: 36.07 years) in the 0–300 m layer. The calculated residence time, about 28.7–40.9 years, in this study is comparable with that in the North Pacific (54–96 years) [55,56], Southeastern Pacific (95 years) [47], and Atlantic Oceans (15–22 years) [57].

3.4. Atmospheric Input of Seawater-Dissolvable Pb

In order to calculate the atmospheric input of seawater-dissolvable Pb in the Indian Ocean, we used the residence time of dissolved ²¹⁰Pb in this study. The residence time of dissolved ²¹⁰Pb in this region was estimated to be 36.5 ± 6.6 years at a water column depth of 0–300 m using the activity of dissolved ²¹⁰Pb. The average annual atmospheric depositional flux of seawater-dissolvable Pb can be calculated by dividing the inventory of dissolved Pb by the residence time of dissolved ²¹⁰Pb, resulting in 0.08 \pm 0.02 nmol cm⁻² yr⁻¹ in this study (Figure 5 and Table 3). The residence time of ²³⁴Th or particulate Pb in the upper layer of nearby regions ranges from tens of days to 1 year [30,58,59], which is significantly shorter than the residence time of the dissolved Pb estimated in this study. In this study, the dissolvable Pb flux in the upper layer may be higher because the fine Pb particles that first enter the ocean are rapidly scavenged, and therefore, not detectable in the deeper layers.

The calculated soluble Pb flux (0.08 ± 0.02 nmol cm⁻² yr⁻¹) from the atmosphere in this study was about eight times higher than the previously estimated total Pb flux $(0.019 \text{ nmol cm}^{-2} \text{ yr}^{-1})$ in the northern Indian Ocean [15], see Table 3). This result implies that there has been an increase in the atmospheric input of Pb into the Indian Ocean to this day, in contrast to the Pacific and Atlantic oceans, where Pb inventories are now decreasing due to the ban on the use of leaded gasoline. We also noted that modern Pb input from the atmosphere (unlike the lithogenic dust with coarse particles) has a relatively smaller particle size due to its artificial origin (fine particle sources measuring <1~50 µm (e.g., PM10, PM2.5, etc.)), and seems to be more soluble in seawater. In addition, the soluble Pb fluxes (0.08 ± 0.02 nmol cm⁻² yr⁻¹) in this study were also higher than the fluxes of wet deposition of Pb in remote oceans, including the North Pacific (0.05–0.08 nmol cm⁻² yr^{-1} [10,15]) and North Atlantic (average: 0.4 nmol cm⁻² yr⁻¹ [15,52,60,61]), and also comparable with the marginal sea region near the continent, such as the Arabian Sea (in the Indian Ocean, ~ 0.12 nmol cm⁻² yr⁻¹ [62]) (Table 3). These results imply that radioactive ²¹⁰Pb could be a useful tracer for quantifying actually dissolvable fractions of atmospheric depositional Pb flux into seawater.



Figure 5. A schematic box model accounting for residence time of dissolved Pb, Pb inventory, and atmospheric flux of seawater-dissolved Pb (nmol $\text{cm}^{-2} \text{ yr}^{-1}$) in the equatorial western Indian Ocean.

Study Area	Collection Period	Fraction	Pb Flux (nmol cm ⁻² yr ⁻¹)	Reference
This study	2018.04	Actual dissolvable fraction into the seawater	0.08 ± 0.02	
North Indian Ocean (Arabian Sea)	1986.10–1986.11	Total deposition flux Soluble atmospheric flux	0.483 ~0.116	[62]
North Indian Ocean	1985	Total deposition flux Dry deposition flux Wet deposition flux	$0.024 \\ 0.005 \\ 0.019$	[15]
South Indian Ocean (Kerguelen)	2008.11-2010.10	Total deposition flux	0.003 ± 0.000	[63]
South Indian Ocean (Crozet)	2010.01-2010.11	Total deposition flux	0.009 ± 0.001	
North Pacific Ocean	1979	Wet deposition flux	0.05-0.08	[10,15]
Northwestern	1996.06 1997.05	Dry deposition flux	0.277 0.913	
Atlantic Ocean	1996.06 1997.05	Wet deposition flux	0.022 1.095	[60]
Northwestern Atlantic Ocean	1985–1990	Wet deposition flux	0.030-0.460	[61]
Western Mediterranean	2011.09–2012.08	Dry deposition flux Wet deposition flux	$0.483 - 1.448 \\ \sim 3.089$	[64]
Northwestern Mediterranean	1986–1992	Total deposition flux of dissolved Pb	0.440-0.881	[65]
Red Sea (Gulf of Agaba)	2003–2005	Dry deposition flux	0.063-0.384	[66]
East Sea/ Sea of Japan	2018.01-2018.02	Actual dissolvable fraction	0.98 ± 0.28	[14]
Japan	2008.02-2008.05	Total deposition flux	0.25-3.29	[67]

Table 3. Comparison of depositional fluxes of Pb (nmol $\text{cm}^{-2} \text{ yr}^{-1}$) with other oceans.

4. Conclusions

For the first time, this study quantified the ²¹⁰Pb budget using the mass balance of total ²¹⁰Pb in the equatorial western Indian Ocean, where Pb is expected to be introduced through various pathways (e.g., industrial activities, the late phase-out of leaded gasoline, wildfires from Australia and Indonesia, etc.). Compared with data from the 1970s (the only published ²¹⁰Pb data from the Indian Ocean), ²¹⁰Pb in the Indian Ocean has increased 1.5- to 2-fold in the upper layer (<1000 m depth) of the water column. This suggests a continuous input of Pb of anthropogenic origin into this region. We estimated that the atmospheric deposition flux of ²¹⁰Pb was 0.1–0.5 dpm cm⁻² yr⁻¹, based on the ²¹⁰Pb budget in our study area. Based on this atmospheric input of ²¹⁰Pb, the residence time of ²¹⁰Pb in the Indian Ocean (0–300 m layer) was calculated to be 29-41 years. Applying this ²¹⁰Pb residence time, seawater-dissolvable Pb was quantified to be 0.08–0.10 nmol cm⁻² yr⁻¹. However, since this estimate was calculated using ²²⁶Ra data from nearby areas, ²²⁶Ra analysis should be performed at this study site in the future in order to calculate a more accurate value. Moreover, in order to better understand the behavior of trace elements, including ²¹⁰Pb, it is necessary to analyze stable Pb isotopes or the particulate fraction of Pb to identify the source of anthropogenic Pb, in order to quantify Pb solubility, and to estimate the input of terrestrial and/or lithogenic materials by investigating radioactive ²²⁶Ra in the Indian Ocean.

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