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## Lead (Pb) Heavy Metal Existence in the Eastern Indonesian Seas

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## ABSTRACT

Lead contamination results in environmental degradation, especially in elevated quantities. The Flores Sea, an international maritime corridor, contains a succession of submerged volcanoes. Furthermore, the region is influenced by the Indonesian throughflow, which transports water masses from the Pacific Ocean to the Indian Ocean. This study aimed to determine the distribution of lead in the Flores Sea at five depths and to evaluate the pollution index of the area. Lead contents in the water were quantified utilizing the AAS method, and the pollution index was determined employing the IP formula. The data were subsequently analyzed by correlation to elucidate the relationship between lead dispersion and water depth. The results revealed that the maximum lead concentration (0.8mgL<sup>-1</sup>) was detected at a depth of 150 meters, while the minimum value (0.081mgL<sup>-1</sup>) was seen at 25 meters. Lead contamination is ascribed to maritime activity and the water masses of the Pacific Ocean transported by Indonesian Through Flow (ITF). According to these data, the Flores Sea is classified as moderately contaminated.

# **INTRODUCTION**

Marine pollution implies the spread of living organisms, substances, energy, or other components in the maritime environment by human activities, exceeding established environmental quality limits. This results in environmental degradation and modifications, impairing the ecosystem's ability to function properly (**Russell, 1974; Khathut, 2018; Wong, 2012**). This damage may include harm to biological resources, human health risks, environmental process disturbance, material quality deterioration, and diminished facility functionality (**Masindi & Muedi, 2018**). Substances released into marine waterways are classified as organic or inorganic, including heavy metals

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(Masindi & Muedi, 2018). Heavy metals infiltrate marine ecosystems through two primary sources: natural phenomena and anthropogenic waste (Masindi *et al.*, 2021).

Lead (Pb) is a highly poisonous heavy metal (Widayatno et al., 2017; Kumar et al., 2020). Lead can inflict primary damage, resulting in immediate and discernible effects on the environment, or secondary damage manifested as subtle changes to the equilibrium of the biological food web, which may only become apparent over extended durations (Sharma, 2011; Naggar et al., 2014). Moreover, if lead contamination is not mitigated, it may result in health complications, fatalities, and food chain disruption (Masindi & Muedi, 2018). Frequent ingestion of lead can result in severe repercussions for people. The repercussions encompass neurological damage, delayed puberty, diminished cognitive function, hearing impairment in babies, and restricted fetal growth throughout gestation (Ades Pardi, 2017; Kumar et al., 2020). Lead exposure in humans can impair glucose metabolism, leading to hypoglycemia, characterized by diminished concentration, hyperactivity, impulsivity, and erratic behavior (Bánfalvi, 2011). Koller and Saleh (2018) assert that lead-induced neurological and behavioral alterations are especially detrimental to children, resulting in central nervous system impairment.

The Flores Sea is a significant marine region often navigated by vessels, constituting a segment of an international commerce route. Foreign vessels, including commercial ships, tankers, and warships, threaten multiple sectors: economic, environmental, social, legal, political, and security (Hutagalung, 2017). These vessels contribute to pollution via ballast water discharge and fuel emissions. Kurniawan *et al.* (2022) assert that ballast water systems introduce pollutants, including microorganisms (plankton, microalgae) and heavy metals. Moreover, fuel emissions from vessels contribute to the environmental introduction of lead (Khathut, 2018). Additionally, the Flores Sea contains underwater volcanoes created by a succession of volcanic activity initiated by faults or cracks in the Komba Waters (Rangin & Silver, 1990; Sarmili & Hutabarat, 2016).

Nienhuis (1986) discovered lead bioaccumulation in the seagrass ecosystems of the Flores Sea. Nienhuis *et al.* (1989) additionally recorded the bioaccumulation of heavy metals in seagrass and other macrofauna species, including dugongs, turtles, fish, sea cucumbers, and gastropods, in the Flores Sea. Lead contamination in the Flores Sea is attributable to fishing activities, shipping, and oil and gas exploration in the Banda and Arafura Seas, which also exacerbate lead pollution in the Flores Sea (Morrison & Delaney, 1996). The Banda and Arafura Seas influx flows into the Flores Sea due to the southeast monsoon winds (Wirasatriya *et al.*, 2021). Furthermore, the Flores Sea is crossed by the Indonesian Throughflow (ITF), which transports water masses from the Pacific Ocean via the Makassar Strait and Banda Sea. This results in fluctuations in salinity and sea surface temperature (Feng *et al.*, 2018; Wirasatriya *et al.*, 2021), establishing circumstances conducive to the buildup of heavy metal-contaminated water

masses in the Flores Sea. This research aimed to ascertain lead concentrations at different depths and their distribution across the Flores Sea.

## MATERIALS AND METHODS

## 1. Geographic location

Geographically, the Flores Sea is situated north of Flores Island and serves as a natural boundary between the provinces of West Nusa Tenggara and South Sulawesi. To the west, it borders the Bali Sea and the Java Sea. The western part of the Flores Sea is a convergence zone for major ocean currents originating from the Makassar Strait, part of the ITF, which transports water from the Pacific to the Indian Ocean. This sea covers an area of approximately 240,000km<sup>2</sup> and plays a crucial role in regional marine biodiversity. Additionally, eddies and upwelling occur in the Flores Sea and its surrounding areas (**Nuzula** *et al.*, **2016**). The depth of the Flores Sea ranges from 300 to 5,500m (Fig. 1), making it one of the deepest seas in the Indonesian archipelago (Fig. 1).



Fig. 1. Map of geographical area, bathymetry, and Flores Sea station samples

Plumbum (Pb) sampling stations (1-6) were located at water depths of 25, 75, 150, 200, and 750m, with seabed sampling conducted in locations a-c. Seabed samples were taken from three locations, including station 2 at a depth of 1145m, station 3 at a depth of 850m, and station 4 at a depth of 500m. The map originated from **Herawati** *et al.* (2024). Sediment samples were taken from three stations from the shallowest depth. Sediment was only taken from 3 stations due to high current and wave conditions at the research

station and the deep sea's position as at stations 5 and 6, and at station 1, the bottom of the waters was rocky.

## 2. Sampling method

Sampling was carried out in the Indonesian Flores Sea from April to May 2023 as a part of the Jala Citra 3 Expedition. Sea water samples were collected from six stations and 5 depths (25, 75, 150, 200, 750m). Sediment samples were collected from the seabed using an Ekman grab (diameter of 107cm; radius of 53,5cm) with approximately 1kg of sediment collected at each site and were stored in a ziplock plastic bag. Water samples, each about 1L, were collected using a Rosette-CTD (Conductivity, Temperature, Depth) at five depths across six stations including *in-situ* parameters of salinity and temperature. The water samples were preserved with H<sub>2</sub>SO<sub>4</sub> as a binding solution, transported to the laboratory, and cooled-stored for later analysis. The collected samples were processed at the Central Laboratory of Padjadjaran University using the Atomic Absorption Spectroscopy (AAS) method for Pb analysis (Abarca et al., 2001). Metals were extracted from 250mL of each seawater sample with acid digestion using HNO<sub>3</sub> 65% in glass containers with a lid and heated until the volume was reduced to 10-20mL. The Pb concentration in prepared samples was analyzed at 283,3nm using AAS Analyst 400 (Perkin Elmer, USA), which operates based on the Lambert-Beer Law the amount of light absorbed is directly proportional to the concentration of the substance. Since atoms absorb light, heavy metal ions or compounds must be converted into atomic form (Warni et al., 2017). Pb concentrations were calculated based on the regression values displayed by the AAS, which were derived from the calibration curve regression.

### 3. Lead concentration

The results of atomic absorption measurements were recorded, and lead (Pb) concentration was calculated using formula 1a and 1b (Lelifajri, 2009) as follows:

Pb concentration in water  $(mgL^{-1}) = C_{reg} x P x v$  .....(1a) Pb concentration in sediment  $(mgkg^{-1}) = C_{reg} x P x V/G$ .....(1b)

Where, Creg is the calculated concentration (mg/L); P is the dilution factor; v is the sample volume and G is the sample weight (kg).

## 4. Statistical analysis

The results of measurements of water quality parameters and Pb levels were tabulated and compiled in the Microsoft Excel program. Then, the data were arranged in the form of tables and figures. The obtained data were compared with the threshold value of environmental quality standards of the Government Regulation of the Republic of Indonesia No. 22 of 2021 Article VII, concerning the implementation of environmental

management and protection and Australian regulations namely **ANZECC and ARMCANZ (2000)**: Australian and New Zealand Guidelines for Fresh and Marine Water Quality.

Determination of Pb pollution index (IP) was calculated using equation (2) from Marganingrum *et al.* (2013):

$$IP_{j} = \sqrt{\frac{(C_{i}/L_{ij})_{M}^{2} + (C_{i}/L_{ij})_{R}^{2}}{2}}$$

Where, IP is the pollution index; Cij is the Pb calculated concentration  $(mgL^{-1})$ ; L is the Pb concentration limit  $(mgL^{-1})$  specified by the **Guideline for seawater quality of Republic Indonesia/PP No. 22 (2021)**; M is a maximum value; R= average value.

If: 0<IP<1 = good; 1<IP<5 = low; 5<IP<10 = medium, and IP>10 = high

Analysis of variance (ANOVA) was carried out to see the relationship between lead concentration based on depth and the location of the data collection station (**Jia** *et al.*, **2018**).

#### RESULTS

The findings underline several important trends that are important for comprehending the degree of heavy metal pollution in the area. Table (6) provides the average concentrations of lead at different depths together with the standard deviations from the findings of these tests. Evaluating whether the Flores Sea's waters fall within reasonable environmental boundaries depends on these values. The data were matched with accepted regulatory levels, including the international recommendations provided by **ANZECC and ARMCANZ (2000)** and the **Republic of Indonesia government Regulation number 22 (2021)**.

Depth (m)         Average (mgL <sup>-1</sup> )           25         0.2755	STDevP					
25 0.2755	DIDCH					
	0.1237					
50 0.4229	0.2091					
150 0.3705	0.2454					
250 0.3669	0.25822					
270 0.3156	0.11152					
Pb Threshold Value in water (mgL <sup>-1</sup> )						
Government Regulation of Republic Indonesia number 22 of 202	0.008					
ANZECC & ARMCANZ (2000)	0.005					

**Table 1.** The lead (Pb) concentrations (mgL<sup>-1</sup>) in Flores Sea were measured and analyzed to understand the distribution of heavy metal contamination across different depths

At every location, the lead concentrations were studied horizontally across the depth strata. All average lead concentrations surpass the safety limits set by both international guidelines and the 2021 rule of the Indonesian government. At 150 meters, the concentration showed the highest average of  $0.37 \pm 0.24$  mgL<sup>-1</sup>. Fascinatingly, at a lesser depth of 25 meters, the lowest lead content of 0.275mgL<sup>-1</sup> was recorded. Though this is less than in the deeper levels, it is still much above the Government Regulation of the Republic of Indonesia's (0.008mgL<sup>-1</sup>) allowed environmental requirements as well as ANZECC & ARMCANZ's.

To observe the vertical distribution of lead concentration in comparison with water quality parameters, Fig. (2) illustrates the variations in lead levels at different depths, providing insight into how these concentrations relate to key water quality indicators.



**Fig. 2.** Relationship of lead with water parameters: a) Temperature; b) Salinity; c) pH; d) Dissolved oxygen (DO). The dashed curve and square-shaped points represent lead concentration values.

At Station 1, a low pH value was seen at depths of 150 and 250 meters (Fig. 2c). At other stations, the water quality metrics remained below acceptable limits, hence not limiting lead concentrations. Station 6 is located adjacent to Batu Tara Volcano, an underwater volcanic formation. Salinity levels in the Flores Sea at depths of 0-200 meters below the surface vary from 34 to 34.5psu. At depths ranging from 200 to 800 meters, the salinity measured approximately 34.6psu. The temperature ranged from 25 to 30°C at

depths of 0-200 meters, and from 5 to  $15^{\circ}$ C at depths of 200-800 meters. The highest lead concentration was observed at station 1, progressively decreasing toward station 5. The lead content at station 1 varied between 0.8 and 0.85mgL<sup>-1</sup>.

# 1. Pollution index

The data analysis in Table (2) shows that the highest pollution index (IP) value was recorded at station 1, with a range of 7 to 8, categorizing it as mild pollution according to Ministerial Decree No. 113. Station 2 falls within the moderate range, with an IP value of 6 to 7. Station 3 follows a similar trend, with an IP value ranging from 6 to 8, indicating moderate pollution levels. At station 4, the IP values vary significantly, with the minimum recorded value of 4.87 at a depth of 250 meters, classifying it as light pollution. At other depths, the IP values range from 5 to 6, categorizing them as moderate pollution. Station 5 displays the lowest IP value of 4.91 at a depth of 150 meters, indicating light pollution, while at other depths, the IP values remain around 6, classifying them as moderate. Station 6 shows the lowest IP values overall, with readings of 4 at both 25 and 250 meters, categorizing these as light pollution. At depths of 50 and 750 meters, the IP values increase to 5.49 and 5.04, respectively, classifying them as moderate pollution.

Station –	Depth (meter)				
	25	50	150	250	750
1	7.19	8.02	8.48	8.25	7.05
2	6.77	7.48	7.35	7.97	6.95
3	6.26	7.07	6.93	8.19	6.74
4	5.98	6.20	6.48	4.87	NA
5	6.25	NA	4.91	6.32	6.79
6	4.38	5.49	NA	4.30	5.04

Table 2. Pollutant index in each station

The data from Table (2) suggest that the average pollution index in the Flores Sea is classified as moderate. Instances of light pollution are recorded at four distinct depths: 250 meters at Station 4, 150 meters at Station 5, and at both 50 and 250 meters at Station 6. Refer to Fig. (3) for a graphic representation of the distribution of pollution categories in the Flores Sea.



Fig. 3. Graph of IP Values Flores Sea, (a) Station; (b) Depth

Fig. (3) illustrates the categories of IP values based on (a) stations and (b) depths. In Fig. (3a), four stations 1, 2, 3, and 5 are identified as falling into the moderate pollution category, with the highest IP value of 7.80 recorded at station 1. Conversely, stations 4 and 6 were categorized as having light pollution, with the lowest IP value of 3.84 at station 6. Fig. (3b) indicates that all depths fall within the moderate pollution category, with only slight variations in IP values. The highest IP value of 6.65 was observed at a depth of 250 meters, while the lowest value, 5.43, was noted at a depth of 750 meters.

### DISCUSSION

The lead concentrations measured at all five depths exceed both the national and international regulatory thresholds, which is a concerning finding (Table 1). This elevated concentration suggests that deeper layers of the water column might be accumulating more lead, possibly due to the influence of external factors such as currents, shipping activity, and volcanic processes in the region. The relatively higher concentrations at mid-depths (75 and 150 meters) could be attributed to the mixing of water masses and the transport of contaminants via ocean currents such as the ITF, which carries water from the Pacific Ocean into the Indian Ocean, potentially bringing pollutants along its path.

The high standard deviations, especially at 150 meters ( $\pm 0.24 \text{mgL}^{-1}$ ) and 200 meters ( $\pm 0.26 \text{mgL}^{-1}$ ), suggest significant spatial variability in lead contamination, which could be due to localized pollution sources or the uneven distribution of sediments and particulates that bind heavy metals. The elevated concentrations of lead across all depths highlight the ongoing risk of heavy metal pollution in the Flores Sea, particularly in areas influenced by shipping routes, underwater volcanic activity, and possibly land-based sources of contamination. These findings emphasize the need for continuous monitoring and further investigation into the sources and long-term impacts of lead pollution on marine ecosystems in this region.

Previous research indicated that lead contamination can result in a reduction of pH, hence contributing to ocean acidification. The mean pH of the world's seas is roughly 8.1,

and a one-unit reduction in pH results in a tenfold escalation in acidity. Ocean acidification, exacerbated by lead poisoning, presents potential threats to marine organisms and ecosystem interactions. Lead sources may originate from volcanic activity. Notwithstanding this, the aquatic conditions at Station 6 remain comparatively steady. Alkaline pH conditions can reduce lead solubility in water, as elevated pH can convert heavy metals from carbonate to hydroxide forms, resulting in their association with particles in the aquatic environment and subsequent sedimentation. Consequently, lead will swiftly precipitate and amass in sediments. Ongoing surveillance of lead levels in sediments, especially at Station 6, is necessary. The data collection conducted in April, coincides with the eastern monsoon. **Nuzula** *et al.* (2016) reported the presence of eddy currents in the Flores Sea, exhibiting a velocity of 11.4cm/ s this month. Nonetheless, these eddy currents do not substantially influence the distribution of lead in the Flores Sea, as surface currents penetrate to a depth of just around 100 meters.

The Makassar current, which flows into the Sulawesi Sea and ultimately converges with the Indian Ocean, is another factor affecting the concentration and distribution of lead in the Flores Sea (**de Silva** *et al.*, 2005). These current transports water masses from the North Pacific and enters the Flores Sea through the Indonesian Through Flow (ITF) (**Godfrey**, 1996). Stewart (2008) describes the migration of seawater from northern latitudes to southern latitudes, traversing the equator, as thermohaline circulation. This circulation is propelled by variations in temperature and salinity across different water bodies.

At depths of 200-800 meters, the salinity and temperature of the Flores Sea water mass are comparable to those of the Eastern South Pacific Intermediate Water (ESPIW), exhibiting salinity values of 34.0-34.4psu and temperatures ranging from 10 to 12°C (**Emery, 2001**). The water mass in the Flores Sea is significantly affected by the Pacific Ocean, since the Mindanao Current transports water from the Pacific into the Flores Sea through the Indonesian Throughflow at the Makassar Strait (**Godfrey, 1996**). The worldwide water masses that flow through a specific body of water can influence its chemical features, including dissolved oxygen levels and nutritional composition, while also establishing distinct temperature and salinity profiles (**Triyulianti** *et al.*, **2018**). The dynamics of contemporary currents and the distribution of lead, as the Flores Sea acts as a principal channel for water flow from the North Pacific Ocean through the Makassar Strait to the south (**Rochyatun, 2013**).

Lead concentrations have been thoroughly recorded in the Pacific Ocean. Schaule and Patterson (1981) reported that lead concentrations in Pacific waters are approximately 1 x  $10^{=6}$  mgL<sup>-1</sup>, occurring at depths beyond 3500 meters. This concentration is anticipated to disseminate from the Northwest Pacific to the North Pacific, intensifying over time as a result of industrial activity and fuel emissions (**Zurbrick** *et al.*, 2017). Lead contamination has been detected in the interior of the Pacific Ocean, with evaluations suggesting that industrial emissions from Asia are a primary source of lead in the northwestern and central Pacific areas (Schaule & Patterson, 1981).

Sediments in the Flores Sea contain lead, as lead tends to settle and accumulate within sediments (**Panca** *et al.*, **2019**). This occurs because water conditions can reduce the solubility of lead, leading to its rapid deposition (**Palar**, **2004**; **Rezki** *et al.*, **2013**). In this study, lead concentrations in sediments were measured at three stations: stations 2, 3, and 4, due to challenges encountered with the equipment used for data collection. At station 2, the lead concentration was 0.9862mg/ kg, at station 3 it was 5.6352mg/ kg, and at station 4 it was 0.1497mg/ kg. These concentrations are well below the threshold set by **ANZECC and ARMCANZ (2000**), which ranges from 50-220mg/ kg. However, monitoring of lead concentrations at station 6 is recommended, as it is located near a series of underwater volcanic mountains.

Additionally, large vessels traversing the Flores Sea emit fuel pollutants. Hansen (2012) suggests that ship emissions can contribute to heavy metal contamination due to unburned fuel oil. Station 1 is also frequently navigated by fishing boats and domestic cargo ships, as the Flores Sea serves as both a fishing zone and a domestic shipping corridor (Cheng & Hu, 2010; Panca *et al.*, 2019). The lead concentration at station 1 does not appear to affect the levels at other stations, likely due to the surface current dynamics in the Flores Sea, which flow westward past the Selayar Islands and then southward into the Sawu Sea.

# CONCLUSION

Lead (Pb) levels in the Flores Sea exceed the established thresholds for environmental protection. The lowest concentration recorded was 0.083mg/ L, while the highest measured value was 0.81mg/ L. Given that the Flores Sea is located along a major shipping route, international maritime activity is the primary source of lead pollution in this area. Additionally, the adjacent Banda Sea and undersea volcanic activity also contribute to the elevated lead levels. Station 3 recorded the highest lead concentration in sediment samples at 5.63mg/ kg, though this value remains below levels of environmental concern. Overall, the Flores Sea is considered to have moderate lead pollution.

The distribution of lead at different depths is influenced by water masses from the Pacific Ocean, which are driven by the Indo-Pacific Throughflow (ITF) current. However, since this study was conducted during the western season, the effect of monsoon winds on the horizontal dispersion of lead was minimal. Key factors, such as water quality and pH levels, are also crucial in preventing the spread of lead in the marine environment.

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