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Mapping Atmospheric Mercury in Lampung Province, Indonesia Using Bark of Multipurpose Tree Species

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Abstract: The use of mercury in gold refining causes air pollution and results in the contamination of multipurpose tree species (MPTS). Tree bark has properties that cause it to store mercury for quite a long time. The purpose of this study was to determine mercury contamination of MPTS and map the mercury contamination distribution in the atmosphere using tree barks as bioindicators. Sampling was performed using purposive sampling. The mercury concentration was obtained by atomic absorption spectroscopy, and the highest THg contents were analyzed using a scanning electron microscope. The analysis was carried out by gauging total mercury (THg), distance, elevation to THg, and interpolation of THg at the research site. The results showed that there were 10 types of MPTS trees whose bark could accumulate mercury. The bark of the *Tamarindus indica* tree stored the greatest amount of THg (74.4 µg dry weight (DW)), followed by *Persea americana* (58.7 µg DW), and *Annona muricata* (44.2 µg DW), respectively. This result was influenced by the roughness of the bark and the location of the plants. No correlation was found between distance and elevation to THg on tree bark. The mercury interpolation in the atmosphere showed that mercury moves from the purification point to the southeast of the purification location.

Keywords: environmental pollution; interpolation; mercury; MPTS plants; tree bark

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1. Introduction

Refining gold using mercury certainly pollutes the environment. Mercury is used because it can separate gold from other materials [1]. However, the use of mercury pollutes the environment and has serious impacts on health. Thus, artisanal small scale gold mining causes environmental problems, as a lot of mercury waste is disposed into water resources [2].

Multipurpose tree species (MPTS) are usually not far from inhabitants, who tend to use them for wood or nontimber products [3]. Moreover, the fruits of MPTS are consumed and they help in increasing community income [4]. Thus, it is dangerous when mercury is accumulated in trees and eventually consumed by humans. Mercury can lead to body poisoning through several mechanisms. Additionally, mercury toxicity can trigger neurological damages, such as cognitive impairments and autoimmune dysfunctions [5].

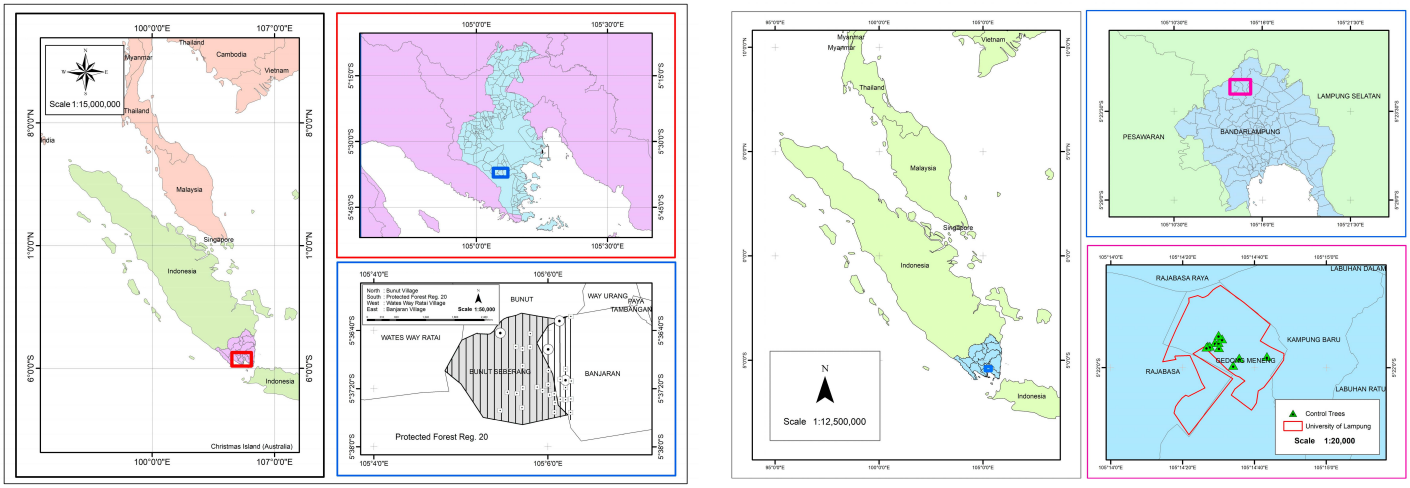
Mercury used for gold refining is volatile; it evaporates at 20 °C [6]. The gaseous mercury will travel with the direction of the wind. Mercury vapor enters human bodies through the respiratory tract, and this is very risky, especially in residential areas. Thus, gold miners, refiners, etc., are at a risk of experiencing detrimental health effects [7]. In

The atmosphere, mercury can move as far as 2500 km in 72 h [8], and it can also stick to tree bark. Based on [9], it was shown that mercury can be deposited in forests, especially in trees. This study aimed to determine the mercury contamination in the atmosphere, and the barks of MPTS trees were used as indicators to estimate the pollution and the distribution levels of mercury through air. This study used tree bark as an indicator because bark can accumulate mercury from the atmosphere continuously [10].

2. Materials and Methods

2.1. Study Location

Bunut Seberang Village, Way Ratai District, Pesawaran Regency, Lampung Province (Figure 1a) was the sampling location, and University of Lampung (Figure 1b), located in Bandar Lampung city, Indonesia, was the control sampling location. The research was conducted in October 2020 for the sample in the Bunut Seberang Village, and it was conducted in December 2020 for the control sample. The distance between the contaminated area and control area was about 32 km (19.9 miles).



(**a**) (**b**)

Figure 1. The research location (**a**) Bunut Seberang Village and (**b**) University of Lampung.

2.2. Sampling Method

There were 10 types of MPTS plants from 28 samples taken at the research site: *Persea americana*, *Tamarindus indica*, *Lansium domesticum*, *Durio zibethinus*, *Spondias dulcis*, *Gnetum gnemon*, *Artocarpus heterophyllus*, *Parkia speciosa*, *Leucaena leucocephala*, *and Annona muricata*. In the control sample, there were 3 types of plants from 11 samples: *T indica*, *Syzygium aqueum*, and *Mangifera indica*.

The research samples were taken by purposive sampling at the study site (Bunut Seberang Village), which was divided into 20 sampling lines with a distance between the lines of 150 m from west to east of the research location (Figure 1a). The research location was a village with a residential area, so not every line had MPTS plants.

Bark samples of 10 cm × 10 cm were taken from trees with a minimum diameter of 20 cm at a height of 1.3 m (Figure 2) [9]. Most of the evaporated mercury was found on tree barks at an altitude of 1.3 m above the ground [11]. The bark extraction led to the nearest gold refining site from the study site.

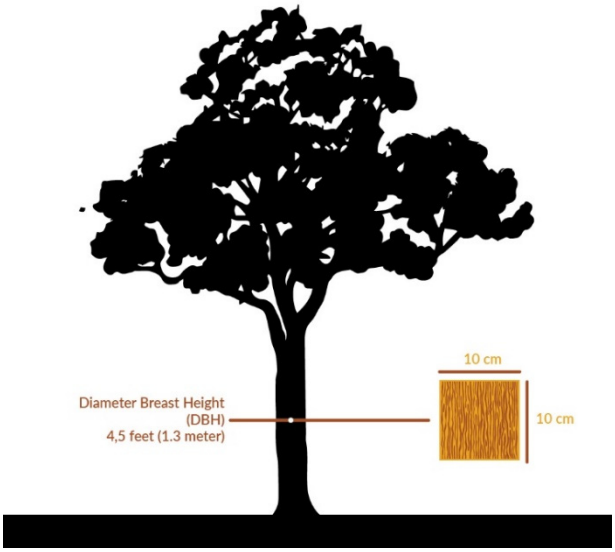


Figure 2. Sampling technique.

The sampling method at the University of Lampung was performed using the purposive sampling method on the same tree species as the trees used for sampling in the Bunut Seberang Village. The locations of the plants used for control sampling are shown in Figure 1b.

2.3. Sample Preparation

2.3.1. Atomic Absorption Spectroscopy Analysis

The sample was dried using an oven at a temperature of 80 °C for 48 h. The sample was weighed and then put into an oven. Afterward, the sample diameter was imaged using a camera to measure the surface area using ImageJ. The samples whose surface areas had been calculated were uniformed with an average area width of 13–18 cm^2. This was to uniform the surface area of the sample and avoid errors. Furthermore, the tree bark samples were mashed using a Philips Plastic HR2115/00 blender with a rotational speed of 13,000 rpm. Then, the first sample was put into a bottle for analysis using AAS.

The ground samples were then analyzed in the laboratory using (AAS) type Agilent 240FS-VGA 7, Agilent Technologies Inc., Australia. AAS was used to determine the mercury content in the samples, and the method used was standard heavy metal contamination mercury (Hg) in food (Indonesian National Standard 01-2896-1998) with a tested sample weight of 5 g.

2.3.2. Scanning Electron Microscopy Analysis

Scanning electron microscopy is a microanalysis that analyzes metal compositions [12]. This method uses SEM type ZEISS/EVO MA 10, The Carl Zeiss Foundation, Germany. The highest sample that accumulates mercury will be scanned by scanning electron method. SEM is used to see bark structure and zoom in on the surface morphology. SEM can show a substrate of heavy metal [13].

2.4. THg Calculation

Total mercury is an accumulation of metallic mercury, which evaporates and can survive in air for 0.4 to 3 years [6]. The accumulation of mercury can be measured by its total weight (THg). Determining the THg values is very useful because various mercury gases are more abundant in tree bark [14]. The following formulation was obtained from [15].

$$THg = (DW \times CHg) \times \frac{(FD)}{(RS)} \quad (1)$$

THg is obtained from the dry weight (DW (gram)) of a sample, multiplied by the concentration of the contained mercury (Concentration of Hg (mg/g DW))) multiplied by the sample area (Fragment Dimension (*FD*)) (100 cm^2). CHg is the concentration of the contained Hg, and real square (cm^2) is the surface area of the sample. THg was used in estimating the mercury content because there are no methods or tools that can predict the form of the contained mercury in samples.

2.5. Statistical Analysis

The performed data analysis tests are a normality test, and a homogeneity test, which used R Studio Version 1.4.1106. The normality test uses the Shapiro–Wilk method, which is an effective and valid method that is used for small samples with data < 50 data [16]. After the normality test was performed, the homogeneity test was performed using the Levene Test method, which is a test method for variance from several populations. This test is an alternative to the Bartlett test [17]. If the data is normally distributed, it is better to use the Bartlett test. If the data is not normally distributed, the Levene Test is used [18]. Bartlett's test shows the Chi-Square Count and Chi-Square Table. If Chi-Square Count < Chi-Square Table, then H0 is accepted. [19].

After knowing the analysis results of the two data, the correlation test can be performed. Correlation analysis is used to determine the relationship between two variables and the direction of this relationship.

3. Results

MPTS Plants and Mercury Content in the Measured Tree Barks

The mercury contents in the analyzed tree barks of the two test locations are presented in Table 1.

Table 1. Mercury and THg concentrations in the tree barks of the MPTS species.

No	Scientific Name	Surface Dimension (cm^2)	Hg Concentration (mg/g-DW *)	Distance (m)	Elevation (m)	THg (µg DW *)
1	*P. americana* 1	15.2	0.26	510	268	15.2
2	*P. americana* 2	14.5	0.57	413	267	58.7
	Mean	14.8	0.41	461	267	36.9
3	*T. indica* 1	15.5	0.79	418	270	40.6
4	*T. indica* 2	17.2	0.85	240	122	74.4
	Mean	16.3	0.82	329	196	57.5
5	*L. domesticum* 1	18.8	0.36	416	270	7.7
6	*L. domesticum* 2	15	0.35	120	268	18.5
	Mean	16.9	0.36	268	269	13.1
7	*D. zibethinus* 1	20.5	0.15	1500	137	6.76
8	*D. zibethinus* 2	19.3	0.06	820	280	2.17
9	*D. zibethinus* 3	16.8	0.02	425	272	0.81
10	*D. zibethinus* 4	18.1	0.04	1073	276	2.44
11	*D. zibethinus* 5	18.9	0.08	570	271	4.38
12	*D. zibethinus* 6	14.3	0.04	730	275	2.58
13	*D. zibethinus* 7	13.1	0.07	950	274	4.34
14	*D. zibethinus* 8	16.5	0.07	105	268	5.25
15	*D. zibethinus* 9	13.1	0.3	610	261	20.9
16	*D. zibethinus* 10	14.4	0.55	700	276	26.7

	Mean	16.5	0.14	748	259	7.63
17	*S. dulcis* 1	18.4	0.33	389	270	7.12
	Mean	18.4	0.33	389	270	7.12
18	*G. gnemon* 1	14.6	1.17	370	274	40
19	*G. gnemon* 2	18.4	0.38	163	269	8.33
	Mean	16.5	0.78	266	271	24.2
20	*A. heterophyllus* 1	16.8	0.19	465	273	8.02
21	*A. heterophyllus* 2	18.6	0.19	410	269	8.25
22	*A. heterophyllus* 3	14.3	0.26	684	258	7.12
	Mean	16.6	0.21	519	266	7.80
23	*P. speciosa* 1	16.1	0.34	520	275	19
24	*P. speciosa* 2	14.6	0.46	244	284	25.1
	Mean	15.3	0.40	382	279	22.0
25	*L. leucochepala* 1	17.6	0.03	1250	277	1.06
26	*L. leucochepala* 2	16.4	0.3	435	271	10.9
	Mean	17.0	0.17	842	274	5.98
27	*A. muricata* 1	17.6	0.53	385	271	30
28	*A. muricata* 2	12.6	0.62	115	120	44.2
	Mean	15.10	0.58	250	195	37.1

* DW = Dry Weight.

The results of the correlation between the tree distance and THg showed a negative trend. This shows that there is a relationship between the two variables. The farther the purification distance from the analyzed tree affects the total amount of the contained mercury in the bark, whether the tree is large or small. The correlation value between the distance variable and THg was −0.41, which indicates that the interpretation of the correlation between the distance variable and THg is moderate.

Elevation of the tree showed different results to THg, which is a negative relationship. Altitude has no effects on the THg amount in the tree barks, while the given rho value was −0.48, which indicates that the interpretations of the height and THg variables are moderate.

A total of 28 samples from the barks of MPTS trees cultivated by the community in Bunut Seberang Village were analyzed for mercury content. The peel of *T. indica* can accumulate 74.4 μg DW of mercury from air at a distance of 122 m from the gold refining center. Furthermore, the peel of *P. americana* accumulated 58.7 μg DW of mercury at a distance of 267 m, and *A. muricata* accumulated 44.2 μg DW at a distance of 120 m. The research control data is presented in Table 2.

Table 2. Mercury amounts in the control samples.

No.	Scientific Name	Surface Dimension (cm^2)	Hg Concentration (mg/g) DW *	THg (μg DW)
1	*T. indica*	17.7	0.0002	0.03
2	*T. indica*	16.9	0.0002	0.02
3	*T. indica*	17.4	0.0002	0.02
4	*S. aqueum*	17.3	0.0002	0.03
5	*S. aqueum*	20.0	0.0002	0.02
6	*S. aqueum*	19.9	0.0002	0.03
7	*S. aqueum*	14.8	0.0002	0.04
8	*M. indica*	15.2	0.007	1.42
9	*M. indica*	14.9	0.023	2.32
10	*M. indica*	16.0	0.011	1.51

11	*M. indica*	15.5	0.0002	0.04

* DW = Dry Weight.

Based on the performed experiments, it has been observed that, at the control sampling location, the distribution of mercury in the atmosphere is below the threshold. The threshold of the mercury content in fruits and vegetables is 0.03 ppm [20]. *T. indica* is an MPTS plant whose fruit is directly consumed or mixed with other food products.

4. Discussion

4.1. Bark and Mercury Contamination

Generally, communities need MPTS plants that provide food and resources, such as fruits, timber, etc. [21]. *P. americana* and *T. indica* have textured and rough skin [22]. The bark of the two trees can accumulate more THg compared to other MPTS plants. *P. americana* can accumulate THg with a range from 15.2 to 58.7 µg DW. *T. indica*, has an accumulation rate of 40.6–74.4 µg DW. The skin surface has an important role with regard to the amount of accumulated THg by the skin [15]. A surface magnification of the *T. indica* tree bark is presented in Figure 3.

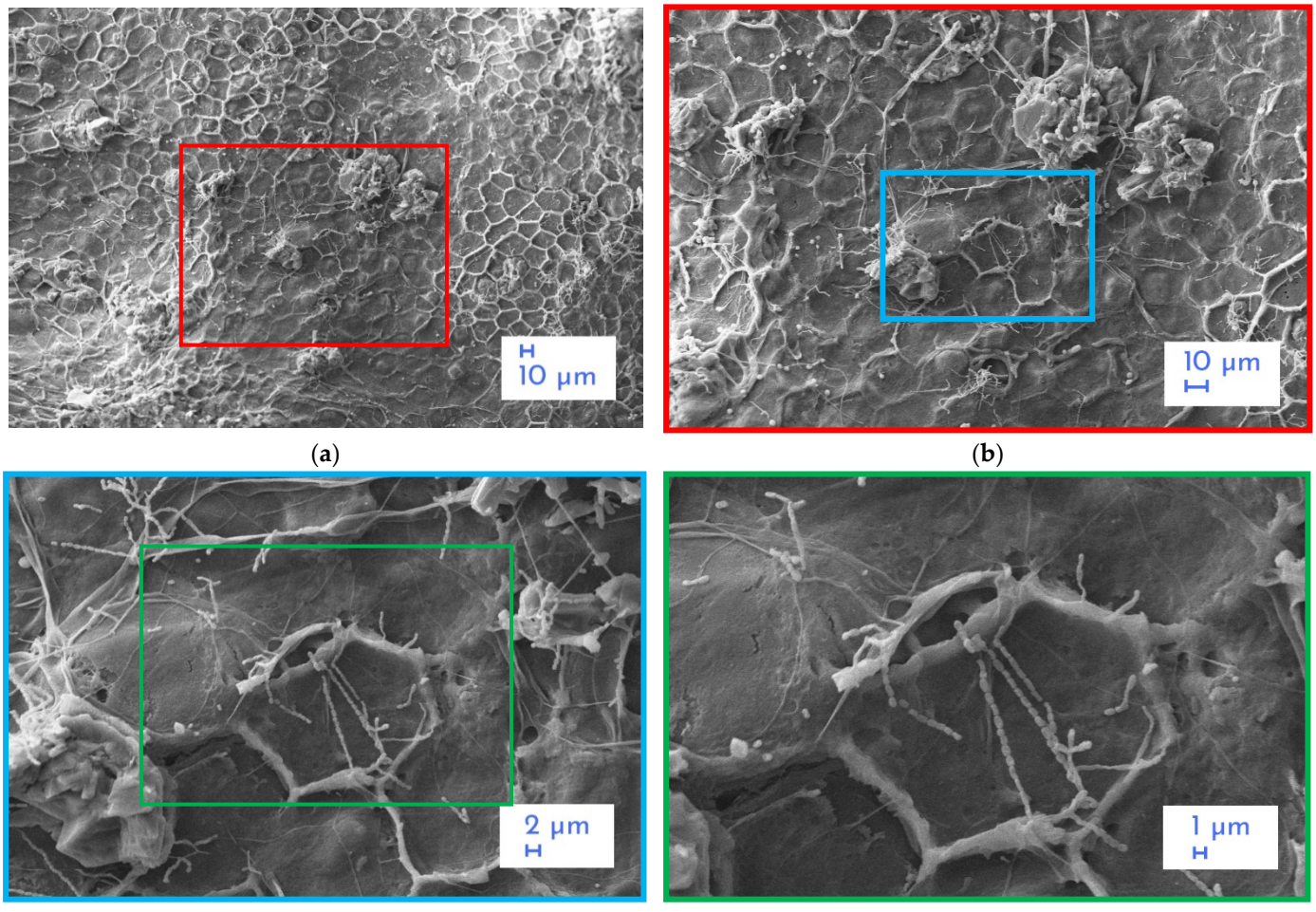


Figure 3. Bark surface of *T. indica* with the magnifications of (**a**) 500× (**b**) 1000× (**c**) 3000× and (**d**) 5000×.

The bark of the *T. indica* tree accumulates the most mercury. As seen from the SEM results, the surface of this tree bark has a faceted shape, so it can be said to be rough. Since tree barks are always exposed to mercury contamination in air, they can be appropriate

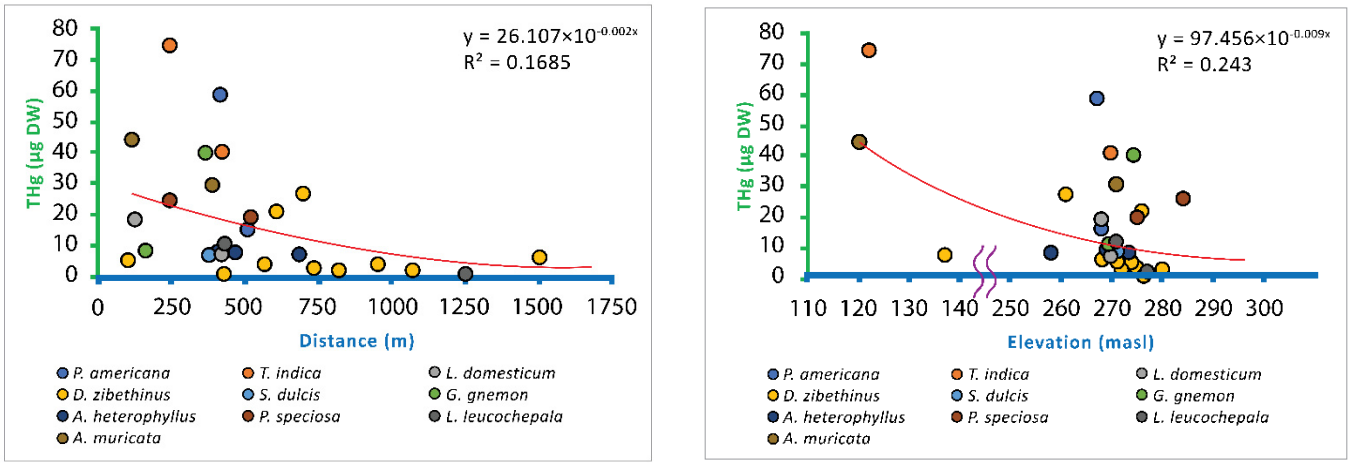
bioindicators [23]. The rough surfaces and high porosity of tree barks make it difficult for pollutants to be washed off them by rain. In this case, mercury, which is a heavy metal, can be mechanically captured by tree barks as particulate and accumulated as gas. Then, it becomes a stable chemical and lasts on tree barks for a long time [24].

It has been proven that the rough surfaces of MPTS plants can accumulate more THg compared with their smooth surfaces. Since the rough surfaces of MPTS plants have more bark texture, they can accumulate aerosol particles efficiently [24]. Additionally, it has been proven that *A. muricata* is among the MPTS plants that can accumulate THg. The range of the accumulated THg by *A. muricata* does not differ much from the ranges of *P. americana* and *T. indica*. *A. muricata* can accumulate30.0–44.2 µg. The surface of the *A. muricata* tree bark is rough and gray [25]. Additionally, the bark of this tree is used in traditional medicine to produce methanol, which is used in treating diseases such as cysts, cholesterol, and high blood pressure [26]. This plant can also inhibit the growth of cancer cells [27].

Thus, cultivating MPTS plants for health purposes near gold refining sites can be very dangerous. Mercury is classified as a hazardous and toxic material (B3). The safe threshold for mercury content in fruits and vegetables is 0.03 ppm, while its threshold in natural mineral water and bottled drinking water is 0.001 ppm [20]. Mercury exposure in humans should not exceed the threshold of 1–2 ppm [28]. The United States Environmental Protection Agency (US EPA) stipulates that the mercury exposure threshold (mercury chloride) for humans is 0.3 g/body weight/day. Furthermore, the thresholds for methyl mercury contamination and elemental mercury are 0.1 ppm/body weight/day and 0.3 g/m3, respectively [29].

4.2. Correlation of THg to Distance and Elevation

The correlation analysis of the distance and THg showed that correlation (r) has is a negative value. A medium r-value of −0.41 indicates a moderate interpretation between the two variables. This shows that the distance between MPTS plants and gold refining sites has a moderate effect on the THg content of the tree barks of MPTS plants. This can be seen in *P. americana* 1 and *P. americana* 2, which are located at a distance of 510 m and 413 m. These two *P. americana* have THg contents that differ quite a lot, which are between 15.2 and 58.7 µg DW. At a distance of 413 m, *P. americana* contains larger THg when compared with a distance of 510 m. This shows that the farther MPTS Plants are from the gold refining sites, the lower the THg (negative correlation). Additionally, *A. muricata* has a significant difference compared with *P. americana*. The distance between these two plants is not much different from the purification location. *A. muricata* 1 has a distance with gold purification of around 385 m, and *A. muricata* 2 has a distance of about 115 m, and, since the THg of each *A. muricata* is not much different, 30 µg DW and 44 µg DW, respectively, the distance has a negative relation with THg. The THg of *D. zibethinus* indicates that there are no significant differences with distance. Based on the R^2 value, the relationship between elevation and THg is stronger than distance and THg. The distribution patterns of THg and the MPTS plant spacing (distance) are presented in Figure 4a.

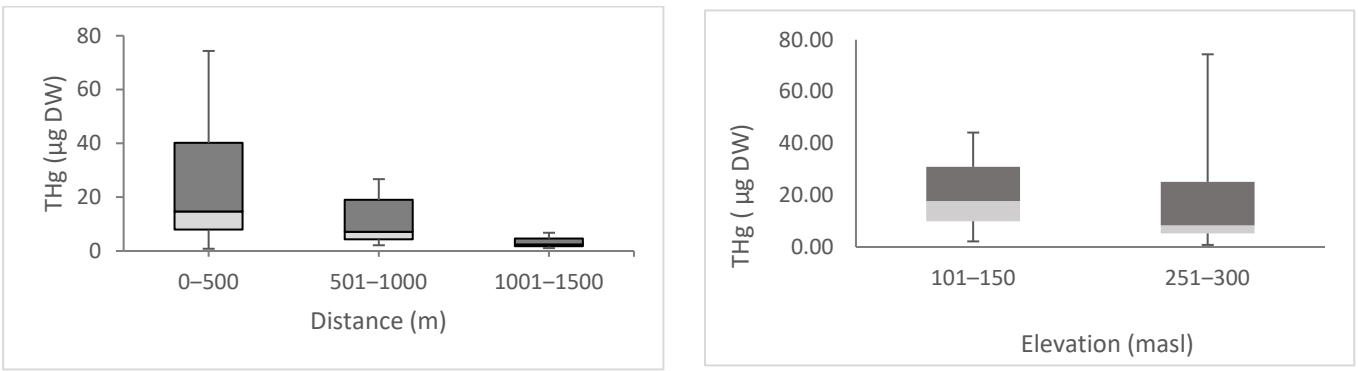


(**a**) (**b**)

Figure 4. Distribution of the MPTS plant correlations: (**a**) Distribution of the distribution patterns on distance and THg variables; (**b**) Distribution of the distribution pattern on the variables of height and THg.

The *T. indica* plants showed the same behaviors. The THg of *T. indica* 1 measured at a distance of 240 m was greater than that observed at a distance of 418 m (Figure 4a). This supports the correlation value of the two variables, which are negative and have a moderate interpretation. A decrease was seen at a distance of 1000 m, but some plants had a fairly high THg above 500 m. The difference in THg in the barks of MPTS plants is mostly caused by factors such as skin roughness, which affects the absorption of mercury, as it evaporates and is carried by wind. Additionally, it can evaporate with the help of the water inside the trees [30]. Furthermore, the environmental conditions around MPTS plants affect the stored amount of THg in their barks [31]. During day or night, mercury in the gaseous form does not affect the sorption levels on the leaves and barks, but it is influenced by the air movement [14]. The high and low mercury concentrations are also affected by strong wind, which forces mercury to circulate at great distances from its original location. Mercury can be carried by wind and move all the way from the Arctic Ocean to the Antarctic Continent [32].

In contrast to the height and THg variables, the results of the correlation analysis showed a negative value. At elevations between 101 and 150 m, the THg content in the tree barks was only ~20 g DW, and this also occurred in the 250–300 m high range. This shows that altitude has no effect on the THg in tree bark (Figure 5b). Rho (r) showed a value of −0.48. These two variables show a moderate interpretation. The height of mercury in the atmosphere can reach 7 km from the surface [33], which shows that mercury can be carried at different heights. The distribution pattern of the altitude and THg variables can be seen in Figure 4b.



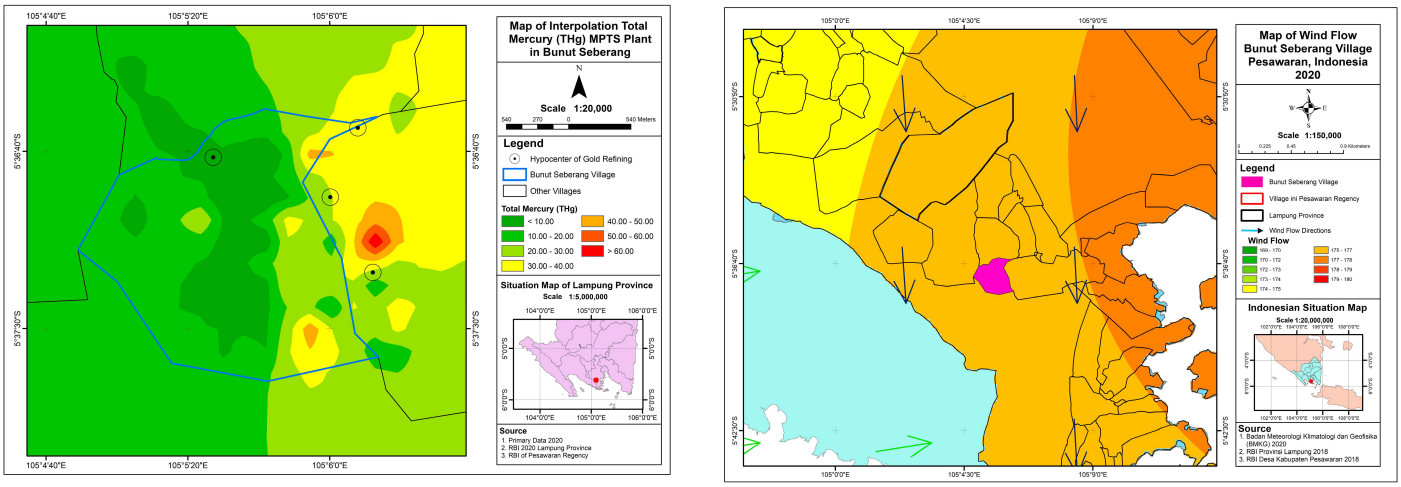
(**a**) (**b**)

Figure 5. Boxplots distribution of THg in tree barks with variable distances (**a**) and elevations (**b**).

4.3. Spatial Distribution of the Atmospheric THg and Wind Direction

The distribution pattern shows that there are several types of plants that can accumulate mercury even though they are below the height of 150 m. In contrast to other species, the plants at an altitude of >250 m accumulate mercury at a range of 0–60 µg. Based on the correlation analysis between the distance and THg, and the correlation between height and THg, some factors that affect the distribution of mercury in tree barks are distance, wind direction, conditions around MPTS plants, and tree bark roughness.

The MPTS barks containing mercury can be used as bioindicators of mercury pollution in the atmosphere. The content of THg in tree bark is higher than that on leaf surfaces [14]. The zones that contain more mercury can be identified by obtaining the distribution patterns of mercury from the barks of the analyzed trees. The distribution pattern of mercury contamination in air is presented in Figure 6a.



(**a**) (**b**)

Figure 6. Map situation of Bunut Seberang Village: (**a**) interpolation of THg in atmospheric with tree barks as bioindicators; (**b**) map of wind and flw at Bunut Seberang village.

The red zone has a larger THg distribution pattern, and the green zone has a smaller THg distribution pattern. The shape of the distribution pattern can be influenced by several factors, such as wind direction and speed. Local winds affect the mercury movement in the atmosphere, and it is a factor that helps tree barks accumulated mercury [34]. The wind direction and speed observations at the research location are presented in Figure 6b. The wind direction and speed can be seen at the research location at 175–177 rad in 2020, and the wind direction moves to the south and leans slightly toward east (southeast). Based on the interpolation of THg and the distribution pattern (Figure 6a), the distribution direction is influenced by the wind movement (Figure 6b). The movement of mercury in the atmosphere can be discovered by observing the movement of wind and using tree bark as a mercury pollution bioindicator. Tree barks are excellent bioindicators because of their porous structures, which efficiently accumulate aerosol particles [24].

5. Conclusions

Based on this research, it was found that the barks of the MPTS plants in Bunut Seberang Village can accumulate mercury with concentrations of up to 1 ppm. The MPTS plants that accumulated the most mercury is *P. americana*, *T. indica*, *G. gnemon*, and *A. muricata*. The mercury absorption rates of the tree barks in the village ranged from 0.81 to 74.4 µg. Additionally, the distribution of THg in the atmosphere, based on the obtained tree bark samples, was influenced by wind movements. Moreover, it was found that the mercury in the atmosphere around and in the village moves to the south and southeast directions.

This is influenced by several factors, including the surrounding environment of the trees, the wind, and the roughness of the respective tree barks.

Author Contributions: All authors contributed to the work presented in this manuscript. T.R. as the principal researcher, undertaken in association with his Master's Program at Lampung University. M.R., S.B.Y. and H.P. acted as supervisors. E.L.W. and S.B. acted as reviewers and advisers. A.T., assisting the principal researcher in obtaining data. All authors have read and agreed to the published version of the manuscript.

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