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Health Risk Assessment of Heavy Metals on Total Suspended Particles in Semi Urban, Urban, and Industrial Areas of Bandung Metropolitan Area, Indonesia

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ABSTRACT

Total Suspended Particulates (TSP) in ambient air contain metallic elements and can be inhaled by humans. Inhaled metal elements risk public health with long-term exposure because metallic elements are carcinogenic in the human body. This study aimed to analyze metallic elements and assess their risk to public health in the Bandung area and its surroundings. The metal elements analyzed in this research are Pb, Al, and Mn. Ambient air sampling was carried out in five industrial areas: Bandung Wetan, Buahbatu, Cibeureum, Padalarang, and Cimahi. High Volume Air Sampler (HVAS) is for sampling TSP in ambient air, and X-Ray Fluorescence (XRF) was used to analyze metallic elements. The results of the study showed that the Cimahi location gave the highest TSP level (126.7 g/m^3) and the lowest is Padalarang (15.1 μg/m^3). The metal elements concentrations, Pb, Al, and Mn were varied widely of 7.1–29.21 ng/m^3, 1054.9–1700.1 ng/m^3, and 8.91–14.79 ng/m^3, respectively. Risk analysis was determined by calculating ADD_{inhale}, Hazard Quotient (HQ), and Hazard Index (HI) to determine whether there is a potential non–carcinogenic effect on public health. Each industrial area gives an HI value > 1.0 and Buahbatu has the highest HI. This study proves that Pb significantly contributes to the increased risk of community exposure to non-carcinogenic effects.

Keywords: ambient air, industrial estate, metallic elements, risk analysis, TSP.

INTRODUCTION

The use of coal and fossil fuels in industrial activities impacts increasing levels of TSP in ambient air, especially in urban areas, which affects public health (Arditsoglou and Samara, 2005; Kang et al., 2009). The presence of TSP that exceeds quality standards in ambient air can pose a health risk because TSP contains several hazardous components such as fine particulates ($PM_{2.5}$

and PM_{10}) (Rita et al., 2016), chemical components (Gonçalves et al., 2016), black carbon (Santoso et al., 2020), and metallic elements (Basha et al., 2010). According to (Shah et al., 2012), motor vehicle emissions, industrial activities, combustion processes, and dust in the atmosphere cause metal elements in TSP. A study conducted by (Shah et al., 2012) indicated that TSP in urban areas of Islamabad, Pakistan, had shown the presence of metallic elements Cd, Co, Cr, Fe, Mn, Pb, Sb, and Zn. After 24 hours of sampling, the results were that Fe and Zn dominate over other metals. However, the dominant metals produced for anthropogenic activities are Sb, Cd, and Zn. Meanwhile, in Brazil, Mn, Fe, Zn, Cu, Ni, and Al metals exceed the US-EPA air quality standards (Quiterio et al., 2004).

In China, a study (Hu et al., 2012) identified Cu, Cd, Zn, Mn, Pb, Ni, Co, and Cr in TSP and $PM_{2.5}$. Therefore, the higher the TSP, the potential for humans to be exposed to metal elements. Through inhalation, both children and adults get higher and can cause respiratory problems (Rahman et al., 2021). Among metal elements, Pb is often highlighted because it is associated with motor vehicle emissions, which are increasing every year. Pb in ambient air is related to leaded gasoline, about 70% of which will be burned and emitted into the air. As a result, it can affect Pb levels in the human body to exceed the standard of > 5 µg/dL (Girsang, 2008). A study by (Hill et al., 2021) found that the concentrations of metallic elements in ambient air derived from Risk Screening Environmental Indicators (RSEI) were a significant predictor of blood metal levels associated with blood metal findings. RSEI concentrations also predicted dysfunction and cardiovascular risk, including changes in blood metal concentrations. Left ventricular mass, blood pressure, and heart rate, so children are positively correlated (more susceptible) to exposure to Pb, Hg, and Cd from air pollution. In addition to Pb, Al metal is also in the spotlight of its presence in ambient air, especially those close to industrial areas, with risk characterization for workers exposed to Al metal having a 31 times higher risk of disease than people not exposed to aluminum (Buranatrevedh, 2010). Based on a study (Du et al., 2019) in mining areas, besides being potentially inhaled, Pb and Mn can contaminate agricultural products, soil and surface water due to metal elements deposition from the air to land, either through dry deposition or wet deposition.

A recap of metal content from studies of metal elements in TSP from various countries is available in Table 1. In Medan and Jakarta, Indonesia, Pb levels are higher than in other countries and have exceeded the quality standard. As for the metallic elements Al and Mn, the quality of the metal elements in the ambient air are also high compared to other countries. However, it has yet to exceed the quality standard. Analysis of human health risks associated with exposure to metals bound to TSP can be calculated using the HQ and HI to determine the level of danger and risk of the presence of a metal element in the ambient air TSP (Chalvatzaki et al., 2019). Because analyses of public health risks from ambient air TSP metal elements are still very limited in Indonesia, this study examines and determines the Al, Mn, and Pb levels in industrial areas in Bandung and surrounding areas of public health or workers in the area.

Table 1. Comparison of the presence of metallic elements in TSP between countries

Country	Metal element (ng/m^3)			TSP (µg/m^3)	Reference
	Al	Mn	Pb		
Kosovo	206–2944	34–191	25–247	80–672	Arditsoglou and Samara, 2005
Amazonia	18–34	1.2–6.4	0.21–0.51	11–43	Gonçalves et al., 2016
Jakarta, Indonesia	803.4–2286.6	39.9–77.31	381.1–531.8	-	Santoso et al., 2020
Mithapur, India	-	13.6–179	10.2–74.3	220.5–350.6	Basha et al., 2010
Islamabad, Pakistan	-	14.1	4.2	29.1	Shah et al., 2012
Rio de Janeiro, Brazil	5774	1216	101	53–155	Quiterio et al., 2004
Medan, Indonesia	-	-	889–3228	-	Girsang, 2008
Bangkok, Thailand	2000	< 100	-	-	Buranatrevedh, 2010
Edmonton, Canada	-	3.58	0.58	-	Bari and Kindzierski, 2017
Standard	-	500	150	60	US EPA, 2009
	15000	-	-	-	ASTDR, 2021

METHODOLOGY

Study area and time

This study is situated in West Java, Indonesia. Bandung area consists of mountains with a rainfall of 30.30–336.60 mm, during 2020 the average temperature is 25.86°C with wind speeds varying from 1.34 to 2.65 m/s and air pressure ranging from 921.98 to 924.09 millibar with 33.66% to 81.50% solar irradiance. There are five industrial locations for air sampling in Bandung and its surroundings in this study, namely Bandung Wetan, Buahbatu, Cibeureum, Padalarang, and Cimahi (Figure 1). Environmental conditions at the Padalarang, Bandung Wetan, and Buahbatu locations are industrial areas in the pharmaceutical sector. In Cibeureum and Cimahi, sampling locations are carried out in industrial areas in the textile sector.

Sample collection

TSP samples were collected using the HVAS method for 1 hour at each sampling location. The principle of HVAS is using a vacuum pump where ambient air is sucked in and passed through a 20.3 × 25.4 cm or (8 × 10 inches) filter with a minimum filtering efficiency percentage of 98.5%, equivalent to the porosity of 0.3 μ at a speed of 1.1 m^3/min to 1.7 m^3/min. Before use, the filter is identified and stored in a room with a temperature of 15°C to 35°C. Ensure the relative humidity is 50% for 24 hours, weigh the filter sheet with analytical balance glassine, then wrapped in plastic during transportation to the field. After sampling, the filter is removed carefully, so no sample is blown or released. The filter is folded with the sample position inside the fold and then stored the filter in the filter storage container. At the laboratory, store the filter in a conditioned room with a temperature of 15°C to 35°C and relative humidity of 50% for 24 hours, then weigh the and record the weight of the filter (W2). The W1 and W2 filter weights were recorded until they were constant, with the difference between the last weighing and the previous one being 4% or 0.5 mg.

TSP analysis

Before obtained the TSP concentration, the flow rate has to be corrected under standard conditions, then calculate the volume of air sampled using Equation 1 and 2, respectively.

$$Q_s = Q_0 x \left[\frac{T_s \text{ x } P_0}{T_0 \text{ x } P_s}\right]^{\frac{1}{2}} \quad (1)$$

$$V_{std} = \frac{\sum_{s=1}^{n} Qs}{n} \text{x } t \quad (2)$$

where: Q_s – the volume flow rate corrected under standard conditions (Nm^3/min);
Q_0 – flow rate of test volume (m^3/min);
T_s – standard temperature (298 K);
T_0 – actual average temperature ($273+T_{measuring}$);

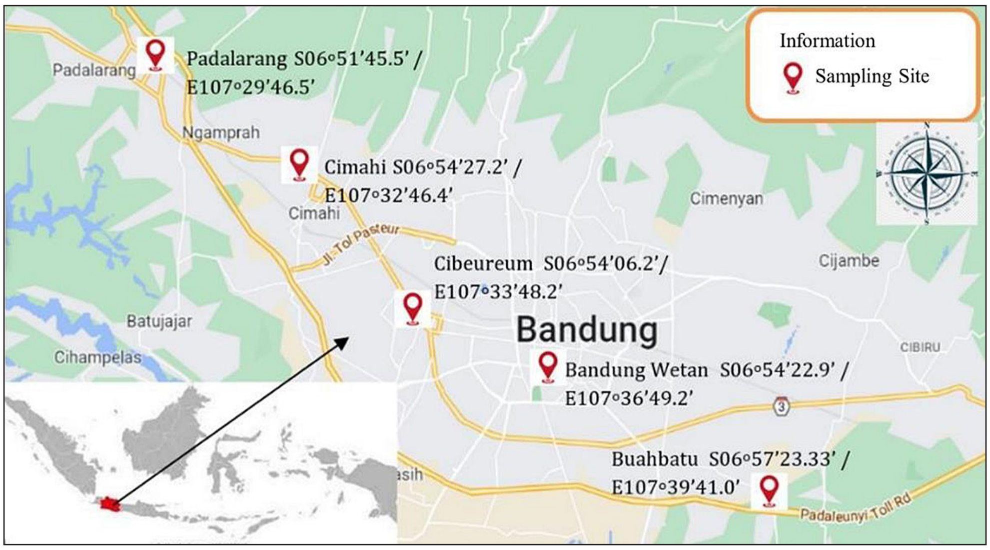

Figure 1. Sampling site

P_s – standard barometric pressure (101.3 kPa);
P_0 – average barometric pressure during sampling;
V_{std} – volume of the test sample air under standard conditions (Nm^3);
n – number of recorded flow rates;
t – duration of taking the test sample (minutes).

The TSP concentration is determined according to the equation 3 and converted into 24 hours measurement with equation 4.

$$C_1 = \frac{(W_2 - W_1) \text{ x } 10^6}{V_{std}} \quad (3)$$

$$C_2 = C_1 (\frac{t_1}{t_2})^p \quad (4)$$

where: C_1 – TSP concentration ($\mu g/m^3$);
C_2 – standard mean concentration ($\mu g/m^3$);
W_1 – initial filter weight (g);
W_2 – final filter weight (g);
V_{std} – volume of standard air test sample (Nm^3);
10^6 – conversion of grams (g) to micrograms (µg);
t_1 – sampling time (hours);
t_2 – standard exposure time (hours);
p – conversion factor (0.17).

Metal analysis

This study's analysis of metallic elements in TSP used an X-Ray Fluorescence (XRF) instrument. The principle of this instrument is that electrons move from a higher energy level to fill the vacancy due to the release of electrons so that it causes an energy difference which then appears as X-rays and is emitted by atoms. The calculation result will be in units of ng/cm^2, so it must be converted to ng/m^3 with equation 5.

$$\frac{ng}{m^3} = \frac{A}{V_{std}} \text{ x } \frac{ng}{cm^2} \quad (5)$$

where: A – filter surface area (cm^2),
V_{std} – standard volume (m^3).

In order to test the differences in parameter concentrations in different location, the Pearson Correlation is used in this study using the RStudio Cloud software.

Health risk assessment

The average daily dose equation (ADD, mg/kg.day) can be used to calculate heavy metal exposure through inhalation using Equation 6.

$$ADD_{inhale} = \frac{C \times InhR \times ET \times EF \times ED}{BW \times AT} \quad (6)$$

where: C – concentration of metal elements (mg/m^3);
$InhR$ – inhalation rate (m^3/hour);
ET – length of outdoor work (hours/day);
EF – days worked per year (days/year);
ED – unit years exposed (years);
BW – body weight (kg);
AT is (365×ED) days.

The InhR value is set according to the working time of the respondent, BW is set at 60 kg (average weight of Indonesian male respondents) and AT (365 × 72 years) (US EPA, 2022; Naseri et al., 2021). Meanwhile, information on the length of time working outdoors (hours/day) and working days (days/year) is known from the results of questionnaires for people who have activities around industrial areas. Each sampling location was conducted by distributing questionnaires to 5 respondents so that the total in this study involved 25 respondents. After obtaining the ADD_{inhale} value, the HQ and HI are calculated. HQ is the ratio of the potential exposure of a substance to the expected level of no adverse effects (calculated as exposure) divided by the appropriate chronic or acute value. At the same time, HI is the "Amount" of the HQ for the poison affecting the same target organ or organs in the body system. Different air toxins can cause similar health effects, combining the harm of different toxins (Du et al., 2019). HQ and HI values less than or equal to 1 indicate that side effects are unlikely and thus can be considered to have negligible harm. However, if the HQ and HI values are greater than > 1, it indicates the potential for non-carcinogenic side effects (Naseri et al., 2021; Goumenou and Tsatsakis., 2019; Huang et al., 2020). The HQ equation is available in equation 7.

$$HQ = \frac{ADD_{inhale}}{RfD} \quad (7)$$

where: HQ – hazard quotient;
ADD_{inhale} – inhaled metallic element dose (mg/kg.day);
RfD – reference dose (mg/m^3). RfD is obtained from equation 8 (Buranatrevedh, 2010). After knowing the HQ value for each metal element, the total HQ in each location is added, and then the HI value is obtained with Equation 8.

$$RfD = \frac{RfC \times InhR}{BW} \quad (8)$$

where: RfC – reference concentration (mg/m^3);
BW is respondent's body weight (60 kg);
$InhR$ – volume of air inhaled by workers during the entire working period (29 L/min × 60 min/hour × ET × 10^{-3} m^3/L). After knowing the HQ value for each metal element, the total HQ at each location is added, and then the HI value is obtained with Equation 9.

$$HI = \sum HQ_i = \sum_{i=1}^{x} \frac{ADDi}{RfDi} \quad (9)$$

RESULTS AND DISCUSSION

Characterization of TSP

After standardized to 24 hours, as shown in Table 2, the Cimahi location gave the highest TSP content of 126.7 µg/m^3, followed by Bandung Wetan of 87.4 µg/m^3, Buah Batu of 50.1 µg/m^3, Cibeureum of 39.1 µg/m^3 and the lowest was Padalarang of 15.1 µg/m^3. It showed that industrial areas located in urban areas such as Cimahi and Bandung Wetan have higher TSP levels. According to Nurmaningsih (2018), the level of community activity in urban areas is very high, with the dense number of vehicles which Scorrelates with the concentration of TSP. Based on Indonesian Government Regulation No. 22 of 2021, the TSP quality standard limit is 230 µg/m^3 within 24 hours. Thus, from the five sampling locations, they were still below the quality standard that had been set. However, even below the quality standard, continuous exposure to TSP-containing metallic elements can increase health problems such as lung cancer (Sari, 2013; Wulansari et al, 2016; Yulaekah and Adi, 2017).

Characterization of heavy metals on TSP

The results of metal elements levels (Al, Mn, and Pb) were presented in Table 3. For metals Al, Mn, and Pb, the highest is in Bandung Wetan, with a value of 170.1 ng/m^3, 14.79 ng/m^3, and 29.1 ng/m^3, respectively. However, the levels of these three metals are still below the specified quality standards. What is interesting from the calculation of metal elements from TSP in this study is that although the Cimahi area contains the highest TSP, the metallic elements Al, Mn, and Pb are still lower than Bandung Wetan, which has a TSP of 87.4 µg/m^3. It proves that the higher TSP does not necessarily correlate significantly correlation with the metal content. The correlation between TSP and metal elements is analyzed using the Pearson correlation test with r table 0.8783. A significant relationship occurs if the value of r count (correlation coefficient) > r table. Calculation of the Pearson correlation for TSP against Al, the coefficient value is (r) = 0.368, smaller than < 0.8783, meaning that there is a tendency that the higher the TSP, the higher the Al, but not significant. It is because Al is naturally sourced from soil minerals and will be high if there are aluminum factories around the area. Hence, the presence of Al is also strongly

Table 2. Calculation of TSP

Location	Volume of test sample in standard state			C_1 TSP (µg/m^3) 1 hour sampling	C_2 TSP (µg/m^3) 24 hours sampling
	Q_0	Q_s	V_{std}		
Padalarang	1.3	1.284	38.54	25.9	15.1
Cimahi	1.3	1.241	37.27	217.5	126.7
Cibeureum	1.3	1.242	37.23	67.1	39.1
Bandung Wetan	1.3	1.243	37.29	150.1	87.4
Buahbatu	1.3	1.239	37.19	86.0	50.1

Table 3. Concentration of metallic elements in TSP

Location	Convert ng/cm² to ng/m³							
	A (cm²)	Vstd (m³)	Al (ng/cm²)	Al (ng/m³)	Mn (ng/cm²)	Mn (ng/m³)	Pb (ng/cm²)	Pb (ng/m³)
Padalarang	17.34	38.54	2779	1250	19.8	8.91	24.3	10.9
Cimahi	17.34	37.27	2661	1238	29.8	13.86	15.2	7.1
Cibeureum	17.34	37.23	2265	1054	27.7	12.90	7.72	3.6
Bandung Wetan	17.34	37.29	3656	1700	31.8	14.79	62.5	29.1
Buahbatu	17.34	37.19	2473	1153	26.3	12.26	58.5	27.3
US EPA, 2009	-	-	-	-	-	500	-	150
ASTDR, 2021	-	-	-	15000	-	-	-	-

influenced by the soil type in the sampling area (Buranatrevedh, 2010). The correlation between TSP and Mn through Pearson's test shows that the value (*r*) = 0.7816 is still < 0.8783, meaning that the relationship is quite strong but not significant. The presence of Mn in TSP can occur due to other substances in TSP, such as hexane, styrene, 2,2,4-trimethylpentane, and naphthalene which are associated with fossil fuels (Bari and Kindzierski, 2017). The correlation between TSP and Pb through the Pearson correlation test shows that the value (r) = 0.0721 is still < 0.8783, meaning that the relationship is weak and insignificant. Therefore, this study found that the ambient air's TSP level did not influence Pb. The level of Pb in the TSP depends on the number of motorized vehicles that pass at the time of sampling (Ankar-Brewoo et al., 2020). According to (Fang et al., 2018; Hou et al., 2019), other human activities that can contribute to metallic elements in the air are burning waste and smelting iron and steel. A recap of the correlation of TSP with metallic elements is provided in Figures 2, 3, and 4.

Health risk assessment

Public health risk analysis of metal elements in ambient air TSP provided an overview of the potential hazards for people living or working in industrial areas by determining ADD_{inhale}, HQ, and HI values from each sampling location. The RFC value of Aluminum used is 6.1×10^{-4} mg/m³, while the RFC of Mn is 9×10^{-6} mg/m³. (Buranatrevedh, 2010). The RFC value of Pb has not been evaluated, so it goes directly to the RfD value, which is 1×10^{-7} mg/kg.day (Abadin et al., 2007). The results of calculating the risk analysis of the metal elements Al, Mn, and Pb from the five locations are available in Table 4–8.

Respondents from the Padalarang community have an average ET = 7.2 hours/day, EF = 247.2 days/year, ED = 7 years and InhR = 12.5 m³/hour. In the Cimahi area, community respondents have an average ET = 9.6 hours/day, EF = 295 days/year, ED = 7.4 years and InhR = 16.7 m³/hour. In the Cibeureum area, community respondents have an average ET = 10 hours/day, EF = 228 days/year, ED = 7 years and InhR = 17 m³/hour. In the

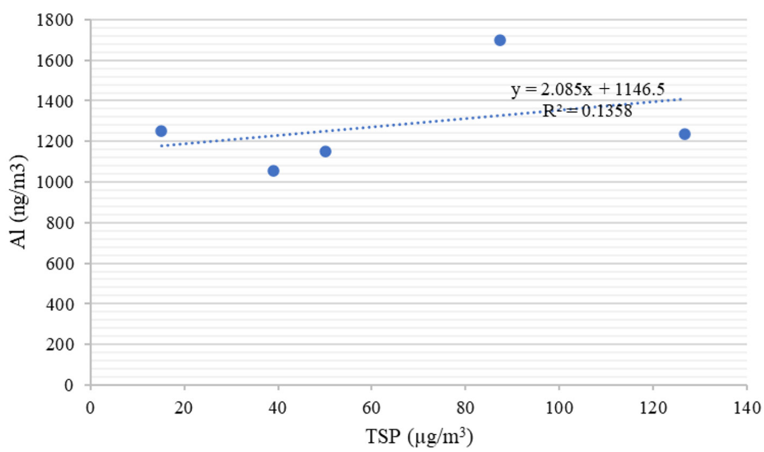

Figure 2. Correlation between TSP and Al

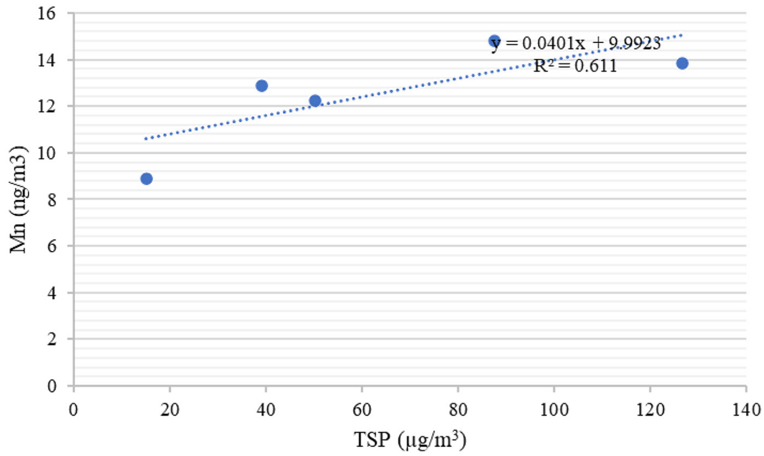

Figure 3. Correlation between TSP and Mn

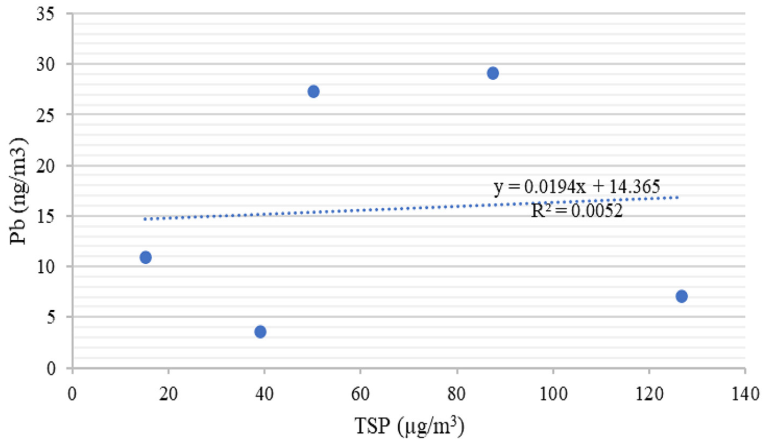

Figure 4. Correlation between TSP and Pb

Bandung Wetan area, community respondents have an average ET = 9.6 hours/day, EF = 295 days/year, ED = 3 years and InhR = 16.7 m³/hour. In the Buahbatu area, community respondents have an average ET = 9.6 hours/day, EF = 238 days/year, ED = 7.6 years and InhR = 16.7 m³/hour. The results show that Al, Mn, and Pb give HI values > 1 in all industrial areas. There is potential for non-carcinogenic side effects from exposure to Al, Mn, and Pb on public health. The longer people are outside and exposed to metal elements, the higher the risk and potential side effects on their health.

Meanwhile, the HI calculation accumulates HQ values from each sampling location, which values varying between 2.8 to 8.9. All sampling locations in this study gave values of more than 1. It means that only metal elements (Al, Mn, and Pb) have given potential non-carcinogenic effects so that they have the potential to disrupt the health of people who work outdoors around industrial areas. It is because ordinary people working outdoors around industrial areas can be exposed to metal elements from TSP for 7.2–10 hours/day, ranging from 228 to 295 days/year. Respondents studied in this study were men aged 19–43 years,

Table 4. Calculation of risk analysis of metal element exposure in Padalarang

Metal element	Risk analysis N = 5			
	ADD_{inhale}	HQ	HI	Conclusion
Al	1.27×10^{-4}	0.91 < 1	2.9 >1	HI >1 there is potential for non-carcinogenic side effects from exposure to metallic elements, especially Pb
Mn	9.0×10^{-7}	0.40 <1		
Pb	1.1×10^{-6}	1.58 > 1		

Table 5. Calculation of risk analysis of metal elements in Cimahi

Metal element	Risk analysis N = 5			
	ADD_{inhale}	HQ	HI	Conclusion
Al	$2.43×10^{-4}$	1.5 > 1	4.6 >1	HI >1 there is potential for non-carcinogenic side effects from exposure to the public by metallic elements Al, Mn and Pb
Mn	$2.70×10^{-6}$	1.1 >1		
Pb	$1.4×10^{-6}$	2.0 >1		

Table 6. Calculation of risk analysis of metal elements in Cibeureum

Metal element	Risk analysis N = 5			
	ADD_{inhale}	HQ	HI	Conclusion
Al	$1.80×10^{-4}$	1.01 > 1	2.8 >1	HI >1 there is a potential for non-carcinogenic side effects from exposure to the public by metal elements, especially Al
Mn	$2.20×10^{-6}$	0.85 < 1		
Pb	$6.0×10^{-7}$	0.88 <1		

Table 7. Calculation of risk analysis of metal elements in Bandung Wetan

Metal element	Risk analysis N = 5			
	ADD_{inhale}	HQ	ADD_{inhale}	Conclusion
Al	$1.72×10^{-4}$	0.95 < 1	5.70 >1	HI >1 there is potential for non-carcinogenic side effects from exposure to metallic elements, especially Pb
Mn	$1.50×10^{-6}$	0.60 < 1		
Pb	$3.00×10^{-6}$	4.21 > 1		

as many as 25, with 36% working as security guards, 28% as traders, 20% as drivers, and 16% as parking attendants.

CONCLUSIONS

The TSP studied at the site was the highest in the Cimahi location, with 126.7 g/m^3. Compared to the TSP air quality standard based on the Government Regulation of the Republic of Indonesia Number 22 of 2021, the concentration of TSP in the study location is still below the quality standard, which is <230 µg/m^3. However, the presence of metal elements in TSP can affect public health. The metal elements analyzed in this study are Al, Mn, and Pb, metallic elements from fossil fuel combustion, and traffic density at each sampling location. The sampling locations in urban areas with heavy traffic intensity, such as Bandung Wetan and Buahbatu, had higher Pb concentrations (29.1 and 27.3 ng/m^3) than other locations. One of the impacts on human health is that Pb can be inhaled and then settles in the bloodstream, causing it to be absorbed into a bone mass, which can cause heart disease and high blood pressure. The highest Al and Mn elements were found in Bandung Wetan, with 170.1 ng/m^3 and 14.79 ng/m^3, respectively. Although the highest metallic elements were in Bandung Wetan, the highest TSP levels were measured in the Cimahi area. This study proves through the Pearson correlation test that high concentrations of TSP do not correlate significantly with high metallic elements. In this investigation, every sampling site produced HI result greater than or equal to 1. Only the metal elements (Al, Mn, and Pb) have the potential non-carcinogenic effects that could be harmful to those who work outside in industrial regions. It is recommended that people who

Table 8. Calculation of risk analysis of metal elements in Buahbatu

Metal element	Risk analysis N = 5			
	ADD_{inhale}	HQ	ADD_{inhale}	Conclusion
Al	$2.10×10^{-4}$	1.20 > 1	8.90 >1	HI >1 there is potential for non-carcinogenic side effects from exposure to metal elements, especially Al and Pb
Mn	$2.20×10^{-6}$	0.83 < 1		
Pb	$4.90×10^{-6}$	6.95 > 1		

work outdoors in industrial areas use masks to reduce the metal elements inhaled during working hours. For companies, it is better to provide routine monitoring facilities for outdoor workers. It is advisable to check the Pb content in the blood every six months, and the company provides mask facilities as health protection for its employees.

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