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Trace metals pollution in ambient air of urban and rural coastal environments

Haryono Setiyo Huboyo[1]*, Slamet Isworo[2], Poerna Sri Oetari[2], Bimastyaji Surya Ramadan[1] and Okto Risdianto Manullang[3]
[1]Department of Environmental Engineering, Faculty of Engineering, Diponegoro University, Semarang, Indonesia
[2]Environmental Health Study Program, Faculty of Environmental Health, Dian Nuswantoro University, Semarang, Indonesia
[3]Department of Regional and Urban Planning, Faculty of Engineering, Diponegoro University, Semarang, Indonesia
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*to whom all correspondence should be addressed: e-mail: huboyo@lecturer.undip.ac.id
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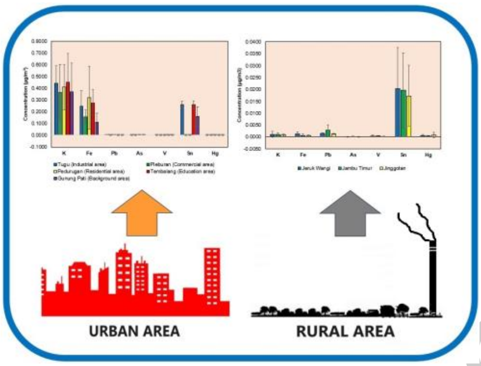
Graphical abstract



Abstract

Air pollution in coastal areas will have a different pattern from mountainous areas due to different wind patterns during the day and night. The city of Semarang, as a representation of urban areas, and Jepara Regency, as a representation of the rural regions, were chosen for this sampling. 24-h fine particulate matter ($PM_{2.5}$) was collected within 3 months of sampling. In this study, seven metal elements derived from this $PM_{2.5}$ were quantified using ICP MS. The analytical method used is a descriptive analysis of the obtained metal element data, enrichment factor (EF), Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) modeling and sources pollutant estimation. The results of the study showed that the stannum (Sn) metal was found in almost all study locations. Sn, K, and Fe are the dominant metals found in urban areas, whereas, in rural areas, only the Sn metal is quite abundant. Nevertheless, the results of EF calculations show that the most significant enrichment in both villages and cities is Sn. Allegedly, this enrichment occurred because of dense activity in the transportation sector and metal industry in urban areas, which brought metals to three villages in Jepara Regency. This research can be used as the main reference in determining health risks associated with metal pollution in atmospheric particulate matter

Keywords: Air pollution, coastal, enrichment factor, HYSPLIT model, metals element

1. Introduction

Air pollution in big cities is mainly caused by motorized vehicles, whereas in rural areas it depends on the activities that surround them. Notably, in big cities, air pollution is caused by various factors, including topography, population, climate, and weather, as well as the level or number of socioeconomic development and industrialization (You *et al.* 2019). The United Nations estimates up to 55% total population in the world live in urban areas (Ritchie and Roser 2019). Air pollution only worsens as the urban population grows, which, in turn, increases the likelihood of communities being exposed to air pollution. For instance, Jakarta and Hanoi are reportedly two of the most polluted cities in Southeast Asia. The report indicated that, in 2018, the average annual concentration of particulate matter $PM_{2.5}$ reached 45.3 and 40.8 μg/m^3 in Jakarta and Hanoi, respectively (Ardiansyah 2019). That is, the level of $PM_{2.5}$ in Jakarta is up to nine times higher than the annual safe limit set by the WHO air quality guidelines, which is only 5 μg/m^3. Jakarta's $PM_{2.5}$ exceeds the yearly safe limit stated in the ambien national standards in Government Regulation No. 41/1999 (updated to Regulation No. 22/2021) which is only 15 μg/m^3.

Industrial residual smoke and motor vehicle exhaust emissions are a source of pollution from immovable and mobile sources, which results in a decrease in air quality and a negative impact on health (Khedidji *et al.* 2017). The remaining industrial process and motor vehicles emissions contain $PM_{2.5}$. The PM in the atmosphere is a complex mixture of elements and organic carbon, mineral dust, and trace elements of water. Particulates in the atmosphere are in the form of suspensions, consisting of solid and liquid particles, measuring from 100 to less than 0.01 μm (Hu *et al.* 2014). On the other hand, rural areas which are close to coal-fired power plants, have increased health risks for the entire population, particularly for children (Artun *et al.* 2017). This is because they are exposed to a $PM_{2.5}$ concentration that is far above the limit set by the WHO air quality guidelines (15μg/m^3 of the 24-hour mean).

Indonesia's government provided 62,400 MW of energy by 2018, but only 12.4% of that came from renewable energy. Coal dominates the sources of energy in Indonesia because of its low cost and extensive availability (Sharvini *et al.* 2018). The government policy of Indonesia caused its performance on climate change conditions to be classified as low, which is currently at 38th position from 60th in 2019 (Burck *et al.* 2019). Looking at the health risks caused by the metal content in the ambient air, we find that it is necessary to carry out a related study of metal concentration found in urban environments and in rural areas that have a power plant in their coastal ecosystem.

Many urban and rural environments have been contaminated with metals (e.g., Cd, Cu, Pb, and Zn) as a result of industrial operations, transportation activities, and other anthropogenic activities (Chantara *et al.* 2019). The release of these contaminants can pose significant potential environmental and human health threats to people who live within the vicinity of the environment. Air pollution due to heavy metal contamination is a severe problem because they are toxic, and their bioaccumulation capacity is hazardous in terms of their effects on the food chain (Mohmand *et al.* 2015). Because many human activities involve the industrial sector, in both rural and urban environments, one or more heavy metals can accumulate. Many metals, such as copper and selenium, are important elements for the growth of plants and living organisms, but at high concentrations, these elements become toxic. Industrialization, urbanization, and agricultural practices are the three main metal sources in ambient air (Power *et al.* 2018 and Sodango *et al.* 2018).

This paper discusses the contribution of metal pollutants from ambient air to the coastal environment, especially in urban and rural areas. This study also compared and analyzed markers (metals) found in ambient air in the two regions using enrichment factors (EFs). The city of Semarang is a representation of a coastal urban environment. In contrast, the villages around the Jepara Regency close to a power plant are a representation of a coastal rural environment affected by power plant activities. This study has been carried out as part of efforts to conserve the coastal environment that is mainly affected by the presence of trace metal elements to create a healthy atmospheric ambience. It is hoped that the knowledge of enriching trace metals in ambient air can become a baseline for further mitigation strategy and management of emission sources.

2. Materials and methods

2.1. Sampling location

The sampling locations in Semarang were chosen based on the areas they represent: (1) industrial area, (2) commercial area, (3) educational area, (4) residential area, and (5) remote area as background. The villages at Jepara Regency are considered as a representation of rural regions. These villages are the closest to a coal-fired power plant, so are predicted to be the most affected by industrial activities. In this rural site, the power plant is about 9 km away to the north of the sampling location. Figure 1 shows the locations of our sampling sites, and Table 1 presents their information, including coordinates and the area they represent.

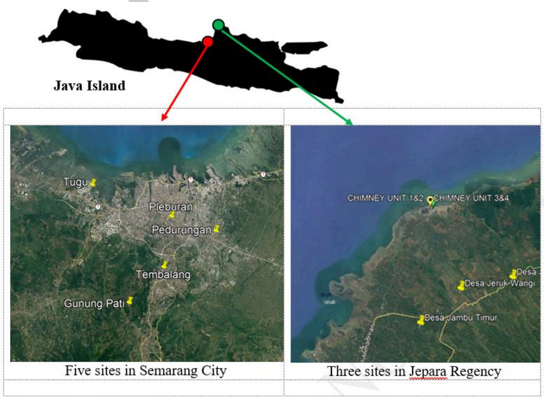


Figure 1. Measurement sites in Semarang city (urban) and Jepara Regency (rural)

Table 1. Sampling sites in urban areas and rural areas

No	Site Name	Coordinates	Remark
		Semarang City (Urban Area)	
1	Tugu	06°58'02.90"S, 110°20'12.50"E	Industrial area
2	Pleburan	06°59'39.71"S, 110°25'28.12"E	Commercial area
3	Pedurungan	07°0.0'21.99"S, 110°28'23.82"E	Residential area
4	Tembalang	07°2' 55.13"S, 110°25'13.43"E	Educational area
5	Gunung Pati	07°05'21.17"S, 110°23'09.49"E	Background area
		Jepara Regency (Rural Area)	
1	Jambu Timur	06°32’15,97”S, 110°44’00,99”E	Rural residential area
2	Jeruk Wangi	06°30’42,11”S, 110°45’53,35”E	Rural residential area
3	Jinggotan	06°30’12,47” S, 110°48’18,89”E	Rural residential area

2.2. Sampling methods

A high-volume air sampler (TFIA 2F Staplex model series) was used to collect $PM_{2.5}$ samples. The $PM_{2.5}$ air sampling time was 23 h, and filter paper and holder preparation took 1 h. The filter used was the EPM 2000 quartz filter (Whatmann Inc.), which has a diameter of 20.3 cm × 25.4 cm and minimum filter efficiency of 98.5%. After the filter was installed in the air sampler, the air flowrate was set to be in the range of 1.1–1.7 m^3/min by Indonesian National Standard 7119.14.2016 on how to measure $PM_{2.5}$. Then, the filter of sampled $PM_{2.5}$ was analyzed using the gravimetric method. Measurement of sample weight was carried out using the Mettler Toledo MS 205P4 model, where each sample was taken three times at each location. The measurements in rural areas were carried out in the period June - September 2018, while measurements in urban areas were performed in August – September 2018. This represents the dry season period in Indonesia.

2.3. Data analysis

$PM_{2.5}$ samples were analyzed for metal elements components using an inductively coupled plasma mass spectrometer (Agilent Technologies 7900 model). The metal element analysis method used is based on USEPA IO-3.4. Seven metals, including, K, Fe, Pb, As, V, Sn, and Hg, were analyzed. Determination of the value of the limit of detection (LoD) is based on USEPA 29 methods in which half the LoD then replaces the concentration of elements that are below the LoD, and arithmetic averages return the missing values. When the elements' concentration level exceeds the LoD, the uncertainty value is calculated as one-third of the LoD value. If the concentration is equal to the LoD, then the uncertainty value is calculated as five-sixths of the LoD and uncertainty analysis. The missing data are computed as four times the arithmetic average.

The EF is an approach used to distinguish metals that come from anthropogenic activities and those from natural sources. The EF for each element X relative to the control/uncontaminated reference material is determined by equation (1):

$$EF = \left(C_n / C_{ref}\right) \Big/ \left(B_n / B_{ref}\right) \quad (1)$$

where EF is the enrichment factor, Cn is the measured metal sample concentration, Cref is the measured reference sample concentration, Bn is the background metal concentration (Earth crust), and Bref is the reference sample background concentration (earth crust). Metal components such as Al, Si, Ti, and Fe are generally used as reference points for EF calculations because they are very abundant in scale material and are not significantly affected by pollution. EF values of <2 indicate minimal enrichment; 2–5, moderate enrichment (moderate); 5–20, significant enrichment; 20–40, very high enrichment; and >40, extremely high enrichment. According to Chen *et al.* (2015), if EF approaches 1, then the metal element can be ascribed to soil particles. Meanwhile, if the EF is more than 10, then it is most likely that the element is derived from human activities (Wan *et al.* 2016).

HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) is used to model the transport and dispersion of metals from particulate matters. It is also used as a back-trajectory analysis to determine the source of air mass and determine the relationship of sources with receptors to strengthen the analysis (Stein et al. 2015 and Rolph *et al.* 2017). HYSPLIT has also been used frequently in various simulations such as atmospheric transport, dispersion, and deposition of pollutants and toxic materials. Examples of its application are detecting and estimating sources of radioactive material, fire smoke, flying dust, contaminants from stationary variations, and causes of moving emissions, such as volcanic ash (Liu *et al.* 2013; Reizer and Orza 2018). Day and night wind roses data are used to see pollutant sources during the day and at night at two sampling sites.

3. Results and discussion

3.1. Metal characteristics

Figures 2 and 3 show the characteristic trends of each metal element in urban and rural areas where several metal elements such as K, Fe, and Sn have concentration values that are quite high compared with those of other metals. The slices are at these two locations, where the concentration of Sn is quite high, indicating pollution due to industrial activities. In Semarang, Sn is most abundant in Gunung Pati, Tembalang, and Tugu area, which represent the background, education, and industrial areas, respectively.

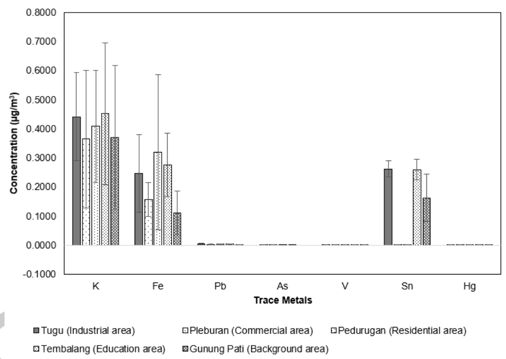


Figure 2. Characteristics of metals found in urban areas

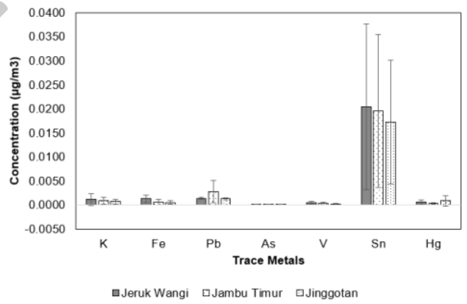


Figure 3. Characteristics of metals found in rural areas

In addition, a high concentration of Fe and K in ambient air is found in almost all locations except the background area, which has the lowest Fe concentration. Most potassium (95%) is used as fertilizer, and the rest is used to make potassium hydroxide (KOH), by electrolysis of a solution of potassium chloride, and then converted to potassium carbonate (K_2CO_3). Potassium carbonate is used to make glass, primarily the kind used to make televisions, whereas potassium hydroxide is used to make soap and liquid detergents. According to Gugamsetty (2012), Fe originates from soil dust and roads, which are suspended again because of the activity of vehicles and living things. Fe is also strongly associated with dust suspended from a crustal material as a potential source of the element in $PM_{2.5}$ (Gugamsetty *et al.* 2012).

Sources of the contribution of stannum (Sn) come from the soil and industrial activities. Stannum enters the environment through both natural and man-made sources. In contrast, organic compounds are mainly released from anthropogenic sources. Inorganic lead is considered relatively immobile in the environment. Exposure of humans to lead can be through breathing, consumption, or skin absorption. Food consumption, especially canned food and drinks, is considered as the primary source of human exposure to inorganic lead forms (Wu *et al.* 2013). At the three locations in the rural area, the value of Sn on average is about 0.0572 ± 0.0153 μg/m^3, which indicates the presence of industrial contamination of the surrounding ambient air at the measurement location. As shown in Figure 3, several other parameters, such as K, Fe, and Pb, appear to be slightly more significant compared with those of As, V, and Hg. This condition shows that industrial pollution is quite worse in those areas, which are close to coastal and industry environments.

3.2. Enrichment factors

EF analysis is used to detect sources of pollutants, which is an analysis used in geochemical studies to determine metal differences from anthropogenic activities and those originating from natural sources (Al-Mur *et al.* 2017). The EF method normalizes the measured heavy metal content concerning a reference metal sample such as Fe (Abrahim and Parker 2008). In this approach, Fe is considered to act as a "proxy" for land. Fe can be used to calculate EFs and is a suitable normalization element because Fe distribution is deemed to be not related to other heavy metals. The EF method will normalize the data from measurements of metal concentrations in the soil with reference samples. Meanwhile, according to Al-Momani (2003), EF values close to 1 indicate that a crustal material is the primary source of an element or that this source has not been mixed with anthropogenic inputs. EF values in the range of 1–10 are considered to be not enriched because of differences between the chemical composition of the soil and the form of the reference crust. Values between 10 and 100 for EF elements are regarded as moderately enriched, indicating higher concentrations of certain elements in the air than those in continental crusts. Lastly, highly enriched conditions are present in EFs > 100, which is a sign of extreme human activity contamination (Al-Momani 2003).

As shown in Table 2, the EF value used as a reference element is Fe. The results show that the EF value for K in Gunung Pati as a background area is quite large. Moreover, Pb, Hg, and Sn also exceed 10, which indicates that all four elements, including K, are caused by human activities. When compared with that of educational and industrial locations, the EF value of elements in Gunung Pati is far higher. This condition indicates that there is pollution caused by human activities in the background area, such as agriculture and other domestic activities. Commercial and residential areas show relatively large EF values compared with those of different locations, indicating pollution from industrial activities and motor vehicle fumes.

Table 2 also shows the existence of enormous EF values for the metal elements Pb, As, V, Sn, and Hg. The high values indicate the presence of substantial metal element enrichment in rural areas. The enrichment of metal elements, notably Pb, As, and Hg, can be caused by the burning of fossil hydrocarbons, vehicle traffic, and emissions from industries related to metals. Also, burning gasoline, coal, and other fuels increases the burden of environmental vanadium (V), which results in environmental pollution and occupational hazards for industrial workers. Sn comes from industrial processes, food and cigarettes, drinking water, and biomass burning. The EF value obtained is also tremendous when compared to that in urban areas. These phenomena indicate pollution from other sources that are near the sampling location in rural areas. However, coal-fired power plants cannot be considered as a source of pollutants; thus, it is necessary to conduct an analysis using HYSPLIT modeling to understand the potential causes of pollutants of several metal components that have been tested (Liu *et al.* 2013).

Table 2. Enrichment factors of metal elements in the sampling locations

Location	K	Fe	Pb	As	V	Sn	Hg
Tugu	7.22	1.00	62.87	29.17	0.90	54,385.12	167.56
Pleburan	8.62	1.00	101.70	52.45	0.89	147.75	169.47
Pedurugan	7.60	1.00	74.20	39.64	0.71	123.12	134.19
Tembalang	5.13	1.00	49.20	28.87	0.93	39,119.29	79.38
Gunung Pati	13.38	1.00	66.16	0.00	0.33	73,979.65	202.12
Jeruk Wangi	1.90	1.00	6,943.09	5,760.99	133.75	21,241.82	21,044,756.54
Jambu Timur	3.91	1.00	44,335.80	16,755.95	163.84	53,954.17	50,396,319.44
Jinggotan	3.93	1.00	16,634.77	11,526.44	162.95	56,195.74	45,091,478.01

3.3. HYSPLIT modeling

This HYSPLIT model uses a back-trajectory analysis to determine the source of pollutants. In this case, wind data such as wind speed and geostrophic altitude of the wind speed are used. The percentage of errors from calculations and the actual location of the source in the calculation of the return path is relatively low, at around 5%.

The back-trajectory model results (Figure 4) indicate that the source of pollutants in the city of Semarang is likely to come from industrial areas and anthropogenic activities located east of the town.

The same results were shown in the rural areas at three sampling points where the direction of the pollution model at these points was due to anthropogenic activities from the southern and eastern parts of the location. This condition shows that the steam power plant that borders these villages does not pollute the surrounding environment. From the pathway formed, pollution possibly comes from the metal industry area around Juwana, Pati District. Most of those metal industries are home-based industry that do not comply with environmental regulations.

3.4. Source of pollution estimation

Wind speed determines the depth of how much air pollutants are initially mixed. The speed and direction of the wind determine the rate of the spread of pollutants. These factors determine whether an area will be polluted and how quickly the level of pollutant thins because they mix with environmental air after the material leaves the source. Meteorological factors will determine the spread of pollutants in ambient air, emitted from both immovable and mobile sources. Meteorological conditions will determine the extent of pollution, the pattern of the spread of pollutants, and the scope and duration of their range. The study location is a coastal area where there is a diurnal wind change. This also leads to the identification of different pollutant sources during the day and night, as shown in the Figure 5.

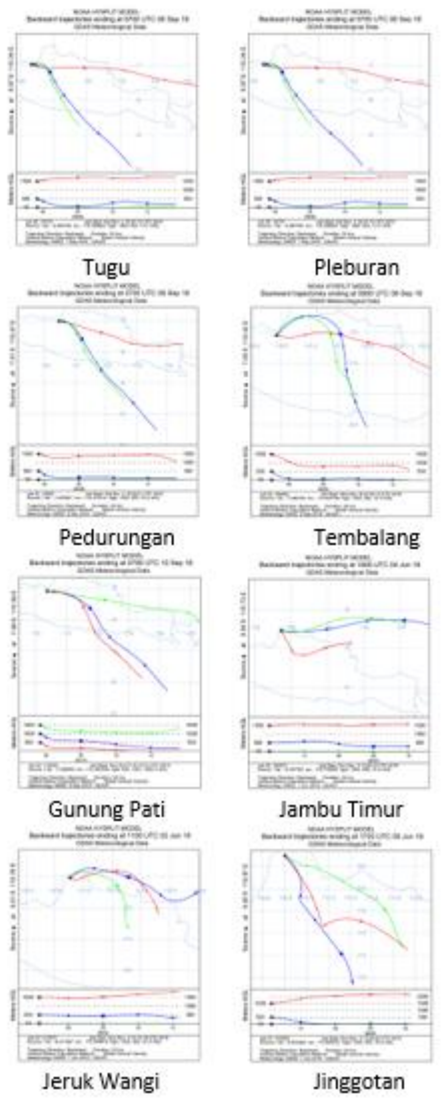


Figure 4. HYSPLIT output of all sampling locations during sampling period

During the sampling period, in the urban area (Semarang city), air pollutants during the daytime came from the East and Southwest while at nighttime they came from the East. This means that pollutants during the daytime are more diverse than those at night due to more diverse source contributors than night time. In the rural area (Jepara regency), the sources of air pollutants during sampling are attributed from sources in the Northeast, East and Southeast regions (during the daytime) and sources from the Southeast direction (at nighttime). So, it is the same as in urban areas, where pollutant sources are more diverse during the daytime than at nighttime. The same conditions can also be seen in the two places when they are extended for a year where, during the daytime, the pollutant sources can be more diverse, which is indicated by the spread of the wind direction of the pollutant sources.

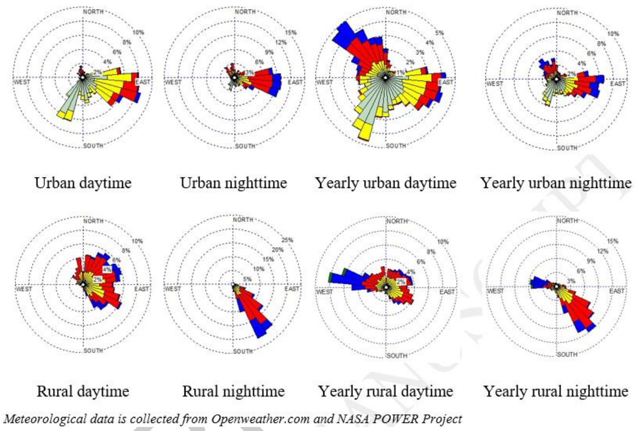


Figure 5. Wind roses pattern during sampling period and yearly period in urban-rural area

The air temperature in the measurement area for 24 h was 30.6°C, and those for 1 h in the morning and 1 h in the evening are 32.52°C and 30.86°C, respectively. Abbasi *et al.* (2017) stated that air temperature directly influences atmospheric stability. In a stable condition, the mass of air pollutants cannot increase but remains in the atmosphere and accumulates, thereby increasing the concentration of pollutants. Conversely, when the air temperature is higher than the ambient air temperature, the mass of air pollutants will grow, and they will spread; thus, there is no precipitation on the surface, and the concentration of pollutants minimizes (Abbasi *et al.* 2017). There were no significant different ambient temperatures between urban and rural areas, thus it should be not a factor to different metal contributions.

Ca, Cu, Fe, K, Mn, Ni, Sr, and Sn are elements that come from the process of soil abrasion carried by dust flying into the air, as reported by Jiang *et al.* (2018) that dust may come from soil dust and road dust. Therefore, extensive traffic activity near the sample location is very likely to cause metal pollution, especially the Pb metal element. The level of urbanization and road age significantly influences the distribution of metals in various urban areas. The roadside soil being the primary source of Pb metal pollution has been studied by Yan *et al.* (2018). In roadside soils, several metals such as Sb, Cu, Cd, Pb, Hg, and Zn were found to be quite high in concentration (Yan *et al.* 2018). Because of its particular physical and chemical properties, vanadium is widely used in the petrochemical industry, smelting iron and steel, welding, catalysts, pigments, storage batteries, and preservatives. Vanadium compounds can be absorbed into the body through the respiratory tract, skin, and digestive tract during mining and metal production. The disposal of waste gas, residues, and industrial wastewater contributes to the massive amounts of vanadium in water, soil, and air (Sun *et al.* 2017). Arsenic has semi-metallic properties, is highly toxic and carcinogenic, and is widely available in the form of oxides or sulfides or as iron salts, sodium, calcium, copper, and others. Arsenic is the 20th element on Earth, and its inorganic forms such as arsenite and arsenic compounds are life-threatening to the environment and living things. Arsenic is a protoplastic toxin because it mainly affects sulphydryl cell groups, which causes dysfunction of cell respiration, cell enzymes, and mitosis (Jaishankar *et al.* 2014).

Although Tugu District is an industrial area, the primary source of its pollutants is motorized vehicles, not the industrial activity itself. This hypothesis is derived because not only are there factories or manufacturing plants in the area, but there are also warehouses and distributing centers. Because they do not deal with production, distribution companies produce emissions from motor vehicles. Another factor is that Tugu Subdistrict is traversed through North Highway Line, which has the densest traffic on the island of Java. One of the most significant sources of anthropogenic trace metal emissions is the metal smelting industry. The pyrometallurgical industrial process produces the most massive emissions from As, Cd, Cu, Ni, and Zn (Zhan *et al.* 2014). Cr, Ba, Mo, Zn, Pb, and Cu are usually associated with motor vehicle emissions and oil combustion, whereas Fe, Pb, and Zn are excluded from municipal waste incinerators (Adgate *et al.* 2007). The impact of generator emissions is PM, which is produced from burning coal and spread through a chimney from a power plant. Enriched elements Pb, Zn, Cu, Ni, and Mn, are mostly from anthropogenic activities, whereas Al, Ca, Mg, and Fe are not enriched and are mainly related to natural sources (Hu *et al.* 2014). Concentrations of geological elements are such as Fe and K, which are strongly correlated to PM2.5 mass analyzed based on the species relationship table with pollutant sources obtained from the existing literature. Pb and Hg are pollutant markers sourced from motorized vehicles, whereas F, As, and K are also correlated because they are a source of biomass combustion.

Sourced from motor vehicle emissions, Pb is used as an anti-detonator in gasoline. Andrew *et al.* (2017) inform that iron (Fe) in the air can be sourced from vehicles even in small amounts. Although the primary source of Fe is suspended soil dust, the content of Fe in ambient air can also be sourced from vehicle activities (Venter *et al.* 2017). Meanwhile, Panda and Shiva Nagendra (2018) stated that Fe is strongly associated with dust resuspension from a crustal material as a potential source of these elements in $PM_{2.5}$. Estimated contributions to emissions from mobile sources have the most significant amount for total contributions. This contribution is likely because almost all activities around the measurement area in Pleburan use motorized vehicles. It is also caused by the monitoring area that is quite close to the road. Also, the route is relatively congested all the time. The contribution of various vehicle emissions, the use of brakes and tires, emissions due to friction with asphalt, and emissions that are suspended again (resuspension) are examples of activities that affect ambient air quality in the Pleburan region.

In this study, the small number of measured samples affected the output of the HYSPLIT program, producing an unknown source. Unknown sources by the HYSPLIT program are relatively low. Thus, they may be related to a variety of marker sources that are not routinely measured at the measurement locations (Reizer and Orza 2018). In this study, data that were not analyzed or not identified could be included in organic aerosols, inorganic ions, and sea salt. The source cannot be identified because the pollutant metal element originating from the source is too small, the activity that causes the source is not dominant, or the intensity of the measurement is low.

4. Conclusion

Great stannum (Sn) concentrations are found in both locations, in the urban and rural areas of the coastal environment. However, other metals components, Fe and K, are also found in large concentrations in urban areas. However, the level of Sn in rural areas is much lower than in urban areas, which indicates that ambient water in urban areas is far more polluted compared with that in rural areas. Surprisingly, the Sn pollution that occurred in the village was strongly suspected not to originate from a power plant near the village. This analysis is reinforced by HYSPLIT modeling that directs potential pollutants from the south and east of the village. The power plant is located north of the three sample villages. It could be that metal component pollution comes from the metal industrial area located in the Juwana District, Pati Regency. The high number of metal components such as Fe, K, and Sn in urban areas is thought to originate from fairly dense transportation activities. This allegation is reinforced by HYSPLIT modeling that directs the source of pollutants from several regions with very dense transportation, the North Highway Line, through Semarang city to neighboring areas such as Demak Regency.

Acknowledgments

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