

CHARACTERISTICS OF FINE PARTICULATES ELEMENTAL COMPOSITION OF TWO LARGEST CITIES IN INDONESIA USING ION BEAM ANALYSIS

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Abstract

Urban air pollution is a major problem in Indonesia and has growing recognition especially on the health effect problems resulting from airborne particles exposure. Even some cities have been identified to have high levels of air pollution with an emerging need of source apportionment. Therefore, characterization of the chemical composition of air particulates is a fundamental step for identification of pollutant sources. These chemical compositions are generally at trace levels thus require an accurate and suitable analytical method such as Ion Beam Analysis (IBA) which is fast, effective and sensitive. In this study, a non-destructive IBA method was applied to characterize PM_{2.5} of ambient air samples in the two largest cities in Indonesia, Jakarta and Surabaya, which have different characteristic as urban cities. Several elemental compositions i.e., F, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Br, Rb, Sr, Y, Zr and Pb, were well quantified for both cities. It was found that Surabaya has high concentration of heavy metals such as, Pb, Fe and Zn, while Jakarta has higher level of Cu concentration than other elements. The results can be further utilized to identify their potential sources while at the same time can serve as science-based evidence for particulate matter pollution mitigation program formulation in each city.

1. INTRODUCTION

Fine particulate matter (i.e., PM_{2.5}, particles with an aerodynamic diameter equal to or lower than 2.5 μm in ambient air) is known to have a detrimental impact on human health i.e., cardiovascular disease, pulmonary disease, increasing risk of diabetes, Alzheimer and lower cognitive performance [1-3]. Fine particulate matter has been associated to several million deaths annually. In 2019, air pollution was estimated to contribute to nearly 6.7 million deaths worldwide which was dominantly caused by fine particulate matter [4]. Fine particles are mainly derived from chemical reactions including anthropogenic combustion-related emissions such as vehicular emissions, industrial emissions (e.g., coal), and biomass burning also secondary aerosol which is formed through gas to particle conversion mechanism [5,6]. The assessments of ambient PM concentrations and chemical composition become important to identify the possible sources and their contributions, so the results may support in planning and implementing more effective control policies for PM air pollution [7-9]. Different compositional share of elements to the total PM concentration would likely contribute to different degree of health impacts.

In Indonesia, urban air pollution becomes a major problem and it has growing recognition from the public due to potential detrimental health effects or even premature mortality linked to airborne particles. The country with an estimated population of over 261 million people and becomes the world's 4th most populous country, has experienced increased economic development for the last decades which often led to rapid and unplanned urbanization. Escalation of economic activity associated with increasing intensity in transportation and industrial sectors, lead to the growth of fossil fuel energy use which in turn causing the higher level of air pollution. Several cities in Indonesia have also been identified for their high levels of air pollution, especially in mega and big cities resulting in a public concern on air pollution monitoring in big cities [10]. This problem has been encountered in Jakarta and Surabaya, recognized as the two largest cities in Indonesia in terms of population and administrative areas. As the capital city of Indonesia with total population of nearly 11 million and the largest city in Southeast

Asia, air quality in Jakarta becomes public concern. The city has 4 major industrial estates within the city and surrounded by more than 10 industrial estates within radii of 20–30 km from the city center. Most of them consume diesel oil with high sulphur content and low energy efficiency with lack of air pollution control device. The land transportation is characterized with medium to heavy daily traffic jams in all type of roads even though public transport keeps improving. These conditions coupled with local meteorological condition contribute to the deterioration of urban air quality. Our previous study reported that the annual mean of $PM_{2.5}$ in Jakarta from 2010 to 2019 were measured in the range of 15.5 – 31 $\mu g/m^3$ and were considered to exceed the Indonesian annual mean national ambient air quality standard (NAAQS) of 15 $\mu g/m^3$ [11]. Similar condition has been also encountered in Surabaya which is known as the second largest city in Indonesia. Our previous study showed that the annual mean of $PM_{2.5}$ in Surabaya from 2012 to 2017 were measured in the range of 15 to 20 $\mu g/m^3$ which were well above the Indonesian NAAQS [8]. Another study confirmed the contribution of vehicular emission to the high levels of daily concentration of air pollutants [12]. In addition, the city has busiest trading city ports in Asia and it is also surrounded by several industrial estates in its vicinity.

These two cities have been experiencing high levels of fine PM concentrations. Both cities have different intensities in industrial and other anthropogenic activities, thus the characterization of air particulate chemical composition is significantly needed. These chemical compositions are generally at trace levels thus require an accurate and suitable analytical method. Nuclear analytical techniques such as neutron activation analysis (NAA), X-ray fluorescence (XRF) and ion beam analysis (IBA) are highly suitable for characterizing the elemental compositions of PM samples [13–15]. Ion beam analytical technique is particularly suitable for fine particle filter paper analysis since the particle loading on these filters can be considered to be thin targets for MeV ion beams used in their analysis [16]. The IBA techniques offer some advantages such as being considerably fast with only needing a few minutes of analysis, non-destructive, no sample preparation required, able to analyse sub micrograms of samples, and multi-elemental analysis simultaneously [17].

In this study, IBA analysis was applied for the multi-elemental characterization of aerosol fine particles in cities of concerned of Jakarta and Surabaya. The measurement of the PM elemental composition is a key factor in the utilization of the data for the determination of possible sources, whose identification and apportionment are an important step in the air quality management process. Therefore, the objectives of this study were to compare PM elemental compositions in both cities of Jakarta and Surabaya. To the extent possible, detailed information of the measurement results for both cities affected by the difference in the nature of emission sources was provided for formulation of the precise and effective regulations related to the air quality management plan.

2. METHODOLOGY

2.1. Sampling site

Sampling of ambient $PM_{2.5}$ was done for 24-hours using GENT sampler from 2019 – mid 2020 carried in the two target cities. Note that there are differences in the urban setting for both cities, for example the scale of areas and population, type of major industrial activities, as well as the traffic characteristics and the urban sprawl. The sampling was done at the office of Environmental Protection Agency of Jakarta Provincial (Jakarta EPA) located in Central Jakarta as shown in Fig. 1a. This site is in the city center and surrounded by arterial roads with heavy traffic volumes especially during the morning and evening rush hours.

There are two sampling sites in Surabaya: East Surabaya (1) and West Surabaya (2) as seen in Fig. 1b. East-South Surabaya was selected due to the domination of industrial estates while West Surabaya site was selected because it is located close to port, but no metal industries are located surrounding the site.

Sampling was carried out using dichotomous sampler Gent stacked filter unit in each site, once a week, at a flow rate of 15–18 L/min at the two sampling sites. The $PM_{2.5}$ air particulate samples were collected on a 0.4 μm pore size filter for 24-hours using GENT sampler from 2019 – mid 2020. We successfully collected totally 105 samples which consists of: 32 samples from Jakarta, 35 samples from East-South Surabaya and 38 samples from West Surabaya. The samples were delivered back to BATAN laboratory for mass and BC analysis.

2.2. Particulate Matter (PM) and Black Carbon (BC) Analysis

Determination of mass concentration $PM_{2.5}$ was carried out by gravimetric method using Mettler Toledo micro balance. The filters were weighed before and after the sampling period to determine the particulate mass

collected and then divided by the total volume of air that passed through the filter to obtain the particulate mass concentration. Before weighing, the filter must be conditioned in a room with humidity between 45-55% and a temperature of 18-25°C. Fine APM ($PM_{2.5}$) is obtained from the weighing of the sample weights on the fine filter.

Determination of carbon in the filter was based on the process of light reflection. The absorption and reflection of visible light by airborne PM in the filter depends on particle concentration, density, refractive index and size. The black carbon concentrations of the samples were determined by reflectance measurement using a Digital Smoke Stain Reflectometer EEL Model 43D. Measurement of BC in the filter was done based on the light absorption principle to quantify the amount of light that is absorbed by the filter sample using the assumed value of the average coefficient of particle mass absorption of $5.7 \text{ m}^2/\text{g}$.

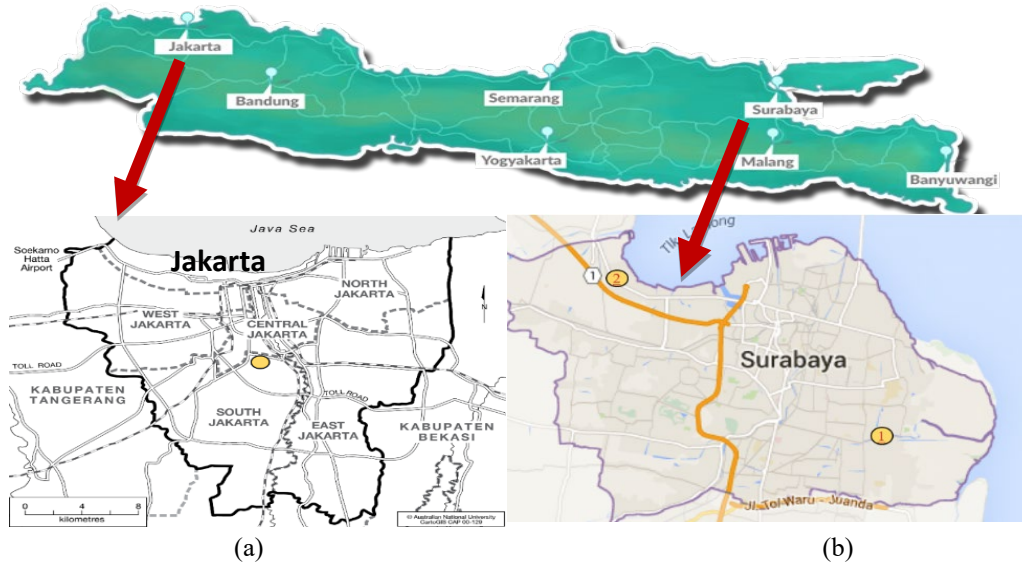


FIG.1. The sampling locations in Jakarta (a) and Surabaya (b)

2.3. Elemental Analysis

The samples were analysed using a non-destructive IBA techniques including proton induced x-ray emission (PIXE), proton induced gamma-ray emission (PIGE), and proton elastic scattering analysis (PESA) at the Australian Nuclear Science and Technology (ANSTO) facility, which has the capability of multi-element analysis with detection limits in the order of ng/cm^2 . The IBA technique uses an 8 mm diameter beam of 2-3 MeV protons with 10-15 nA target current. A total of 18 elements were successfully detected such as Na, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Br, and Pb.

3. RESULTS AND DISCUSSIONS

3.1. PM and BC mass concentration

Comparative analysis was done for the mass concentration (i.e. $PM_{2.5}$ and PM_{10}) and the concentration of black carbon in fine particulate in Jakarta and Surabaya and the results are presented in Table 1. The mean concentrations of $PM_{2.5}$ and PM_{10} in Jakarta were 16.13 and $34.75 \mu\text{g}/\text{m}^3$, respectively whereas for Surabaya were 18.52 and $45.18 \mu\text{g}/\text{m}^3$, respectively. The results of mean concentrations of $PM_{2.5}$ and PM_{10} in Surabaya were higher than those measured in Jakarta. The 24-hour average $PM_{2.5}$ and PM_{10} mass fraction results for the two cities are shown in Fig. 2. The box represents 25-75% of the distributions of the $PM_{2.5}$ and PM_{10} concentrations while the horizontal bar in the box indicates the median. The points lying outside the range defined by the whiskers (extreme events) are plotted as outlier dots. Second to the previous finding, Fig. 2 showed that the level of average $PM_{2.5}$ and PM_{10} concentrations in Surabaya were also higher than those measured in Jakarta. Note that, PM

concentrations in Surabaya were obtained by averaging the data from 2 sampling sites; East and West Surabaya, which shows the potential contribution of more emission sources to the two sites while only one site was considered for Jakarta. It seemed that contributions of industrial and port emissions to the measured PM were crucial. Especially the shipping emission caused by continuous burning of marine fuel oil (MFO) with sulfur content of more than 10,000 ppm with no control device installed in the tail pipe. Industry also uses heavy diesel oil (HDO) for back-up generator mainly as well as coal.

Strict implementation of Large-Scale Social Restriction (LSSR) policy due to Covid-19 pandemic in the early of 2020 was promulgated and it was reported to significantly reduce the PM concentration especially in Jakarta (Santoso et al., 2021). Jakarta implemented the LSSR policy earlier than Surabaya since March 2020 while the latter implemented the LSSR policy in the end of April 2020. During the implementing months of LSSR, the concentration of $PM_{2.5}$ in Jakarta reduced significantly, reached almost 40% compare to the period months in the previous year [18]. In Surabaya the reduction of $PM_{2.5}$ concentration only 7-15% but has significant decrease in the heavy metal concentration [19]. In addition, Jakarta is very strict in implementing the LSSR because the first Covid case was found in Depok, about 30 km from Jakarta. The implementation of the LSSR in 2020 has affected fuel consumption in Jakarta, including for the industrial sector. Annual fuel consumption for all types of fuel tends to decrease from 2019 to 2020. The reduction rate for diesel consumption reaches 23% which results in a reduction in air pollutant emissions from industrial activities (<https://ditppu.menlhk.go.id/simpel/gis/konsumsienergi>). LSSR also limits the intensity of mobile transportation, especially on the main roads in the city. The distribution of fuels including gasoline and diesel in 2020 has decreased due to less traffic mobility which is one of the reasons for the low $PM_{2.5}$ in Jakarta. Therefore, it can be concluded that transportation contributed significantly to the measured PM in Jakarta site, while in Surabaya it was mainly contributed by the industrial smelter. Compared to previous study by Santoso et al (2020), the concentrations of $PM_{2.5}$ and PM_{10} measured during the period of 2019-2020 in Jakarta site are lower than those measured in the similar period for the two sites in Surabaya. However, $PM_{2.5}$ values measured for both cities exceeded the Indonesian NAAQS of $15 \mu g m^{-3}$ and according to the United States PM_{10} annual standard, both values of PM_{10} from Jakarta and Surabaya are still below the standards of $50 \mu g m^{-3}$ [10].

TABLE 1. PM mass and BC concentrations measured in Jakarta and Surabaya

	Surabaya ($\mu g/m^3$) ^a		Center Jakarta ($\mu g/m^3$)	
	Average	Range	Average	Range
$PM_{2.5}$	18.52 ± 8.09	3.17-33.35	16.13 ± 6.22	5.00-29.29
PM_{10}	45.18 ± 19.92	11.42-92.51	34.75 ± 12.39	10.89-66.74
BC	3.76 ± 1.63	0.53-6.85	3.39 ± 0.75	1.31-5.02

Note: ^a Values were averaged from two sites

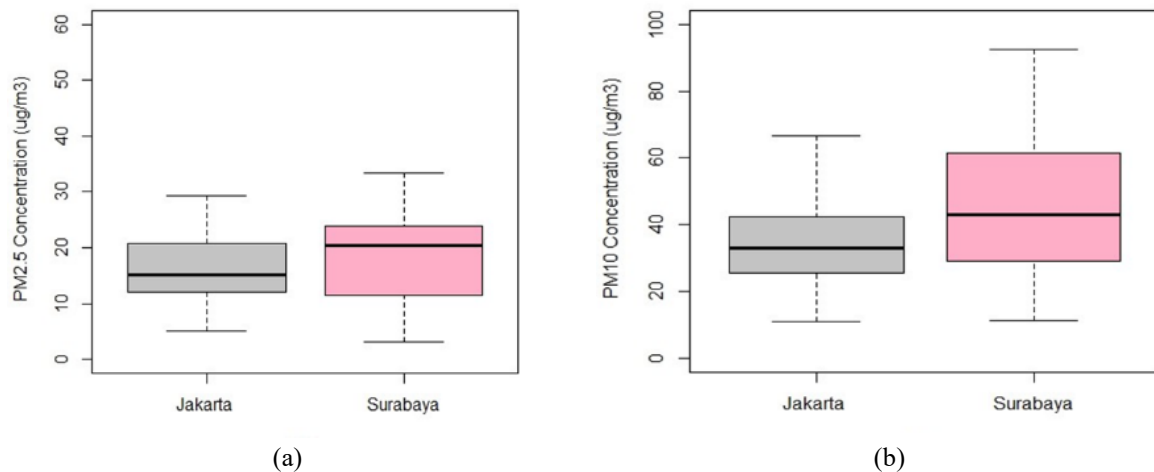


FIG. 2. Box and whisker plot of $PM_{2.5}$ (a), and PM_{10} (b) concentrations of Jakarta and Surabaya 2019 - mid 2020

The box and whisker plot for the $PM_{2.5}$ Black Carbon (BC) measurements at the Jakarta and Surabaya cities are presented in Fig.3. BC is generally emitted into the atmosphere due to incomplete combustion. In this study, BC measurement was conducted using reflectance method. To avoid inaccuracies in reflectance and absorbance for large particle sizes, the BC measurements were only done for fine APM. Black carbon is a form of impurity from carbon that results from incomplete combustion of fossil fuels or biomass burning. Black carbon (BC) is a major contributor to the fine particulate matter in the atmosphere and responsible for the light absorption by particles [20]. The results of BC concentration are presented in Table 1. The average BC concentrations for Jakarta and Surabaya were 3.39 and 3.76 $\mu\text{g}/\text{m}^3$, respectively. Similar to the PM concentration, BC concentrations in Surabaya were measured higher than those measured in Jakarta. The first site in Surabaya was close to the industrial areas within 10 km radii with the seasonal reoccurring open burning practice. The second site is close to the port which emits substantial BC emission from the shipping call activity. In addition to the local traffic, the above-mentioned emission sources were potentially contributing to the higher levels of BC measured at the two sites.

The BC concentration of Surabaya site during this study period was the highest compared to other cities in Java Island such as Jakarta, Bandung, Semarang, Tangerang and Yogyakarta obtained from the previous study by Santoso et al. [10]. BC concentration in Jakarta during 2019-2020 tends to increase compared to the 2010-2017 period. Notably, BC concentrations measured at the two cities in this study were far higher compared to the other cities inside and outside Java Island [10]. The BC sources in Jakarta were potentially contributed to motor vehicles as the sampling site in Jakarta is located near one of the busiest arterial roads in the town [21].

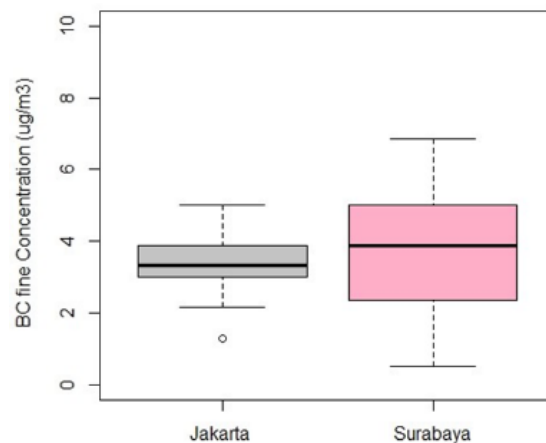


FIG. 3. Box and whisker plot of $PM_{2.5}$ Black Carbon (BC) concentrations of Jakarta and Surabaya 2019 - mid 2020

3.2. $PM_{2.5}$ elemental concentration

Elemental characterization of $PM_{2.5}$ was done using IBA facilities at ANSTO, Australia. IBA techniques of PIXE, PIGE, RBS and PESA were used simultaneously to measure elemental concentrations of more than 20 different elements found in fine particle ($PM_{2.5}$) air pollution, which can be used for characterisation and fingerprint of pollution sources in order to estimate their contributions to the total mass loading. These methods have been demonstrated world-wide for aerosol pollution studies [15,16,22]. These data together with their errors and minimum detectable limits were used in Positive Matrix Factorisation (PMF) analyses to quantitatively determine source fingerprints and their contributions to the total measured fine mass [15,22]. In this study, IBA techniques was applied to fine particulate samples from Surabaya and Jakarta for elemental characterization. The elemental composition of $PM_{2.5}$ of Jakarta and Surabaya are presented in Table 2. The results obtained indicated that the chemical composition of fine particulates of $PM_{2.5}$ in Jakarta was dominated by Sulphur, followed by Aluminium and Silicon as crustal elements which may come from the road dust. The major composition of $PM_{2.5}$ in Surabaya, either from site 1 or site 2, is also Sulphur. The use of fuels containing high concentration of S is one of the main sources of S emissions which may be contributed by industry and shipping. There are several types of diesel fuel or biodiesel that have a sulphur content ranging from 300 to 2500 parts per million (ppm) [23] but

MFO used for shipping certainly has higher range above 10,000 ppm. This is one of the reasons of the high level of S in PM_{2.5} in urban areas of Indonesia. The potassium (K) concentrations in Surabaya were slightly higher than those characterized in Jakarta. Potassium is a key element /finger print of biomass burning. Besides transportation and industry, the higher in K was probably due to open burning activities that are still being carried out in Java including in Surabaya, especially to reduce the volume of household waste or crop residue [24]. Parts of elemental concentrations in PM_{2.5} Surabaya are higher than in Jakarta Al, Si, Zn, Fe, Pb. There are several industrial activities in the vicinity of Surabaya which are located much closer to the 1st sampling site. While in Jakarta, the industrial activities are mainly located more than 15 km away from the sampling site in central Jakarta.

TABLE 2. Elemental concentration of PM_{2.5} in Surabaya and Jakarta 2019 – mid 2020 using IBA techniques

	Center Jakarta $\mu\text{g}/\text{m}^3$	East Surabaya (1) $\mu\text{g}/\text{m}^3$	West Surabaya (2) $\mu\text{g}/\text{m}^3$
Na	0.30 ± 0.09	0.39 ± 0.14	0.35 ± 0.17
Al	0.67 ± 0.05	0.72 ± 0.10	0.72 ± 0.14
Si	0.92 ± 0.14	1.05 ± 0.22	1.07 ± 0.37
S	1.74 ± 0.67	2.09 ± 0.59	1.64 ± 0.55
Cl	0.23 ± 0.02	0.23 ± 0.03	0.21 ± 0.01
K	0.39 ± 0.12	0.56 ± 0.23	0.47 ± 0.32
Ca	0.20 ± 0.10	0.16 ± 0.07	0.27 ± 0.25
Ti	0.010 ± 0.005	0.016 ± 0.007	0.011 ± 0.008
V	0.00003 ± 0.0002	0.00006 ± 0.0002	0.0004 ± 0.0008
Cr	0.0033 ± 0.0012	0.0046 ± 0.003	0.0031 ± 0.0010
Mn	0.009 ± 0.008	0.019 ± 0.02	0.008 ± 0.005
Fe	0.18 ± 0.07	0.33 ± 0.21	0.22 ± 0.15
Ni	0.0012 ± 0.0006	0.0014 ± 0.001	0.0011 ± 0.0006
Cu	0.017 ± 0.016	0.010 ± 0.005	0.006 ± 0.003
Zn	0.14 ± 0.07	0.61 ± 0.52	0.15 ± 0.19
As	0.0018 ± 0.0015	0.0026 ± 0.003	0.0018 ± 0.0018
Br	0.010 ± 0.010	0.012 ± 0.006	0.006 ± 0.004
Pb	0.044 ± 0.07	0.34 ± 0.38	0.035 ± 0.06

The time series of elemental (S, K, Cu, Mn, Fe, Zn, Pb) concentrations of PM_{2.5} in Jakarta and Surabaya are presented in Fig. 4. It can be seen that S concentrations of Jakarta and Surabaya were in the similar range as well as in between 1st and 2nd sites in Surabaya. This showed strong signal of fuel combustion of diesel fuel from the industry or other activities. Sulphur concentration in Jakarta was measured higher compared to the previous study in 2008-2009 (1.177 $\mu\text{g}/\text{m}^3$) [11]. Notably, sulphur concentrations measured at the two sites were far higher compared to the other cities in Indonesia [10]. Potassium concentrations in Surabaya 1 were higher than those measured in Surabaya 2 and Jakarta which may be sourced from open burning of domestic waste. The concentrations of Fe, Zn, Pb and Mn in Surabaya 1 were higher than those measured in Surabaya 2 and Jakarta due to the close location of the smelter industrial area. As can be seen in Fig. 4, the Surabaya 1 Zn and Pb concentration were peaking at several days during the time period observed. Pb concentrations measured in Surabaya 2 and Jakarta seems to have similar range with the concentrations of Zn, Pb and Mn, except for Fe. Cu concentration measured in Jakarta was 2-3 times higher than Surabaya describing the possibility of the sources from non-ferrous industries. The Cu concentrations of PM_{2.5} in Jakarta were lower than that measured in Iran (up to 167 ng/m^3) or even with the measurement conducted in South Korea (35.7 ng/m^3). However, the levels were measured higher than those measured in Switzerland and Netherland (<10 ng/m^3) [25-27].

Comparison of heavy metal elements Zn, Fe, Mn, and Pb of PM_{2.5} concentration in Jakarta to Surabaya are presented in Fig. 5. The elemental characteristics of Zn Mn, Fe and Pb are known as tracers of steel industry, while Cu and Pb are for the metal industry, and Pb is for acid batteries. The concentrations of Fe, Zn, Pb at Surabaya were significantly higher than those measured in Jakarta. Compared to the steel industry area in the Yangtze River Delta region China, the Fe and Zn concentrations at Surabaya were still lower, except for Pb where the concentration was measured nearly double [28]. The high concentrations of Pb in Surabaya were likely come from Lead Battery smelter in Surabaya's vicinity. Similar results with high level of Mn, Ni, As, Cd, Pb were also found for a scrap iron and steel smelting industry in Nigeria and the results also showed that the source

contribution from metallurgical production reached 6% of the total mass of $PM_{2.5}$ [29]. The IBA techniques were successfully applied to characterize the elemental compositions of $PM_{2.5}$ in Jakarta as well as in Surabaya and showed different characteristics of air quality of each city. The results obtained can then be used to determine the source apportionment in Jakarta and Surabaya to formulate appropriate strategies and policies for PM reduction in each city.

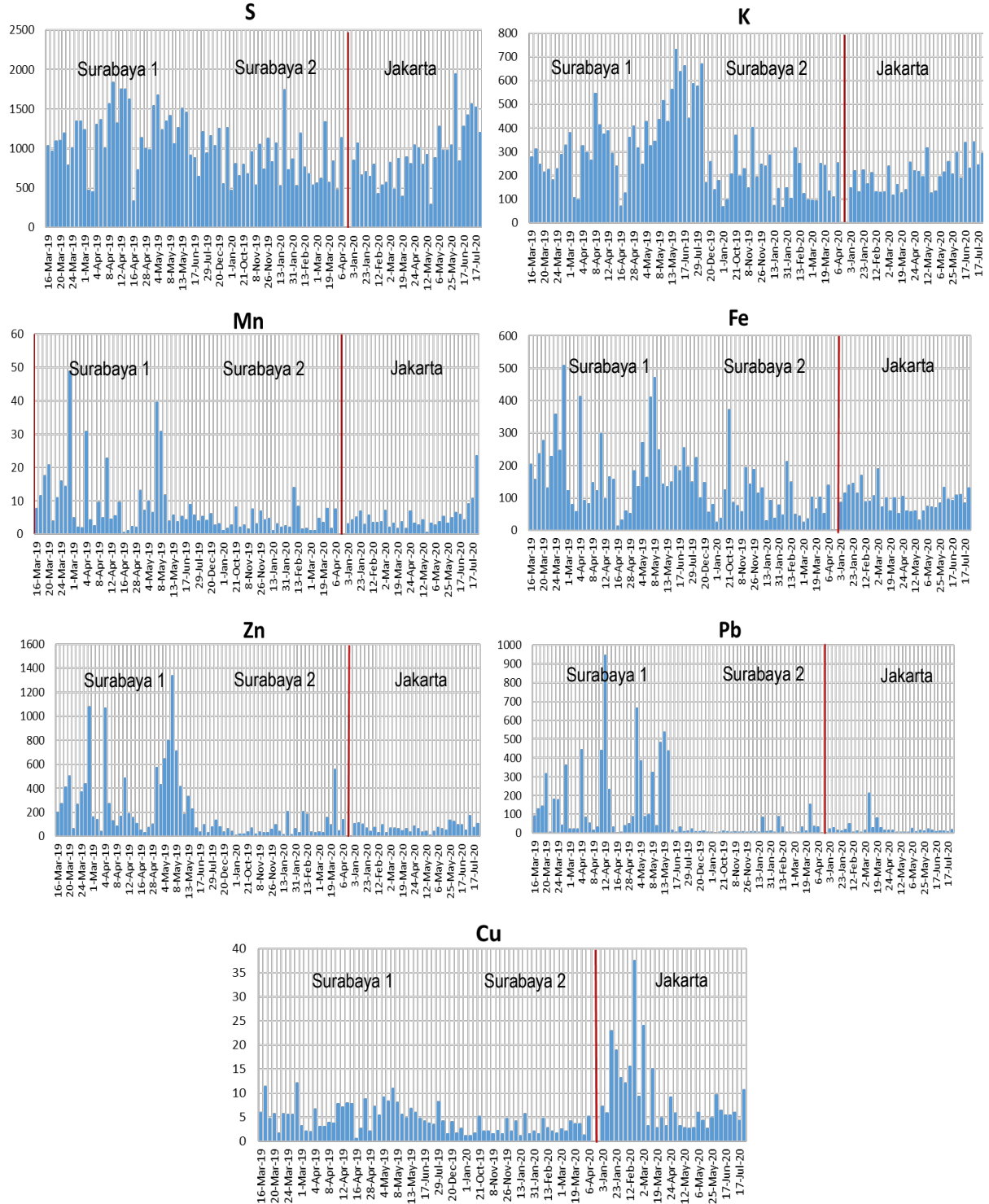


FIG. 4. Time series of S, K, Mn, Fe, Zn, Pb and Cu concentrations of $PM_{2.5}$ (in ng/m^3) of Jakarta and Surabaya 2019-mid 2020

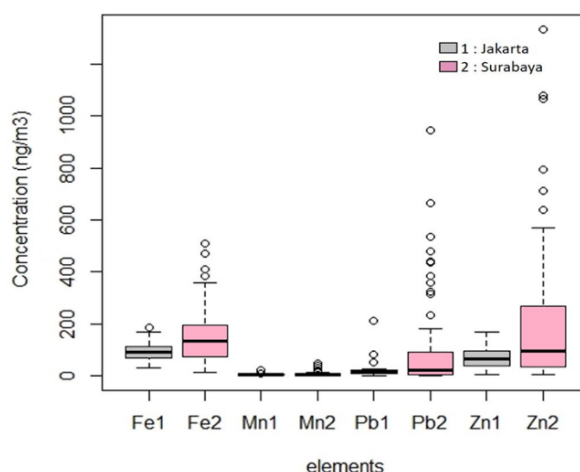


FIG. 5. Box and whisker plot of Fe, Mn, Fe, Pb and Zn concentrations of $PM_{2.5}$ Jakarta and Surabaya 2019 – mid 2020

4. CONCLUSIONS

The results of $PM_{2.5}$ elemental composition in the ambient air using IBA technique analysis was able to provide good and comprehensive results in term of urban PM air pollution characterization. Three sampling locations from Jakarta and Surabaya gave different results according to the characteristics of the area especially the nearby emission sources. The multi-element identification from the IBA analysis was also able to provide an overview of the contribution of emissions from industry to air quality in the surrounding environment. Heavy metal concentrations (Fe, Zn, Mn, Pb) in East Surabaya (1) were higher than those measured in West Surabaya (2) and Jakarta, while higher Cu was found in Jakarta compared to the 2 sites in Surabaya. These results indicated that the concentration of heavy metal in East Surabaya was likely to be correlated with industrial activities in the vicinity. Although the concentration of heavy metals was still below the NAAQS, the problem of its pollution cannot be ignored because it is very harmful to human health even in a low concentration. These results can be used as an early warning to formulate appropriate strategies and policies so that greater losses can be avoided in the two concerned cities.

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