Characterization of Fine Particulate Matters Collected in the Vicinity of Coal-fired Power Plants in Java using ED-XRF

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Abstract. The number of coal-fired power plant in Java has been increased because of the government policy to supply the electricity in Indonesia to meet the electricity needs. This mega project is assumed to have an impact on the environmental and human health. In this study, the assessment of the air quality near the coal fired power plant has been carried out through characterization of the fine particulate matters. The particulate matters PM$_{2.5}$ were collected in four sites in the vicinity of coal-fired power plant in Banten, West Java and Central Java, Indonesia. The elemental concentrations in PM$_{2.5}$ such as Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br and Pb have been identified using energy dispersive X-ray Fluorescence-EDXRF. The validation of XRF measurement was applied using standard reference material NIST SRM 2783 Air Particulate on Filter Media. The elemental concentration of PM$_{2.5}$ especially for the toxic elements as well as the mass and black carbon were compared between four sites. For some sites, the level of elemental concentrations was lower than other industrial sites in Java, due to the sampling was carried out before the commissioning/normal operation. Further and periodical monitoring will be needed to obtain a better comprehensive result to assess the impact of coal power plant to the surrounding environment.

1. Introduction
The energy consumption in Indonesia especially in Java has risen due to the rapid growth of industrial and economic activities. Perusahaan Listrik Negara (PLN), Indonesia’s state-owned electricity company has a plan to build an additional power plant; the energy industry in Indonesia is currently dominated by coal-based power plants to meet this need. Indonesia government plan to develop an additional 35,000 MW of new power capacity [1,2]. The program comprises more than 40 power plant mostly coal fired in Java and Bali. Coal fired power plant covers about 53% of total electricity generation capacity in Indonesia, and it is estimated to be increased up to 66% [3]. Completion of the 35,000-Megawatt (MW) plant program is expected to decline from the original target in 2026. During 2015 to 2019, PLN had completed the construction of a 14,792.5 MW power plant from the planned 35,000 MW [4].

This huge and rapid development of coal fired power plant should be taken into consideration related to the environmental impact. The process of coal combustion from coal fired power plant is significant source of pollutant since it produces massive amounts of CO$_x$, SO$_x$, NO$_x$ and particulate matter [3,5]. The local coal fired power plant in Indonesia use low rank coal subbituminous coal, however its sulfur content is often low, sometimes less than 1 percent by weight. These emissions increase the ambient concentration of PM less than 2.5 microns in diameter (PM$_{2.5}$) over hundreds to
thousands of kilometers downwind of the plants [6,7]. Exposure to PM$_{2.5}$ has been associated with cardiopulmonary diseases, lung cancer and numerous other respiratory illnesses [8]. The concentration of elements such as mercury, lead, cadmium, arsenic, chromium, and other hazardous elements should be monitored. The high concentration of heavy metals in the fine particulate matter could be harmful to human health.

Nuclear analysis techniques such as ED-XRF are analytical techniques that are very suitable for characterizing airborne particulate samples. ED-XRF has been applied to characterization of airborne particulate samples in various pollutant source identification research. It has high capability, simultaneous measurement, multi-element, high sensitivity, short analysis time and its non-destructive which makes it ideal for this type of work [9,10]. Air particulate samples in the media filters have a mass that is very small up to 100 µg and the trace elements in it are also in trace level, making it difficult to analyze using other conventional methods. ED-XRF is an effective tool and allows the characterization of multi-element compositions and can produce large data sets for source identification [11].

There are limited numbers of publications related assessment of environmental impact from coal-fired power plant in Indonesia, with respect to the chemical composition of the fine particulate matter. Previous study has been published related the chemical composition on feed coal and fine particulate matter in Cilacap area[12]. In this study, characterization of fine particulate matter in the vicinity of a coal-fired plant in Java island covering Banten, West Java and Central Java was carried out. The objectives of this study are to provide characteristic of mass, black carbon and elemental concentrations of fine particulate matter emitted from coal-fired power plant.

2. Method

2.1. Sampling

Sampling was conducted at 4 sampling sites near the coal fired power plant Suralaya, Labuan, Cilacap and Tanjung Jati in Java island covering Banten, West Java, and Central Java provinces. The sampling locations for these sampling sites are shown in Figure 1. Airborne particulate matter was collected using Gent stacked filter unit dichotomous sampler that capable for collecting particulate matter in the PM$_{2.5,10}$ (coarse) fraction and PM$_{2.5}$ size (fine) fraction [13]. Sampling was collected on flow rate of 15-18 L/min.

2.2. PM, BC and elemental analysis

The particulate matter mass of each fine and coarse fractions was determined by gravimetric using a microbalance. The filters were stored for 24 hours prior to weighing in controlled environmental condition room. Mass concentrations of PM$_{2.5}$ and PM$_{2.5,10}$ (µg.m$^{-3}$) were obtained by dividing the gravimetric mass by the volume of air that passed through the filter. The concentration of black carbon (BC) in the samples was determined by reflectance measurement using an EEL Smoke Stain Reflectometer model 43D. Secondary standard of known reflectance were used to calibrate the reflectometer. Elemental analysis was performed using an energy dispersive Xray fluorescence ED-XRF Epsilon 5 (Panalytical, Ltd) by optimizing the five existing secondary targets (Al, CaF$_2$, Fe, Ge, and Zr) [11].
2.3. Reconstructed Mass (RCM)

The measurement of so many different elemental species allowed estimates of significant fine particle components like sea salt, ammonium sulphate, soil and smoke. We could define the RCM as a sum from sea salt=Na+Cl, ammonium sulfate=4.125*S, soil=2.20*Al+2.49*Si+1.63*Ca+1.94*Ti+2.42*Fe and smoke=K-0.6*Fe, and BC [7].

3. Results and Discussions

PM$_{2.5}$, PM$_{10}$ and black carbon concentrations for each sampling site are summarized in Table 1 and shown in Figure 2. Twenty-four hours average concentration of PM$_{2.5}$ at four sampling sites in the vicinity of coal power plant in Cilacap, Labuan, Suralaya and Tanjung Jati were 8.24, 17.06, 14.65 and 14.46 $\mu$g.m$^{-3}$, respectively. The highest concentration of PM$_{2.5}$ was found in Labuan and the lowest was in Cilacap. Even though, this PM$_{2.5}$ concentration was still below the 24 hours of Indonesian national air ambient standard (65 $\mu$g.m$^{-3}$), but by assuming that these weeks of sampling can represent the annual mean of PM$_{2.5}$ level, the PM$_{2.5}$ concentrations in sampling site Labuan, Suralaya dan Tanjung Jati would exceed the Indonesian national ambient air quality standard of an annual mean PM$_{2.5}$ value of 15 $\mu$g.m$^{-3}$ (Government Regulation, 1999).
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Table 1. The sampling data, mean mass and BC concentrations (µg.m\(^{-3}\))

| Sampling site (n) | PM\(_{2.5}\)  | PM\(_{10}\)  | BC          |
|------------------|-------------|-------------|------------|
| Cilacap (2) [12] | 8.24±0.02   | 44.50±18.89 | 1.35±0.14  |
| Labuan (3)       | 17.06±2.32  | 28.89±4.79  | 2.11±0.26  |
| Suralaya (3)     | 14.65±5.93  | 37.43±5.44  | 2.65±0.73  |
| Tanjung Jati (3) | 14.46±5.11  | 19.37±6.51  | 2.52±0.71  |

In Cilacap sampling site, the samples were collected before the coal fired power plant was not operating and still under construction. Therefore, the PM\(_{2.5}\) concentration was low due to the emission of coal power plant is not contributed significantly. On the contrary, the PM\(_{10}\) concentration in Cilacap site was higher compared to other sites since the pollutant source such as road dust or crustal element were higher during the construction activity. The ratio of PM\(_{2.5}\)/PM\(_{10}\) in Cilacap was 18.5\% which indicated that the coarse particulate was dominant compared to fine particulate. Compared to other PM\(_{2.5}\) data in several cities in Java island, PM\(_{2.5}\) concentrations in Labuan, Suralaya and Tanjung Jati were in similar level with PM\(_{2.5}\) reported by Santoso et al, 2020 for seven urban cities in Java in average of 14-18 µg.m\(^{-3}\) except for Yogyakarta [14]. In this study, we are unable to identify the high concentration or increasing of concentration of PM\(_{2.5}\), PM\(_{10}\) or BC from time to time, due to the short-term monitoring.

Black carbon, BC is commonly emitted to the atmosphere due to incomplete combustion, the major source of BC are transport related emissions such as diesel emission and long-range transport from fossil fuel related sources such as coal fired power plant emission, and biomass burning could be other substantial sources of BC [15]. The average BC fine contributed 12-19\% of the PM\(_{2.5}\) mass concentration in the four sampling sites. These values do not differ significantly with the values reported in industrial area in Serpong area in 2008 (13-26\%) which used the same sampling method and analytical procedure [16]. The lowest BC was found in Cilacap, with similar reason for PM\(_{2.5}\). It is should be noted that the activity of coal combustion due to the operation of coal fired power plant in Cilacap site was not begun yet at the sampling time. This data could be used as preliminary data or background data before the emission of coal combustion will take place.

Table 2. The elemental concentrations in PM\(_{2.5}\) in four sampling sites

| Element (ng.m\(^{-3}\)) | Cilacap [9] | Labuan | Suralaya | Tanjung Jati |
|-------------------------|-------------|--------|----------|-------------|
| Na                      | 218.33      | 2.63   | 24.41    | 105.35      |
| Mg                      | 112.15      | 16.27  | 19.15    | 19.21       |
| Al                      | 137.19      | 16.21  | 72.47    | 40.35       |
| Si                      | 160.21      | 17.87  | 128.45   | 50.84       |
| S                       | 172.35      | 392.24 | 528.74   | 589.16      |
| Cl                      | 383.18      | 5.11   | 7.27     | 5.69        |
| K                       | 174.11      | 168.51 | 188.99   | 174.19      |
| Ca                      | 87.35       | 14.42  | 57.61    | 42.02       |
| Ti                      | 8.49        | 3.64   | 5.72     | 4.59        |
| Cr                      | 1.68        | 1.94   | 2.93     | 5.29        |
| Mn                      | 4.96        | 4.33   | 11.90    | 5.17        |
| Fe                      | 269.47      | 187.36 | 302.04   | 259.18      |
| Ni                      | 1.35        | 1.23   | 2.45     | 1.58        |
| Cu                      | 0.07        | 1.42   | 1.24     | 1.04        |
| Zn                      | 8.28        | 21.25  | 52.15    | 12.08       |
| Br                      | 11.13       | 3.56   | 4.80     | 6.40        |
| Pb                      | 3.19        | 8.27   | 6.22     | 4.57        |
Elemental concentration of fine particulate matters collected in four sampling sites are summarized in Table 2. Elements such as Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Br and Pb were identified. Elemental concentration results in fine particulate matter, PM$_{2.5}$, shown that sulfur has highest concentrations in three sites of Labuan, Suralaya and Tanjung Jati with average of 392.24, 528.74 and 589.16 ng.m$^{-3}$, respectively. Sulfur in airborne particulate matter is generally present because of the atmospheric conversion of SO$_2$ to sulfate particulate phase through homogeneous processes. Hanoi also reported high sulfur in PM$_{2.5}$ with average value of $3810 \pm 2700$ µg.m$^{-3}$ due to the emission of coal power plant [7]. Compared to Hanoi study, the sulfur emission in the three sampling sites were still lower, this could be happened due to the short monitoring and limited number of samples. Even though the emission of sulfur is lower than other study, it is worth noted that the major emission of the coal power plant identified was sulfur. Long monitoring to identify the emitted pollutant from coal power plant emission should be considered to monitoring the impact to the air quality and surroundings. In Cilacap sites, the major elements were the crustal elements such as Al, Si, Ca, and Fe which indicated the road dust/soil sources are dominant. Heavy metals such as Cr, Ni, Cu, Zn and Pb were in low concentrations compared to other industrial sites in Indonesia reported by Santoso et al [16].

The calculation of RCM, the average of measured PM$_{2.5}$ and its ratio are resumed in Table 3. It can be seen that the average RCM for fine particulates reaches 25.7 to 51.0%, which means that reconstruction represents maximum only 51% of the mass of the fine particulate and the remaining 49% cannot be reconstructed due to unmeasured of organic matter such as nitrates and organic carbon. The remaining missing mass more than 49% was mainly nitrates and organic matters. Other study reported that organic matter contributes about 28±11% of mass and nitrates are between 5 to 15% of the mass [7].

![Figure 3](image-url)

*Figure 3.* Each component of RCM in each sampling site
Table 3. Reconstructed mass vs measured PM$_{2.5}$

| Sites      | RCM (ng.m$^{-3}$) | Measured PM$_{2.5}$ (ng.m$^{-3}$) | Ratio RCM to measured PM$_{2.5}$ |
|------------|-------------------|-----------------------------------|---------------------------------|
| Cilacap    | 4196.18           | 8236.11                           | 51.0%                           |
| Labuan     | 4355.87           | 17056.55                          | 25.7%                           |
| Suralaya   | 6472.50           | 16903.50                          | 40.8%                           |
| Tanjung Jati | 6009.19         | 14459.66                          | 42.6%                           |

By the calculation of RCM as a sum of sea salt, ammonium sulphate, soil, smoke and BC, the percentage of RCM components in the calculated RCM values for fine particulate matter samples are shown in Figure 3. Figure 3 showed that the average RCM of PM$_{2.5}$ mass at Labuan, Suralaya and Tanjung Jati was composed of 42.49% black carbon, 33.9-40.3% sulphate, 13-20% soil, 0.1-2.0% seasalt and 0.1-1.3% smoke. While for Cilacap, the RCM was composed of 32.4% black carbon, 17.1% sulphate, 34.5% soil, 15.3% seasalt and 0.7% smoke. Black carbon and sulphate give the largest contribution to fine particulate RCM with a contribution of a total more than 79% for three sites Labuan, Suralaya and Tanjung Jati except for Cilacap, the major contribution of BC and soil was dominant.

4. CONCLUSION

This study presented the PM$_{2.5}$, BC and elemental concentration from fine particulate matter samples collected in the vicinity of four coal power plant in Java island. The 24 hours of mass concentration of PM$_{2.5}$ were still below the threshold value of twenty-four hours of PM$_{2.5}$ concentration of Indonesia. Elemental analysis of PM$_{2.5}$ revealed that sulfur has a highest concentration for Labuan, Suralaya and Tanjung Jati sites, while in Cilacap was dominated by crustal matter. The major component of RCM in PM$_{2.5}$ in those three sites were black carbon and sulphate which dominates more than 79%, while in Cilacap were soil and with average of 34.5%. For some sites, the level of elemental concentrations was lower than other industrial sites in Java, due to the sampling was carried out before the commissioning/normal operation. Further and periodical monitoring will be needed to obtain a better comprehensive result to assess the impact of coal power plant to the surrounding environment.

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