Identification of Potential Source Location of Sulfur at Urban Area Bandung using Conditional Probability Function (CPF)

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Abstract. Bandung area is known as the center of economic growth in West Java. Bandung is also surrounding by mountains, therefore the morphological form of its territory is like a giant bowl. The previous study showed that the concentration of Sulfur in air particulate matter from Bandung is high. The objective of this study is to examine the use of conditional probability function (CPF) to identify directions of Sulfur sources using data collected from Bandung. CPF method is an advanced tool that analyzes pollutant source contributions in relation to the winds at the site and has been applied to find source directions. Data collection were done by air particulate sampling using GENT Sampler for 24-hours, once a week in Bandung during 2012-2014. EDXRF technique was applied for Sulfur analysis. The CPF values for Sulfur presented in polar plots, for fine and coarse particles. From this polar plot, the contributions of Sulfur in fine particles were coming from all sectors in Bandung whereas, in coarse particles, Sulfur dominated from the northern sector of Bandung.

Keywords : conditional probability function, sulfur, wind, particles.

1. Introduction
Sulfur (S) is the tenth most abundant element in the universe. The atmosphere plays a complex and critical role in the global cycle of Sulfur. The presence of sulfur in the atmosphere is a few days with wide variations dependent on meteorological and other factors [1]. The atmosphere is also the recipient of the majority of anthropogenic sulfur, which considered as one of the air pollution sources due to its possibility to oxidized and form sulfur dioxide (SO$_2$). The compound can be transformed into sulfuric acid and mixed with water vapor in the atmosphere to form acid rain, which is one of the serious environmental problems of transboundary nature [2]. Furthermore, SO$_2$ emissions that lead to its high concentrations in the air could also form other sulfur oxides (SO$_x$) that can react with other compounds in the atmosphere to form small particles. These particles contribute to particulate matter pollution. Airborne particulate matter (APM) consists mostly of organic matter, metals, water-soluble ions and mineral oxides [3]. This particle may penetrate deeply into sensitive parts of the lungs and cause additional health problems.

Air pollution studies in Indonesia have been conducted for health effect examination as well as source apportionment using multivariate receptor models such as positive matrix factorization (PMF). The technique was developed to provide a flexible modeling approach that can effectively use the information contained in the data [4,5]. The process of identification and apportionment of pollutants to their sources is an important step in air quality management. Source apportionment studies in several urban areas Indonesia have been reported. Santoso et al. (2008) presented the composition and source apportionment of fine and coarse particle samples collected in Bandung and Lembang, Indonesia.
between 2002 and 2004 [6]. This study also showed that the concentration of Sulfur in air particulate matter from Bandung during that period was high. Beside sulfur, Lestari and Muliadi (2009) also reported that concentration of sulfate ($\text{SO}_4^{2-}$) in fine and coarse particle from Bandung during 2001 – 2007 also high [7].

However, understanding the nature and sources of airborne particulate matter where there are limited or few impacts from a local source is useful. The conditional probability function (CPF) method is an advanced tool that analyzes pollutant source contributions in relation to the winds at the site and has been applied to find source directions [8]. The conditional probability function (CPF) has been successfully used to analyze point source impacts and to identify source direction in multiple studies, and most of it used in conjunction with PMF [9-12]. CPF plots constructed from the PMF modeling results by Lee and Hopke (2006) clearly showed factor specific wind directions consistent with the locations of corresponding industrial point sources identified from emission inventories [11]. In Indonesia, only limited publications related to CPF utilization to identify the local source direction of air pollution.

In this study, the Conditional Probability Function (CPF) method was utilized to identify the local source directions of Sulfur in Bandung area, which were known to be high from the previous study by Santoso et al. (2008). Air particulate matter (APM) sampling in Bandung area was conducted during 2012-2014, and the Sulfur concentration in APM during the time will be determined and compared with the previous study. Bandung is located at latitude 6°91’ S, longitude 107°60’ E and 630 m above sea level. It is the provincial capital of West Java and is categorized as an industrial city with a population which is more than 2.6 million inhabitants in an area of approximately 167.67 km$^2$ [6]. Bandung area is known as the center of economic growth in West Java. Many small-scale factories are also located around the city. Bandung is also surrounded by mountains, therefore the morphological form of its territory is like a giant bowl. Hourly measurements of APM mass and Sulfur along with CPF are used to identify the directionality of sources contributing to observed pollutant concentrations in urban area Bandung from 2012-2014.

2. Experimental methods and data analysis

2.1. Air particulate sampling

The sampling was located at the rooftop of Environmental Agency of West Java, Naripan Street, Bandung City (latitude: 6°92’ S and longitude: 107°61’ E) as shown in Figure 1. The sampling site represents an urban mixed site (commercial, offices and traffic areas). The samples were collected with a Gent stacked filter unit sampler capable of collecting particulate matter in the PM$_{2.5-10}$ and PM$_{2.5}$ size fractions [13]. The fine and coarse fraction samples are collected on a 0.4 μm pore size filter and coated 8 μm pore size filter, respectively. Samples were collected for 24 h once a week during 2014, at a flow rate of 15–18 L/min.

![Figure 1. Maps of Bandung, West Java and sampling location.](image-url)
2.2. Sample Analysis

After sampling, all exposed filters were stored in plastic petri dishes with diameter 47mm for 24 h prior to weighing in controlled environmental condition at 18–23°C and 40–60 % humidity. The particulate matter mass of each of the coarse and fine fractions was determined by gravimetric method. Mass concentrations were obtained by dividing the gravimetric mass by the volume of air that passed through the filter to obtain the concentration of PM$_{2.5}$ and PM$_{2.5-10}$ (µg/m$^3$). Determination of Sulfur was carried out by X-Ray Fluorescence Spectrometry (PANalytical Epsilon 5 XRF Spectrometer). The method of the validation was performed by measuring Standard Reference Material (SRM) NIST 2783 Air Particulate on Filter Media.

2.3. Wind rose diagram

The wind rose is graphically presenting the wind conditions, direction and speed, over a period of time at a specific location. Wind speed (WS) and wind direction (WD) data are needed for creating wind rose diagram. Data values for WS and WD were obtained from the nearby Airport. Software WRPLOT was utilized for generating the wind rose diagram. The collected wind data is then sorted by wind direction so that the percentage of time that the wind was blowing from each direction can be determined. The wind direction data is sorted into twelve equal arc segments, 30° each segment, in preparation for plotting a circular graph in which the radius of each of the twelve segments represents the percentage of time that the wind blew from each of the twelve 30° direction segments.

2.4. Conditional Probability Function (CPF)

The CPF method was developed to assess the impacts of sources from various wind directions using the source contributions coupled with the surface wind direction data [8]. The CPF predicts the probability that a given source contribution from a given wind direction will exceed a predetermined threshold criterion [14]. The same daily contribution was assigned to each hour of a given day to match to the hourly wind data. CPF is a simple model, but it provides a powerful approach for identifying the directionality of point sources and/or source areas whose emissions significantly impact the receptor site [15]. The CPF is defined with the following equation:

$$CPF_{\Delta\theta} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}}$$  \hspace{1cm} (1)

where $m_{\Delta\theta}$ is the number of occurrence from wind sector $\Delta\theta$ that exceeded the threshold criterion and $n_{\Delta\theta}$ is the total number of data from the same wind sector. In this study, 12 sectors ($\Delta\theta = 30^\circ$) were used. Calm winds (<1ms$^{-1}$) periods were excluded from the calculation due to the isotropic behavior of the wind vane for such conditions. The threshold criterion was set at the upper 25 percent quartile of the fractional source contributions for each source. In urban areas, the sources, which are close to the receptor site, can be clearly identified by CPF [11].

3. Results and Discussion

Sampling site at Naripan, Bandung located in the center of Bandung City. The area represents an urban mixed site (industrial, commercial, tourism and traffic areas). Total of 210 samples which consist of fine and coarse particulate matter were collected on weekdays, once a week during 2012-2014. Determination of Sulfur in the fine particulate matter (FPM) and coarse particulate matter (CPM) was carried out using X-Ray Fluorescence (XRF) Spectrometer at Center for Applied Nuclear Science and Technology Laboratory Bandung. Quality control (QC) of the method was also applied by measuring SRM NIST NIST 2783 Air Particulate on Filter Media in the same conditions as the samples. Table 1 presents the analytical accuracy and precision, described as %recovery and % coefficient of variance (%CV). The value was acceptable, compared to AOAC International guidelines [16]. Figure 2 shows the periodic measurement of SRM for method validation.
The analysis results of mass concentration PM$_{2.5}$ and PM$_{10}$ and Sulfur concentration in FPM and CPM from Bandung during this study period were shown in Table 2. Most of the filters contain Sulfur. The sulfur in FPM and CPM are associated with the sources of pollution in Bandung. Natural sources of pollution that have significant effects on the city are the secondary aerosol, soil dust, road dust, lime dust, and volcanic dust. Anthropogenic sources consist of industry, biomass burning, and vehicular emissions [7]. Time variation of Sulfur concentration in FPM and CPM during 2012-2014 is shown in Figure 3. The significant increasing average occurred for FPM in 2014. That might be correlated with volcanic activity from the eruption of several mountains in Indonesia. Mount Sinabung in Karo land, North Sumatra, was erupted by spraying hot lava and pyroclastic into the atmosphere on January 21$^{st}$ and released a hot cloud on 3$^{rd}$ February 2014. Mount Kelud near the town of Kediri in East Java erupted on February 13. Bursts of volcanic dust clouds and other materials reach a height of several kilometers [18]. The volcanic ash from Kelud spreading through Java and Madura Island [19]. Fine particles can remain airborne for long periods and travel hundreds of miles. It can be seen from Table 2 that mass concentration of fine particles in 2014 is highest compare to 2012 and 2013 due to the traveling volcanic dust. The variation of PM and Sulfur concentration from 2012 – 2014 in Table 2 shows the same pattern, therefore the high sulfur concentration is related to the high PM concentration. The concentration of Sulfur in 2012 is higher than 2013, due to the increasing activity of Tangkuban Perahu Mountain. The status of the mountain was also increased to an alert level since August 23, 2012 [28].

### Table 2. The average of PM$_{2.5}$, PM$_{10}$ and Sulfur concentration in FPM and CPM Bandung during the period of study

| Year | PM$_{2.5}$ (µg/m$^3$) | PM$_{10}$ (µg/m$^3$) | S in FPM (ng/m$^3$) | S in CPM (ng/m$^3$) |
|------|----------------------|---------------------|---------------------|---------------------|
| 2012 | 17.28±7.02           | 44.33±14.79         | 745.69 ± 341.95     | 501.17 ± 254.58     |
| 2013 | 14.39±5.30           | 38.62±10.97         | 624.86 ± 270.33     | 427.73 ± 229.01     |
| 2014 | 20.48±7.83           | 45.50±14.42         | 1341.60 ± 789.58    | 743.68 ± 305.51     |
Comparison of Sulfur in FPM and CPM in Bandung with previous study in Bandung and with other countries in Asia was also presented in Table 3. The average Sulfur in FPM Bandung in 2012 and 2013 are lower than previous study (Bandung 2002-2004), but higher in 2014 due to the volcanic activity from other cities. The range of Sulfur in FPM from Serpong is generally higher than Bandung as a result of the high anthropogenic activities whereas in CPM is lower than Bandung. The source apportionment of FPM in Serpong showed that the contribution of Sulfur was from the formation of lead sulfate in old batteries. The emissions of diesel vehicles were also make contribution since Indonesia still has a high sulfur content in diesel fuel (~5 000 ppm) [6]. High values of Sulfur probably arises from multiple fuelled power plants that burn coal as their main fuel and residual oil. There are two power plants within 50 km of the sampling site in Serpong [20]. Sulfur in FPM and CPM in Ulanbataar, Mongolia was higher than Bandung due to the coal combustion. The emissions from coal combustion sources consist of two distinct source types, the first identified as a high–temperature combustion source originating from power station emissions in the western part of Ulaanbaatar and the other from coal combustion used for domestic heating during winter [21].

Sulfur in FPM from two sites (AQM and AEA) in Srilanka almost similar with this study. Sulfur mostly comes from motor vehicle particularly that is using diesel fuels [22]. In Thailand, both average of Sulfur in FPM and CPM were higher than Bandung. Traffic and biomass burning were contributed since the sampling area was in Bangkok. Industrial or coal–fired power plant as well as diesel vehicles burning high Sulfur fuel possibly also made contribution to this factor [23]. The concentration of Sulfur in FPM from Kuala Lumpur, Malaysia is higher than Bandung whereas Sulfur in CPM almost similar with CPM Bandung. High concentration of Sulfur in FPM from Kuala Lumpur are correlated with anthropogenic activity such as biomass burning, industry as well as exhaust from motor vehicles. It was reported that the city of Kuala Lumpur was among the Asian cities that were heavily affected by vehicle emissions [24]. The average Sulfur in FPM from Dhaka, Bangladesh is higher than Bandung 2012-2013 but lower than Bandung 2014 whereas in CPM is higher. Study by Begum et al (2010) was found that the pollution contributions from the brick kiln increased over the years. Because of the increasing economic activities mainly around the Dhaka city, there has been a significant population growth, which created extra demand on transportation and infrastructures. The building boom has led to establishment of additional brick kilns. Currently, over 1,000 brick kilns are operating around Dhaka, and primarily use coal as fuel [25].
### Table 3. The average and range of Sulfur in FPM and CPM in Bandung sites 2012-2014, comparison with previous studies and comparison with other studies.

| Year              | Fine Fraction (ng/m³) | Coarse fraction (ng/m³) |
|-------------------|-----------------------|-------------------------|
|                   | Sulfur (average)      | Sulfur (range)          | Sulfur (average) | Sulfur (range) |
| Bandung 2012      | 745.69 ± 341.95       | 25.68 – 1168.64         | 501.17 ± 254.58 | 49.57 – 1040.66 |
| Bandung 2013      | 624.86 ± 270.33       | 145.97 – 1100.41        | 427.73 ± 229.01 | 115.53 – 1329.38 |
| Bandung 2014      | 1341.60 ± 789.58      | 264.38 – 2918.90        | 743.68 ± 305.51 | 278.82 – 1212.15 |
| Bandung 2002-2004 | 1049.20 ± 684.30      | -                       | -               | -               |
| Serpong (Setu)    | 681–1409              | 456–746                 |                 |                 |
| Serpong (EMC)     | 892–1533              | 146–697                 |                 |                 |
| Serpong (BSD)     | 687–1362              | 113–674                 |                 |                 |
| Ulaanbaatar, Mongolia | 1969±3978            | 125 – 40079             | 1458±1659       | 129 – 13831     |
| Colombo,Sri Lanka | 709.3 ± 511.4         | 585.8 – 3583.7          |                 |                 |
| Colombo,Sri Lanka | 484.7 ± 304.8         | 436.4 – 1990.9          |                 |                 |
| Thailand (Bangkok) | 1546 ± 1218           | 946 ± 1002              |                 |                 |
| Malaysia          | 1935 ± 949            | 479 ± 288               |                 |                 |
| Dhaka, Bangladesh | 1200 ± 793            | 961 ± 839               |                 |                 |

*This study

Bandung has a tropical weather with average temperature of 23°C and rainfall intensity of 120 mm h⁻¹ [7]. Identification of the local source directions of Sulfur in Bandung area using CPF needs metrological data particularly wind speed and wind direction. In this study, the metrological data during 2012-2014 were obtained from nearby airport. Wind rose and wind class frequency distribution were generated using WRPLOT software. Figure 4, 6 and 8 presents wind rose that shows characteristics of wind speed and direction per year in Bandung during the study. Wind rose shows that the frequencies of wind directions and are a simple and popular way to represent local transport information [17]. The major wind blowing in 2012 was from east with majority speed, average about 2.10 – 5.70 m/s and from west with majority speed, ranged from 3.60 – 5.70 m/s. Wind direction in 2013-2014 mostly comes from west and north west direction, with speed average about 3-5 m/s. Wind direction frequency from east increases in 2014 than 2013, although it was not dominant compared to 2012. In general, Indonesia has two seasons which are dry season (occurs during April–October) and wet season (occurs during October–April). Major wind blows during dry season from east direction while during wet season major wind blows from west direction [7].

![Figure 4. Wind rose of 2012.](image1)

![Figure 5. Wind class frequency distribution.](image2)
A CPF method is needed to find the directions of high values of estimated source contributions that are likely to be related to the directions of sources. Sources are likely to be located in directions that have high CPF values. CPF was applied to fine and coarse particulate matter. The CPF values for Sulfur presented in polar plots, for fine and coarse particles per year (2012 – 2014) as presented in Figure 10 – Figure 15. The contributions of Sulfur in fine particles for each year shows the different pattern. In 2012, the major source directions of S were coming from west, south-west and north-west from the sampling sites whereas in 2013 were coming from east, north, and west direction. CPF polar plot of FPM in 2014 shows that the Sulfur sources dominantly comes north, south and south-east. The sampling site at local EPA of West Java is located at the centre of Bandung city and surrounded by the commercials, offices, and roads that near the busy intersection with traffic coming from multiple directions that could be one of the emission sources of Sulfur. The gas station in the east side of the sampling site could also become one of the sulfur source that is emitted during the refueling activity. In the west side of sampling site, there are several industries such as pharmacy, textiles, etc. that could be release sulfur during the production process. The distinguishable directionality for Sulfur in coarse particles were detected where Sulfur is dominated from the northern sector of Bandung.

Fine particulate matter usually dominantly comes from anthropogenic sources whereas coarse particulate matter dominantly comes from natural sources. Previous study by Santoso et al. (2008) related to source apportionment of fine and coarse particulate matter in Bandung using PMF suggest that Sulfur as major constituent identified as secondary sulfate. This factor partly contributed from refuse burning or transport of biomass burning from the limestone kilns that are located in Padalarang district, approximately 25 km north-west side away from the sampling site. The Large amount of wood is burnt in those kilns. Other industry in Bandung such as textile industries using coal as the main energy supply
for the dying process [26]. The high concentration of sulfur is likely the result of high sulfur concentration in motor vehicle fuels. Generally, the motor vehicle fuels consist of hydrocarbon and contain sulfur since the fuels are produced from underground extraction [27]. Thus, the locally formed sulfate is condensing onto the surface of the existing particles [6].

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**Figure 10.** CPF for FPM Bandung 2012.

**Figure 11.** CPF for CPM Bandung 2012.

**Figure 12.** CPF for FPM Bandung 2013.

**Figure 13.** CPF for CPM Bandung 2013.

**Figure 14.** CPF for FPM Bandung 2014.

**Figure 15.** CPF for CPM Bandung 2014.
4. Conclusions
Fine particulate matter usually dominantly comes from anthropogenic sources whereas coarse particulate matter dominantly comes from natural sources. The CPF polar plot of Sulfur for fine and coarse particles showed that the contributions of Sulfur in fine particles shows different pattern for each year. From the previous study, the sulfur in fine particle mostly comes from industrial activity and motor vehicle that spread in all sectors of Bandung. Significant increase was found for Sulfur concentration in FPM that probably correlated with volcanic activity from several mountains in Indonesia during 2014 such as Kelud and Sinabung, therefore the CPF in FPM Bandung 2014 shows that the source of S in FPM was from south east, north and northwest of Bandung. In coarse particles, Sulfur dominated from the northern sector of Bandung. This source probably comes from the nearby volcano, Tangkuban Perahu that is 30 km north of Bandung.

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