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# $PM_{10}$ black carbon and ionic species concentration of urban atmosphere in Makassar of South Sulawesi Province, Indonesia

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## ABSTRACT

This paper presents a study on black carbon and ionic species concentrations in $PM_{10}$, sampled from the urban area of Makassar in the Province of South Sulawesi, Indonesia. The samples were collected on a weekly basis within a period of one year from February 2012 to January 2013. Results showed that the mean concentration of $PM_{10}$ black carbon was 2.01±0.93 μg/m$^3$, while ionic species concentrations were found in descending order; $SO_4^{2-}$>$NO_3^-$>$Cl^-$>$NH_4^+$, with mean concentrations of 1.44±1.04 μg/m$^3$, 1.11±1.06 μg/m$^3$, 0.87±0.91 μg/m$^3$, 0.49±0.64 μg/m$^3$, respectively. Black carbon concentration was higher during the dry months, which may be attributed to uncontrolled biomass burning during hot and dry weather conditions. Similarly, $SO_4^{2-}$ and $NH_4^+$ concentrations were higher during the dry month seasons. On the contrary, $Cl^-$ concentration was higher during the wet months, influenced by the marine environment surrounding the area.

***Keywords:*** *Air pollution, particulate pollution, black carbon, ions, $PM_{10}$*

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## 1. Introduction

Currently, urban air pollution has reached to a critical level in many parts of the world. This is particularly evident from researches conducted on the association between airborne particulate matter and human health (Birch and Cary, 1996; Colette et al., 2008; Aurela et al., 2010; Bautista et al., 2014). The urban areas in Indonesia are developing where transportation and industrial activities are becoming important. However, the rapid exploitation of natural energy resources in the country is not easily contained (Soehodho and Taufick, 2005; Santosa et al., 2008). Thus, the impacts on the existing and future air quality require serious attention.

In general, the most critical part of air pollution in many cities in Indonesia, especially Makassar, is the airborne particulate matter (PM). Airborne PM is unique among air contaminants because of its complexity in the terms of physical properties and chemical composition. Human respiratory health is most affected by finer particulate size fractions with aerodynamic diameter less than 10 μm that finds its way to the blood by penetration through the alveolar tissue in the lungs (Zanobetti et al., 2003; Mar et al., 2004). Evidently, studies have found that the particles with size less than 10 μm, or designated as $PM_{10}$, have long been associated with increasing mortality and morbidity (Dockery et al., 1992; Dockery and Pope, 1994).

Many countries have set standards for ambient $PM_{10}$ concentrations. For example, the Korean Ministry of Environment has promulgated an ambient air quality standard of 50 μg/m$^3$ and 100 μg/m$^3$ for annual and 24–h average, respectively (Lee et al., 2013). The World Health Organization set a stringent limit of 20 μg/m$^3$ for annual average (WHO, 2000). In Indonesia, the limit is set to 150 μg/m$^3$ for 24–h (DOE Indonesia, 1999), but there is still no limit stipulated for the annual standard for $PM_{10}$.

Black carbon (BC) and ionic species are categorized under $PM_{10}$. These particles are mainly produced by man–made activities such as transportation, industry, and biomass burning (Quraishi et al., 2009; Tiwari et al., 2009; Invernizzi et al., 2011; Pereira et al., 2012; Tsai et al., 2012). In many cases, these components have strong associations (positively correlated) with the particle number concentrations in the range of 100–180 nm (Cheng et al., 2014) and 56–180 (Cheng et al., 2013). BC is a nonvolatile fraction known as black carbon or elemental carbon (EC) (Begum and Biswas, 2009). BC is also a term used to describe a range of carbonaceous materials produced and formed through incomplete combustion of fossil fuels, biofuels, and residues from biomass burning, and emitted by both anthropogenic and naturally occurring soot (Masiello, 2004; Hammes et al., 2008). In global scale, BC aerosol is considered to be the second largest radiative forcing agent after carbon dioxide (Ramanathan and Carmichael, 2008). Bond et al. (2011) concluded that 85% of the BC total forcing is anthropogenic.

This study was conducted to investigate the BC and ionic component in the airborne $PM_{10}$ particulate sample collected in Makassar. Studies on BC and ionic species in Makassar are still

limited. Therefore, it is essential to study the impact of these components on air quality at the observation site.

## 2. Materials and Methods

### 2.1. Sampling site

Makassar city is strategically located near the south and north of the provinces of Sulawesi. It ranges from the western region to the eastern region of Indonesia and from the northern to the southern region of Indonesia. Makassar is located at 119° 24′ 38″ East longitude and 5° 8′ 19″ South latitude; bound by the Maros regency at the north and eastern side, Gowa Regency at the South Side, and Makassar Strait at the West Side. Makassar city covers an area of 17 577 $km^2$, constituting fourteen districts. The sampling was conducted at Daya Makassar, known to be a mixed urban, residential–commercial, and industrial area. The site is close to Makassar Industrial Estate which is surrounded by residential areas and commercial malls as depicted in Figure 1.

### 2.2. Sampling

Sample was collected using a standard size selective high volume air sampler (Andersen, SA321A) using a glass fiber (TFAGF810) type filter (20.3 cm x 25.5 cm) as the collection medium. The glass fiber filters have a number of specialties: have higher density than paper filters, able to tolerate a wider temperature range, has high retention capacity for particulate material, and has efficiency to collect 99% of submicron particles as small as 0.3 µm and over (Michael and Annunziata, 2012). The sampler was calibrated and operated with volumetric air flow rates of 1.13 $m^3$/min for a 24–hr basis. The selection of the sampler was based on several properties such as the ability to measure the mass concentration of the particulate matter with an aerodynamic diameter less than or equal to a nominal 10 micrometer ($PM_{10}$) in ambient air over a 24–hour period, the ability to measure the total volumetric air flow rates during the sampling period, and it is equipped with a flow control device that is capable of maintaining the sampler's operating flow rate within the flow rate limits specified for the sampler inlet over normal variations in line voltage and filter pressure drop. These properties makes the sampler fit to attain and maintain the ambient quality standard of the particulate matter.

### 2.3. Analysis of samples

The BC concentration in $PM_{10}$ sample was measured using a Smoke Stain Reflectometer (M43D EEL; Diffusion Systems Ltd., London, UK) based on a reflectance method (Biswas et al., 2003; Begum et al., 2004; Lestiani et al., 2007; Kothai et al., 2011; Lestari and Mauliadi, 2009). The measurement was done based on the darkness of the smoke stains on the sample. This was carried out using a photo–electric reflectometer. Firstly, a steady light was emitted onto the carbon or smoke stain by the photo–electric reflectometer. The light will be reflected back from the smoke stain to a photo–sensitive element. The reading was made by amplifying the electrical response. Darker stain obtained means lesser light is reflected, therefore a low meter reading corresponds to a dark surface, and a high reading to a light surface. Reflectometer reads on a scale of 0 (black) to 100 (white). The reflectometer reading, together with the measured volume of air sampled, was used to calculate the ambient concentration of BC from the standard calibration (Christolis et al., 1992).

Analysis for ions, e.g., chloride ($Cl^-$), nitrate ($NO_3^-$), ammonium ($NH_4^+$), and sulfate ($SO_4^{2-}$) in the $PM_{10}$ sample filter was performed using Ion Chromatography (Dionex, DX500 Model) equipped with an analytical column IonPac AS15, eluent source of EG40 with a flow rate of 0.5 mL/min, suppressed conductivity for detection, and an eluent suppressor (ASRS–ULTRA). A quarter of the 20.3 cm x 25.5 cm sample filter was cut into small pieces, and then placed into 100 mL conical flask. Twenty five milliliters of deionized water was added into the conical flask. The sample was ultrasonically extracted for at least 30 minutes at room temperature to dissolve the ionic components. Then, the sample was filtered and analyzed for the ionic content. Similarly, the blank filter was subjected to the same experimental procedures. The final concentration of each of the ionic component was calculated taking into consideration of the total effective area of the sampled filter, and divided by the total air volumetric flow rate passing through the filter.

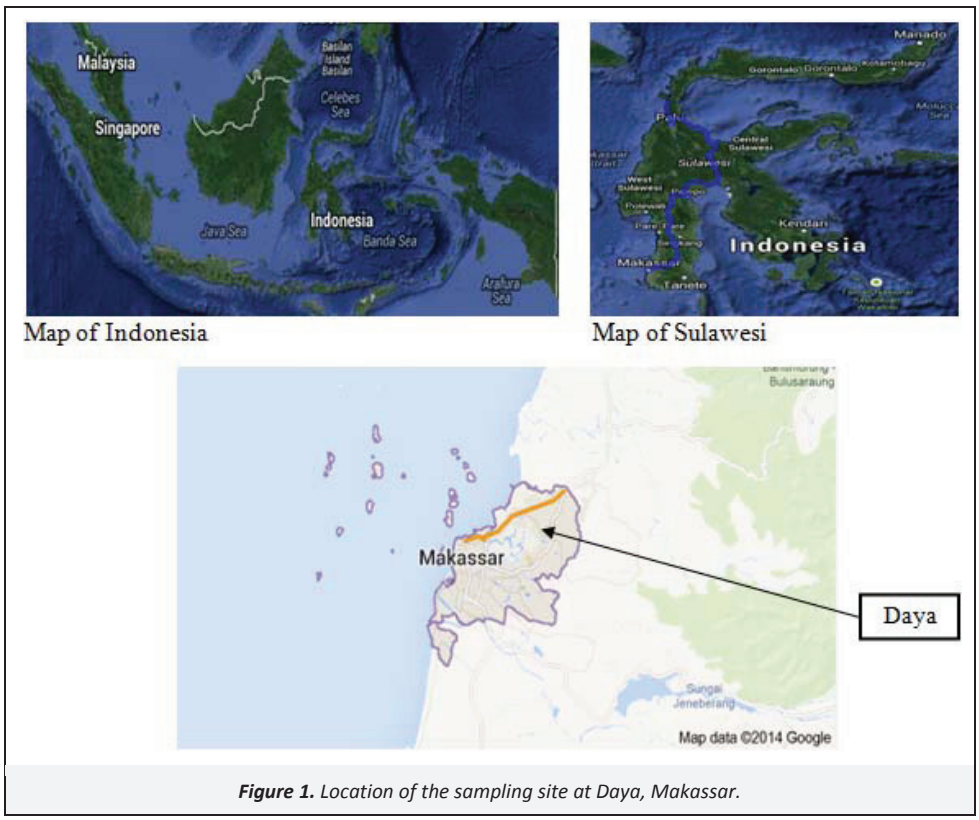


***Figure 1.*** *Location of the sampling site at Daya, Makassar.*

## 3. Results and Discussion

### 3.1. Particulate matter ($PM_{10}$)

Makassar, considered as a developing city among other cities in Indonesia, was found to have $PM_{10}$ concentration of around 32.9 μg/m$^3$. This value is much lower than those found in major cities like Bandung and Serpong, at 61.0 and 51.8 μg/m$^3$ respectively, as reported by Lestari and Mauliadi (2009) and Santoso et al. (2011). $PM_{10}$ concentrations in cities of Indonesia were found to be less than that in Shanghai (Li et al., 2012).

Table 1 presents the monthly variation of $PM_{10}$ concentrations showing that higher $PM_{10}$ concentrations were observed during the wet (October–March) and dry seasons (April–September) experienced in the area. The particulate matter concentration was found to be relatively higher during the dry season compared to the wet season, influenced by uncontrolled biomass burning during hot and dry weather conditions. A similar observation was reported in Bandung where the atmospheric particulate concentration was observed to be higher during the dry season than the wet season (Lestari and Mauliadi, 2009). In a related study, Juneng et al. (2011) reported significant seasonal variations in $PM_{10}$ concentrations in the Klang Valley, Malaysia, where very high concentrations were recorded during the dry period of the summer monsoon season.

### 3.2. Black carbon and ionic constituents of $PM_{10}$

Table 2 presents the means, standard deviations, as well as ranges of BC and ionic concentrations, and their respective contributions to $PM_{10}$ in Makassar. It can be observed that BC constituted the highest percentage in $PM_{10}$. BC and total ions represented only 18.2% of the total mass concentration of the $PM_{10}$ samples at the site [i.e BC 6.1%, followed by $SO_4^{2-}$ (4.5%), $NO_3^-$ (3.4%), $Cl^-$ (2.7%) and $NH_4^+$ (1.5%)], while 81.8% were unidentified constituents that need to be investigated further.

Figure 2 presents the linear relationship between $PM_{10}$ and BC concentration, which showed a significant positive correlation ($p$<0.01, $R^2$=0.52), indicating a strong relationship between these two parameters in the study area. Positive intercept mean that there were other components, namely ionic and elemental components contributing to the $PM_{10}$ at the study site, which is needed to be taken into account in the investigation.

***Table 1.*** *Monthly concentrations (μg/m$^3$, mean±standard deviation) of $PM_{10}$, BC and ions at Makassar*

| | $PM_{10}$ | BC | $Cl^-$ | $SO_4^{2-}$ | $NO_3^-$ | $NH_4^+$ |
|---|---|---|---|---|---|---|
| February 2012 | 17.81±6.37 | 2.46 ±0.97 | 0.86 ±0.86 | 0.88 ±0.93 | 1.87 ±2.63 | 0.51 ±0.72 |
| March 2012 | 28.84±10.86 | 3.22±0.87 | 1.12±0.73 | 1.41±1.32 | 0.44±0.59 | 0.32±0.57 |
| April 2012 | 33.41±5.75 | 2.60±0.14 | 1.22±0.83 | 1.84±1.06 | 0.68±0.78 | 0.82±0.48 |
| May 2012 | 42.38±7.29 | 2.39±1.34 | 0.76±0.39 | 1.23±0.46 | 2.14±1.79 | 1.43±0.59 |
| June 2012 | 49.49±11.46 | 2.96±0.93 | 0.09±0.03 | 2.37±1.17 | 1.15±1.34 | 0.67±0.53 |
| July 2012 | 37.85±4.55 | 2.55±0.38 | 0.33±0.45 | 2.37±0.92 | 0.83±0.26 | 0.29±0.49 |
| August 2012 | 41.58±5.73 | 1.96±0.34 | 0.07±0.01 | 1.49±0.16 | 0.91±1.09 | 0.04±0.02 |
| September 2012 | 35.64±8.39 | 1.56±0.48 | 1.09±1.45 | 1.04±1.57 | 0.92±0.83 | 0.20±0.35 |
| October 2012 | 24.29±2.83 | 1.27±0.31 | 0.49±0.45 | 1.86±1.05 | 0.99±0.68 | 0.64±1.16 |
| November 2012 | 23.41±4.93 | 1.60±0.51 | 1.42±1.40 | 1.10±0.71 | 1.51±1.20 | 0.24±0.44 |
| December 2012 | 23.34±3.88 | 1.04±0.32 | 0.99±1.35 | 0.76±1.32 | 0.96±0.48 | 0.37±0.51 |
| January 2013 | 25.63±5.41 | 1.06±0.26 | 1.11±0.82 | 1.25±0.89 | 1.11±0.47 | 0.27±0.49 |

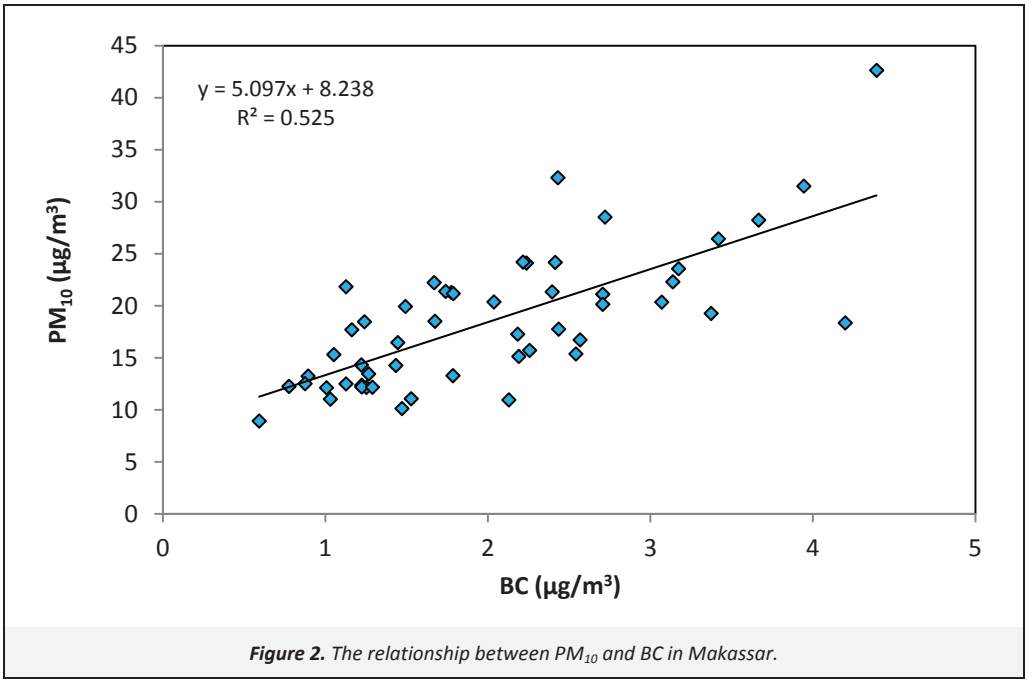


***Figure 2.*** *The relationship between $PM_{10}$ and BC in Makassar.*

***Table 2.*** *Mean, standard deviations, minimum and maximum concentration of constituents in $PM_{10}$ in Makassar (Feb 2012–Jan 2013)*

| Element | Mean | Std Deviation | Minimum | Maximum |
|---|---|---|---|---|
| $PM_{10}$ | 32.9 [100] | 11.1 | 13.3 | 63.0 |
| BC | 2.01 [6.1] | 0.93 | 0.59 | 4.39 |
| $Cl^-$ | 0.87 [2.7] | 0.91 | 0.06 | 3.47 |
| $SO_4^{2-}$ | 1.44 [4.5] | 1.03 | 0.08 | 4.00 |
| $NO_3^-$ | 1.11 [3.4] | 1.06 | 0.01 | 4.71 |
| $NH_4^+$ | 0.49 [1.5] | 0.64 | 0.02 | 2.71 |

*Number of samples=53; Unit=μg/m$^3$ ; [ ]=percent of $PM_{10}$*

As depicted in Table 2**,** the percentage of the BC observed in this study was ~6.1%, which is quite similar as reported elsewhere (Dotse et al., 2012). In addition, the authors found that there was a higher percentage of the BC in a finer size fraction of $PM_{2.5}$ with an average of 18.4%. A similar finding in Bandung, Indonesia showed a high concentration of carbon black in the $PM_{2.5}$ fraction of ~25.1% (Lestari and Mauliadi, 2009). However, no comparison on the percentage of BC concentration in $PM_{10}$ could be made in this case, as the analysis of BC in $PM_{10}$ was not conducted by the authors.

As presented in Table 2, sulfate ($SO_4^{2-}$) represents the most dominant component among the ionic species, followed by $NO_3^-$, $Cl^-$, and $NH_4^+$ with the concentration of 1.44±1.04 μg/m$^3$, 1.11± 1.06 μg/m$^3$, 0.87±0.91 μg/m$^3$, 0.49±0.64 μg/m$^3$, respectively. Sulfate is a major constituent of pollutant at four monitoring sites in China, comprising 23.8–46.7% in $PM_{2.5}$ and 18.3–30.7% in $PM_{10}$, originating from sea salt and non–sea salt (Wang et al., 2006). Rashid et al. (1997) also reported that $SO_4^{2-}$ which originates from both the natural or sea salt constitutes from 0.37–0.50%, while anthropogenic or non–sea salt $SO_4^{2-}$ constitutes 3.31–8.28% of the $PM_{10}$ sampled in Kuala Lumpur, Malaysia. It was then assumed that the concentration is due to the location of Makassar which is surrounded by seas, thus the contribution of sea salt sulfate could be significant in the area.

### 3.3. Seasonal variation of black carbon and ions species

Table 1 presents the monthly mean of BC and ionic species concentration along with $PM_{10}$ in Makassar which coincided with the wet and dry seasons experienced at the study site. Figure 3 presents the mean of the BC and ionic species concentration during dry and wet season, which showed that a higher concentration of BC was observed during the dry season (i.e., 2.34 μg/m$^3$) compared to that during the wet season (i.e., 1.78 μg/m$^3$). The BC mass concentrations in the Taipei urban area recorded in the spring, summer, autumn, and winter were 3.3 μg/m$^3$, 3.2 μg/m$^3$, 2.4 μg/m$^3$ and 3.1 μg/m$^3$, respectively. Similarly, sulfate and ammonium recorded higher concentrations in the dry season than the wet season (Cheng et al., 2014). Kim Oanh et al. (2006) studied $NO_3^-$ and $SO_4^{2-}$ in $PM_{2.5}$ and $PM_{10}$ in Beijing, China. They reported that both ionic species were of secondary particles; partly linked to the burning activity which was more frequent during the dry season as compared to the wet season. The authors also observed that chloride had a lower concentration during the dry season compared to the wet season because it is easily vaporized in dry and hot days. The authors concluded that a depletion of chloride occurred during the dry and hot months experienced in the region.

Table 1 presents the monthly variation of BC, showing the highest monthly mean concentration of BC in June 2012 (3.22± 0.87 μg/m$^3$) and the lowest in December 2012 (1.04±0.33 μg/m$^3$). The presence of BC is partly believed to be attributed to vehicular emissions, particularly from the diesel fuelled vehicles. The sampling site was close to the main bus station of the city. In addition, uncontrolled biomass burnings were frequently observed during the hot and dry weather conditions in the region.

Chloride is normally derived from sea salt (SS) and non–sea salt (NSS) sources, probably related to the industrial emissions like coal combustion (Wang et al., 2006). Studies have shown that nitric acid can react with sodium chloride on sea salt (Pryor and Sorensen, 2000; Saul et al., 2006). As seen in Table 1, the monthly mean of $Cl^-$ concentration in Makassar recorded the highest concentration in November 2012 (1.42±1.40 μg/m$^3$), and the lowest was in August 2012 (0.07±0.01 μg/ m$^3$) with its concentration ranging from 0.06 to 3.47 μg/m$^3$. The presence of $Cl^-$ is believed to be carried through the winds passing through the Makassar Sea bringing sea sprays along with.

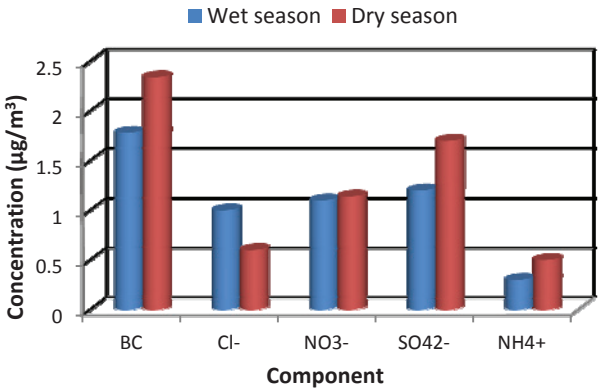


***Figure 3.*** *Black carbon and ionic species during the wet and dry seasons.*

The concentration of $NH_4^+$ observed in this study ranged from 0.02 to 2.71 μg/m$^3$. Ammonium, as investigated in this study, originated mainly from gaseous substances such as combustion products. Unlike chloride, ammonium concentration and its size distributions appeared to be location dependent and strongly influenced by localized sources. Ammonium and ammonia nitrogen are closely related (Walworth, 2013), with $NH_4^+$ predominantly in fine particles, with an aerodynamic diameter less than 2 μm (Wall et al., 1988). In addition, $NH_4^+$ aerosols aggressively affect the earth's radiative balance, both directly by scattering incoming radiation and indirectly by acting as cloud condensation nuclei (CCN). They also contribute to the long–range transport of acidic pollutants because the atmospheric lifetime of ammonia is short (<24 h), while $NH_4^+$ salt has an atmospheric lifetime of a few days (Alves et al., 2007). Long range–transport can result in deposition of $NH_4^+$ salts far away from the emission sources. Upon deposition, $NH_4^+$ aerosols can contribute to soil acidification, forest decline, and eutrophication of waterways (Marley and Gaffney, 2004).

The concentration of nitrate and sulfate were found to be the highest in May 2012 (2.14 μg/m$^3$) and July 2012 (2.37 μg/m$^3$). It was observed that the sulfate concentration was higher during the dry season than the wet season at the study site. However, no significant seasonal difference for nitrate was observed in this case. Nitrate and sulfate (sea salt is also a main sulfate contributor) in the atmosphere generally originate from high temperature combustion processes, produced from conversion of $NO_X$ and $SO_X$ gases.

A significant portion of nitrate results from the atmospheric conversion of nitrogen oxides and ammonia (Seinfeld and Pandis, 1998; Han et al., 2006). In addition, nitrate also originates from both primary and secondary sources, with the coarse mode nitrate generated from sea salt spray and the fine mode nitrate produced by photochemical reaction. Sulfate in coarse mode may be attributed to the reaction of $CaCO_3$ with $H_2SO_4$ to form $CaSO_4$ in coarse particles (Tsai et al., 2012), apart from sea salt origin (Wang et al., 2006). Ammonium nitrate and ammonium sulfate are said to be the dominant constituents in the particle size of $PM_{10}$ (Tsai et al., 2012).

## 4. Conclusion

Airborne particulate matter (with aerodynamic diameter of less or equal than 10 µm or $PM_{10}$) samples were collected on a weekly basis for one year from February 2012 to January 2013 at one site of Makassar, Province of South Sulawesi Indonesia. Results showed that the average particulate mass concentration was 32.9±11.1 µg/m$^3$. The concentration of $PM_{10}$ found in this study was lower than those measured in more developed cities like Bandung and Serpong of Indonesia.

The mean concentration of BC was found to be 2.01± 0.93 µg/m$^3$, while the ionic species was found in descending order $SO_4^{2-}$>$NO_3^-$>$Cl^-$>$NH_4^+$ with mean concentrations of 1.44± 1.04 µg/m$^3$, 1.11±1.06 µg/m$^3$, 0.87±0.91µg/m$^3$, 0.49±0.64 µg/m$^3$, respectively. BC and ionic species constituted 6.1% and 12.1% of the $PM_{10}$ mass concentration, respectively. Both BC and ions contributed 18.2%, while 81.8% of the particulate matter is still needed to be identified. The BC concentration was higher during the dry months, which may be attributed to uncontrolled biomass burning during hot and dry weather conditions. Similarly, $SO_4^{2-}$ and $NH_4^+$ concentrations were higher during the dry months. On the contrary, $Cl^-$ concentration was higher during the wet month period, suggesting the intrusion of the sea sprays during rainy season, which carries chloride (i.e. sea salt component) along with.

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