CHEMICAL COMPOSITION AND SOURCES ATTRIBUTION OF RAINWATER IN BANDUNG AREA, INDONESIA

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ABSTRACT: Data of international and national monitoring stations have showed evidences of rainwater acidification in Indonesia. This research aimed to investigate the small-scale variations and the influence of urban activities to rainwater chemical composition in Bandung, Indonesia, as well as to determine the contributing sources to its chemical variation. Rainwater bulk samples had been collected weekly at 4 (four) locations representing differences in altitude and local land use, from February to September 2016. Samples were analyzed for pH, conductivity, and major ions (SO_4^{2-}, NO_3^-, Cl^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, and NH_4^+). The results showed that the pH of rainwater were in the range of acidic to normal (average 5.42 ± 0.72), where ammonium acted as the major neutralizing factor (NF). Small-scale spatial variability of nss-SO_4^{2-}, NO_3^{2-}, nss-Cl^-, nss-Ca^{2+}, nss-Mg^{2+}, and NH_4^+ within 4 (four) locations were observed. Sulphate and NO_3^- as well as Ca^{2+} and Mg^{2+} were significantly correlated, with r = 0.870 and r = 0.929, respectively. Principal Component Analysis (PCA) indicated three main sources of rainwater chemistry. Firstly, a mix of natural sources (e.g., sea-salt aerosols, crustal elements and waste decomposition) which consisted of sea-salt SO_4^{2-}, ss-Cl^-, Na^+, ss-Ca^{2+}, nss-Ca^{2+}, ss-K^+, nss-K^+ and NH_4^+. Secondly, fossil fuel combustion/vehicular emissions, which consisted of nss-SO_4^{2-}, NO_3^-, H^+, and nss-Mg^{2+}; and thirdly, nss-Cl^+ presumably from other acid reaction. These three components accounted for 80.25% of the total variance. The results suggested that local and long-range transported emissions of natural and anthropogenic sources contributed to the rainwater chemistry in the Bandung area.

Keywords: Acid Rain, Wet Deposition, PCA, Rainwater Chemistry

1. INTRODUCTION

Chemical compositions of rainwater have been studied in many urban areas worldwide, such as to evaluate the impact of air pollution. Source analysis and contribution of anthropogenic and natural sources to rainwater contamination are important in the context of air quality management [1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18].

The pH of rainwater could be used as an indicator of atmospheric pollution because in normal conditions, based on the value in the equilibrium of pure water and atmospheric CO_2, pH of rainwater is 5.6[19]. Acidity variability occurs due to atmospheric chemistry processes involving air pollutants absorbed from various sources and being wash out by rain droplets. For examples, the alkaline pH in rainwater in the Pune Region of India mainly caused by the abundance of Ca^{2+} originated from vehicle driven roadside dust [2]. Acidic rainwater in Shihwa Basin Korea was influenced by the dominant ions of SO_4^{2-} and NO_3^- which were neutralized by NH_4^+ and Ca^{2+} [17].

Elevation differences and local anthropogenic activities might account for the variation in the rainwater chemical composition [13], causing, e.g. monthly variations of rainwater chemistry in the Yulong Mountain Region in Southwestern China.

Bandung area is located in a mountainous region of West Java Indonesia where the lowest level is at an elevation of 768 meters above sea level (asl). Bandung is the third most populous city in Indonesia after Jakarta and Surabaya. It is the capital of the West Java Province, located about 140 km from Jakarta as the national capital. Its urban activities potentially affect its air quality, and hence rainwater, to some extent. Bandung has a tropical, wet, and dry climate with average temperature ranges from 18.1°C to 31.9°C during 2015 and an average rainfall of 184.75 mm with the ranges of 0.3 mm/month (in July during the dry season) to 322.4 mm/month (in November in the wet season).

Due to its characteristics, rainwater quality in the Bandung area might be influenced by air pollution. This paper explores rainwater characteristics in Bandung and identifies the likely sources that determine its chemical composition. Samples were collected during the rainy events from February to September 2016.

2. METHODOLOGY

2.1 Sampling Area

Rainwater samples were collected at 4 (four) locations, representing different elevation and land use within the Bandung area, as shown in Figure 1 as follow :

1) Lembang (06°49'35,01"S 107°37'04,54"E) with an altitude of approximately 1.272 m asl lies on the hillside of the mountain area. The site is located approximately 12 km away from the center of Bandung City, with the land use predominantly being agricultural, livestock, and tourism activity.
2) Coblong (06°55'11,07"S 107°36'04,54" E) with an altitude of approximately 803 m asl. It is an urban area with high traffic activity.
3) Sumur Bandung (06°55'11,07"S 107°36'39,34"E) with an altitude of approximately 742 m asl. It is the center of major urban activity such as offices complex, trading, and traffic area.
4) Buah Batu (06°53'26,69"S 107°36'43,73"U) with an altitude of approximately 642 m asl is a sampling site in urban settlement area with high population density and their associated activities.
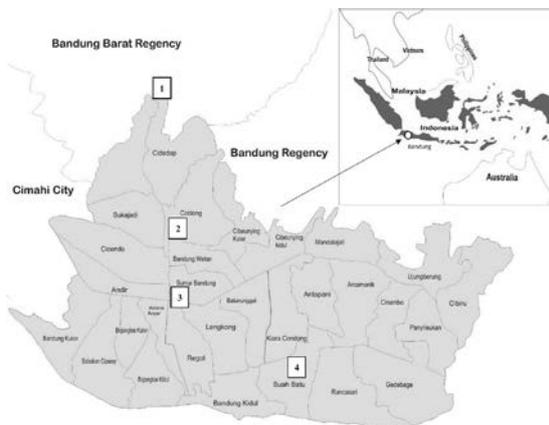
Fig. 1. Sampling Site

2.2 Samples Collection and Analysis

Rainwater samples were taken on a weekly basis from February to September 2016. The rainwater samples were collected using a polyethylene bottle (1.000 mL). Rainwater passed through a polyethylene funnel (12 mm in diameter) equipped with a 1 mm filter plug that was put inside a stainless steel cylinder. The sampler bottles, funnels and filters were cleaned and rinsed with deionized water before each use. Rainwater samples were then transferred to 100 mL PE bottles for ion analysis.

The pH values and conductivity for the rainwater samples were analyzed with a pH meter (Sartorius Professional Meter PP-20), and a conductivity meter (Agilent Technologies 3200 C). Major ions in rainwater of sulphate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), and ammonium (NH_4^+) were determined using Ion Chromatography (DIONEX ICS 500 DP).

Statistical characteristics of rainfall, pH, conductivity, and Precipitation-Weighted Mean (PWM) of ion concentration were analyzed. The spatial variation between the sampling sites was analyzed using One-way ANOVA. Source attribution was analyzed using correlation and Principal Component Analysis (PCA).

2.1 Quality Control of Samples Analysis

The quality control of the samples was checked by Ion Balance (IB) defined as Ion Percent Difference (IPD), as IPD = [(C − A) / (C + A)] × 100, where, C and A represents anion and cation equivalents (μeq L^{-1}), respectively [20]. Precipitation weighted mean (PWM) was carried out by calculating the sum of the product of ion concentration and the precipitation amount, obtained from rainwater sample, for each data pair, and dividing the result by the sum of precipitation amount for a certain period [21].

3. RESULT AND DISCUSSION

3.1 Rainfall Amount, pH and Conductivity of Rainwater

Rainfall amount at 4 (four) sampling sites was largely varied, ranging between 40.69 – 347.66 mm/month. High rainfall events during 2016 were suggested to be impacted by the La Nina event (Fig.2), with the highest events recorded in March, April, and May.
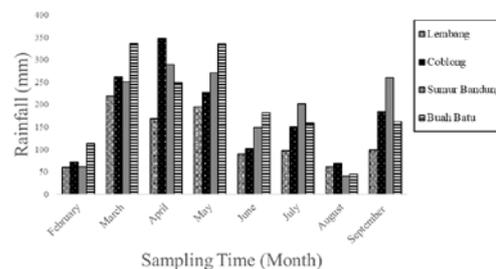
Fig.2 Rainfall Distribution at Four Sampling Sites

The pH of the rainwater ranged from 4.74 to 6.20 (average 5.45), lower than the pH of typical natural rainwater (5.6), indicating the influence of acidic substances in the rainwater [19].

Rainwater from Buah Batu showed a tendency of more acidic compared to other sites, particularly the Lembang site which is located in the rural area at the highest altitude. This condition is contrary to the acidic orographic enhancement found in Northern England [23], suggesting local urban emission sources might have a more significant influence in the chemistry of rainfall.

Nearly 60% of acid precipitation pH was between 5.0 to less than 5.6, while 42% had pH less than 5.0 (which would have significant effects on environmental acidification). Within this 42%, 11% were strongly acidic with pH < 4.5 (Fig. 3).
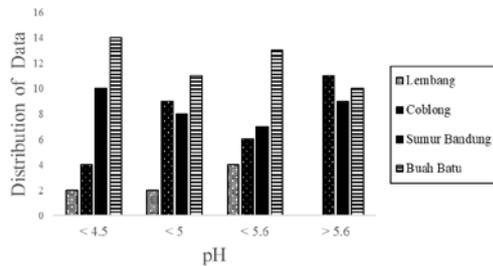
Fig.3 Rainwater pH Distribution

The major precursors of acidity in rainwater were SO_2 and NO_x as were found in the sources attribution analysis (See Section 3.5). Soil particles and sea spray origin species might play an essential role in neutralizing SO_2 and NO_x.

The abundance of acid precursors is seen in the rainwater conductivity, the value being related to the total sum of anions and cations. Rainwater conductivity is mainly contributed by water-soluble ions. The rainwater electric conductivity ranged from <0.5 to 100 µS/cm (Fig. 4). There is no clear pattern as to whether conductivity was related to the rainwater amount, as the temporal variation of conductivity did not resemble the pattern of rainfall amount as seen in Fig. 2.
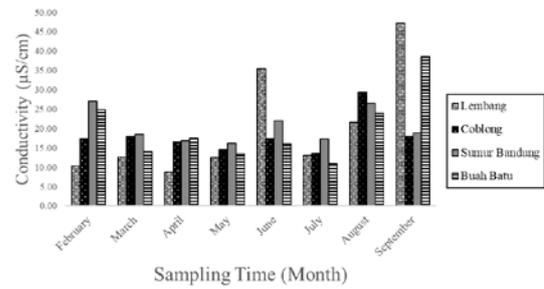
Fig.4 Temporal Variation of Conductivity (µS/cm)

3.2 Temporal Variation of Precipitation-Weighted Mean (PWM) Concentration

Precipitation-Weighted Mean (PWM) concentrations were calculated to eliminate the dillution effect of rainwater quantity during high rainfall. The temporal variations of PWM concentration of major ions that reflected the variation due to acid chemistry processes in rainwater are shown in Figure 5.

While rainfall monthly variation showed clear pattern of seasonal influence of lowest amount during June – August at all sites, SO_4^{2-} and NO_3^- PWM concentrations showed irregular patterns. Particularly for SO_4^{2-} and NO_3^-, the PWM concentrations were detected during low and high rainfall, suggesting that rainfall was not the sole factor affecting the concentration. Instead, the variation might be due to the strength of acid precursor emissions from local and regional sources. On the other hand, increasing ammonium (NH_4^+) as well as Cl^-, Na^+, Ca^{2+}, Mg^{2+}, and K^+ ions showed in the PWM concentration on lower rainfall suggested the dilution effect of these natural-origin ions. In addition, the strong temporal variation of anion SO_4^{2-} and cation NH_4^+ were presumably caused by the neutralization reaction in the rainout and washout deposition processes.
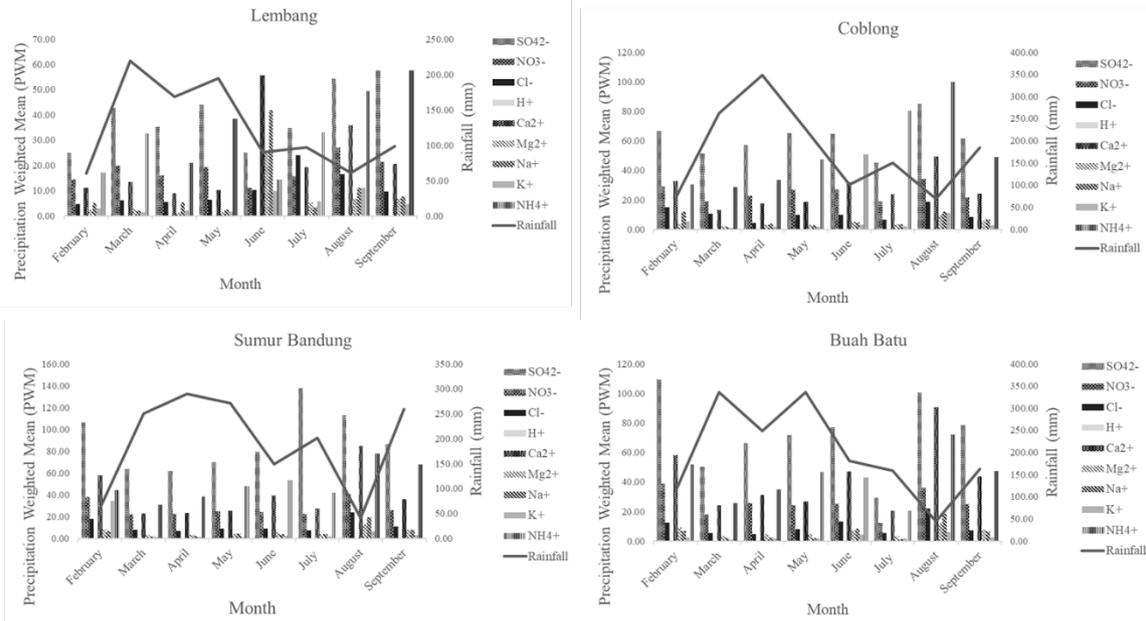
Fig.5 Temporal Variation of Precipitation Weight Mean (PWM) Concentration Ions

3.3 Neutralizing Factors

Neutralizing factor (NF) was used to evaluate the neutralization of major anions by crustal components (Ca^{2+} and Mg^{2+}) and NH_4^+. The neutralization of acids by base cations were evaluated by calculating the neutralization factors using the formula: NF (x) = $((X^+)/(SO_4^{2-} + NO_3^-)$ where X is the Ca^{2+}, Mg^{2+} and NH_4^+.

The NF observed for Ca^{2+}, Mg^{2+} and NH_4^+ were 0.31, 0.09, 0.56 (Lembang); 0.26, 0.05, 0.56 (Coblong); 0.32, 0.05, 0.49 (Sumur Bandung) and 0.38, 0.06, 0.43 (Buah Batu). Hence, NF showed that NH_4^+ was the dominant neutralizing ion in the Bandung area.

This result was similar to what was found in Chiang May Thailand [1], Dalian China [6], Ya'an, Southwestern China [9], and Nanpin Mangdang Mountain China [25].

3.4 Sea salt and Non-Sea Salt Contribution

PWM concentration of sea salt (ss) and non-sea salt (nss) contributions were described in Figure 6. Data used for analyzing ss-ions and nss-ions were the long-term monitoring by EANET (Acid Deposition Monitoring Network in East Asia). Non-sea salt SO_4^{2-} and nss-Ca^{2+} were the dominant ions for all sampling sites. Non-sea salt sulfate was the highest anion PWM concentration in the Bandung area for more than ten years of EANET monitoring. The trend of SO_2 emission during the 2000 – 2014 period in Indonesia increased by 50% (+ 0.12 ppb year^{-1}) [23]. The observation found in this study was in line with the EANET finding.
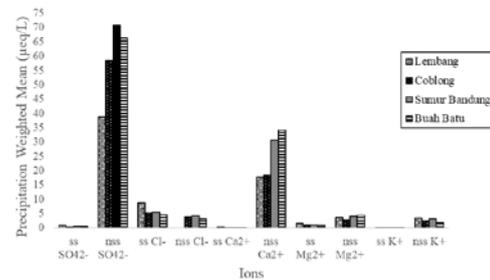
Fig.6 PWM Concentration of Sea Salt and Non-Sea Salt Contribution

3.5 Sources Attribution Analysis

3.5.1 One-Way ANOVA

One-way analysis of variance (ANOVA) was used to analyze the spatial variability of the ions in rainwater chemistry, as shown in Table 1. Statistical analysis was applied to log-normal transformed data because the raw data of rainwater amount and ion concentrations had positive skewness distribution.

The F-test statistic was applied to test the null hypothesis; that there is no variance of ions concentration and rainfall amount between different sampling sites ($\alpha = 0.05$).

It was found that the concentration of nss-SO_4^{2-}, NO_3^{2-}, nss-Ca^{2+}, nss-Mg^{2+}, and NH_4^+ ions were significantly different between the 4 (four) sampling sites indicating small-scale spatial

variability.

Table 1 ANOVA of Ion Concentration in Rainfall and Rainfall Amount

	F	Sig.
Rainfall (mm)	1.289	0.281
ss-SO_4^{2-}	0.252	0.860
nss-SO_4^{2-}	**11.353**	**0.000**
NO_3^-	**3.258**	**0.024**
ss-Cl^-	0.076	0.973
nss-Cl^-	1.104	0.351
H^+	0.479	0.698
Na^+	0.252	0.859
ss-Ca^{2+}	0.252	0.860
nss-Ca^{2+}	**7.991**	**0.000**
ss-Mg^{2+}	0.088	0.967
nss-Mg^{2+}	**2.931**	**0.037**
ss-K^+	0.253	0.859
nss-K^+	0.517	0.671
NH_4^+	**2.932**	**0.037**

3.5.2 Ion Correlation

Correlation between ions was used to determine the different sources of anions and cations in the rainwater and their potential linkages. Ions concentration in rainwater was affected by rainfall amount, possibly causing a multicollinearity problem. Partial correlation analysis of the strength and direction of a linear relationship between ions whilst controlling for rainwater amount were calculated and presented in

Table 2.

The acidic ions of non-sea salt SO_4^{2-} and NO_3^- that significantly correlated (r = 0.901) possibly indicate similar sources. Natural rainwater becomes more acidic from the sulfuric and nitric acids that are formed by acidic gases of SO_2 and NO_3 from anthropogenic activities.

Significant correlation between SO_4^{2-} and NH_4^+ (r = 0.431) also NO_3^- and NH_4^+ (r = 0.481) indicated that ammonium exists in the atmosphere as $(NH_4)_2SO_4$ and NH_4NO_3 [19]. On the other hand, a significant correlation between Ca^{2+} and Mg^{2+} (r = 0.801) showed their occurrence from the crustal origin.

3.5.3 Principal Component Analysis (PCA)

Principal components analysis (PCA) was used to identify and associate potential sources attribution of the species in rainwater chemistry. PCA was applied to explore data relationships and source types for rainwater composition in many studies [1, 2, 5, 9, 10, 11, 13, 16, 18]. As a cut-off, factors with an eigenvalue >1 were retained. Factors are extracted to be interpreted as a common source for the components in the same factor. In this study, PCA explained 82.291% of the data variance.

Table 2. Partial Correlation Coefficients of Major Ions in Rainwater

	nssSO_4^{2-}	NO_3^-	ssCl^-	nssCl^-	H^+	Na^{2+}	ssCa^{2+}	nssCa^{2+}	ssMg^{2+}	nssMg^{2+}	ssK^+	nssK^+	NH_4^+
ssSO_4^{2-}	.563	.530	1.000	-.197	.109	1.000	1.000	.619	1.000	.449	1.000	.617	.430
	.000	.000	.000	.055	.291	.000	.000	.000	.000	.000	.000	.000	.000
nssSO_4^{2-}		.901	.563	.079	.178	.563	.563	.780	.563	.611	.562	.389	.431
		.000	.000	.446	.084	.000	.000	.000	.000	.000	.000	.000	.000
NO_3^-			.530	.009	.226	.530	.530	.627	.530	.438	.530	.366	.481
			.000	.929	.027	.000	.000	.000	.000	.000	.000	.000	.000
ssCl^-				-.198	.109	1.000	1.000	.619	1.000	.449	1.000	.616	.430
				.053	.290	.000	.000	.000	.000	.000	.000	.000	.000
nssCl^-					-.009	-.197	-.196	.048	-.197	.124	-.197	.107	-.095
					.930	.055	.055	.646	.055	.227	.054	.300	.358
H^+						.109	.110	.062	.109	.041	.109	.075	-.154
						.291	.288	.549	.290	.691	.292	.470	.134
Na^{2+}							1.000	.619	1.000	.449	1.000	.617	.430
							.000	.000	.000	.000	.000	.000	.000
ssCa^{2+}								.619	1.000	.450	1.000	.617	.430
								.000	.000	.000	.000	.000	.000
nssCa^{2+}									.619	.801	.618	.440	.282
									.000	.000	.000	.000	.005
ssMg^{2+}										.449	1.000	.617	.430
										.000	.000	.000	.000
nssMg^{2+}											.449	.379	.292
											.000	.000	.004
ssK^+												.616	.430
												.000	.000
nssK^+													.204
													.046

Table 3. Component Matrix of the Sources Attribution Analysis using Principal Component Analysis (PCA).

	Component		
	1	2	3
ss-SO_4^{2-}	**0.955**	0.272	0.015
nss-SO_4^{2-}	0.304	**0.900**	0.080
NO_3^-	0.355	**0.853**	-0.008
ss-Cl^-	**0.955**	0.272	0.015
nss-Cl^-	-0.179	0.151	**0.822**
H^+	-0.365	**0.402**	-0.518
Na^+	**0.955**	0.272	0.015
ss-Ca^{2+}	**0.955**	0.272	0.015
nss-Ca^{2+}	**0.639**	0.548	0.341
ss-Mg^{2+}	**0.955**	0.272	0.015
nss-Mg^{2+}	**0.479**	**0.522**	0.427
ss-K^+	**0.955**	0.271	0.14
nss-K^+	**0.760**	0.168	0.326
NH_4^+	**0.430**	0.376	-0.008
Total Variance (%)	**62.750**	**10.780**	**8.761**

Factor 1 explained approximately 60.75 % of the total variance, with strong positive loadings for sea salt SO_4^{2-}, sea salt Cl^-, sea salt Ca^{2+}, sea salt and non-sea salt Mg^{2+}, Na^+, sea salt and non-sea salt K^+ indicating the natural source (sea and soil) contributions to rainwater. In addition, non-sea salt K^+ in rainwater presumably comes from biomass burning [1]. NH_4^+ sources might come from human and animal wastes and other human-made activities (possibly fertilizers, agriculture wastes, and urban activities).

Factor 2 explained approximately 10.78% of the total variance, with strong positive loading for non-sea salt SO_4^{2-}, NO_3^-, H^+, and non-sea salt Mg^{2+} suggesting anthropogenic sources from vehicular emission, most notably fossil fuels combustion. With hydrogen ion being in the same factor, this also points to acidification as a result. In Bandung, there were ± 1.6 million units of a motor vehicle registered in 2015, which likely contributed significantly to SO_2 and NO_3 emissions [24].

Factor 3 explained approximately 8.76 % of the total variance with non-sea salt Cl^- as the sole component, possibly as the result of the acid HCl with alkaline compound (Ca^{2+}, Mg^{2+}, and K^+).

4. CONCLUSIONS

The analysis of rainwater in Bandung in this study found that:

1. The pH of rainwater tends to be acidic with the values ranged between 3.4 to 7.71 (average 5.42 ± 0.72), with the average conductivity of 19.60 µS/cm.

2. The major anion component was SO_4^{2-} (36.2% of the total anion mass), while NH_4^+ was the main cation component (47.7% of total cation mass). The major components found that, SO_4^{2-} and NH_4^+ PWM concentrations were the highest at all sampling sites in almost every month, compared to other ions.

3. Ammonium (NH_4^+) was found to be the dominant neutralizing ion.

4. Small-scale spatial variability found for nss-SO_4^{2-}, NO_3^{2-}, nss-Cl^-, nss-Ca^{2+}, nss-Mg^{2+}, and NH_4^+, indicate the influence of local emission sources.

5. Principal Component Analysis (PCA) demonstrated that rainwater was also affected by natural sources of sea salts and crustal sources as well as biomass burning and local anthropogenic activities (vehicular emission, fossil fuels combustion, human and animal wastes and human made activities (fertilizers and agriculture wastes).

5. ACKNOWLEDGMENTS

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