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Wed, Jun 5, 2019 at 11:30 AM

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# GEOMATE Journal Review and Evaluation

## Paper ID number

87856

## Paper Title

CHEMICAL COMPOSITION AND SOURCES  
ATTRIBUTION OF RAINWATER IN BANDUNG AREA  
INDONESIA

### i. Originality

3

### ii. Quality

3

### iii. Relevance

5

### iv. Presentation

3

### v. Recommendation

4

### Total (sum of i to v)

18

## General comments

This is an interesting research on CHEMICAL COMPOSITION AND SOURCES ATTRIBUTION OF RAINWATER IN BANDUNG AREA INDONESIA . It is a good quality paper. It can be recommended for journal publication after a minor revision.

## Mandatory changes

Please check the attachment to improve the manuscript.

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# GEOMATE Journal Review and Evaluation

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CHEMICAL COMPOSITION AND SOURCES  
ATTRIBUTION OF RAINWATER IN BANDUNG AREA  
INDONESIA

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3

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4

### v. Recommendation

1

### Total (sum of i to v)

14

## General comments

1. The authors have very poor command on english language. This paper need to be completely re-written.
2. Test results should be compared with some standard or other sources.
3. The results should be validated.
4. Paper Title and the analysis do not march - abstract and conclusions are not clear
5. Revise abstract and conclusion
6. Itemize conclusion in simple english

## Mandatory changes

Should be completely re-written

## Suggested changes

The title should match with the analysis and findings. It appears that the authors are analyzing the rainwater chemical composition data and relating its contribution from various sources- vehicle emission... etc.

Due to poor english, the paper is not clear to readers what the authors have done.

Suggested Title: Statistical/???analysis of chemical composition and their attributing sources of rainwater in Bandung, Indonesia.

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

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**ABSTRACT** : Chemical composition and sources attribution of rainwater in Bandung area, Indonesia was conducted at 4 (four) locations during February to September 2016. The pH of the rainwater ranged from 4.74 to 6.20 (average of 5.45) with Precipitation Weighted Mean (PWM) pH of 4.80 to 4.91. The acidic ions of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were significantly correlated ( $r = 0.870$ ), so were  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ( $r = 0.929$ ) indicating similar sources. Principal component analysis applied to the studied variables extracted three main components accounted for 80.25% of the total variance of the pooled data. Sea-salt  $\text{SO}_4^{2-}$ , ss- $\text{Cl}^-$ ,  $\text{Na}^+$ , ss- $\text{Ca}^{2+}$ , ss- $\text{K}^+$ , nss- $\text{K}^+$  and  $\text{NH}_4^+$  showing a high influence of natural sources related to the contribution of sea salts and crustal sources, suggesting the influence of long-range atmospheric transport anthropogenic activities (human and animal wastes, and biomass burning); the second group consist of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{H}^+$  and nss- $\text{Mg}^{2+}$  likely from vehicular emissions and fossil fuels combustion and the third group contained nss- $\text{Cl}^-$  presumably from other acid reaction. It is concluded that rainwater quality in Bandung is influenced by air pollution from man-made activities.

**Keywords:** acid rain, wet deposition, PCA, rainwater chemistry

## 1. INTRODUCTION

Chemical composition of rainwater was studied in many urban area worldwide e.g. for evaluating the impact of air pollution. Source analysis and contribution from both 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 pollutant because normally pH of rainwater is 5.6, as the value that is expected on the equilibrium of pure water and atmospheric  $\text{CO}_2$  [19]. Acidity variability occurs due to atmospheric chemistry processes involving various sources of air pollutants that are being absorbed and washout by rain droplets.

Alkaline pH in rainwater in Pune region India mainly originated from the abundance of  $\text{Ca}^{2+}$  caused by vehicle driven roadside dust [2]. While acidic rainwater in Shihwa Basin Korea were influenced the dominant ions of  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  and neutralized by  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  [17].

Elevation differences and local anthropogenic activities might account for the variation in the chemical composition of rainwater [13], causing e.g. monthly variations of rainwater chemistry in the Mountain region of Yulong in Southwestern China. Bandung area is located in mountainous region of West Java Indonesia at elevation of 768 meter above sea level. 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 national capital. Its urban activities and various accesses from different cities potentially affect the air quality and hence rainwater to some extended.

Bandung area has a tropical wet and dry climate with average ranges temperature of 18.1 to 31.9°C during 2015 and rainfall average of 184.75 mm with the ranges 0.3 mm/month (in July during the dry season) to 322.4 mm/month (in November in the wet season).

Due the its characteristics, rainwater quality in Bandung area is suggested to be influenced by air pollution. The purposes of the study is to characterize the chemical composition of rainwater. Samples were collected during rainy event in February to September 2016 and analyzed for sources attribution.

## 2. METODOLOGY

### 2.1 Sampling Area

Rainwater samples were collected at four locations, representing different elevation and landuse within Bandung area as are shown in Figure 1 as follow :

- 1) Lembang (06°49'35,01"S 107°37'04,54"E) with altitude about 1.272 m asl lies on the hillside of the mountain area. The site is located approximately 12 km away from the centre of



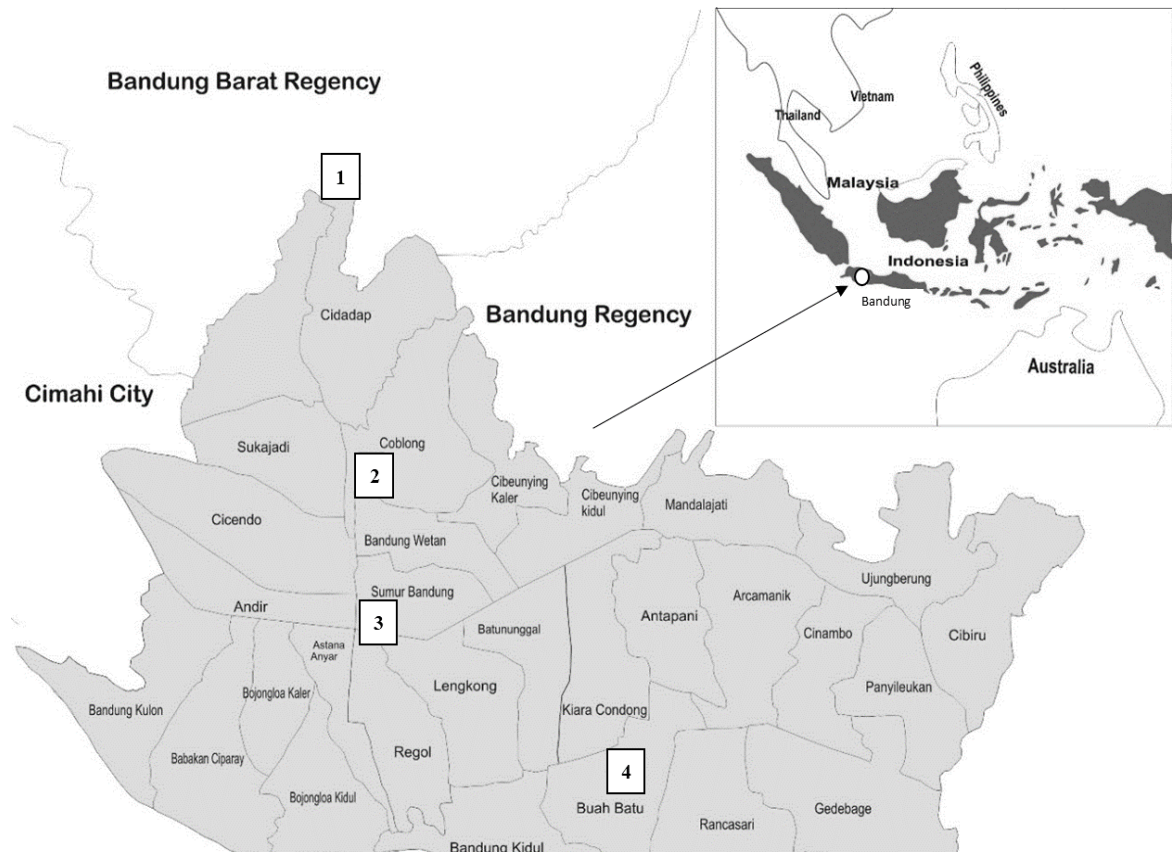


Fig. 1. Sampling Site

- Bandung City, with landuse dominated by agricultural, livestock and tourism activity.
- 2) Coblong (06°55'11,07"S 107°36'04,54"E) with altitude about 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 altitude about 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 altitude about 642 m asl is a sampling site in urban settlement area with high population density and their associated activities.

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. Spatial variation between sampling site was analyzed using *One-way* ANOVA. Sources attribution was analyzed using correlation and Principal Component Analysis (PCA).

## 2.2 Sample Collection and Analysis

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

The pH values and conductivity for the rainwater samples were analyzed with a pH meter (Sartorius Professional Meter PP-20) and a

## 2.1 Quality Control

The quality control of the samples was checked by ionic balance defined as : Ion Percent Difference =  $[(C - A) / (C + A)] \times 100$ , where C and A represents anion and cation equivalents ( $\mu eq L^{-1}$ ), respectively [20].

Precipitation weighted mean (PWM) was carried out by summing the product of ion concentration and the precipitation amount, obtained from rainwater sampler, 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 were largely varied, ranging between 40.69 – 347.66 mm/month. High rainfall events during 2016 suggested as due La Nina impact in this year (Fig.2), with the highest events recorded in March, April and May.

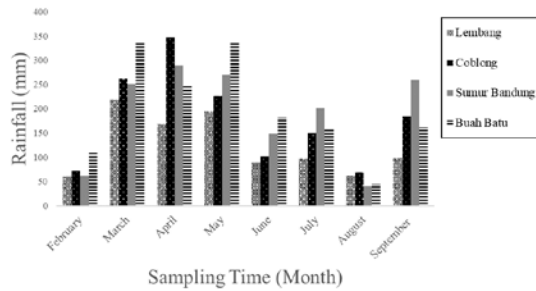


Fig.2 Rainfall Distribution at Four Sampling Site

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 influence of acidic substances in the rainwater [19].

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

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

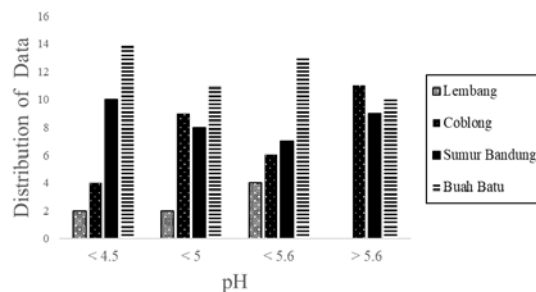


Fig.3 Rainwater pH Distribution

The major precursors of acidity in rainwater were  $SO_2$  and  $NO_x$  as found in sources attribution analysis. Soil particles and sea spray origin species might play an important role in neutralizing  $SO_2$  and  $NO_x$ .

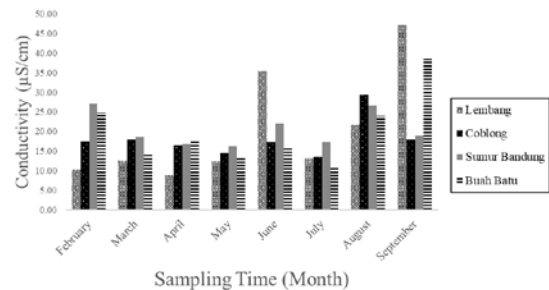
Rainwater conductivity is mainly contributed by water-soluble ions, the value being related to the total sum of anions and cations. The range of electric conductivity for rainwater was from < 0.5 to 100  $\mu S/cm$  (Fig. 4). There is no clear pattern whether conductivity was related to the rainwater amount as the temporal variation of conductivity did not resemble the pattern of rainfall amount in Fig. 1.

Fig.4 Temporal Variation of Conductivity ( $\mu S/cm$ )

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

Precipitation Weighted Mean (PWM) concentrations was calculated to eliminated the dilution effect of rainwater quantity during high rainfall. Temporal variation of PWM concentration of major ions that reflect the variation due to acid chemistry processes in rainwater were shown in Figure 5.

The  $SO_4^{2-}$  and  $NO_3^-$  PWM concentrations showed irregular pattern where their high PWM



concentration were detected in low and high rainfall, might be due to the strength of acid precursor emissions from local and regional sources. Ammonium ( $NH_4^+$ ) PWM concentration and  $Cl^-$ ,  $Na^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$  and  $K^+$  ions showed the increasing trend with decreasing rainfall amount suggesting the dilution of these ions that originated from natural sources.

The strong temporal variation showed by anion  $SO_4^{2-}$  and cation  $NH_4^+$  presumably caused by the neutralization reaction in rainout and washout deposition processes.

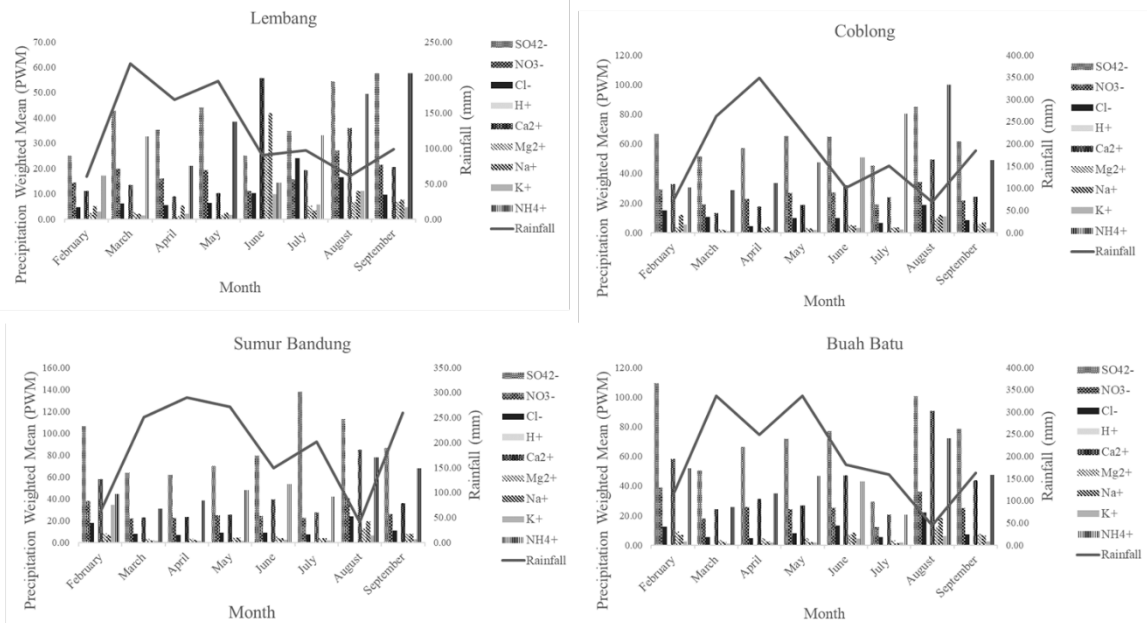


Fig.5 Temporal Variation of Precipitation Weight Mean (PWM) Concentration Ions

### 3.3 Neutralizing Factors

Neutralizing factor (NF) were used to evaluate the neutralization of major ions by crustal components ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) and  $\text{NH}_4^+$ . The neutralization of acids by base cations were evaluated by calculating the neutralization factors using the formula:  $\text{NF}(x) = ((X^+) / (\text{SO}_4^{2-} + \text{NO}_3^-))$  where X is the  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$ .

The NF observed for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$  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). The NF shows  $\text{NH}_4^+$  was dominant neutralizing ion in Bandung area.

### 3.4 Sea salt and Non-Sea Salt Contribution

PWM concentration of sea-salt and non-sea salt contribution were described in Figure 6. Non-Sea salt  $\text{SO}_4^{2-}$  and non-sea salt  $\text{Ca}^{2+}$  were the dominant ions for all sampling site.

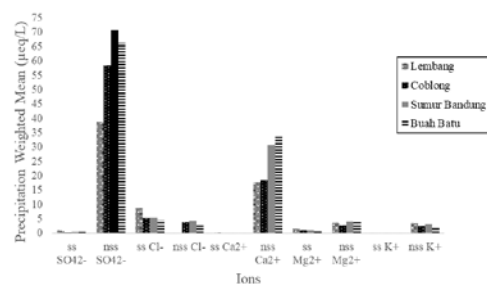


Fig.6 PWM Concentration Sea Salt and Non-Sea Salt Contribution

Non-sea salt sulfate was the highest anion PWM concentration in Bandung area for more than ten years monitoring by EANET (Acid Deposition Monitoring Network in East Asia). The trend of  $\text{SO}_2$  emission 2000 – 2014 in Indonesia increased by 50% (+ 0.12 ppb year<sup>-1</sup> [23]). The observation at the in this study was in line EANET finding.

### 3.5 Sources Attribution Analysis

Statistical analysis were done using log-normal transformed data because the raw data of rainwater amount and ions concentration had positive skewness distribution.

#### 3.5.1 One-Way ANOVA

One-way analysis of variance (ANOVA) was used to analyzed spatial variability of the ions in rainwater chemistry as is shown in Table 1.

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 site ( $\alpha = 0.05$ ).

It was found that concentration of ions nss- $\text{SO}_4^{2-}$ ,  $\text{NO}_3^{2-}$ , nss- $\text{Cl}^-$ , nss- $\text{Ca}^{2+}$ , nss- $\text{Mg}^{2+}$  and  $\text{NH}_4^+$  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
ssSO <sub>4</sub> <sup>2-</sup>	0.252	0.860
nssSO <sub>4</sub> <sup>2-</sup>	<b>11.353</b>	<b>0.000</b>
NO <sub>3</sub> <sup>-</sup>	<b>3.258</b>	<b>0.024</b>
ssCl <sup>-</sup>	0.076	0.973
nssCl <sup>-</sup>	1.104	0.351
H <sup>+</sup>	0.479	0.698
Na <sup>+</sup>	0.252	0.859
ssCa <sup>2+</sup>	0.252	0.860
nssCa <sup>2+</sup>	<b>7.991</b>	<b>0.000</b>
ssMg <sup>2+</sup>	0.088	0.967
nssMg <sup>2+</sup>	<b>2.931</b>	<b>0.037</b>
ssK <sup>+</sup>	0.253	0.859
nssK <sup>+</sup>	0.517	0.671
NH <sub>4</sub> <sup>+</sup>	<b>2.932</b>	<b>0.037</b>

### 3.5.2 Ion Correlation

Correlation between ions is used to determine the sources of different anions and cations in the rainwater and their potential linkages. Ions concentration in rainwater was affected by rainfall amount and that might cause 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<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> that significantly correlated (r = 0.901) might indicated similar sources. Natural rainwater becomes more acidic because sulfuric and nitric acids are formed from acidic gases of SO<sub>2</sub> and NO<sub>3</sub> from anthropogenic activities.

Significant correlation between SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> (r = 0.431) also NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> (r = 0.481) explained that ammonium compound in the atmosphere generally occurs as (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> [19].

On the other hand, significant correlation between Ca<sup>2+</sup> and Mg<sup>2+</sup> (r = 0.801) showed their occurrence from crustal origin.

### 3.5.3 Principal Component Analysis (PCA)

Principal components analysis (PCA) was used to identify and associate possible sources attribution of the species in rainwater chemistry .

As a cut off, factors with an eigenvalue >1 were retained. PCA explained 82.291% of the data variance. Factors are extracted to be interpreted as a common source for the components in the same factor.

Factor 1 explained about 60.75 % of the total variance, with strong positive loadings for sea salt SO<sub>4</sub><sup>2-</sup>, sea salt Cl<sup>-</sup>, sea salt Ca<sup>2+</sup>, sea salt Mg<sup>2+</sup>, Na<sup>+</sup>, sea salt K<sup>+</sup>, and non-sea salt K<sup>+</sup> showing a high influence of natural sources (sea and soil)

Table 2. Partial Correlation Coefficients Major Ions in Rainwater

	nssSO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	ssCl <sup>-</sup>	nssCl <sup>-</sup>	H <sup>+</sup>	Na <sup>2+</sup>	ssCa <sup>2+</sup>	nssCa <sup>2+</sup>	ssMg <sup>2+</sup>	nssMg <sup>2+</sup>	ssK <sup>+</sup>	nssK <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>
ssSO <sub>4</sub> <sup>2-</sup>	.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 <sub>4</sub> <sup>2-</sup>		.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 <sub>3</sub> <sup>-</sup>			.530	.009	.226	.530	.530	.627	.530	.438	.530	.366	.481
			.000	.929	.027	.000	.000	.000	.000	.000	.000	.000	.000
ssCl <sup>-</sup>				-.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 <sup>-</sup>					-.009	-.197	-.196	.048	-.197	.124	-.197	.107	-.095
					.930	.055	.055	.646	.055	.227	.054	.300	.358
H <sup>+</sup>						.109	.110	.062	.109	.041	.109	.075	-.154
						.291	.288	.549	.290	.691	.292	.470	.134
Na <sup>2+</sup>							1.000	.619	1.000	.449	1.000	.617	.430
							.000	.000	.000	.000	.000	.000	.000
ssCa <sup>2+</sup>								.619	1.000	.450	1.000	.617	.430
								.000	.000	.000	.000	.000	.000
nssCa <sup>2+</sup>									.619	.801	.618	.440	.282
									.000	.000	.000	.000	.005
ssMg <sup>2+</sup>										.449	1.000	.617	.430
										.000	.000	.000	.000
nssMg <sup>2+</sup>											.449	.379	.292
											.000	.000	.004
ssK <sup>+</sup>												.616	.430
												.000	.000
nssK <sup>+</sup>													.204
													.046

contribution to Bandung area rainwater. Non-sea salt  $K^+$  in rainwater probably comes from biomass burning [1].  $NH_4^+$  sources from human and animal wastes, also originated from manmade activities (possibly fertilizers, agriculture wastes and urban activities).

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

	Component		
	1	2	3
ss-SO <sub>4</sub> <sup>2-</sup>	<b>0.955</b>	0.272	0.015
nss-SO <sub>4</sub> <sup>2-</sup>	0.304	<b>0.900</b>	0.080
NO <sub>3</sub> <sup>-</sup>	0.355	<b>0.853</b>	-0.008
ss-Cl <sup>-</sup>	<b>0.955</b>	0.272	0.015
nss-Cl <sup>-</sup>	-0.179	0.151	<b>0.822</b>
H <sup>+</sup>	-0.365	<b>0.402</b>	-0.518
Na <sup>+</sup>	<b>0.955</b>	0.272	0.015
ss-Ca <sup>2+</sup>	<b>0.955</b>	0.272	0.015
nss-Ca <sup>2+</sup>	<b>0.639</b>	0.548	0.341
ss-Mg <sup>2+</sup>	<b>0.955</b>	0.272	0.015
nss-Mg <sup>2+</sup>	0.479	<b>0.522</b>	0.427
ss-K <sup>+</sup>	<b>0.955</b>	0.271	0.14
nss-K <sup>+</sup>	<b>0.760</b>	0.168	0.326
NH <sub>4</sub> <sup>+</sup>	<b>0.430</b>	0.376	-0.008
Total Variance (%)	<b>62.750</b>	<b>10.780</b>	<b>8.761</b>

Factor 2 explained about 10.78% of total variance, with strong positive loading for non-sea salt SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, H<sup>+</sup> and non-sea salt Mg<sup>2+</sup> were indicating the anthropogenic sources from vehicular emission, most notably fossil fuels combustion. With hydrogen ion being in the same factor, also acidification as a result. In Bandung area there were ± 1.6 million units of motor vehicle in 2015 which very likely significantly increase emission of SO<sub>2</sub> and NO<sub>3</sub> [24].

Factor 3 explained about 8.76 % of total variance for non-sea Cl<sup>-</sup> probably as a result of reaction of the acid HCl with alkaline compound (Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup>).

#### 4. CONCLUSION

A study of the chemical characteristics of rainwater in Bandung area, Indonesia was conducted in 2016. The rainwater was acidic with an average pH of 4.81 and average conductivity of 20.01 μS/cm. The major anion component was SO<sub>4</sub><sup>2-</sup> (36.2% of the total anion mass), while NH<sub>4</sub><sup>+</sup> was the main cation component (47.7% of total cation mass). The temporal variation of anion SO<sub>4</sub><sup>2-</sup> and cation NH<sub>4</sub><sup>+</sup> PWM concentration

showed these two ions as the highest in almost every month at all sampling sites, compared to other ions. Ammonium (NH<sub>4</sub><sup>+</sup>) was the dominant neutralizing ion in rainwater chemistry in Bandung area.

Sources attribution analysis demonstrated that rainwater in Bandung area were also affected by natural sources with the contribution 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 manmade activities (fertilizer and pesticides wastes). Further studies should be conducted to determine the sources and control of air pollution and implication of the rainwater quality towards its potentials as one of the water resources in urban area.

#### 5. KNOWLEDGEMENTS

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Response by Authors to Reviewer's Remarks/Comments

# Chemical Composition and Sources Attribution of Rainwater in Bandung Area Indonesia

Authors: Yuniarti Hasan, Driejana, Aminudin Sulaeman, and Herto Dwi Ariesyady

The authors have summarized their replies to the Reviewers' comments in this response letter in a two column format. A revised manuscript is submitted addressing all the comments to the Journal of GEOMATE for possible publication.

	<i>Reviewer A's Comments Authors Response</i>	<i>Reviewer A's Comments Authors Response</i>
	Rewrite the Abstract to reflect : Brief Introduction, Brief Justification, Brief Methodology, Brief Result (s) and Brief Conclusion	The authors appreciate the comment from the reviewer A : the abstract has been revised in manuscript
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	Paper Title and the analysis do not march - abstract and conclusions are not clear.	The abstract has revised in manuscript
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The authors appreciate the valuable comments from the Reviewers.





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- a) **Nia Yuniarti Hasan:** conception, design, acquisition, analysis, and interpretation of data and drafting the article.
- b) **Driejana:** general concept, some result interpretation, and refining the article.
- c) **Aminudin Sulaeman:** critical reviewing and final approval of the version to be submitted.
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# CHEMICAL COMPOSITION AND SOURCES ATTRIBUTION OF RAINWATER IN BANDUNG AREA, INDONESIA

Nia Yuniarti Hasan<sup>1</sup>, \*Drijeana<sup>2</sup>, Aminudin Sulaeman<sup>3</sup>, and Herto Dwi Ariesyady<sup>4</sup>

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\*Corresponding Author, Received: 20 May 2019, Revised: 13 Aug. 2019, Accepted: 28 Aug. 2019

**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 ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ , and  $\text{NH}_4^+$ ). The results showed that the pH of rainwater were in the range of acidic to normal (average  $5.42 \pm 0.72$ ), where ammonium acted as the major neutralizing factor (NF). Small-scale spatial variability of  $\text{nss-SO}_4^{2-}$ ,  $\text{NO}_3^{2-}$ ,  $\text{nss-Cl}^-$ ,  $\text{nss-Ca}^{2+}$ ,  $\text{nss-Mg}^{2+}$ , and  $\text{NH}_4^+$  within 4 (four) locations were observed. Sulphate and  $\text{NO}_3^-$  as well as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  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  $\text{SO}_4^{2-}$ ,  $\text{ss-Cl}^-$ ,  $\text{Na}^+$ ,  $\text{ss-Ca}^{2+}$ ,  $\text{nss-Ca}^{2+}$ ,  $\text{ss-K}^+$ ,  $\text{nss-K}^+$  and  $\text{NH}_4^+$ . Secondly, fossil fuel combustion/vehicular emissions, which consisted of  $\text{nss-SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{H}^+$ , and  $\text{nss-Mg}^{2+}$ ; and thirdly,  $\text{nss-Cl}^-$  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  $\text{CO}_2$ , 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  $\text{Ca}^{2+}$  originated from vehicle driven roadside dust [2]. Acidic rainwater in Shihwa Basin Korea was influenced by the dominant ions of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  which were neutralized by  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  [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  $18.1^\circ\text{C}$  to  $31.9^\circ\text{C}$  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.

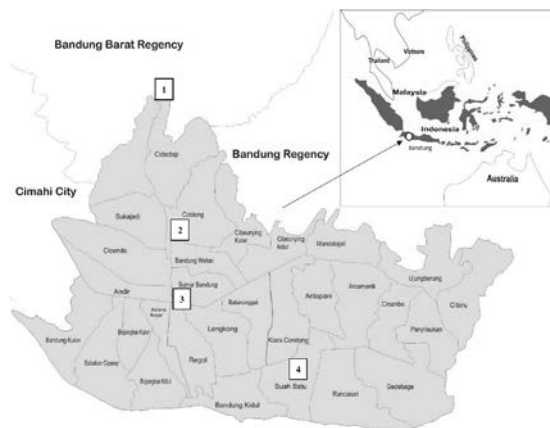


Fig. 1. Sampling Site

### 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 ( $\text{SO}_4^{2-}$ ), nitrate ( $\text{NO}_3^-$ ), chloride ( $\text{Cl}^-$ ), calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ), sodium ( $\text{Na}^+$ ), potassium ( $\text{K}^+$ ), and ammonium ( $\text{NH}_4^+$ ) 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  $\text{IPD} = [(C - A) / (C + A)] \times 100$ , where, C and A represents anion and cation equivalents ( $\mu\text{eq L}^{-1}$ ), 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.

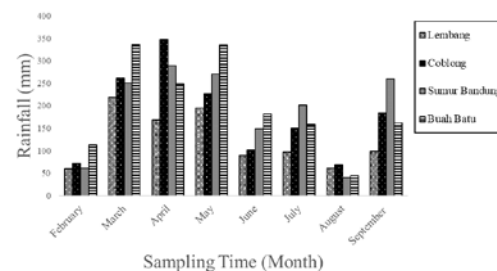


Fig.2 Rainfall Distribution at Four Sampling Sites

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).

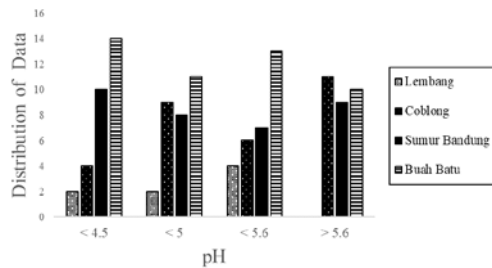


Fig.3 Rainwater pH Distribution

The major precursors of acidity in rainwater were SO<sub>2</sub> and NO<sub>x</sub> 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<sub>2</sub> and NO<sub>x</sub>. 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.

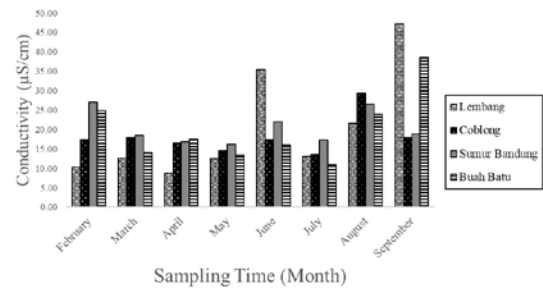


Fig.4 Temporal Variation of Conductivity (μS/cm)

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

Precipitation-Weighted Mean (PWM) concentrations were calculated to eliminate the dilution 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<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> PWM concentrations showed irregular patterns. Particularly for SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>, 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<sub>4</sub><sup>+</sup>) as well as Cl<sup>-</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup> 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<sub>4</sub><sup>2-</sup> and cation NH<sub>4</sub><sup>+</sup> were presumably caused by the neutralization reaction in the rainout and washout deposition processes.

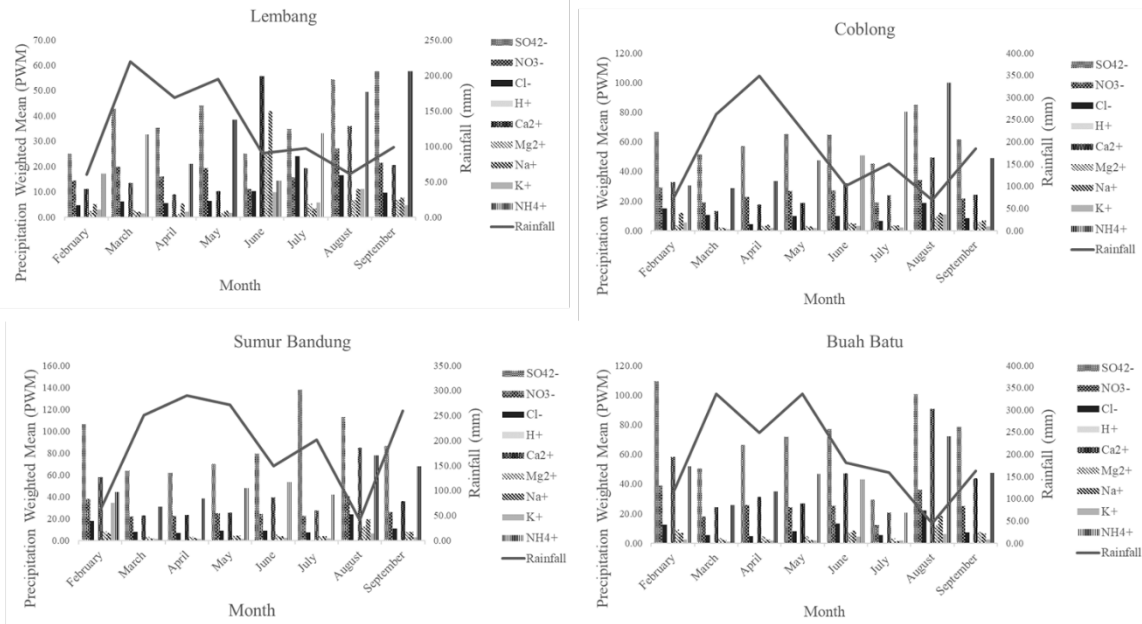


Fig.5 Temporal Variation of Precipitation Weight Mean (PWM) Concentration Ions

### 3.3 Neutralizing Factors

Neutralizing factor (NF) was used to evaluate the neutralization of major anions by crustal components ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) and  $\text{NH}_4^+$ . The neutralization of acids by base cations were evaluated by calculating the neutralization factors using the formula:  $\text{NF}(x) = ((X^+) / (\text{SO}_4^{2-} + \text{NO}_3^-))$  where X is the  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$ .

The NF observed for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$  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  $\text{NH}_4^+$  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  $\text{SO}_4^{2-}$  and nss- $\text{Ca}^{2+}$  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  $\text{SO}_2$  emission during the 2000 – 2014 period in Indonesia increased by 50% (+ 0.12 ppb year<sup>-1</sup>) [23]. The observation found in this study was in line with the EANET finding.

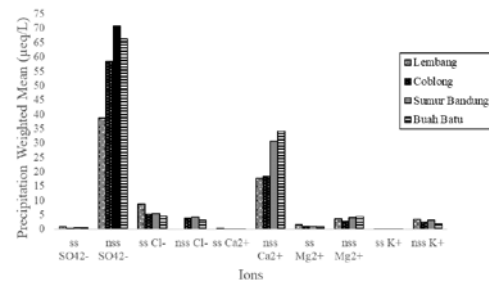


Fig.6 PWM Concentration of Sea Salt and Non-Sea Salt Contribution

### 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- $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , nss- $\text{Ca}^{2+}$ , nss- $\text{Mg}^{2+}$ , and  $\text{NH}_4^+$  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 <sub>4</sub> <sup>2-</sup>	0.252	0.860
nss-SO <sub>4</sub> <sup>2-</sup>	<b>11.353</b>	<b>0.000</b>
NO <sub>3</sub> <sup>-</sup>	<b>3.258</b>	<b>0.024</b>
ss-Cl <sup>-</sup>	0.076	0.973
nss-Cl <sup>-</sup>	1.104	0.351
H <sup>+</sup>	0.479	0.698
Na <sup>+</sup>	0.252	0.859
ss-Ca <sup>2+</sup>	0.252	0.860
nss-Ca <sup>2+</sup>	<b>7.991</b>	<b>0.000</b>
ss-Mg <sup>2+</sup>	0.088	0.967
nss-Mg <sup>2+</sup>	<b>2.931</b>	<b>0.037</b>
ss-K <sup>+</sup>	0.253	0.859
nss-K <sup>+</sup>	0.517	0.671
NH <sub>4</sub> <sup>+</sup>	<b>2.932</b>	<b>0.037</b>

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<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> 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<sub>2</sub> and NO<sub>3</sub> from anthropogenic activities.

Significant correlation between SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> (r = 0.431) also NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> (r = 0.481) indicated that ammonium exists in the atmosphere as (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> [19]. On the other hand, a significant correlation between Ca<sup>2+</sup> and Mg<sup>2+</sup> (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 <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	ssCl <sup>-</sup>	nssCl <sup>-</sup>	H <sup>+</sup>	Na <sup>2+</sup>	ssCa <sup>2+</sup>	nssCa <sup>2+</sup>	ssMg <sup>2+</sup>	nssMg <sup>2+</sup>	ssK <sup>+</sup>	nssK <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>
ssSO <sub>4</sub> <sup>2-</sup>	.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 <sub>4</sub> <sup>2-</sup>		.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 <sub>3</sub> <sup>-</sup>			.530	.009	.226	.530	.530	.627	.530	.438	.530	.366	.481
			.000	.929	.027	.000	.000	.000	.000	.000	.000	.000	.000
ssCl <sup>-</sup>				-.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 <sup>-</sup>					-.009	-.197	-.196	.048	-.197	.124	-.197	.107	-.095
					.930	.055	.055	.646	.055	.227	.054	.300	.358
H <sup>+</sup>						.109	.110	.062	.109	.041	.109	.075	-.154
						.291	.288	.549	.290	.691	.292	.470	.134
Na <sup>2+</sup>							1.000	.619	1.000	.449	1.000	.617	.430
							.000	.000	.000	.000	.000	.000	.000
ssCa <sup>2+</sup>								.619	1.000	.450	1.000	.617	.430
								.000	.000	.000	.000	.000	.000
nssCa <sup>2+</sup>									.619	.801	.618	.440	.282
									.000	.000	.000	.000	.005
ssMg <sup>2+</sup>										.449	1.000	.617	.430
										.000	.000	.000	.000
nssMg <sup>2+</sup>											.449	.379	.292
											.000	.000	.004
ssK <sup>+</sup>												.616	.430
												.000	.000
nssK <sup>+</sup>													.204
													.046

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

	Component		
	1	2	3
ss-SO <sub>4</sub> <sup>2-</sup>	<b>0.955</b>	0.272	0.015
nss-SO <sub>4</sub> <sup>2-</sup>	0.304	<b>0.900</b>	0.080
NO <sub>3</sub> <sup>-</sup>	0.355	<b>0.853</b>	-0.008
ss-Cl <sup>-</sup>	<b>0.955</b>	0.272	0.015
nss-Cl <sup>-</sup>	-0.179	0.151	<b>0.822</b>
H <sup>+</sup>	-0.365	<b>0.402</b>	-0.518
Na <sup>+</sup>	<b>0.955</b>	0.272	0.015
ss-Ca <sup>2+</sup>	<b>0.955</b>	0.272	0.015
nss-Ca <sup>2+</sup>	<b>0.639</b>	0.548	0.341
ss-Mg <sup>2+</sup>	<b>0.955</b>	0.272	0.015
nss-Mg <sup>2+</sup>	<b>0.479</b>	<b>0.522</b>	0.427
ss-K <sup>+</sup>	<b>0.955</b>	0.271	0.14
nss-K <sup>+</sup>	<b>0.760</b>	0.168	0.326
NH <sub>4</sub> <sup>+</sup>	<b>0.430</b>	0.376	-0.008
Total Variance (%)	<b>62.750</b>	<b>10.780</b>	<b>8.761</b>

Factor 1 explained approximately 60.75 % of the total variance, with strong positive loadings for sea salt SO<sub>4</sub><sup>2-</sup>, sea salt Cl<sup>-</sup>, sea salt Ca<sup>2+</sup>, sea salt and non-sea salt Mg<sup>2+</sup>, Na<sup>+</sup>, sea salt and non-sea salt K<sup>+</sup> indicating the natural source (sea and soil) contributions to rainwater. In addition, non-sea salt K<sup>+</sup> in rainwater presumably comes from biomass burning [1]. NH<sub>4</sub><sup>+</sup> 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<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, H<sup>+</sup>, and non-sea salt Mg<sup>2+</sup> 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<sub>2</sub> and NO<sub>3</sub> emissions [24].

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

#### 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<sub>4</sub><sup>2-</sup> (36.2% of the total anion mass), while NH<sub>4</sub><sup>+</sup> was the main cation component (47.7% of total cation mass). The major components found that, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> PWM concentrations were the highest at all sampling sites in almost every month, compared to other ions.
3. Ammonium (NH<sub>4</sub><sup>+</sup>) was found to be the dominant neutralizing ion.
4. Small-scale spatial variability found for nss-SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>2-</sup>, nss-Cl<sup>-</sup>, nss-Ca<sup>2+</sup>, nss-Mg<sup>2+</sup>, and NH<sub>4</sub><sup>+</sup>, 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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