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Research article

Rainwater chemistry of acid precipitation occurrences due to long-range transboundary haze pollution and prolonged drought events during southwest monsoon season: climate change driven

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ABSTRACT

The purposes of this research were to study the characteristics chemistry of pH, anions and cations in rainwater, and to identify the possible sources that contributing to the acid precipitation during southwest monsoon season with occurrence of extreme drought event. During the southwest monsoon season, it normally occurs along with haze phenomenon that every year will hit Southeast Asia. This condition will aggravate with high acidic particles in the atmosphere due to the prolonged drought. The analysed parameters which involved pH, anions (NO_3^-, SO_4^{2-} and Cl^-) and cations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) were analysed using pH meter, Hach DR 2800, argentometric method and ICP-OES. From the findings, it showed that acid rain occurred during the southwest monsoon season with the range of pH values from 4.95 ± 0.13 to 6.40 ± 0.03 and the total average of pH 5.71 ± 0.32. Anions NO_3^-, SO_4^{2-} and Cl^- were found to be the dominant compositions of the acid rain occurrences with higher concentrations detected. In overall, rural area recorded with higher acidity of precipitation at total average of pH 5.54 ± 0.39 compared to urban area at pH 5.77 ± 0.26. Rural area surprisingly recorded higher frequency occurrences of acid rain with pH lesser than 5.6 and below compared to urban area. As for public health and safety, all rainwater samples during the acid rain event were found exceeded the allowable limits of NWQS and WHO standards, that shown not suitable for skin contact, recreational purposes even for drinking purposes.

1. Introduction

Acid precipitation is defined as the atmospheric acids which are deposited on the earth in the form of wet deposition or dry deposition [1]. In this research, the monitored acid rain is referring to wet deposition. The occurrence of acid rain is determined by the pH of the rainfall. The rain that falls to the ground having the pH lower than 5.6 is considered acid rain [2]. For anthropogenic emissions, the typical pH values of acid rain will be in the range of 3.5–5 [3]. The sources of acidity in rain can come from anthropogenic or natural activities. Even so, anthropogenic sources are the main contributors of acid rain in urban area. Anthropogenic activities such as burning of fossil fuels from transportation or industrial activities will release sulphuric acid and nitric acid into the atmosphere that are known as the inorganic acids [2, 4, 5, 6]. Meanwhile, the sources of rain acidity in rural area will come from biogenic volatile organic carbon emissions are such as isoprene, propene, acetic acid, formic acid and oxalic acid that are more organic acids [4]. According to [7], biogenic volatile organic carbon emissions may not directly emit in the form organic acids, but they may form in the atmosphere through secondary reactions. From that point, it means that the acidity of rainwater may not be necessarily from the urban like pollutions such as automobile emissions and industrial combustions, but it can come from natural source such as the biogenic volatile organic emissions from forests, vegetation and livestock manure [8]. These sources will increase the atmospheric acidity in the rural area that usually under-estimated and rarely been discussed by other researcher, as it always assumed that acid rain phenomena only occur in a busy city by heavy traffic congestions and industries activity.

Rain is a natural phenomenon that falls to the earth in the form of water droplets. Rain can range from light to heavy and sometimes occur as extreme event. The occurrence of rain in a region is largely affected by monsoon, a prevailing wind blown from a place in globe depending on

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the season [9]. During southwest monsoon season, the condition is expected to be drier compared to northeast monsoon season [10]. The Australian continent which experiences winter has low air temperature hence creates high pressure in contrast with the Asian continent that experiences summer during southwest monsoon. This causes the wind from Australia to blow crossing the equator and deflecting towards northeast, ultimately arriving in Southeast Asia. Furthermore, Sumatra and Kalimantan, Indonesia are experienced on large-scale fire outbreaks during El-Nino – Southern Oscillation (ENSO) phenomenon due to dry conditions and warm temperatures [11, 12]. According to [13], the annual climate during southwest monsoon season is largely impacted by ENSO phenomenon causing prolonged drought event. The wind which blows southerly from Sumatra and Kalimantan, Indonesia facilitates long range transportation of smokes from the fires event and prolong duration of poor air quality that contribute to haze phenomenon. According to [14] and [15], haze particles contain acid aerosols such as SO_2 and NO_2. In the meantime, normal fires event may also emit acidic particles such as NOx to the atmosphere. Thus, this show dry periods with prolonged drought during southwest monsoon put high at risk with acid rain occurrence.

Determining the composition of rainwater provides a major input sources of several elements to from the earth's surface [16]. Besides, study of the rainwater chemistry is important in order to quantify and qualify atmospheric contaminants to prevent damages to natural ecosystems through degradation of air quality, acidification of water bodies and soils, causing imbalances in the nutrient cycle and ecosystem productivity, as well as health issues [17]. The composition of rain is controlled by complex processes and it is influenced by both natural and anthropogenic activities [18]. Anions which includes nitrate (NO_3^-), sulphate (SO_4^{2-}), and chloride (Cl^-) are negatively charged substances that present in aqueous solution. According to [19], the presence of acidic substances in rainwater is indicated by concentration of anions. The amount of acid compound in rainwater would affect the cycling of nutrients in the environment [20]. Meanwhile, cations are positively charged substances of calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+) and potassium (K^+), that presence of more alkaline substances in rainwater that indicated by concentration of the cations. The anthropogenic sources of anions and cations can be from fossil fuel combustion, biomass burning, soil properties and rock weathering, including fertilizers, forest fires, wind-blown dust and sea-water breeze. In [21, 22] explained that concentrations of major ions in rainwater samples were higher in the dry season compared to rainy season, due to the high loads of compound trapped in the atmosphere for a longer period during dry season such as southwest monsoon season.

Deterioration of forests, acidification of lakes and grounds, marble decaying, and the degradation of building and ancient monuments are the potential ecological deterioration of acid rain phenomenon [23]. Besides, the effect of acid rain which includes the disruption in leaf physiology and plant growth due to leaching of nutrients in the soil [24]. Soil acidification can cause microbial activity in soil reduced or completely inhibited [25]. Acidification of lakes and rivers can cause loss of aquatic organism population [26]. Sudden changes in pH can cause birth defects to aquatic species such as amphibian, fish and insects. Aquatic organism will stop hatching due to acidity in aquatic environment affecting their population ratios and ecosystem [27]. Plants are also affected by acidification of aquatic ecosystems where acidification changes the rates and amounts of primary production, nutrient cycling and decomposition. Whilst, acid rain can affect human health by causing respiratory problems such as asthma, dry coughs, headache and throat irritation [27]. Human will experience brain damage, kidney problems, Alzheimer's disease by consuming the toxic plants or animals affected by acid rain pollutants [28]. Moreover, it is important to note that until today there is still no standard enacted in the law or regulation in the world for acid rain or at least a guideline for reference of the acidity of precipitation. There is also very low interest and only few studies been done on the effects of acid rain and it is very limited. Therefore, there is a great concern in need for this type of research to be conducted; to determine the occurrence of acid rain especially during dry season with prolonged drought and haze event. At the same time, by determining the composition of the rainwater chemistry, it will provide an insight on the possible sources of air pollutant that contributing to the acid precipitation.

2. Materials and methods

2.1. Study area

The study area is focussed in the Northern part of Borneo in Sabah. There were six major sampling areas involved in this research that were divided into two main clusters throughout the state represented the urban and rural areas as shown in Figure 1 and Figure 2. The study area of Sabah was chosen as it is located right at the centre of the haze event where the phenomena can be found reoccurring. At the same time, the study area is exposed to prolonged drought yearly during southwest monsoon season. Meanwhile, majority of Sabah's boundary were

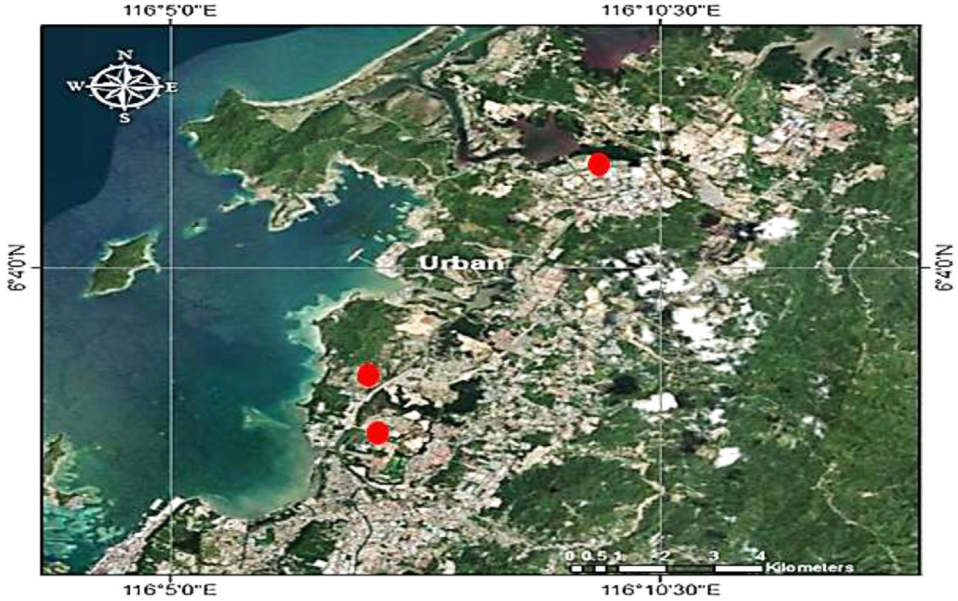

Figure 1. The 3 sampling areas for urban of this research (Source: ArcMap 10.4.1, 2015).

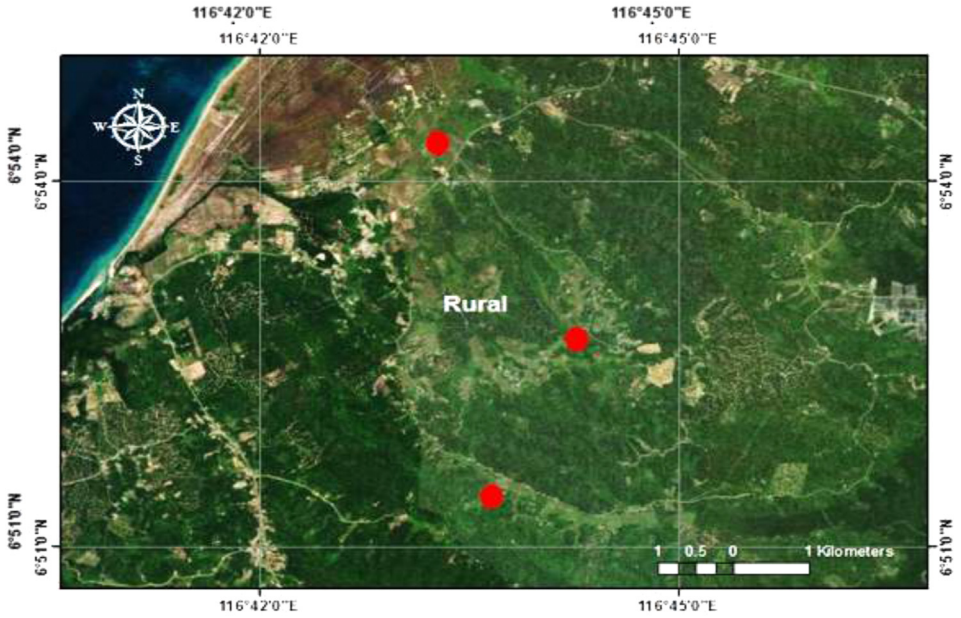

Figure 2. The 3 sampling areas for rural of this research (Source: ArcMap 10.4.1, 2015).

surrounded by sea. This makes the study area more exposed to the long-ranged transportation of pollutants from other continent, and it was interesting to determine whether the rainwater composition of the studied area was contributed by trans-boundary pollution during southwest monsoon. Subsequently, sea-spray can also be a contributor to acidity in atmosphere [16, 26]. Apart from that, Sabah is also largely populated, urbanized with many man-made structure, high traffic congestions, many industrial activities and the ambient temperature could be having an extreme change during the day, which may cause resulting effect on recirculation and build-up concentration of the urban air pollutants [29]. At the same time, due to the wider and bigger stretched island of the study area, in few hundred kilometres away from capital city, rural area that are less urbanized with low traffic can be found. Such area would be the focus of the study to determine the natural origin and other sources such as agricultural burning and forest fires that contribute to acid rain phenomenon in the region. The sampling at each sites of study area was done when rain events occurred.

2.2. pH, anions and cations of rainwater analysis

The pH, anions (NO_3^-, SO_4^{2-} and Cl^-) and cations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) were analysed. For In-situ parameter, pH of the rainwater was measured using electrometric method to detect small pH changes to the sampled rainwater. Separate polyethylene sampling bottles for each different parameter such as anions and cations were used. Other than that, further analysis was carried out using argentometric method for Cl^- and Hach method for NO_3^- and SO_4^{2-} to measure anions and Inductively Coupled Plasma- Optical Emission Spectrometry (ICP-OES) analysis was performed to measure the cations compositions of Na^+, K^+, Mg^{2+} and Ca^{2+} in the sampled rainwater. For Hach method analysis, the preparation of sample for NO_3^- analysis started with filling 10 ml of the rainwater samples stored in 4 °C storage into a square sample cell. Then, the sample was added with NitraVer 5 Nitrate Reagent Powder Pillow and stoppered with a parafilm. The stopper was set to 1 min and shaken vigorously during the period. When the timer had expired, the sample cell left for 5 min for reaction to take place. The blank preparation was prepared in a second square cell of 10 ml distilled water. Both sample cells wiped. The blank sample was inserted into cell holder and then followed by the first sample. Result was obtained. The same steps were applied for SO_4^{2-} analysis, but no preservation solution was added. For sulphate, the reagent used was SulfaVer 4 powder pillow and the sample was swirled after reagent addition [30, 31]. For Cl^- analysis which electrometric method was performed, where the rainwater sample from 4 °C storage and a blank sample (distilled water) was prepared. The volume for both samples was measured using measuring cylinder and the reading was recorded. Then, 50 ml of rainwater and blank samples were poured into separate 250 ml size conical flask. 1 ml of K_2CrO_4 (potassium chromate) solution was added into each conical flask as indicator and a blank white paper was put under the conical flask on the retort stand. In this study, since the rainwater had pH around 5, 1 ml of NaOH solution was added into the rainwater sample in the conical flask. After that, the solution in each conical flask was titrated with (silver nitrate) $AgNO_3$. While titrating, the conical flask was stirred slowly to mix the solutions and stopped when solutions turning reddish brown. The needed volume of $AgNO_3$ to be titrated into the sample solutions to turn from clear to reddish brown was recorded. The chloride concentration was determined using the formula [31]:-

$$\textbf{Chloride} = [(\mathbf{S} - \mathbf{B}) \times \mathbf{M} \times \mathbf{35450}] \cdot \mathbf{V}$$

S = volume of titration for sample (ml)
B = volume of titration for the blank (ml)
V = volume of sample used (50 ml)
M = molarity of $AgNO_3$ (0.0141 M)

For the Na^+, K^+, Mg^{2+} and Ca^{2+} analyses, the samples were measured using ICP-OES analysis, cylinders were measure and the readings were recorded. The water sample was filtered through a 0.45 μm of sterile membrane filter [32] and transferred into centrifuge tubes. The rainwater samples were then measured by ICP-OES and the absorbance value for samples were recorded. The concentration of Na^+, K^+, Mg^{2+} and Ca^{2+} for each sample was determined through the absorbance value by the standard calibration graph. The wavelengths of the cations are Na^+ (589.592), K^+ (766.490), Mg^{2+} (285.213) and Ca^{2+} (317.933). After analysis, the data obtained were analysed using statistical analysis SPSS version 26 to determine the relationship between rainwater ions compositions and the origin sources of the acid precipitation, thus the air pollutants. The data obtained was statistically analysed by Pearson Correlation Coefficient using SPSS to see the significant contributions of the different ions' concentrations with occurrence of acid precipitation in different locations of studied area for comparison [26]. Backward trajectory analysis by Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was run to identify the sources of air masses and to build source-receptor relationships. Back trajectory analysis in HYSPLIT

model allows determination of long-range transportation of pollutants including such transboundary pollution. In this study, HYSPLIT trajectory model was used and archive trajectory was computed [12, 13, 26]. Meteorological data used was GDAS (1°, global, 2006-present). After that, the coordinates of the location to analyse were key in and the archive trajectory is shown. The data on desired date was selected. Model run details used are backward trajectory direction, total run time 72 h, height of above ground level (AGL) at 500 m. Fire hotspots map was also generated to see which regions with active fires on specific date and correlated with backward trajectory analysis. Further analysis of the obtained data has been conducted for the sampled rainwater, by comparing to the National Water Quality Standard (NWQS) and World Health Organization (WHO) Standard to detect whether the rainwater from acid precipitation event is rather safe for skin contact, recreational purposes even for drinking purposes that is very important to public health assessment, especially during Southwest Monsoon Season with Prolonged Drought and Haze Event.

3. Results and discussions

3.1. Rainwater pH, ions concentrations and compositions

Overall, the findings show the acid rain occurred during the southwest monsoon season with the total average of pH recorded at 5.71 ± 0.32, with range of pH values from 4.95 ± 0.13 to 6.40 ± 0.03. In overall, rural area recorded with higher acidity of precipitation at total average of pH 5.54 ± 0.39 compared to urban area at pH 5.77 ± 0.26. This summarise the highest acidity with pH 4.95 ± 0.13 was recorded at rural area and in contrast, the lowest acidity with pH 6.40 ± 0.03 was reported at urban area. Table 1 summarize the findings of the research. For the total average of ions concentrations were recorded at NO_3^- (3.75 ± 3.20 mg/L), SO_4^{2-} (1.49 ± 0.63 mg/L) Cl^- (3.70 ± 2.82 mg/L) for anions and Na^+ (1.17 ± 0.70 mg/L), K^+ (0.38 ± 0.21 mg/L), Mg^{2+} (0.15 ± 0.11 mg/L) and Ca^{2+} (1.16 ± 0.62 mg/L) for cations concentrations. Whilst, in overall total percentage of ions compositions in the acid precipitation is shown in Figure 3, with anions NO_3^-, Cl^- and SO_4^{2-} were found to be the dominant compositions of the acid rain occurrences, at 32%, 31% and 13% respectively, followed by Ca^{2+} (10%), Na^+ (10%), K^+ (3%) and Mg^{2+} (1%).

Anions NO_3^- and SO_4^{2-} were found to be the dominant compositions of the acid rain occurrences for both studied areas, urban and rural areas, with higher concentrations detected. According to [6], the possible reasons that lead to acidity in rain of urban area are rapid industrialisation, urbanisation and rapid increasing of automobiles, through fossil fuel combustion, vehicles emit SO_2 and NO_x into atmosphere [33]. Large amount of SO_2 and NO_x in the atmosphere can lowering the rainwater pH during precipitation event [34].

Further discussion on the phenomenon of acid rain occurrence in the rural area which was found unlikely from anthropogenic activities such as vehicular and industrial activities that emit SO_2 and NO_x that were found at low concentrations. In rural, Cl^- found higher due to the HCl that was formed in the atmosphere if a cloud has a higher temperature especially during drought period and more acidic gaseous molecules in the environment, and HCI is highly dissolvable in water, and thus subsequent absorption of HCl in raindrops may reduce the pH of rainwater [35]. Subsequently, Cl^- derived from sea salt particles was found to react with SO_3 and NO_2 generating HCl in the atmosphere [36]. In addition [37], added that HCl emitted from tropical biomass burning during dry periods when there is less rain occurrence. Further [38], mentioned that warming effect could be produced from the mixing of Cl^- with light-absorbing substances such as soot and nitroaromatic compounds, humic-like substances.

Moreover, according to [2, 33], rainwater acidity and pH can be affected by the amount of rainfall due to the ion's concentrations dilution effect from the rain event. On the other hand, other possibility might cause from the cations that acts as neutralisation agent that balance the concentrations between the anions in rainwater. Apart from that [39], also stated atmospheric scavenging which is the clean-up process of atmospheric aerosols by precipitation may cause alteration of the rainwater pH. With this regard, it's added that normal rainwater pH tends to be around 5.6 due to dissolution carbon dioxide (CO_2) in rain droplets even when the atmosphere is clean [21], and below than 5.6 is called acid rain phenomenon. In Figure 1, it also shows the percentage of rainwater pH by the acidity frequency, and as for total overall it shows pH below than 5.6 recorded at 33% for pH < 5 and 55% for pH 5.0 to 5.9. It also shows that rural area had pH lower than 5 (33%) compared to urban area that had mostly normal rainwater pH range 5.6 and above. As for public health and safety, all rainwater samples when compared with the allowable limits of NWQS and WHO standards (pH ranged from 6.5 to 8.5), were found exceeded the permissible level, and this shown during the acid precipitation is not suitable for skin contact, recreational purposes even for drinking purposes.

3.2. Relationship of acid precipitation and ions concentrations

In this study statistical evaluation between the pH and ions composition in rainwater were performed using Pearson Correlation Coefficients analysis, in order to characterize the effect of major ions

Table 1. The pH values and ions concentration (mg/L) of cations and anions in rainwater.

Parameter	Min Value	Max value	Mean	
pH	5.50 ± 0.02	6.40 ± 0.03	5.77 ± 0.26	Urban
NO_3^-	0.80 ± 0.23	8.29 ± 5.42	4.13 ± 3.18	
SO_4^{2-}	0.33 ± 0.00	9.56 ± 11.35	1.86 ± 0.03	
Cl^-	0.50 ± 0.00	6.00 ± 4.36	2.91 ± 2.19	
Ca^{2+}	0.31 ± 0.17	8.85 ± 3.00	1.39 ± 0.05	
Mg^{2+}	0.032 ± 0.01	0.27 ± 0.02	0.14 ± 0.11	
Na^+	0.32 ± 0.05	2.24 ± 0.78	0.97 ± 0.55	
K^+	0.13 ± 0.00	1.68 ± 1.23	0.41 ± 0.27	
pH	4.95 ± 0.13	6.01 ± 0.28	5.54 ± 0.39	Rural
NO_3^-	0.78 ± 0.29	6.04 ± 7.44	2.80 ± 1.08	
SO_4^{2-}	0.33 ± 0.00	1.11 ± 0.19	0.58 ± 0.22	
Cl^-	2.67 ± 1.26	10.00 ± 4.82	5.65 ± 3.24	
Ca^{2+}	0.20 ± 0.08	1.90 ± 1.18	0.60 ± 0.55	
Mg^{2+}	0.09 ± 0.01	0.32 ± 0.02	0.18 ± 0.08	
Na^+	0.72 ± 0.14	3.28 ± 0.29	1.55 ± 0.77	
K^+	0.15 ± 0.07	0.40 ± 0.47	0.30 ± 0.17	

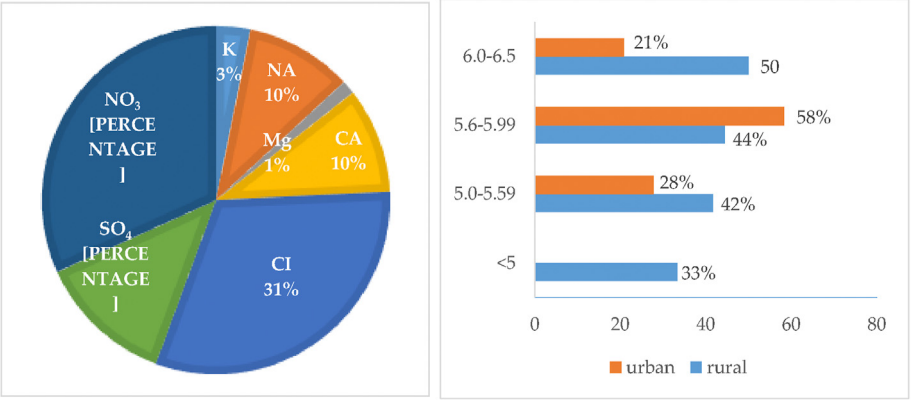

Figure 3. Overall total percentage (%) of ions compositions (a); pH frequency (%) of rainwater (b).

concentrations in rainwater with the pH, and at the same time to identify the sources of measured ions [29] as shown in Table 2 for urban studied area and Table 3 for rural.

From the study, it is shown the main contributors of anions and cations onto the acid precipitation in the urban area studied are highly from, Mg^{2+} (0.903**), Na^{+} (0.849**), K^{+} (0.814**), Ca^{2+} (0.740*) and Cl^{-} (0.709*) respectively. From this point, cation Mg^{2+} shown the dominant contributor to the rain ion composition for urban area. It can be seen the major pollutants ions from NO_3^- and SO_4^{2-} for all the urban sampling locations indicated no significant contributor to the rainwater ions composition. This indicated that the behaviour in precipitation and the co-emission of the precursor NO_x and SO_2 from vehicles and industrial emissions are not one of the anthropogenic sources in the urban area. Meanwhile, reported by [40] stated that the reaction of sea salt particles (NaCl) in the atmosphere form $NaNO_3$ will release Cl^{-}, thus this explained the higher correlations coefficients of Na^{+} and Cl^{-} ions in rainwater. In [41] also added that the Mg^{2+} may come from $MgCl_2$ salt, of the sea vicinity origin, this explained higher Mg^{2+} in the sampled rainwater which derived mostly from sea particles same as Cl^{-}. Past study by [42] showed high correlations of K^{+}, Ca^{2+}, Mg^{2+}, Na^{+} and K^{+} are resulting from the occurrence of biomass burning, earth crust materials, soil dust, road dust from constructions activity and natural process such as oxidation by microbes in soil releasing nutrient gaseous to the atmosphere. In contrast with the rural area, NO_3. (0.956**) turns out to be significant and contribute to ions composition in the rainwater compared to urban area. Subsequently, significant strong correlation of Cl^{-} (-0.985**), Na^{+} (-0.984*) and Mg^{2+} (-0.958*) in the rural area indicates that largely contributed from marine origin, and strong correlation of SO_4^{2-} (-0.515), K^{+} (-0.667*), Cl^{-} (-0.985**) and Ca^{2+} (-0.721*) resulting from biomass burning throughout the drought season and haze event.

3.3. Backward trajectory and fire hotspots analysis

In this study, the rainwater observed with pH below 5.00 was further analysed with backward trajectory analysis, in order to identify the origins of sources by long-range transportation, air mass trajectories of 72-hours were computed using Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT). The 72-hours backward trajectory time was chosen as [43] mentioned that air masses usually takes two to three days transport time. In the HYSPLIT analysis, 500 m above ground level (AGL) was used. According to [17], trajectories with lower than 500-meter altitudes are subjected to interferences from hills and valleys and whereas trajectories at higher altitudes exceed the mixing layer of compounds of interest. Subsequently, fire hotpot map which shows active fires areas also generated. The data used in generating the fire hotspot map is VIIRS Active Fire from NASA. In Figure 4(a) it shows the air masses were influenced by southwest monsoon where it travelled from Bau, Sarawak (Western part of Borneo) and blown to the North towards the ocean. Figure 4(b) shows the air masses took 30 h on the ocean before being blown towards northeast direction passing through the region of Sarawak and Brunei to finally reach studied area.

Subsequently, it shows significantly from Figure 4 that the air masses were blown from west Kalimantan, Indonesia and blown north-westerly towards the ocean. The air masses took 18 h on the ocean before being blown easterly and bend north-easterly towards the studied area. Based on the HYSPLIT analysis projected at different time, it shows the air masses might pick up some particles in the atmosphere from the region of Sarawak, Brunei, and West Kalimantan. In the meantime, it was possible that the air masses also pick up some sea particles and landed at the studied area. Figure 5 shows fire hotspots analysis, by yellow colour grid on the map indicates fire counts lower than 100. By comparing both Figures 4 and 5, it shows the air masses have reached the studied area that originated from area with fires, primarily due to biomass burning. Therefore, there

Table 2. Correlation coefficients between major ionic concentrations in rainwater of urban area.

	NO_3^-	SO_4^{2-}	Cl^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	pH
NO_3^-	1.000							
SO_4^{2-}	0.765*	1.000						
Cl^-	0.096	-0.024	1.000					
Ca^{2+}	-0.180	-0.159	0.482	1.000				
Mg^{2+}	0.096	0.183	0.617*	0.868**	1.000			
Na^+	0.392	0.481	0.687*	0.603*	0.879**	1.000		
K^+	0.406	0.458	0.614	0.735*	0.855**	0.829*	1.000	
pH	**0.290**	**0.178**	**0.709***	**0.740***	**0.903****	**0.849****	**0.814****	**1.000**

*: Correlation is significant at $P < 0.05$ (two-tailed). **: Correlation is significant at $P < 0.01$ (two-tailed).

Table 3. Correlation coefficients between major ionic concentrations in rainwater of rural area.

	NO_3^-	SO_4^{2-}	Cl^-	Ca^{2+}	Mg^{2+}	Na^+	K^+	pH
NO_3^-	1.000							
SO_4^{2-}	-0.744	1.000						
Cl^-	-0.992	0.655	1.000					
Ca^{2+}	-0.893	0.965	0.830	1.000				
Mg^{2+}	-1.000**	0.740	0.993	0.890	1.000			
Na^+	-0.993**	0.661	1.000**	0.834*	0.994**	1.000		
K^+	-0.856*	0.982**	0.785*	0.997*	0.853**	0.790*	1.000	
pH	**0.956****	**-0.515**	**-0.985****	**-0.721***	**-0.958***	**-0.984***	**-0.667***	**1.000**

*: Correlation is significant at $P < 0.05$ (two-tailed). **: Correlation is significant at $P < 0.05$ (two-tailed).

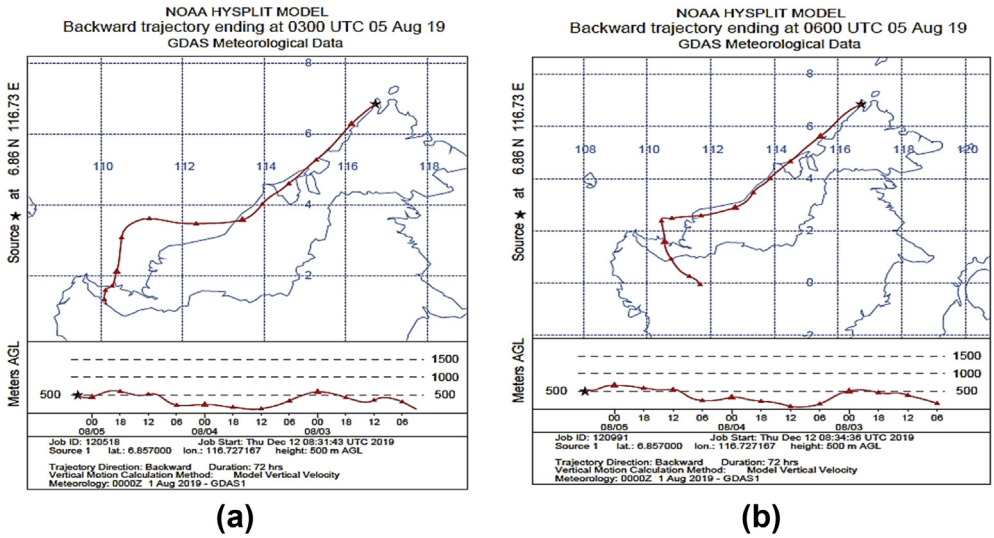

Figure 4. Backward trajectory analysis (Source: https://www.ready.noaa.gov/HYSPLIT.php).

may be probability the air masses landed and picked up particles from the smokes of biomass burning from Sarawak and West Kalimantan.

According to [44], it was found biomass burning at near site have enough alkaline compounds in the atmosphere to neutralise acidic compounds while with distance from burning site, the alkaline compounds may decrease and affecting the neutralisation processes by alkaline compounds. Therefore, further away from the burning site could have higher acidic components in atmosphere and hence may contribute to acid rain. Thus, in this study it may explain the significant acid rain phenomena in the rural area, of pH 4.95 that is too acidic that might impact by long-range transportation of pollutant from the biomass burning. The air masses that passed through the ocean may also picked up some sea particles and brought to the atmosphere of the rural area. This may explain the high Cl^- content in the sampled rainwater could also contributed from long range transportation. As mentioned earlier, Cl^- are probable in causing rainwater acidity from the production of HCl. In addition, it should be noted that Cl^- derived from biomass burning could retained in the atmosphere for long time, up to 520 days [39].

Figure 5. Fire hotpots map (Source: https://firms.modaps.eosdis.nasa.gov/map/).

4. Conclusions

In conclusion, the study showed acid rain had occurred during the southwest monsoon season. In overall, rural area recorded with higher acidity of precipitation at total average of pH 5.54 ± 0.39 compared to urban area at pH 5.77 ± 0.26. From the frequency occurrences of acid rain with pH lesser than 5.6 and below, rural area surprisingly recorded higher compared to urban area. Meanwhile, anions NO_3^-, SO_4^{2-} and Cl^- were found to be the dominant compositions of the acid rain occurrences with higher concentrations detected. The trajectory analysis and hotspots found that there is long-range transportation of air masses and transboundary pollutants from the neighbour regions, primarily due to haze events from the biomass burning and peat fires and aggravated by prolonged drought. From the correlation coefficient analysis, also found that the main sources and contributors of the acid rain occurrences were from ions of Mg^{2+} (0.903**) and Na^+ (0.849**) for urban studied area; and Cl^- (-0.985**) and Na^+ (-0.984*) for rural area respectively. This shows that the acidity of rainwater mainly come from natural source such as earth crust materials, sea salt particles, marine origins, biomass burning, and the biogenic volatile organic emissions, and not dominantly from typical air pollutants from automobile and industrial combustions. As for public health and safety, all rainwater samples during the acid rain

events were found exceeded the allowable limits of NWQS and WHO standards, that shown not suitable for skin contact, recreational purposes and drinking purposes. This study is very important as a next step to formulate a policy or at least a guideline for reference in the process of developing a standard for acid precipitation inside our law and regulation, as it can be seen posing high risk to public health when the acid rain occur especially during dry periods with prolonged drought during southwest monsoon and haze events.

Declarations

Author contribution statement

Carolyn Payus: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Celine Jikilim: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Justin Sentian: Conceived and designed the experiments.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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