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ASSESSMENT OF IONIC SPECIES IN RAINWATER TOWARD pH VARIABILITY BETWEEN HIGHLANDS AND LOWLAND IN MALAYSIA

(Penilaian Spesis Ionik Dalam Air Hujan Terhadap Kepelbagaian pH di Antara Tanah Tinggi dan Tanah Rendah di Malaysia)

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Abstract

This study investigated the variability of ionic species and pH in rainwater chemistry during wet fallout events in Highlands (Cameron Highlands, CH) and lowlands (Petaling Jaya, PJ) in 2019. A total of 1,040 datasets were obtained from the Malaysia Meteorological Department. Descriptive analysis, Mann-Whitney U test, Spearman correlation test, and discriminant analysis were employed. The selected variables included pH, H^+, Na^+, $K^{+,}$ Ca^{2+}, Mn^{2+}, Zn^{2+}, Cl^-, NO^{3-}, and SO_4^{2-}. Mn^{2+} exhibited a significant difference (p <0.05) between CH and PJ. Rainfall at PJ was more acidic due to higher concentrations of Cl^-, SO_4^{2-}, and NO^{3-}. pH in CH showed a strong positive correlation with Na^+, K^+, and Ca^{2+} as compared to PJ, attributed to atmospheric neutralizing reactions between acids (HNO_3 and H_2SO_4) and alkaline compounds ($CaCO_3$, NaCl, and KNO_3) present in suspended fine particles during wet fallout. Discriminant analysis, by using the confusion matrix, demonstrated significant differences in the overall performances of anions, cations, and pH in rainwater chemistry between CH and PJ, achieving 94.32% classification accuracy. This study highlighted the variability of ionic species and pH in rainwater chemistry between highlands and lowland areas. The findings contributed to understanding atmospheric heterogeneous reactions and factors that influenced rainwater chemistry at different geographic locations.

Keywords: wet fall-out, ionic species, statistical analysis, Petaling Jaya, Cameron Highlands

Abstrak

Kajian ini mengkaji kebolehubahan spesies ionik dan pH dalam kimia air hujan semasa berlakunya hujan di tanah tinggi (Cameron Highlands, CH) dan tanah rendah (Petaling Jaya, PJ) pada tahun 2019. Sebanyak 1,040 set data diperoleh daripada Jabatan Meteorologi Malaysia. Analisis deskriptif, ujian Mann-Whitney U, ujian korelasi Spearman, dan analisis diskriminasi telah digunakan. Pemboleh ubah yang terpilih termasuk pH, H^+, Na^+, K^+, Ca^{2+}, Mn^{2+}, Zn^{2+}, Cl^-, NO^3-, dan SO_4^{2-}. Mn^{2+} menunjukkan perbezaan yang signifikan (p <0.05) antara CH dengan PJ. Hujan di PJ lebih berasid sebab disumbangkan oleh kepekatan Cl^-,

SO_4^{2-}, dan NO_3^- yang lebih tinggi. pH di CH menunjukkan korelasi positif yang kuat dengan Na^+, K^+, dan Ca^{2+} berbanding dengan PJ, disebabkan oleh tindak balas peneutralan atmosfera antara asid (HNO_3 dan H_2SO_4) dengan sebatian alkali ($CaCO_3$, NaCl, dan KNO_3) yang terdapat dalam zarah halus yang terampai semasa hujan. Analisis diskriminasi, menggunakan matriks kekeliruan, menunjukkan perbezaan ketara dalam prestasi keseluruhan anion, kation, dan pH dalam kimia air hujan antara CH dengan PJ, mencapai ketepatan pengelasan 94.32%. Kesimpulannya, kajian ini mengetengahkan kebolehubahan spesies ion dan pH dalam kimia air hujan antara kawasan tanah tinggi ddengan tanah rendah. Penemuan ini menyumbang kepada pemahaman tindak balas heterogen atmosfera dan faktor yang mempengaruhi kimia air hujan di lokasi geografi yang berbeza.

Kata kunci: hujan, spesies ionik, analisis statistik, Petaling Jaya, Cameron Highlands

Introduction

Rainwater chemistry is a heterogenous atmospheric reaction, whereby the conversion from gas to particle occurs. The gaseous and aerosol released can originate from natural (marine spray, haze, resuspension) or anthropogenic activities (industrial process, transportation, agricultural pesticide spray that can be transported by winds from elsewhere [1]. The repetitive process from dry to wet deposition in the atmosphere is important with the nutrient cycle cleansing the pollutant in our ambient air [2]. The continuity of these processes occurs through day and night, regardless of the geolocation or even the altitude, including highland or lowland. According to Li et al. [3], the chemical composition of highlands would provide us with regional information regarding atmospheric pollution as compared to what the lowland offers as the local condition. Do et al. [4] reported that industrial development has resulted in an increase of SO2 and NOx emissions that finally led to significant disruption in atmospheric conditions, and thus affected the pH value in rainwater.

In Malaysia, a lot of highland areas were spotted, but a few were explored in the tourism sector such as Cameron Highlands, Pahang, and Kundasang, in Sabah. The Highlands and lowlands are both crucial for natural ecosystems. With reference to Razali et al., areas located at more than 1,000 m above sea level are categorized as mountains and classified as reserved forests or catchment forests, which serve as major sources of hydroelectric production for lowlands [5]. Lowland areas are experiencing higher population growth than high plateau areas, as they offer greater opportunities to stimulate the economy. In this study, rainwater samples were acquired from the Malaysian Meteorological Department (MMD) from two different sites, namely Cameron Highlands (CH) and Petaling Jaya (PJ) in the year 2019. The aims of this study are: (1) to assess the difference of pH, cation, and anion in rainwater between CH and PJ, representing highlands and lowland, respectively; (2) to investigate the relationship between the contribution of chemical composition (anion and cation) toward the fluctuation of pH value throughout 2019.

Materials and Methods

The chemical composition dataset

This study selected CH as the highland area and PJ as the lowland area. The 2019 rainwater data for the two sites were acquired from the Malaysian Meteorological Department (MMD) on a weekly basis which accounted from January to December. Since this study focused on the differences between highland and lowland areas, the total days of rainfall between these sites most likely differed. PJ recorded a total of 50 weeks available for the rainwater volume, while CH only provided 44 weeks out of 52 weeks in 2019. Therefore, the total data extracted at PJ and CH were 750 (50 weeks × 15 variables) and 440 (44 weeks × 10 variables), respectively. For CH, the variables involved in this study were H^+, Na^+, Ca^{2+}, Mn^{2+}, Zn^{2+}, Cl^-, NO_3^-, SO_4^{2-}, and pH. However, PJ recorded a higher number of variables, which included H^+, Na^+, Ca^{2+}, NH_4^+, Mg^{2+}, Ca^{2+}, Cu^{2+}, Mn^{2+}, Pb^{2+}, Zn^{2+}, Fl^-, Cl^-, NO_3^- SO_4^{2-}, and pH. Only the variables available in the two domains were selected and analyzed for a better understanding of statistical analysis. From the enumerated variables, only pH, H^+, Na^+, K^+, Ca^{2+}, Mn^{2+}, Zn^{2+}, Cl^-, NO_3^-, and SO_4^{2-} were selected for additional univariate and multivariate statistical analysis for both zones.

Study area

Ghosh et al. [6] described CH as the location of the borderline between a humid rainforest climate and a subtropical upland climate. CH (4° 29’ 3’’N, 101° 22 17’’E) is situated at 912m – 1,960m above sea level with a temperature range between 16°C and 22˚C, and total average rainfall fluctuated between 1800 and 3000 mm, a perfect setting for an agricultural area [7]. The local community's commitment mainly depends on tea estate farmlands such as strawberry farms, and vegetable farms as well as flower gardens like lavender gardens and rose gardens [5]. The major drawback in air quality and rainwater chemistry was probably contributed by the uncontrolled routine application of chemical pesticides in CH. The farms and estates had the possibility of excessive consumption of pesticides, herbicides, and fertilizers that may influence the concentration of ionic species in rainwater, resulting in unstable pH readings in the rainwater.

Petaling Jaya (3° 06’ 07’’N, 101° 38; 42’’E) with location at 556m above sea level is one of the busiest cities in Klang valley, which is encircled by Kuala Lumpur, Sungai Buloh, Shah Alam, and Puchong. According to Morissey et al. [8], PJ with a size of 97.2 km^2 recorded a temperature between 31°C and 33°C, making June and July being recorded as the driest month. In rainfall volume, PJ received heavy annual rainfall which accumulated up to 3300 mm. Shabanda et al. [9] emphasized that Petaling Jaya is an urban area involved in various industries, high traffic, and commercial and residential areas. Petaling Jaya industries include manufacturing, electrical and electronics, machinery, food and beverage, plastics, and chemical industries. Unlike CH, the variability in chemical composition in PJ rainwater might differ due to higher contributions from fossil fuel combustion, municipal waste, and industrial activities, which are exacerbated by the meteorological conditions [2]. Figure 1 illustrates the variation of land use activities in CH and PJ.

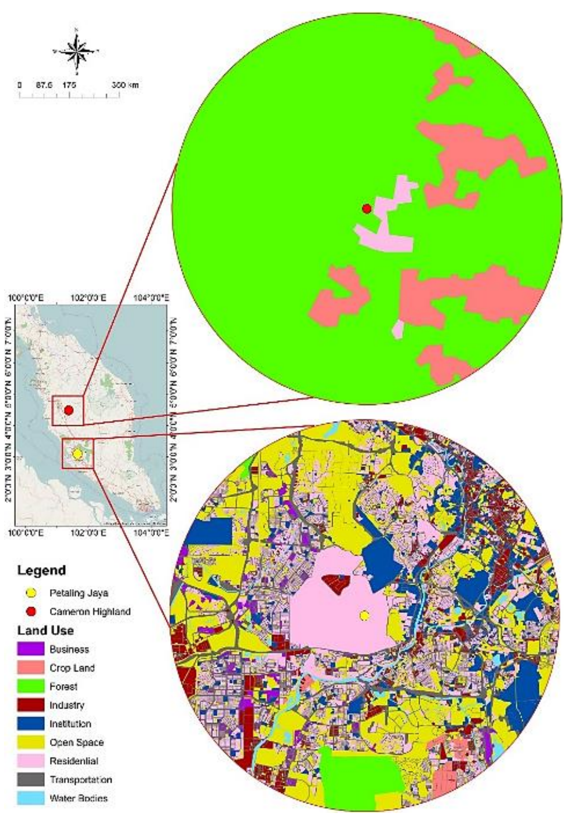

Figure 1. The illustration of land use activities in Petaling Jaya and Cameron Highlands

Statistical analysis: Pre-treatment dataset

Any missing data in this study were identified and removed from the datasets to increase data accuracy and uniformity. The data were mostly indicated as "Not Available" because an inadequate sample was recorded within that particular week. Not only that, but any variable was also recorded lower than the detection limit (LDL), and any results that showed no variation between weeks in 2019 were removed. The raw data received from MMD contained redundant and inconsistent data that had to be reduced to improve data coherence and reduce data corruption and database size. In this study, data transformation is a process of changing the format or structure of data. In this study, standardization methods were used to change the scale of data across a range. To proceed with multivariate statistical analysis like discriminant analysis, the datasets had to be standardized first. The transformation dataset process benefited this study in two ways; firstly, it canceled the unit with different variables and secondly, it reduced the range limit between variables. Data with a substantial gap such as between H^+ (e.g., 0.000005 µmol/L) and NO_3^- (100 µmol/L) could make H^+ seemed less important, yet both ions played an important role in determining the acidity in rainwater. Therefore, having a transformation dataset range (e.g., -1,1 or 0 -1) helps to reduce issues in this study [12]. Therefore, this study employed the standardization techniques, and the formula is written as below:

$$Z = \frac{x-\mu}{\sigma} \quad (1)$$

Where, x = individual data, μ = mean (average), and σ = standard deviation

Descriptive analysis

The data visualization in this study was performed by using R Studio (R development, 4.2.2). Martins et al. [10] substantiated the role of descriptive analysis in assessing activities that influence the characteristics of a given source. It entails employing techniques such as the box-whisker plot (boxplot) to draw informed conclusions. In the context of the standard boxplot, an issue arises when dealing with data that exhibit asymmetry or heavy-tailedness, resulting in an inflated identification of values as atypical. Notably, Cable and Deng [11] previously conducted a comprehensive study, quantifying the concentration of ionic species and pH in the Detroit Metropolitan area. The boxplots depicted the upper and lower limits, corresponding to 1.5 times the interquartile range (IQR) away from the box, while the median was represented by a vertical line within the box. This insightful methodology facilitated a thorough understanding of data distribution, identification of outliers, and exploration of the relations between ion concentrations and pH in CH and PJ.

Normality test, comparative, and correlation analysis

The normality test employed in this study utilized the Shapiro-Wilk test, with a sample size of less than 500 ($n < 500$), implemented by using XLSTAT (ver. 2019, USA). To visualize the Spearman Rank correlation analysis, the corrplot library function within R Studio (R development, 4.2.2) was utilized. Prabhaker et al. [12] conducted a statistical analysis and reported a *p*-value of greater than 0.05, indicating acceptance of the null hypothesis (H_0), and thus suggested a normal distribution. Conversely, for the alternative hypothesis (H_a), a *p*-value of below 0.05 was observed. In this study, the Shapiro-Wilk test rejected the null hypothesis ($p > 0.05$) for all variables, indicating that non-parametric analysis was necessary due to the non-normal nature of data. Consequently, given the non-parametric nature of the data, this study employed appropriate non-normal data analysis techniques. To compare independent datasets, the Mann-Whitney U test was employed, which utilized medians as the basis for comparison of each individual variable between the CH and PJ groups.

Spearman correlation analysis was conducted to explore the relation between pH, cation, and anion in the CH and PJ groups, with a significance level at 0.05. The strength of the correlation was classified as strong ($r > 0.70$), moderate ($0.30 \leq r \leq 0.69$), or weak ($r < 0.30$), as per the classification proposed by Ibrahim et al. [13]. The Spearman correlogram provided a visual representation of the correlations observed in the data. Positive correlations were depicted by blueish cubic representations, while inversible correlations were indicated by reddish cubic representations. Blank (white) cell areas indicated correlations that were statistically insignificant with a 95% confidence interval (p >0.05).

Results and Discussion

Variation of pH, cations, and anions in rainwater

Figure 2 illustrates the pH fluctuated between 4.18 and 5.92 in both areas during 2019. From the times series analysis, the overall mean pH measurement for CH and PJ were 4.87 ± 0.27 and 4.59 ± 0.76, respectively. For the record, pH in CH ranged 4.27-5.58 while CH showed a greater variance of 4.18-5.92, with the lowest value being recorded in Week 8 (February). In comparison, there were only two noticeable circumstances in that CH showed a greater pH value as compared to PJ: Week 11 – Week 18 and Week 36 – Week 44. The study conducted by Park et al. [14] provided evidence which supported the significant role of precipitation quantity in influencing rainwater acidity and pH. This influence was attributed to the dilution effect on ion concentrations during precipitation events. Regions with higher rainfall, such as high plateaus of Cameron Highlands, underwent more pronounced dilution of ionic species as compared to areas with lower rainfall, like Petaling Jaya, resulting in lower total mean concentrations of ionic species in the Cameron Highlands relative to Jaya Petaling. In contrast, Payus et al. [15] postulated the occurrence of neutralizing reactions between cations and anions within rainwater, leading to an equilibrium of ionic species concentrations. Furthermore, the study highlighted the substantial impact of atmospheric scavenging, a process involving the removal of airborne particles and gases from the atmosphere through precipitation, on the concentration of ionic species in rainwater and consequent modulation of rainwater pH levels.

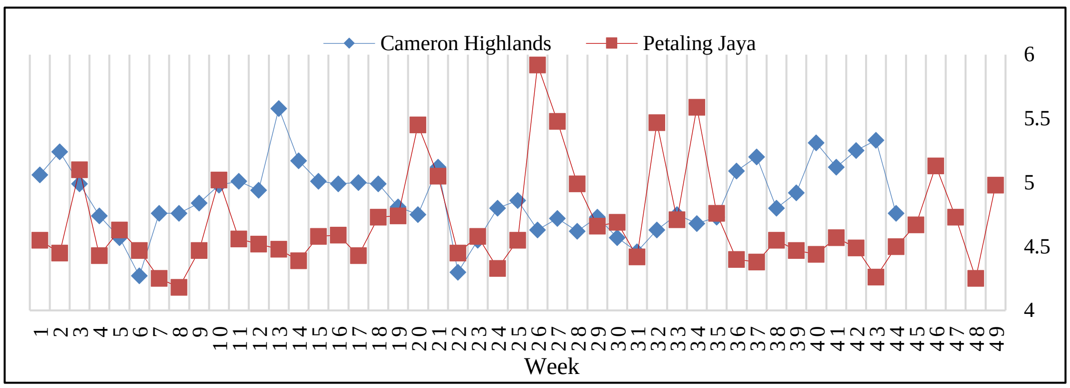

Figure 2. Time series analysis on pH variation in Cameron Highlands and Petaling Jaya during 2019.

Additionally, a higher temperature in the cloud led to an increased concentration of Cl^-, resulting in a decreased pH level of rainwater. This phenomenon was attributed to the heightened availability of gaseous molecules and high solubility of HCl in water. While the study placed considerable emphasis on pH, it was essential to direct attention toward the diverse array of cations and anions requiring investigation [14]. In the comparison of all variables, H^+ was determined to be the lowest concentration in both areas, with individual means that ranged from 0.000016 µmol/L - 0.00002628 µmol/L. The presence of H^+ ions in rainwater inflicted the rainwater acidity level. The higher the H^+ concentration present in rainwater, the lower the pH value in rainwater it gets. However, it will also depend on the influence of the external factor, including the type of anthropogenic activities and meteorological conditions, exacerbated by uncontrolled on vehicle possession in Malaysia especially in urban areas [16]. In this study, Na^+, NH_4^+, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-} were among the abundance of species discovered in rainwater. Table 1 tabulates the rudimentary descriptive analysis which showed the min, max, mean, and standard deviation of individual species found in CH and PJ.

Together, the Mann – Whitney U test was integrated into the table to verify the differences in each individual -

species at different lowlands and highlands. NO_3-, NH_4^+, SO_4^{2-}, Cl^-, Na^+, and Ca^{2+} were the concentrated ions as compared to others, with PJ showing a higher concentration than CH. Despite having differences in terrain structure, agricultural activities, as well as industrial and urbanization effects, Khan et al. [2] confirmed that the only explanation that PJ had concentrated ions was due to fractional acidity (FA). The divergence in FA between developed and less developed areas suggested a significant variation in the neutralization of alkaline ions in rainwater, primarily due to the considerable influence of NO_3- and SO_4^{2-} on rainwater acidification. For instance, the average NO_3- found in PJ was approximately 69.7% lower as compared to what was recorded in PJ. This pattern was similar with SO_4^{2-}, Cl^-, Na^+, Ca^{2+} were all 66.7%, 53.4%, 51.9%, and 49.7% lower, respectively. Not only on the average value for individual species, but also the observation from the max values also proved that the differences between types of anthropogenic activities operated in both areas proved the hypothesis constructed in this study.

Table 1. Statistic description of rainwater anion, cation (μmol/L) and pH in Petaling Jaya and Cameron Highlands

	Petaling Jaya				Cameron Highlands				Mann –
Variable	**Mean**	**Min**	**Max**	**Std. Dev**	**Mean**	**Min**	**Max**	**Std. Dev**	**Whitney U test (*p*-value)**
NO_3^-	41.50	5.95	127.57	21.73	12.56	0.02	43.79	9.71	<0.0001
NH_4^+	36.79	0.10	140.67	27.11	*	*	*	*	NA
SO_4^{2-}	19.73	2.97	51.36	10.78	6.58	0.15	25.08	5.67	<0.0001
Cl^-	14.54	3.65	39.68	8.99	6.78	0.08	25.79	5.69	<0.0001
Na^+	9.45	1.66	27.01	6.11	4.55	0.55	26.19	4.73	<0.0001
Ca^{2+}	7.37	0.90	23.14	4.64	3.71	0.43	18.1	3.42	<0.0001
K^+	2.69	0.40	13.52	2.22	1.93	0.37	8.02	1.53	0.033
Mg^{2+}	1.07	0.20	2.62	0.70	*	*	*	*	NA
Fl^-	0.80	0.05	1.70	0.40	*	*	*	*	NA
Zn^{2+}	0.63	0.01	9.21	1.37	0.16	0.01	0.51	0.11	<0.0001
Mn^{2+}	0.17	0.00	1.06	0.17	0.21	0.00	1.31	0.29	**0.4690**
Cu^{2+}	0.13	0.01	4.34	0.61	*	*	*	*	NA
Pb^{2+}	0.02	0.00	0.59	0.08	*	*	*	*	NA
H^+	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.0000
pH	4.59	0.00	5.92	0.76	4.87	4.27	5.58	0.27	0.0000

Asterisk (*): the individual datasets was recorded below the detection limit.
NA: The analysis can't perform due to incompatibility comparison between CH & PJ
Bolded result: The result is not significantly differed between CH and PH with 95% confidence interval.

From the result, NO_3^- and NH_4^+ were the only variables which showed a huge gap in the range. The NO_3^- in CH ranged between 0.02 µmol/L - 43.77 µmol/L, whereas PJ recorded 5.95 µmol/L - 127.57 µmol/L and 0.10 µmol/L - 140.67 µmol/L for both ions, respectively. The possible reasons that caused acid rain in urban areas were rapid industrialization, urbanization and rapid increase in automobiles, fossil fuel combustion, and the emission of SO_2 and NO_x from vehicles into the atmosphere [17]. A higher concentration of SO_2 and NO_x in the atmosphere could lower the pH of rainwater during wet falls because these were acidic species. Higher levels of Cl^- observed at Petaling Jaya may be associated with the formation of HCl in the atmosphere. Since the data showed non - parametric type, the comparison analysis was performed by using Mann – Whitney U Test. The independent comparison test between these two locations proved that there was a significant difference (p <0.05) for all variables, except for Mn^{2+}. In other words, the existence of manganese ions in both rainwaters was found comparable even though there was fluctuation in overall concentration throughout 2019. The finding revealed in Mann – Whitney result was supported by the Discriminant Analysis (DA), whereby the classification between CH and PJ unquestionably differed with a total of 94.32% dissimilarities.

The influence of cations and anions on the pH variability

In this study, the second objective of the study was achieved as the statistical evaluation between the pH and the group of cations and anions was performed by Spearman's rank correlation analysis. It helped to characterize the effect of these ionic species concentrations in rainwater with the pH as it discovered the strength of correlation. Among all, there were a few that had a significant correlation toward pH. H^+ was the only variable that had a significant correlation (*p* <0.05) in both areas, with Cameron Highlands having an inverse strong correlation with CH than Petaling Jaya, with r = -1.00 and -0.88, respectively. pH and H^+ had an inversely strong proportional relationship, the higher the concentration of H^+, the lower the concentration of pH [18]. This relationship could be described in the equation of pH = -log [H^+]. Other than H^+, other variables such as Zn^{2+}, NO_3^-, and SO_4^{2-} were the ions that demonstrated a significant correlation toward pH, and these were only found in CH. Despite H^+, other variables did not possess a significant correlation in PJ.

For the record, SO_4^{2-} was the only ion which showed the highest correlation indication with *r* (SO_4^{2-}/pH) = -0.75, followed by *r* (NO_3^-/pH) = -0.53, and *r* (Zn^{2+}/pH) = -0.40. The observation derived from this investigation aligned closely with the research conducted by Zeng and Han, wherein the ratio of SO_4^{2-} to NO_3^- exceeded the value of 1. This noteworthy observation suggested the influence of stationary sources of pollutant emissions, such as the transportation and dispersion of pollutants emitted during coal combustion activities. An inverse–strong correlation was observed, which might probably be due to the availability of H_2SO_4 in the atmosphere, forming acidic precipitation in CH [19]. For both locations, there was a strong correlation of *r* (Na^+/K^+) = 0.73 – 0.82, whereby CH had a higher correlation as compared to PJ. Zeng and Han, and Zeng et al. explained that a stronger correlation of Na^+ with K^+ and Ca^{2+} was due to atmospheric neutralizing reactions between the acids (HNO_3 and H_2SO_4) with alkaline compounds ($CaCO_3$, NaCl, and KNO_3) that existed in suspended fine particles [20,21]. The strength and direction of correlation between different ions indicated the effects of ions concentration on pH concentration, which resulted in the acidity or alkalinity of the rainwater of the particular area [22]. Figure 3 illustrates the overall correlation analysis between chemical composition toward pH variability in rainwater for CH and PJ, representing highland and lowland as hypothesized in this study.

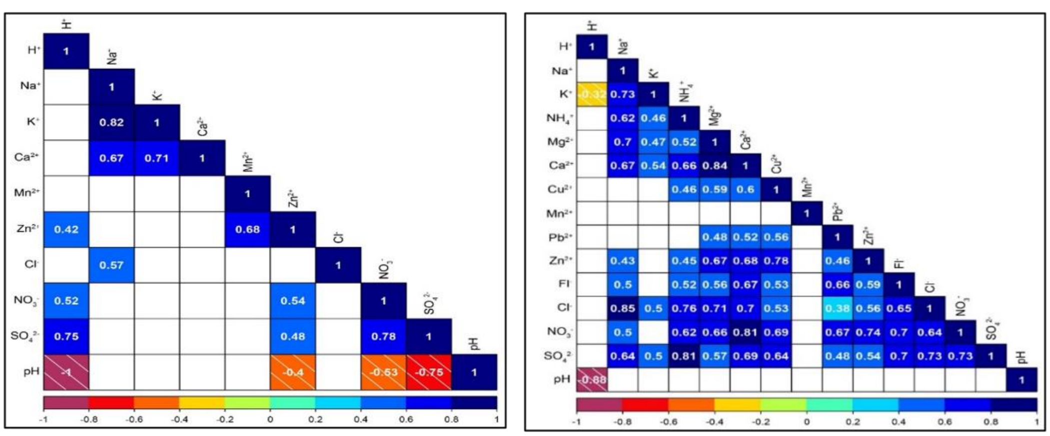

Figure 3. Spearman's correlation coefficient between pH and ionic species at Cameron Highlands (left) and Petaling Jaya (right). The colored cells denoted with a significance level of *p* <0.05, while white cells denote statistically non – significant correlation (*p* >0.05)

Conclusion

In brief, the study revealed that rainwater in both highland (CH) and lowland (PJ) regions could be classified as acidic, with the pH threshold for acid rain established at 5.6. Comparatively, the average pH value in PJ (4.59) was lower than in CH (4.87). The lowest pH values were observed in February 2019 during the Northeast Monsoon (NEM) period, characterized by increased precipitation on the Peninsula's east coast, although the study focused on the west coast, which might have been influenced by the nearby Titiwangsa mountain range. Statistical analysis demonstrated that various ions, including Na^+, NH_4^+, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}, significantly influenced the H^+ concentration and pH value. The Mann-Whitney U Test revealed significant differences among all variables, except for Mn^{2+}. Discriminant Analysis indicated substantial dissimilarity (94.32%) in overall performance between CH and PJ. Spearman's rank correlation analysis highlighted significant correlations between pH and ionic species, with three ionic species that affected pH value in CH, while none were significant in PJ. The fluctuation in pH variability throughout 2019 was attributed to interactions between ions such as NO_3^- and SO_4^{2-} and alkaline compounds ($CaCO_3$, NaCl, and KNO_3) during the dry deposition process. To advance research and practical applications, future studies should adopt a holistic approach, encompassing factors like climate change, trans boundary haze pollution, and monsoon patterns, conducting spatial and temporal surveys in Malaysia, considering variations in altitude, rainfall volume, population density, traffic levels, and land use types. Regular monitoring on a weekly, monthly, or annual basis would offer valuable insights.

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