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RESEARCH ARTICLE

Comparison of rainwater quality before and during the MCO using chemometric analyses

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Abstract

This study aimedto classify the spatiotemporal analysis of rainwater quality before and during the Movement Control Order (MCO) implementation due to the COVID-19 pandemic. Chemometric analysis was carried out on rainwater samples collected from 24-gauge stations throughout Malaysia to determine the samples' chemical content, pH, and conductivity. Other than that, hierarchical agglomerative cluster analysis (HACA) and discriminant analysis (DA) were used to classify the quality of rainwater at each location into four clusters, namely good, satisfactory, moderate, and bad clusters. Note that DA was carried out on the predefined clusters. The reduction in acidity levels occurred in 11 stations (46% of overall stations) after the MCO was implemented. Chemical content and ion abundance followed a downward trend, indicating that Cl^- and Na^+ were the most dominant among the anions and cations. Apart from that, NH_4^+, Ca^{2+}, NO_3^-, and SO_4^{2-} concentrations were evident in areas with significant anthropogenic activity, as there was a difference in the total chemical content in rainwater when compared before and during the MCO. Based on the dataset before the MCO, 75% of gauge stations were in the good cluster, 8.3% in the satisfactory cluster, 12.5% in the moderate cluster, and 4.2% in the bad cluster. Meanwhile, the dataset during the MCO shows that 72.7% of gauge stations were in the good cluster, 9.1% in the satisfactory cluster, 9.1% in the moderate, and 4.5% in the bad cluster. From this study, the chemometric analysis of the year 2020 rainwater chemical composite dataset strongly indicates that reduction of human activities during MCO affected the quality of rainwater.

Keywords Rainwater quality · COVID-19 pandemic · HACA · DA · MCO · Malaysia

Introduction

The Malaysian government's traffic control ordinance, commonly referred to as the Movement Control Order (MCO), is a series of national quarantine and health management measures introduced by the Malaysian government beginning on March 18, 2020, in response to the COVID-19 pandemic. The earliest COVID-19 cases in Malaysia were detected on January 25, 2020 (Hashim et al. 2021), and the number of cases has since increased exponentially. Therefore, the Malaysian government announced the implementation of the MCO to isolate the source of the COVID-19 outbreak. The MCO is the nationwide ban on movements and mass gatherings, including all religious, sports, social, and cultural activities. Therefore, all local or federal industries were closed except for infrastructure services and supermarkets, wet markets, grocery stores, and multi-functional stores selling daily necessities, including all educational institutions and schools (Awang et al. 2021).

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As states have followed social distancing and quarantine standard operating procedure (SOP) for more than a year, reduced human interference in the natural environment has given nature a "healing time." One significant impact observed is the air quality, experienced by everyone and recorded in various official reports (Bashir et al. 2020). As a result, reducing the movement of people in this situation has improved air quality with a good change in the Malaysia 2020 air quality index (Abdullah et al. 2020). The MCO has led to decreased consumption of fossil fuels by the urban transportation system. Meanwhile, the suspension of factory operations and other industries has reduced the emission of pollutants, such as gaseous SO_4^{2-}, NO_3^-, CO, CO2, CH4, NMHC, and O3; by-products of the photochemical process; and also particles into the atmospheres (Budiwati et al. 2016).

Other than that, air quality improvement also affects rainwater quality as it relates to each other. Previous studies have shown that polluted gases and particulate are removed through rainwater as it dissolves and is absorbed by raindrops during rainfall (Leong et al. 2018). The chemical composition of rainwater is a particularly sensitive indicator of air pollution, and it can provide information on the sources of chemicals and characteristics of rainwater in various areas (Calvo et al. 2010). Note that gases emitted by urban transportation and industries and the location factors will have a mesoscale impact on the rainwater's chemical composition in surrounding areas (Wang et al. 2019). Rainwater's chemical content is also influenced by the direction of wind and clouds that can transfer pollutants in every direction (Szép et al. 2019). The highest chemical concentration in rainwater is mainly due to adverse meteorological conditions, rainfall, and increased anthropogenic emissions (Meng et al. 2019).

Complex processes control rainfall composition, and natural and anthropogenic activities influence it, as Norela et al. (2013) noted. Studying rainwater precipitation in Malaysia is crucial as it helps uncover the source of pollution that affects precipitation chemistry (Uygur et al. 2010). In this regard, the research reported here was to study the chemical characteristics of precipitation in Malaysia. During the period from January to July 2020, we conducted this study. Rainwater parameters analyzed are as follows: pH, conductivity (EC), ammonium (NH_4^+), calcium (Ca^{2+}), fluoride (F^-), magnesium (Mg^{2+}), potassium (K^+), sodium (Na^+), nitrate (NO_3^-), sulfate (SO_4^{2-}), acetate (CH3COO −), chloride (Cl^-), formate (HCO2 −), methylseleninic acid (MSA) (CH_4O_2Se), oxalate ($C_2O_4^{2-}$), copper (Cu^{2+}), iron (Fe^{2+}), manganese (Mn^{2+}), mercury (Hg^+), nickel (Ni^{2+}), cadmium (Cd^{2+}), lead (Pb^{2+}), and zinc (Zn^{2+}) for a total of four hundred and sixty (460) samples.

We must consider rainwater quality of utmost importance because it predicts atmospheric quality to some extent and governs the ecosystem's health. Therefore, we consider rainwater quality to be paramount because it predicts the quality of the atmosphere and impacts some ecosystem health. Hence, this study analyzed rainwater's chemical composition data to detect changes in rainwater quality in Malaysia after MCO was implemented and classify the data according to the rainwater's level of quality at each gauge station. Research on rainwater chemistry is essential to prevent damage to natural ecosystems like acidification of water bodies and soils and imbalances in the ecosystem's productivity, cycle, and health problems (Payus et al. 2020). This information can also help authorities identify the source of rainwater pollution. Therefore, we can take appropriate action to overcome the problem of rainwater pollution, such as acid rain, which can harm humans and the local environment.

Methodology

Sample collection

Rainwater samples were collected from 24-gauge stations throughout Malaysia (Fig. 1) that the Meteorological Department of Malaysia manages, and the Department of Chemistry Malaysia analyzed the samples. Samples were collected from these gauge stations from October 22, 2019, to June 23, 2020. We considered 460 samples and divided them into two categories based on dates, before the Movement Control Order (MCO) and during the MCO. There were 244 samples with 23 parameters (5612) for the period before the MCO and 216 with 23 parameters (4968) for the period during MCO. The 24-gauge stations are located in various landforms such as hilly forests, coastal areas, and areas involved in various economic activities such as industrial areas, rubber plantations, rubber processing areas, big cities, and small towns.

Consequently, the samples were brought to the laboratory to determine their chemical composition and characteristics. The outline of the methodology used in this analysis can be seen in Fig. 2, and five main processes are included in this study. The rainwater pH was analyzed using pH Accumet AB150, and the conductivity was measured using a conductivity meter (Seven easy S30-K). The anions (F, Cl, NO_3^-, SO_4^{2-}) and organic acids (formate, acetate, oxalate) were examined utilizing the Ion Chromatography Dionex ICS 200 Anion System. At the same time, the cations (Na, NH_4, K, Mg, Ca) were determined employing the Ion Chromatography Dionex ICS2000 Cation System. On the other hand, the IC anions were investigated by applying the analytical column AS17, guard column AG17C, Suppressor ADRS600, eluent KOH and IC Cation analytical column CS12A, guard column CG16, suppressor CDRS600, and eluent methylseleninic acid MSA.

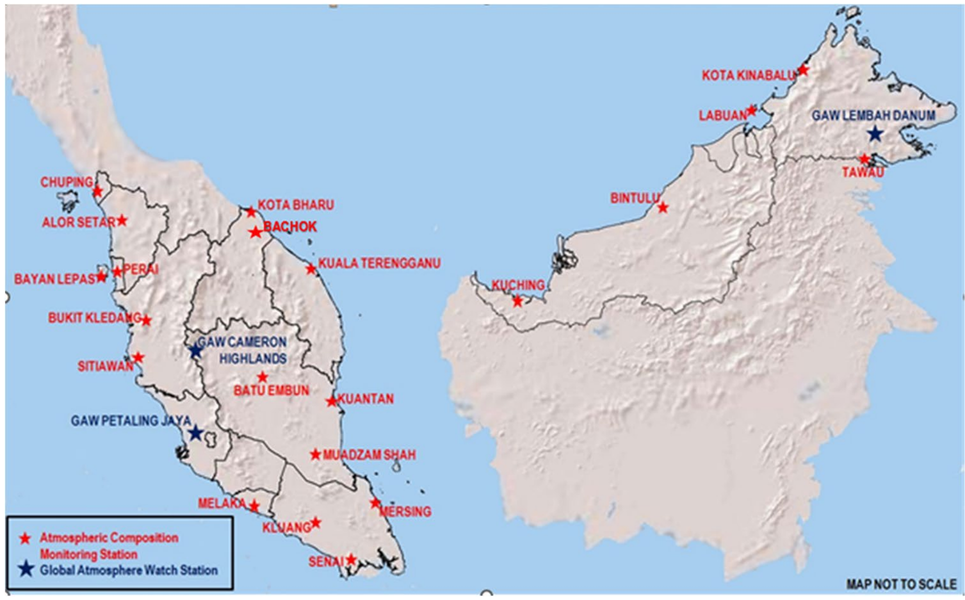


Fig. 1 Location of 24-gauge stations throughout Malaysia

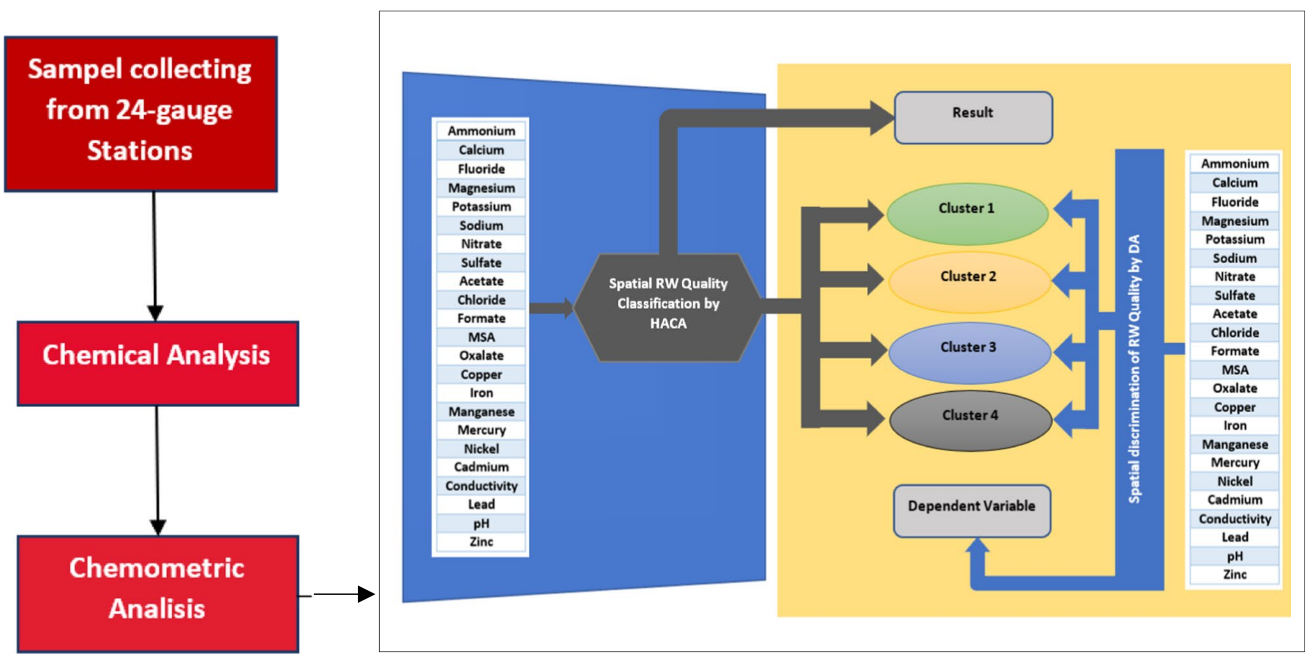


Fig. 2 The outline of rainwater quality study using chemometric analyses

QA/QC

According to the technical specifications established by the Department of Chemistry Malaysia, we performed rainwater sampling and analytical procedures. We conducted the chemical analysis according to Standard Methods (APHA 2017) and continuously monitored the quality measurements from the rain gauges. The METROHM 881 Compact Ion Chromatograph analyzed the cations and anions. We used a Perkin Elmer NexION 300Q ICP-MS (Perkin Elmer Inc., USA) fitted with an integrated autosampler, cross-flow nebulizer, and quartz torch for trace metal analysis. A pH meter (Mettler Toledo) calculated the pH of rainwater, and a conductivity meter calculated its conductivity.

The relative standard deviations (RSD) for the ionic analysis were better than 5%. Moreover, the data quality for rainwater samples was checked by ionic balance. All the rainfall data obtained from the Department of Chemistry, Malaysia, were pre-treated for subsequent statistical analysis. Data were statistically analyzed using the Microsoft

Excel 2019 software and XLSTAT 2021. It encompassed 23 physicochemical ions and metal ions variables, such as ammonium (NH_4^+), calcium (Ca^{2+}), fluoride (F^-), magnesium (Mg^{2+}), potassium (K^+), sodium (Na^+), nitrate (NO_3^-), sulfate (SO_4^{2-}), acetate (CH3COO −), chloride (Cl^-), formate (HCO2 −), MSA (CH_4O_2Se), oxalate ($C_2O_4^{2-}$), copper (Cu^{2+}), iron (Fe^{2+}), manganese (Mn^{2+}), mercury (Hg^+), nickel (Ni^{2+}), cadmium (Cd^{2+}), conductivity (EC), lead (Pb^{2+}), pH, and zinc (Zn^{2+}).

Statistical and chemometric analysis

Statistical analysis and chemometric (multivariate) techniques consisting of the hierarchical agglomerative cluster analysis (HACA) and discriminant analysis (DA) were utilized to analyze rainwater data to assess the characteristics of the harvested rainwater before and after MCO to investigate and classify the datasets obtained from the gauge stations. This was done to determine the rainwater's quality and compare these two situations.

Hierarchical agglomerative cluster analysis (HACA)

HACA is the most common approach and explains the similarity in relationships between any one sample and the entire dataset in a dendrogram (Juahir et al. 2010). In this study, HACA was employed to group the sampling sites according to the chemical contents of rainwater. To achieve these objectives, HACA was performed on data by creating columns representing 24 chemical composites and characteristics in rainwater and rows representing sampling sites using Ward's method with a squared Euclidean distance as the linkage distance used as a measure of similarity in the data. To group the similar sampling sites, cluster rows were created, while similarity among individual compounds for source identification was determined by cluster columns (Ananty et al. 2013).

Discriminant analysis

A discriminant analysis determines the variables that discriminate between two or more naturally occurring groups/clusters. It constructs a discriminant function (DF) for each group (Dominick et al. 2012; Juahir et al. 2017), which is calculated using Eq. 1:

$$f = \left(G_i\right) = k_i + n_j = 1 w_{ij} P_{ij} \quad (1)$$

where

i is the number of groups (G);

ki is the constant inherent in each group;

n is the number of parameters used to classify a dataset into a particular group; and

wj is the weighting coefficient assigned to a particular parameter (pj) in a DF analysis.

In this study, DA was applied to determine whether groups differed concerning the mean of a variable and to use that variable to predict group membership. Note that DA was applied to the transformed data using the standard, forward stepwise, and backward stepwise modes. These were used to construct DFs to evaluate the variation in pollutants found in the rainwater. In the stepwise forward mode, variables were included stepwise, starting with the most significant variable until no significant changes were detected. In the backward stepwise mode, variables were removed stepwise, starting with the least significant variable, until no significant changes were observed (Osman et al. 2012). Other than that, the user can eliminate redundant variables that best separate the group and then add one variable at a time: the variable that is added is the one that provides the maximum additional separation in the group. Alternatively, backward elimination starts with all variables and eliminates one variable at a time until the one that contributes the least to group separation remains. In stepwise selection, the two techniques are combined as variables are added one at a time. The variables are re-examined at each step to determine if any have become redundant and will be dropped (Juahir et al. 2018).

Results and discussion

Main compositions of rainwater

The concentration of chemical contents in Malaysian rainwater for 2020 is summarized in Table 1. It describes the chemical content, also known as parameters, in rainwater before the Movement Control Order (MCO) directive was announced and after the MCO was implemented. There are 24 parameters that determine the composition of rainwater in this study. Apart from that, concentrations of some ions show high dispersions based on their mean values and standard deviations (Table 1), which indicates a large variation of ion concentrations in the rainfall. It provides inputs on the source of several elements on the earth's surface in study areas. Overall, the condition after the MCO has increased the quality of rainwater in Malaysia in some areas. However, in some areas, the analysis presents that the chemical composites and character of the rainwater samples are unaffected, and some are deteriorating the rainwater quality.

Table 1 Concentration of chemical compositions in rainwater in Malaysia for 2020 (μmol/L)

Before the MCO					During the MCO				
Variable	Minimum	Maximum	Mean	Std. deviation	Variable	Minimum	Maximum	Mean	Std. deviation
Ammonium	0.281	47.941	9.246	11.997	Ammonium	0.100	53.707	8.420	11.859
Calcium	1.415	38.503	5.855	7.503	Calcium	1.240	19.261	4.181	4.048
Fluoride	0.100	2.880	0.691	0.615	Fluoride	0.100	3.649	0.699	0.921
Magnesium	0.420	29.762	4.593	6.589	Magnesium	0.500	7.318	1.783	1.717
Potassium	1.503	80.475	6.344	15.850	Potassium	1.336	11.626	3.461	3.008
Sodium	6.916	158.945	39.378	45.186	Sodium	4.103	761.811	50.657	163.433
Nitrate	1.473	40.220	10.270	9.620	Nitrate	0.010	53.868	10.041	11.437
Sulfate	1.312	24.290	7.619	5.285	Sulfate	1.825	21.077	6.895	4.719
Acetate	0.040	2.486	0.439	0.669	Acetate	0.040	3.356	0.473	0.776
Chloride	9.121	284.443	50.415	66.100	Chloride	5.880	794.625	54.639	169.995
Formate	0.017	1.218	0.116	0.243	Formate	0.020	2.127	0.249	0.468
MSA	0.002	1.194	0.104	0.250	MSA	0.002	1.992	0.171	0.434
Oxalate	0.023	0.869	0.134	0.175	Oxalate	0.010	0.722	0.091	0.151
Copper	0.003	0.201	0.036	0.060	Copper	0.003	0.589	0.046	0.128
Iron	0.035	0.232	0.071	0.046	Iron	0.035	0.144	0.060	0.035
Manganese	0.038	0.148	0.082	0.031	Manganese	0.012	0.466	0.092	0.093
Mercury	0.0003	0.0005	0.0003	0.0001	Mercury	0.00025	0.00033	0.00025	0.00002
Nickel	0.001	0.054	0.005	0.012	Nickel	0.002	0.017	0.002	0.003
Cadmium	0.001	0.001	0.004	0.016	Cadmium	0.001	0.219	0.027	0.062
Conductivity	0.447	5.318	1.468	1.056	Conductivity	0.429	10.106	1.555	2.029
Lead	0.001	0.867	0.039	0.176	Lead	0.001	0.008	0.038	0.002
pH	4.319	5.704	5.146	0.314	pH	4.391	5.598	5.053	0.301
Zinc	0.077	5.085	0.578	1.132	Zinc	0.039	4.795	0.486	1.034

Some of the chemical ions that show a declining trend are NO_3^- and SO_4^2 after MCO. Based on the overall data, the average relative magnitude of concentration in the precipitation before the MCO is $Cl^- > Na^+ > NO_3^- > NH_4^+ > SO_4^{2-} > Ca^{2+} > K^+ > Mg^{2+} >$ CH3COO – $> Zn^{2+} > F^- >$ HCO-2 + H $> CH_4O_2Se > C_2O_4^{2-} > Cu^{2+} > Mn^{2+} > Cd^{2+} > Fe^{2+} > Pb^{2+} > Ni^{2+} > Hg^+$. Meanwhile, the content of chemical ions when after MCO was implemented was $Cl^- > Na^+ > K^+ > NH_4^+ > NO_3^- > Ca^{2+} > Mg^{2+} > SO_4^{2-} > Zn^{2+} > F^- >$ CH3COO – > HCO-2 + H $> CH_4O_2Se > C_2O_4^{2-} > Fe^{2+} > Cu^{2+} > Mn^{2+} > Ni^{2+} > Pb^{2+} > Cd^{2+} > Hg^+$.

The dominance of Cl^- and Na^+ was mainly observed near the coastline, suggesting the presence of sea salt in the rainwater, while exhibiting maximum values in the before Movement Control Order (BMCO) data for Kota Bharu (284.43 μmol/L, Cl^-; 157.57 μmol/L, Na^+) and during the MCO for Mersing (794.625 μmol/L, Cl^-; 761.811 μmol/L, Na^+). The Bachok, Kuala Terengganu, Kuantan, and Mersing stations exhibited higher concentrations of Na^+ and Cl^- than other locations and remote sites because these areas were all on the coast. After implementing the MCO, these stations experienced a decline in Cl^- and Na^+ concentrations, except for the Mersing station. The presence of this element is due to the surface factors mentioned above. In addition, the analysis results also demonstrate the blocking effect of the mountainous regions in front of air masses loaded with sea spray, such as in Lembah Danum (11.823 μmol/L, Cl^-; 8.802 μmol/L, Na^+) and Sitiawan stations (9.121 μmol/L, Cl^-; 6.916 μmol/L, Na^+), which had low sodium and chloride values.

Other than that, anions NO_3^- and SO_4^{2-} were found to be the dominant compositions of the acid rain occurrences for some studied areas, with higher concentrations detected. According to Chang et al. (2022), the possible reasons that lead to acidity in the rain of urban regions are rapid the industrialization and urbanization and the rapid increase of automobiles through fossil fuel combustion, SO_4^{2-} and NO_3^-, into the atmosphere (Porfírio et al. 2020). Moreover, a large amount of SO_4^{2-} and NO_3^- in the atmosphere can lower the rainwater pH during precipitation events (Szép et al. 2019). A comparison of the mean of NO_3^- and SO_4^{2-} (Table 1) content in rainwater before and after MCO found that the existence of ions of these two types of chemicals is reduced after the MCO situation; NH_4^+, Ca^{2+}, Mg^{2+}, and K^+ are alkaline substances found in rainwater, which indicates a concentration of cations (Li et al. 2015; Metzger et al. 2006; Zhang et al. 2002). Correspondingly, the anthropogenic sources of anions and cations could be fossil fuel combustion, biomass burning, soil properties, and rock weathering, including fertilizers, forest fires, wind-blown dust, or seawater breeze (Budiwati et al. 2016; Leong et al. 2018). After the MCO, the reduced mean value of cations and anions (Table 1) strongly suggests that reducing movement and human activity can improve rainwater quality.

The highest concentration of NH_4^+ was found in Bayan Lepas station (47.94 μmol/L), located in Penang Island, BMCO, and decreased to 20.68 μmol/L after MCO. The Bayan Lepas Free Industrial Zone (FIZ) was established in the southeastern part of the Penang Island in 1976 and consists of four phases to accommodate various light and heavy industries. Besides, the FIZ houses manufacturers of electronics, fabricated metal products, machinery, and precision tools. Approximately 39% of the factories are related to the electronics industry (Khodami et al. 2017). This station also has high mean concentrations of Ca^{2+} (12.02 μmol/L), F^- (1.18 μmol/L), NO_3^- (22.79 μmol/L), and also SO_4^{2-} (11.18 μmol/L). After MCO was implemented, Tanah Rata station recorded the highest NH_4^+ reading of 53.707 μmol/L, followed by Bayan Lepas and Alor Setar.

Tanah Rata is located in Pahang and is a well-known agricultural area. Ammonia and ammonium compounds are excellent sources of plant nitrogen and are present in most fertilizers, either in natural or synthetic fertilizer formulations (Huang et al. 2017). Other than that, increased fertilizer-related plant activities and open burning in agricultural areas are among the main reasons for increased levels of NH_4^+ dispersed into the air, dissolving in the rainwater during rainfall. Human activities involving agriculture, urbanization, tourism, and land clearing activities are expected to increase pollution sources, especially in the case of farming, urbanization, settlement, and infrastructure development. These activities have tremendously impacted natural systems, river basins' dynamics, and their adjoining valley slopes and channel systems (Rozimah and Khairulmaini 2016a, b). This indicates that the rainwater in the study area is more affected by human activity compared to remote mountain areas and oceanic islands (Zeng et al. 2019). This indicates that the rainwater in the study area is more affected by human activity compared to remote mountain areas and oceanic islands (Zeng et al. 2019).

Apart from that, the presence of Ca^{2+} and Mg^{2+} in the precipitation shows the influence of terrestrial sources, such as the dissolution of dolomites and limestones (Keresztesi et al. 2020; Niu et al. 2014; Szep et al. 2019). Note that calcium can also originate from anthropogenic activities, such as open quarries and cement factories, while magnesium can be attributed to marine natural sources (Xiao et al. 2017). The highest mean concentrations of Ca^{2+} and Mg^{2+} before the MCO were found in Kota Bharu (38.503 μmol/L, Ca^{2+}; 29.762 μmol/L, Mg^{2+}) and during the MCO in Tanah Rata (19.261 μmol/L, Ca^{2+}) and Bachok (7.318 μmol/L, Mg^{2+}). Overall, the mean Ca^{2+} and Mg^{2+} in the precipitation declined when compared to before the MCO and during the MCO. Reduction of anthropological activity during the MCO has been shown to reduce the content of pollutants in rainwater.

On the other hand, the mean concentration of F^- was slightly higher than indicated when observed before the MCO compared to during the MCO. The concentration of F^- in rainwater before the MCO was 0.691 μmol/L compared to 0.699 during the MCO, with an SD of 0.0.615 and 0.921, respectively. This indicates that the total F^- content in each sample from all gauge stations has little variation. Readings indicate F^- content in the rainwater sample taken at each station. F^- is a mineral that occurs naturally and is released from rocks into the soil, water, and air, with almost all water containing some F^- (Qian et al. 1999). Since some F^- compounds in the earth's upper crust are soluble in water, dust, and soil particles from the foot of mountains, particularly high in F^- could seep into the groundwater and rainwater by weathering and leaching of bedrock. The highest F^- concentration was recorded in Kota Bharu (2.88 μmol/L; F^-) before the MCO and Sitiawan (3.649 μmol/L) after MCO.

HCO-2 + H, CH3COO –, and $C_2O_4^{2-}$ were the most common organic acids in both cloud water and rainwater, as this study discovers the presence of organic acids in the rainwater's composition. These organic acids are found in the atmosphere and can be analyzed based on observed stormwater samples and classified according to chemical type. Here, HCO-2 + H and CH3COO – concentrations were estimated using ion chromatography on precipitation samples collected in 2020 from all gauge stations in Malaysia. These organic acids are found in the atmosphere and can be analyzed based on observed stormwater samples and classified according to chemical type (Twohy et al. 2021). The mean concentrations for CHO_2 and CH_4O_2Se were 0.116 μmol/L, SD 0.243, and 0.439 μmol/L, SD 0.669, respectively, before the MCO and 0.249 μmol/L, SD 0.468, and 0.473 μmol/L, SD 0.776, respectively, after the MCO. Note that the maximum CHO_2 and CH_4O_2Se concentrations from Sitiawan after the MCO were 2.127 μmol/L and 3.356 μmol/L, possibly due to an increase in CH3COO – from direct industrial emissions involving heavy vehicular traffic load or indirect CH3COO – formation. In addition, widespread local use of biomass as a domestic fuel also contribute to increased CH_4O_2Se (Chang et al. 2022).

Other than that, methanesulfonic acid (MSA) (CH_4O_2Se) was present in aerosol and rain collected at all study areas. The mean CH_4O_2Se value for the precipitation was 0.104 μmol/L before the MCO and 0.171 μmol/L during the MCO. These values are very similar to other marine areas around the world. The CH_4O_2Se ratio is lower than 1% (0.2

and 0.7% for rain and aerosol, respectively), which is well below the North Sea and Atlantic Ocean values reported in the literature (8–10%) (Sun et al. 2016).

Oxalate $C_2O_4{}^{2-}$ is a dicarboxylic acid dianion with the chemical name oxalate PubChem (2004). The mean concentration of this ion was 0.134 µmol/L, SD 0.175 before the MCO and 0.091 µmol/L, SD 0.151 during the MCO period. Hence, there was a decrease in the concentration of this chemical after the MCO was implemented. $C_2O_4{}^{2-}$ is also referred to as ethanedioate. It is obtained by deprotonating both the carboxy groups of $C_2H_2O_4$ and is widely used for extracting derivatives, such as salts of oxalic acid, dimethyl oxalate, or sodium oxalate. Apart from that, it also forms coordination compounds and is sometimes abbreviated as an ox.

Furthermore, the HCO-2 + H to CH3COO – (formic/acetic) ratio shows that primary emissions dominate organic acids in 71.31% of cloud water and overall rainwater samples. The positive matrix factorization (PMF) analysis determined four factors as organic sources. Acids in cloud water include biogenic emissions (61.8%), anthropogenic emissions (15.28%), marine emissions (15.07%), and soil emissions (7.85%) (Sun et al. 2016). Heavy metals are toxic to humans when present in high concentrations. Generally, heavy metal ions in water can become hazardous when their concentrations exceed the permissible limit (Chibueze Izah et al. 2016; Elinge et al. 2011). The mean and SD concentrations for eight chemical contents classified as heavy metal ions in rainwater after MCO are Cu^{2+} (0.036 µmol/L SD 0.060), Fe^{2+} (0.071 µmol/L SD 0.046), Mn^{2+} (0.082 µmol/L SD 0.031), Hg^{+} (0.0003 µmol/L SD 0.0001), Ni^{2+} (0.005 µmol/L SD 0.012), Cd^{2+} (0.004 µmol/L SD 0.016), Pb^{2+} (0.039 µmol/L SD 0.176), Zn^{2+} (0.578 µmol/L SD 1.132), BMCO data and Cu^{2+} (0.046 µmol/L SD 0.128), Fe^{2+} (0.060 µmol/L SD 0.035), Mn^{2+} (0.092 µmol/L SD 0.093), Hg^{+} (0.00025 µmol/L SD 0.00002), Ni^{2+} (0.002 µmol/L SD 0.003), Cd^{2+} (0.027 µmol/L SD 0.062), Pb^{2+} (0.002 µmol/L SD 0.002), and Zn^{2+} (0.486 µmol/L SD 1.034). In addition, heavy metals, such as Cd^{2+}, Hg^{+}, and Pb^{2+}, are highly poisonous, and various types of human activities have increased the concentration of heavy metals in the environment. For example, anthropogenic activities (e.g., industry, agriculture) increase the content of heavy metals in different environmental matrices, such as water, soil, and air. However, studies have demonstrated that although traces are present in Malaysian rainwater, these metallic elements are generally below the health limit guidelines, except in highly industrialized areas.

Stations with the maximum concentrations of copper before the MCO were Kluang, Petaling Jaya, and Bukit Kledang stations, while Bukit Kledang (0.5894 µmol/L), Kluang, and Mersing (0.589 µmol/L) saw maximum concentrations after the MCO was implemented. Besides, other metals that were discovered were Fe^{2+} (Melaka 0.232 µmol/L BMCO and Bukit Kledang 0.144 µmol/L MCO), Mn^{2+} (Seberang Perai 0.148 µmol/L BMCO, Mersing 0.466 µmol/L MCO), Ni^{2+} (Melaka 0.054 µmol/L BMCO, Kluang 0.017 µmol/L MCO), Zn^{2+} (Seberang Perai 5.085 µmol/L, BMCO, Seberang Perai 4.795 µmol/L MCO), Pb^{2+} (Kluang 0.867 µmol/L BMCO, Melaka 0.038 µmol/L MCO), Cd^{2+} (Seberang Perai 0.001 µmol/L BMCO Melaka 0.219 µmol/L MCO), and Hg^{+} (Senai 0.0005 µmol/L BMCO and Kluang 0.00033 µmol/L MCO).

pH and conductivity

According to Fig. 3, the mean pH concentration before the MCO (data from October 22, 2019, to March 17, 2020, 244 N dataset) was 5.146 and after MCO (March 23, 2020, to June 30, 2020, 236 N dataset) was 5.053. This indicates a further decrease in pH variation in rainwater with an increment of 1.8%. Other than that, this demonstrates a mild improvement in the rainwater's acidity because the intern of acidity when the MCO was introduced and the reduction in the human movement has had a positive effect on the environment by reducing the release of pollutants into the air, which in turn led to worsening levels of acid rain. Detailed information about changes in rainwater pH at all gauge stations before the MCO compared to after MCO is shown in Fig. 1. Even if there is a small change in the pH reading, it greatly influences the environment. The impact of changes in the acidity of rainwater is significant even if the number of changes in the pH reading is small and cannot be proven by statistical methods such as the *t*-test (Han et al. 2019). Note that 46% of gauge stations indicated an increased pH reading, yielding a decrease in the rainwater's acidity level. There was a decrease in pH reading in 56% of gauge stations located in industrial areas, cities, and coastal areas. Essential industries such as glove factories, medicine, and food were still not included in the MCO. Therefore, most of the areas with these factories were operating as usual. Some even had to increase production because of the COVID-19 pandemic, especially the dramatic increase in rubber gloves, face masks, and many other medical supplies. This usually occurred in stations located in Bayan Lepas, Seberang Perai, and Muadzam Shah, where there are palm oil mills and oil palm plantations. Another area that experienced increased rainfall acidity was Sitiawan, Perak, where there are rubber plantations and the YTL rubber glove processing plant (Fernandez 2012). Other stations that witnessed increased levels of rainfall acidity after the MCO was implemented were Kuala Terengganu, Kuantan, Mersing, Chuping, Melaka, Batu Embun, Alor Setar, Bukit Kledang, and Petaling Jaya stations.

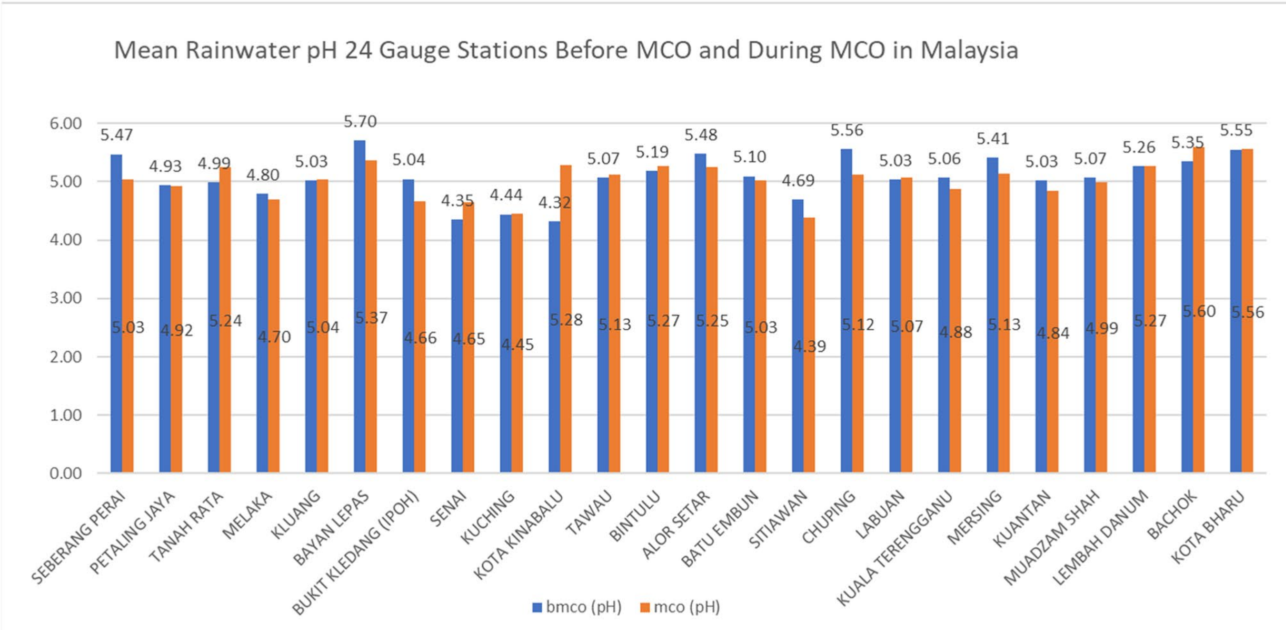


Fig. 3 Mean Rainwater pH at 24-gauge stations before the MCO and after MCO

Based on the comparison of the two sets of data in Table 1, 11 variations in chemical contents saw a decrease in readings compared to 10 chemical contents that increased in readings after MCO.

The reduction in acidity level concentrations occurred at 11 stations (46% of overall stations). The highest reduction was at traffic-dense and industrial areas where emissions had been reduced. In Malaysia, some main sources of air pollution are motor vehicles, industrial emissions, and open burning (Latif et al. 2014). Note that the lowest and highest readings for the conductivity of rainwater in this study before the MCO comes from the Kota Bharu Station, 5.318, and the Mersing Station, 10.106, respectively (Table 1). The mean conductivity before and after MCO was 1.468 and 1.555, respectively (Table 1). There was an increment in the mean conductivity of rainwater before and after MCO was implemented. This indicates that rainwater can have a level of conductivity if certain ions are present in rainwater (O'Donnell 2019).

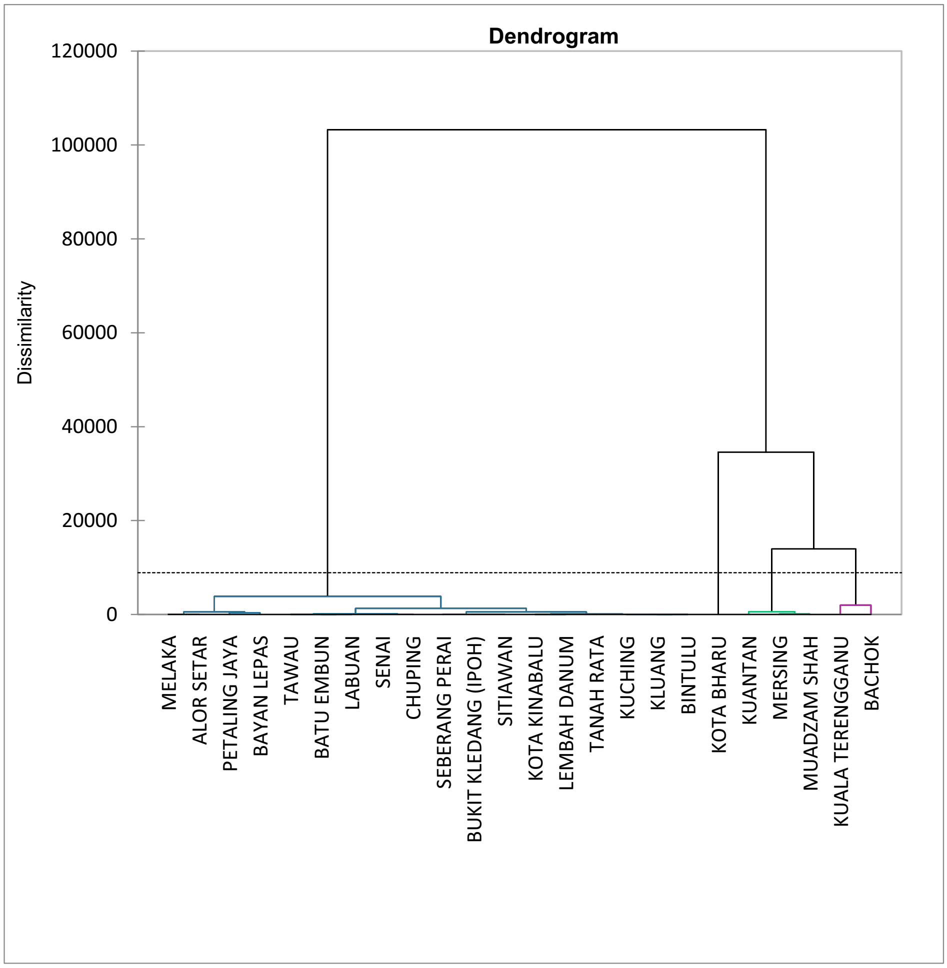
Cluster analyses (CA)

The main purpose of cluster analysis is to categorize rainwater chemicals and data into clusters based on their basic similarities, as shown in Fig. 4a and b. Hierarchical clustering involves the sequential formation of clusters for the 24 rain gauge stations. It can be inferred from Table 2 that CA using the Ward method had identified four clusters from two datasets for rainwater before and after the MCO. Note that CA also collected these datasets to evaluate spatial variation in all the gauge stations in Malaysia, which were classified into good, satisfactory, moderate, and bad clusters.

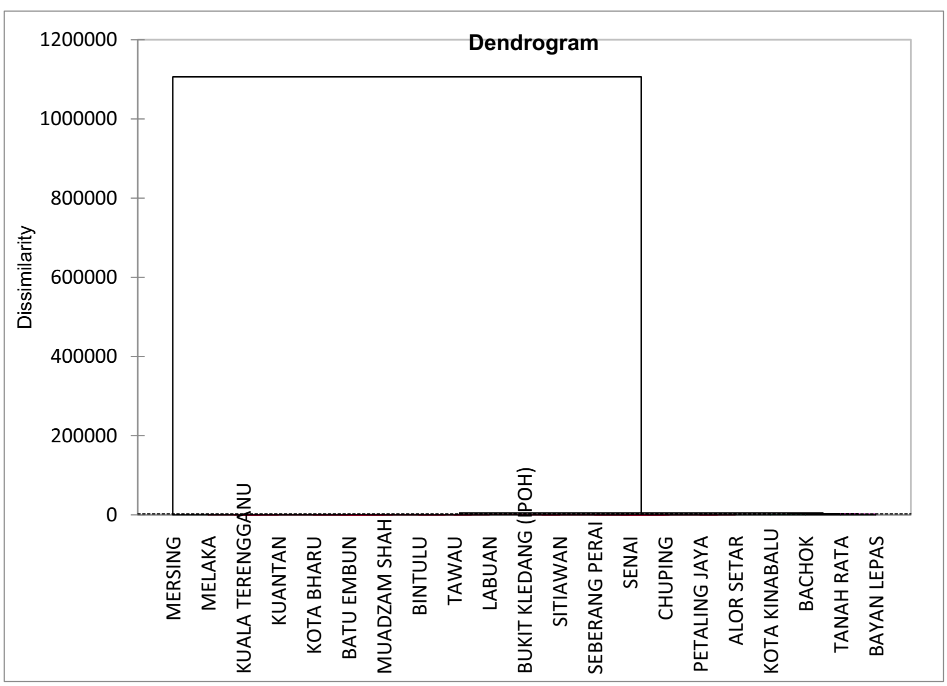
As for the BMCO dataset, cluster 1, which comprised 18 rain gauge stations, was classified as having a good cluster, with the overall samples having the least chemical composites in the raindrops. As for the after MCO dataset, the number of gauge stations in cluster 1 increased to 19 stations, namely stations in Seberang Perai, Petaling Jaya, Melaka, Kluang, Bukit Kledang (Ipoh), Senai, Kuching, Tawau, Bintulu Alor Setar, Batu Embun, Sitiawan, Chuping, Labuan, and Lembah Danum. All these stations maintained a good cluster of rainwater quality before and during the MCO. Other than that, there were 3-gauge stations that indicated a decline in rainwater quality. The Tanah Rata and Bayan Lepas stations changed from the good cluster based on their BMCO data to the satisfactory cluster after MCO. Another substation that saw a decline in rainwater quality was Kota Kinabalu, which moved from the good to the moderate cluster. The main attributable factor for this situation was the two chemical composites in the Kota Kinabalu rainwater sample, sodium and chloride. These chemicals dominated the other chemicals in Kota Kinabalu's rainwater sample, which has developed into an over-concentrated urban area. Recent studies have shown that the air quality in Kota Kinabalu has deteriorated (Abas et al. 2020).

Cluster 2 represents a satisfactory class for 2-gauge stations in this group based on the BMCO data and two stations based on the MCO data. The analysis shows that Kuala Terengganu was in this cluster before the MCO. Still, this gauge station saw an improvement in its rainwater

Fig. 4 Dendrograms of rainwater quality **a**)BMCO **b**) MCO



a) BMCO



b) MCO

Table 2 The mean and variation of Malaysia rainwater pH before and after MCO

Gauge Stations	BMCO (pH)	MCO (pH)	Variation
Seberang Perai	5.47	5.03	-0.44
Petaling Jaya	4.93	4.92	-0.02
Tanah Rata	4.99	5.24	0.26
Melaka	4.80	4.70	-0.09
Kluang	5.03	5.04	0.01
Bayan Lepas	5.70	5.37	-0.33
Bukit Kledang (IPOH)	5.04	4.66	-0.38
Senai	4.35	4.65	0.30
Kuching	4.44	4.45	0.01
Kota Kinabalu	4.32	5.28	0.96
Tawau	5.07	5.13	0.05
Bintulu	5.19	5.27	0.08
Alor Setar	5.48	5.25	-0.24
Batu Embun	5.10	5.03	-0.07
Sitiawan	4.69	4.39	-0.30
Chuping	5.56	5.12	-0.43
Labuan	5.03	5.07	0.03
Kuala Terengganu	5.06	4.88	-0.18
Mersing	5.41	5.13	-0.28
Kuantan	5.03	4.84	-0.18
Muadzam Shah	5.07	4.99	-0.08
Lembah Danum	5.26	5.27	0.01
Bachok	5.35	5.60	0.25
Kota Bharu	5.55	5.56	0.01

quality and moved into a good cluster after MCO. Adherence to MCO instructions has positively affected the environmental quality in this area. The reduction of fuel emissions from vehicles visiting the city and the reduction in industrial activities has positively impacted air quality. Apart from that, the second station in this cluster is Bachok, one of Kelantan's administrative districts. The main economic activity in Bachok is agriculture, besides fishing and business, as well as employment in the government and private sectors (Tai et al. 2021). Bachok and Kuala Terengganu are located on the coast facing the South China Sea, and it has beautiful beaches and an attractive tourist spot. The chemical content of sodium and chloride is predominantly present in rainwater at these two stations due to sublimation from seawater, which is the dominant landform here (Keresztesi et al. 2020). The dominance of chloride and sodium was mainly observed near the coastline, suggesting the presence of sea salts in the rainwater. Tanah Rata and Bayan Lepas stations are in this cluster; both have gone from the good to the satisfactory cluster. Note that land in Tanah Rata is used for agriculture practices and agro-tourism (Ibrahim et al. 2018).

The MCO does not positively affect air quality in this area because although tourism activities are controlled, active agriculture activities are still going on to meet essential food supplies. The FIZ in Bayan Lepas was established in the southeastern part of the Penang Island in 1976 and consists of four phases to accommodate various light and heavy industries as well as several rubber glove and medical item factories that had to continue operations and even increase productivity to meet the current needs during the MCO. Approximately 39% of the factories are related to the electronics and computer compartment industry (Khodami et al. 2017). Essential category industries had to operate as usual during the MCO by applying strict SOPs outlined by the authorities. There are 17 sectors that were allowed to operate, including food and beverage, health services, veterinary care, welfare, and security services (Malaysia Now 2021).

Cluster 3 was classified as having a moderate class. There were 3-gauge stations based on the BMCO dataset and two stations based on the MCO dataset in this cluster. Mersing, Kuantan, and Muadzam Shah are stations identified in this cluster for the BMCO group. However, during the MCO, the Kuantan and Muadzam Shah stations saw an increase in rainwater quality from moderate to good, but Mersing's rainwater quality moved to the bad cluster.

Kota Kinabalu had undergone a cluster change from good, before the MCO, to moderate, during the MCO. In addition to Kota Kinabalu, Bachok also experienced a deterioration in rainwater quality during the MCO and moved from the satisfactory to the moderate cluster. Both these stations are located along the coastline, and the rainwater samples from these stations had very high levels of chloride and sodium compared to other chemical substances.

Cluster 4 consists of the Kota Bharu station for the BMCO dataset, while the Mersing station was reported to have bad PRI after MCO. Kota Bharu is situated on the east coast of Malaysia and is well known for its cultural reserves. Additionally, the surrounding areas mostly comprise old buildings, but lots of development has increased its popularity, attracting locals and foreigners to the area (Reruang et al. 2012). Kota Bharu, which has a high population density, is also one of the interesting places for tourists to visit (Mohamad Rohani 2015). When the MCO was implemented, the bustle of the city was reduced, resulting in the lesser movement of motor vehicles, which ultimately contributed to the reduction in air pollutant emissions and consequently improved the air quality in this station from bad to good. The class centroid BMCO table indicates that Kota Bharu has 13 parameters consisting of 23 parameters related to chemical content in rainwater, which is the highest compared to other stations, and the chemicals present were mainly ammonium, calcium, fluoride, magnesium, potassium, sodium, nitrate, sulfate, and chloride. As for

Mersing's rainwater quality, the condition was influenced by Mersing's high levels of sodium and chloride (Table 3). Meanwhile, other chemicals also contributed to rainwater pollution, such as ammonium, sulfate, and nitrate, were at low levels. This means that Mersing's rainwater quality was influenced by natural oceanic factors involving sodium and chloride (Abbas et al. 1993).

The classification matrices for spatial and temporal variations of rainwater quality based on chemical composites in rainwater samples collected from identified gauge stations in Malaysia using DA are shown in Table 4 (before the MCO) and 4 (after MCO). In Table 4, the spatial discriminant standard mode generated 33.33% of assignments correctly with 24 discriminant variables ($p < 0.05$). Meanwhile, the accuracy of spatial classification using the stepwise forward mode was 100% with three discriminant variables: Ammonium, potassium, and sodium. Meanwhile, the backward stepwise mode discriminant function (DF) analysis was 66.67% with 19 discriminant variables.

As for dissimilarity in MCO data, discriminant analysis (DA) applied the standard, forward stepwise, and backward

Table 3 The classification of rainwater quality for 24-gauge stations in Malaysia before and after MCO by HACA

BMCO		MCO	
Station	Class	Station	Class
Seberang Perai	Good	Seberang Perai	Good
Petaling Jaya	Good	Petaling Jaya	Good
Tanah Rata	Good	Tanah Rata	Satisfactory
Melaka	Good	Melaka	Good
Kluang	Good	Kluang	Good
Bayan Lepas	Good	Bayan Lepas	Satisfactory
Bukit Kledang (IPOH)	Good	Bukit Kledang (IPOH)	Good
Senai	Good	Senai	Good
Kuching	Good	Kuching	Good
Kota Kinabalu	Good	Kota Kinabalu	Moderate
Tawau	Good	Tawau	Good
Bintulu	Good	Bintulu	Good
Alor Setar	Good	Alor Setar	Good
Batu Embun	Good	Batu Embun	Good
Sitiawan	Good	Sitiawan	Good
Chuping	Good	Chuping	Good
Labuan	Good	Labuan	Good
Kuala Terengganu	Satisfactory	Kuala Terengganu	Good
Mersing	Moderate	Mersing	Bad
Kuantan	Moderate	Kuantan	Good
Muadzam Shah	Moderate	Muadzam Shah	Good
Lembah Danum	Good	Lembah Danum	Good
Bachok	Satisfactory	Bachok	Moderate
Kota Bharu	Bad	Kota Bharu	Good

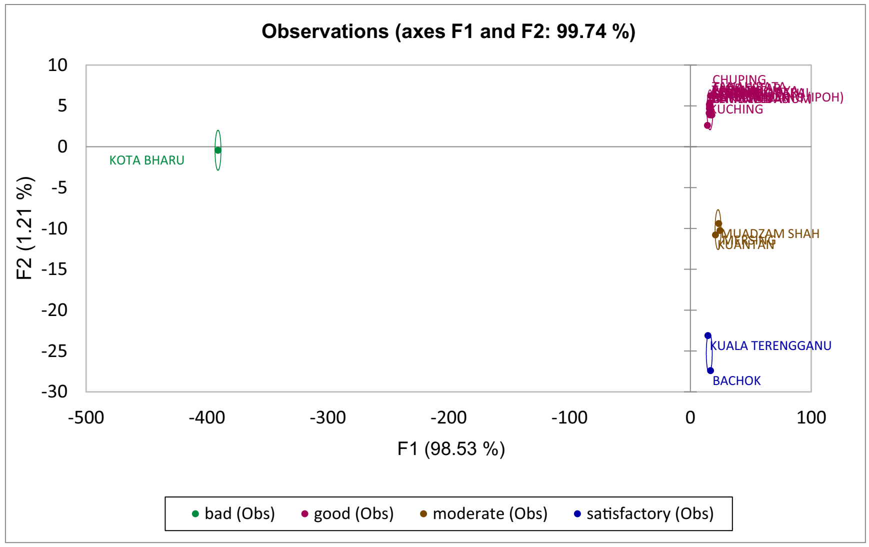
Table 4 Confusion matrix for the cross-validation result (DA) between situation BMCO and After MCO

BMCO						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%
Good	14	1	2	1	18	5.56%
Moderate	3	0	0	0	3	0.00%
Satisfactory	2	0	0	0	2	0.00%
Total	20	1	2	1	24	8.33%
Stepwise forward						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%
Good	0	18	0	0	18	100.00%
Moderate	0	0	3	0	3	100.00%
Satisfactory	0	0	0	2	2	100.00%
Total	1	18	3	2	24	100.00%
Stepwise backward						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%

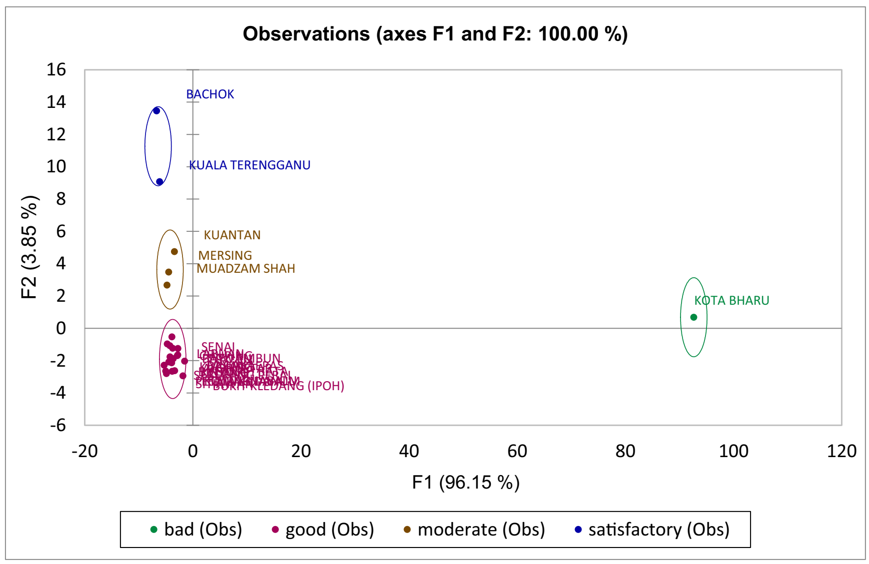
Table 4 (continued)

Good	0	12	3	3	18	66.67%
Moderate	0	2	1	0	3	33.33%
Satisfactory	0	0	0	2	2	100.00%
Total	1	14	4	5	24	66.67%
MCO						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%
Good	11	3	1	1	16	18.75%
Moderate	0	1	0	1	2	0.00%
Satisfactory	0	0	2	0	2	0.00%
Total	12	4	3	2	21	19.05%
Stepwise forward						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%
Good	0	19	0	0	19	100.00%
Moderate	0	0	2	0	2	100.00%
Satisfactory	0	0	0	2	2	100.00%
Total	1	19	2	2	24	100.00%
Stepwise backward						
From \ To	Bad	Good	Moderate	Satisfactory	Total	% Correct
Bad	1	0	0	0	1	100.00%
Good	0	14	2	0	16	87.50%
Moderate	0	1	1	0	2	50.00%
Satisfactory	0	0	0	2	2	100.00%
Total	1	15	3	2	21	85.71%

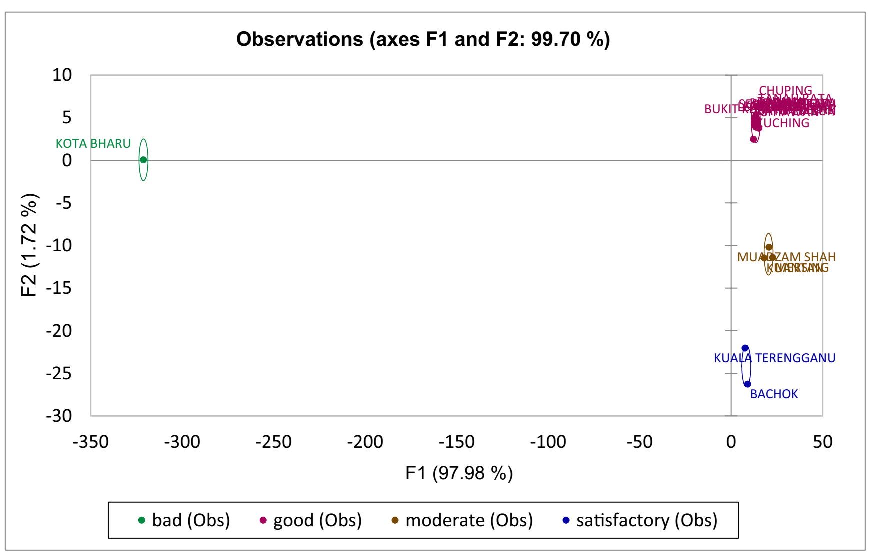
Fig. 5 The score plot 3 modes for BMCO data (**a**), (**b**), and (**c**)



a) Standard Mode BMCO

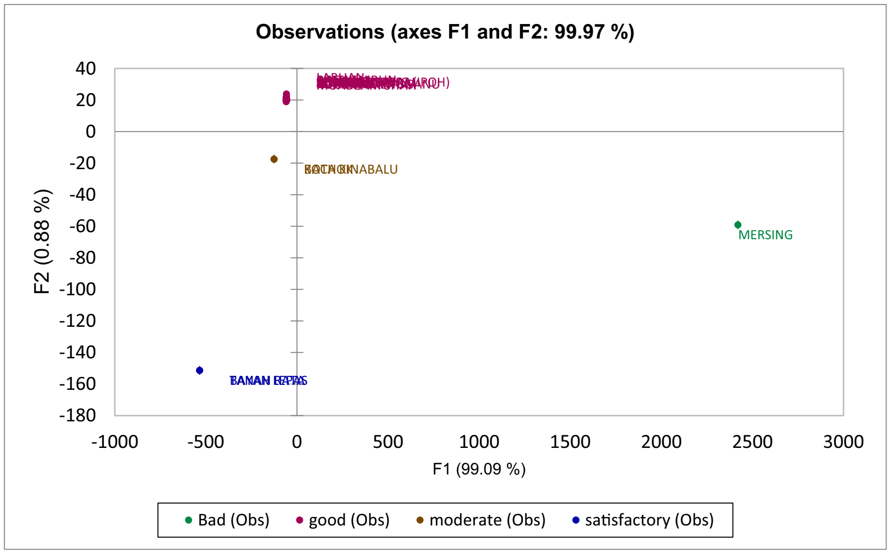


b) Stepwise Forward Mode BMCO

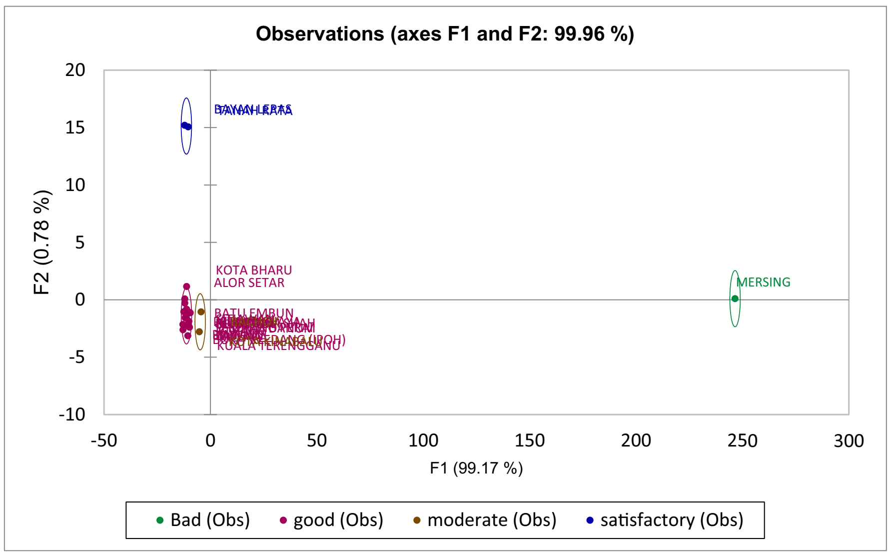


c) Stepwise Backward Mode BMCO

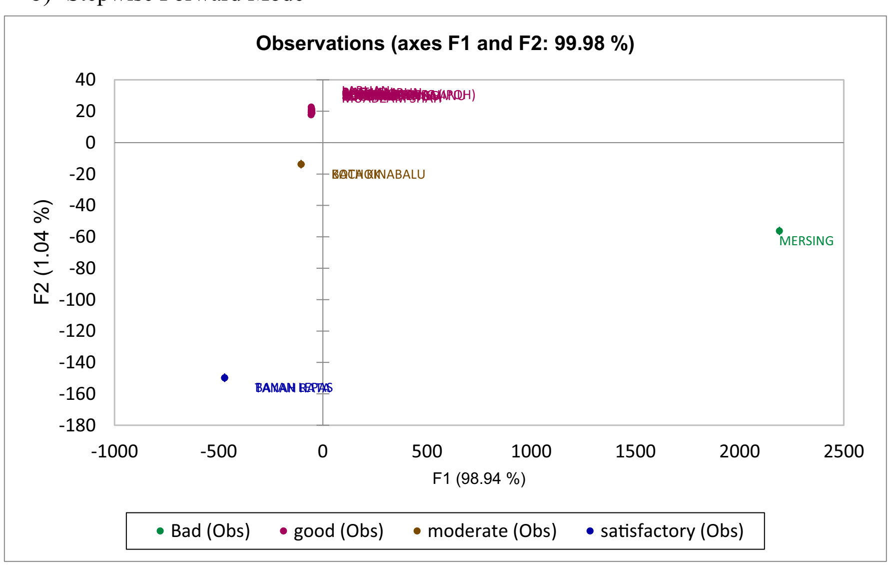
Fig. 6 The score plot by 3 modes for MCO data (**a**), (**b**), and (**c**)



a) standard mode



b) Stepwise Forward Mode



c) Stepwise Backward Mode

stepwise modes (Table 4). The MCO classification accuracy was 66.67% with 24 discriminant variables for the standard mode, whereas the accuracy of rainwater quality classification using the stepwise forward mode was 95.83% with four discriminant variables and the backward stepwise mode with an accuracy of 79.17% with 19 discriminant variables.

The standard mode score plot for different sampling areas (Fig. 5a, b, and c) illustrates that the confidence circle for each cluster (good, satisfactory, moderate, and bad) was separated from each other, and there were no significant similarities between the clusters. This was indicated based on the confidence circles for the stepwise forward mode with three discriminant variables and the backward stepwise mode with 19 discriminant variables.

The score plot for standard and stepwise backward mode (Fig. 6a and b) showed that the mean vector for all clusters differed from one another. However, for the stepwise forward mode, there was a slight difference in the mean vector between the moderate and good clusters, as indicated in Fig. 6c. The dataset was plotted, and the position between these two clusters was found to be very close. This indicates that cluster changes can occur easily between gauge stations found in the moderate and good clusters due to several factors, such as the time the sample was taken or slight changes in chemical compositions in the rainwater sample.

Conclusion

The introduction of the Movement Control Order (MCO) has had a significant impact on the quality of rainwater in Malaysia. Although there was an improvement in the quality of rainwater in some areas, there was a decrease in quality in some other places. Even when the MCO was implemented, essential activities were carried out as usual. In fact, some industries had to increase their production capacity to meet people's needs, mainly to control and treat COVID-19 patients and maintain the supply of essential items. Overall, the rainwater's mean pH value had increased. The rainwater samples were acidic, lower than normal rainwater (pH 5.6). Implementation of the MCO in March 2020 reduced the rainwater's acidity. Other than that, the mean pH for 11-gauge stations had increased, while 13-gauge stations saw a decrease in pH or an increase in the rainwater's acidity. There was a reduction in 12 chemical components in the rainwater after the MCO was implemented. Therefore, the rainwater quality has increased in Malaysia in some areas. But some areas are unchanged or worsening the impact on rainwater quality Apart from that, the content of NO_3^- and SO_4^2 ions from industries and vehicles has decreased after MCO. It is proof that the MCO situation has positively impacted the environment. The cluster analysis divided data into four categories, namely, good, satisfactory, moderate, and bad. Hence, the cluster analysis demonstrated that the stations had an improved rainwater quality by illustrating that four stations have improved the rank of the cluster analysis. All the DA results supported the spatial classification in the cluster analysis with accurate values. It should be noted that the implementation of the MCO affected the chemical content of rainwater. Still, the quality of rainwater was also influenced by many other factors, such location of pollutant sources and other meteorological and environmental conditions.

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Author contribution Nadiana Ariffin conceived the presented idea. Nadiana Ariffin and Hafizan Juahir developed the theory and performed the computations. Hafizan Juahir verified the analytical methods and supervised the findings of this work.

Nadiana Ariffin wrote the first draft manuscript.

Hafizan Juahir, Roslan Umar, and Mokhairi Makhtar supervised the project.

Nadiana Ariffin designed the model and the computational framework and analyzed the data.

Nadiana Ariffin and Munirah Abdul Zali processed the experimental data.

Azimah Ismail and Nur Hanis Mohamad Hanapi contributed to the final version of the manuscript.

Data availability The dataset generated and analyzed during the current study are not publicly available due to the protection of confidential information but are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare no competing interests.

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