ABSTRACT A comparative analysis of trace metal (Cu, Pb, Fe, Mn, Zn, Cd, Ni and Co) concentration and physical parameters (pH, EC, TDS and DO) in rainwater samples collected from two major coastal cities in Malaysian Borneo (Sarawak state) were determined in the present research. Cumulative monthly rainwater samples were collected from the Limbang city and Miri city during October 2016–September 2017. Rainwater collected from the Limbang city shows slightly alkaline nature with a mean pH = 6.07 whereas the rainwater in Miri city is acidic (mean pH = 5.35). Trace metal concentration in rainwater collected from both locations shows slight variation. Mean concentration of trace metals in rainwater samples follows the decreasing order of Fe > Ni > Pb > Mn > Co > Cu > Zn > Cd and Fe > Ni > Pb > Mn > Zn > Co > Cu > Cd in Limbang city and Miri city respectively. Among the trace metals, Fe (1.09 and 0.98 mg/L) and Ni (0.15 and 0.13 mg/L) shows the highest mean concentration in rainwater samples collected from both locations and maximum concentration of trace metals are observed in rainwater samples collected from the Limbang city. Pearson’s correlation test explained the inter-relationship between the parameters whereas the factor analysis confirmed the contributing sources of trace metals (anthropogenic activities such as pollution from vehicles, petrochemical industries, forest biomass burning and dust particles from exposed land area) and its variation in the rainwater samples by showing a total variance of 80.18% with three factor components in the Limbang city and a variance of 93.11% with four factor components in Miri city. High Pb/Zn ratio also indicates the strong influence of anthropogenic activities present in the region. Backward air mass trajectory analysis supports the findings by indicating a contribution from combined marine and crustal sources of air mass trajectories reaching the sampling locations and is heavily controlled by prevailing monsoon characteristics of the region. Overall, it can be concluded that, the major source of trace metals in rainwater in this region is contributed by anthropogenic processes operated in the region.

KEY WORDS Trace metals, Statistical analysis, HYSPLIT, Trajectory, Sarawak

1. INTRODUCTION

Rainfall is one of the important forms of precipitation which removes the pollutants reached in the atmosphere as the product of rapid urbanisation and industrial...
sation (Tiwari et al., 2016; Migliavacca et al., 2005). The composition of deposition actually reflects the composition of the atmosphere through which it falls. During precipitation events, the gases and aerosols are incorporated into raindrops falling down plays an important role in atmospheric deposition mechanisms (Özsoy and Örnektekin, 2009). The chemical characteristics of these gases and aerosols are controlled by their sources of origin i.e. natural or anthropogenic processes. Natural sources of chemical constituents present in the atmosphere are from crustal/marine areas or biological fragments through dust and salt spray whereas the emission processes dominate in the anthropogenic source of contribution (Rusydi, 2018; Sorooshian et al., 2013; Al-Khashman, 2009; Zhang et al., 2007; Kulshrestha et al., 2003). Chemical composition of rainwater is not only influenced by the local and regional transportation of pollutants but also controlled by the cloud formation and amount of rainfall (Ojekunle et al., 2018; Calvo et al., 2010; Sarkar et al., 2006).

Understanding the role of chemical characteristics (enriched load of chemicals) of rainfall over the human health and ecosystem, numerous studies were carried out in different parts of the world to estimate precipitation chemistry i.e. both wet and dry precipitation (Williams et al., 2021; Rivera-Rivera et al., 2020; Silva et al., 2020; Greuling et al., 2019; Khayan et al., 2019; Li et al., 2017; Williams et al., 2015; Cerqueira et al., 2014; Sorooshian et al., 2013; Gunawardena et al., 2012; Calvo et al., 2010). Industrialised and neighbouring areas in most of the countries witnessed severe acid rain, which unfavourably affected aquatic system, soil, vegetation, buildings, etc (Li et al., 2017; Bravo et al., 2000; Zhang et al., 1999).

Among the chemical constituents present in the rainfall, concentration of trace metals acquired special attention due to toxic nature, bioaccumulation and carcinogenic properties of these metals (Siudek and Frankowski, 2017). Anthropogenic activities such as industrial activity including mining, combustion of petro-chemical and coal substances and automobile exhausts are the major processes contribute and control the concentration of trace metals in the atmosphere (Siudek and Frankowski, 2017; Lynam et al., 2014; Moreda-Piñeiro et al., 2014). The rainwater sample collection locations identified in the Sarawak state of Malaysian Borneo lack detailed information about the trace metal concentration in rainwater even the region having number of petrochemical industries and off-shore exploitation platforms. Research reports available from this region given importance to the forest fire related haze and its effect over the atmospheric characteristics (Nadzir et al., 2017; Gomyo et al., 2008; Radojevic, 2003; Radojevic and Tan, 2000). Considering the importance of generating a baseline report on trace metal concentration in rainwater reaching this region, the present research was framed with an objective of comprehending the concentration and source contributors of trace metals in rainwater reaching the spatially separated coastal cities in northern Sarawak, Malaysia through hydro-statistical analysis.

1.1 Study Area

In the present research, rainwater samples collected from two spatially separated coastal cities in northern Sarawak (Malaysia, Borneo), namely Limbang and Miri were analysed to identify trace metal concentration and its variability over a period of one year from October 2016 to September 2017 (Fig. 1). Limbang city is the administrative capital of the Limbang district covers an area ≤ 35 km² with a total urban population less than 15,000, located in the banks of the Limbang River, which drain into the Brunei bay before joining the South China Sea. Miri city is the second largest city (the area of 977 km² with a total urban population of 300,000) in Sarawak and located in between the Miri River and the Baram River, which drains directly into the South China Sea. Though these two places are located in northern Sarawak, both are spatially separated at 118 km. Even though the region shows highly varying geomorphological and geological characteristic, both the sampling locations are situated in the lower peat swamps or lower peat forest regions. In general, elevation in this region varies in the range of 1 m to 2,400 m above the mean sea level, which shows highly varying topographical features and includes low-lying flood plains (peat swamps), residual mounds, elongated denudating structural hills, escarpments, valleys and highland plateaus. Geologically the region is dominated by sedimentary rock formations with occasional very minor basaltic rocks (MGDM, 2013). Tropical rainforests (primary and secondary) dominantly cover the area (more than 90%) with regional oil palm plantations and very local scale agricultural activities such as wet paddy, hill paddy and fruit orchards.

The selected study locations are situated in the northern Borneo facing the South China Sea experiencing 10 months monsoon seasons with very short inter-monsoon periods. May to September considered as southwest
monsoon (SWM) and November to March as northeast monsoon (NEM) and are separated by individual inter-monsoon months of April and October. Though the general monsoonal seasons in the sampling locations are same, both show higher variation in meteorological characteristics such as amount of rainfall and temperature (Table 1).
Considering the sampling locations, Limbang city recorded the highest mean monthly and annual rainfall of 503 mm and 6,039 mm whereas Miri city recorded comparatively lower mean monthly and annual rainfall of 317 mm and 3,809 mm respectively. Monthly rainfall during the study period varies in the range of 257 mm (July 2017) to 794 mm (April 2017) in Limbang city and 149 mm (July 2017) to 459 mm (August 2017) in Miri city (Fig. 2a). It was observed that recorded highest monthly rainfall in these stations shows a difference of 335 mm. Monthly mean temperatures in the study locations varies between 26°C to 27°C in Limbang city and 26°C to 28°C in Miri city (Fig. 2b). Considering the wind direction, wind recorded in the Limbang city shows dominance of southwest and west direction whereas those recorded at Miri city showed dominance of southwest and north direction during the period of research.

2. MATERIALS AND METHODS

In order to analyse and determine the concentration of trace metals in rainwater reaching two coastal cities in northern Sarawak, Malaysian Borneo, monthly cumulative rainwater samples during the periods including October 2016 and September 2017 was collected. A total of 12 samples from each location was collected using rainwater collection apparatus made up of polyethylene barrels (5 L) and funnels (10 cm diameter) which was kept above the ground (80 cm) using a metal frame to avoid the splash effect from the ground surface. Before setting up the collection apparatus, the polyethylene barrels and funnels were thoroughly cleaned using distilled water and a small hosepipe was connected in the top portion of the barrel to minimise the exchange of air within and outside the sample collector (Oza et al., 2020; Jeelani et al., 2018; IAEA, 2014; Deshpande and Gupta, 2008). Later, to prevent the evaporation, paraffin oil (0.5 cm thickness) was put in the container. The cumulative monthly rainwater collected was then transferred into two 1 L polyethylene bottles after filtering using Whatman no. 1 filter paper (11 μm) by end of each month. These samples were preserved in lab condition and were used for the analysis. Physical parameters such as pH, electrical conductivity (EC), total dissolved solids (TDS) and dissolved oxygen (DO) of rainwater were measured in the laboratory using portable pH, conductivity meter (Thermo Scientific Orion Star, 4 Star Plus Meter), and Hach meter (Hach® DR2800 spectrophotometer). Later the water samples were filtered using 0.45 μm Millipore filter paper and the filtered water was then digested using 10 mL concentrated Nitric acid (HNO₃) to lower the pH~2 and make to 10 mL by heating around 60 – 70°C using hotplate (APHA, 2012). The concentrated sample is then remade to 100 mL using ultra-pure water for the analysis of trace metals (Cu, Pb, Fe, Mn, Zn, Cd, Ni and Co) using Atomic Absorption Spectrophotometer (Perkin Elmer Analyst 400).

Further, the analysis results are taken into statistical software to perform Pearson’s correlation and factor analysis to identify the inter-relationships and source contribution of trace metals in rainwater. As part of assessing the contribution of anthropogenic sources in the concentration of trace metals, the elemental ratio analysis of Pb/Zn suggested by Sakata and Asakura (2009) was implemented in the present research. This will assist to identify sources of external inputs of industrial pollution from short (local and regional) and long-range transports (Rivera-Rivera et al., 2020; Sakata and Asakura, 2009; Okuda et al., 2004). Then, to identify and confirm the source contribution of chemical constituents measured in rainwater samples collected from these locations, HYbrid Single-Particle Lagrangian Integrated Trajectory (HYPLIT) transport model based air mass back trajectory analysis managed by the United States National Atmospheric and Oceanic Administration (NOAA) Air

| Sampling period | Rainfall (mm) | Temperature (°C) |
|-----------------|--------------|-----------------|
|                 | Limbang      | Miri            | Limbang      | Miri            |
| Oct-16          | 306.19       | 190.14          | 26.29        | 27.36           |
| Nov-16          | 617.61       | 333.28          | 26.21        | 27.30           |
| Dec-16          | 439.52       | 260.97          | 26.36        | 27.43           |
| Jan-17          | 401.65       | 340.25          | 26.50        | 27.39           |
| Feb-17          | 492.97       | 399.94          | 25.82        | 26.61           |
| Mar-17          | 564.03       | 313.94          | 25.90        | 26.74           |
| Apr-17          | 794.60       | 420.03          | 26.64        | 27.56           |
| May-17          | 641.29       | 377.83          | 27.00        | 28.07           |
| Jun-17          | 274.68       | 219.18          | 26.59        | 27.74           |
| Jul-17          | 257.24       | 149.57          | 26.49        | 27.72           |
| Aug-17          | 664.35       | 459.63          | 26.50        | 27.60           |
| Sep-17          | 585.47       | 344.81          | 25.82        | 26.62           |
| Mean            | 503.30       | 317.47          | 26.41        | 27.43           |
| Annual          | 6039.60      | 3809.63         |              |                 |
Resources Laboratory (ARL) was also analysed for individual months. This will provide reliable information about the transportation of chemical constituents contributing to the rainfall reaching a particular location (Williams et al., 2021; Srivastava et al., 2020; Innocente et al., 2017; Rolph et al., 2017; Stein et al., 2015; Moreda-Piñeiro et al., 2014).

3. RESULTS AND DISCUSSION

3.1 Physical Characteristics of Rainwater

Table 2 shows the results of parameters like pH, EC, TDS and DO measured from rainwater samples collected from Limbang city and Miri city. Fig. 3 shows comparison of monthly values of parameters analysed. In Limbang city, rainwater pH varies in the range of 5.33 to 7.14 with a mean of 6.07. EC varies from 58.17 to 117.16 μS/cm with a mean of 87.56 μS/cm. TDS shows variation from 40.72 to 82.01 mg/L with a mean of 61.29 mg/L. DO in the rainwater during the period of study range from 3.40 to 9.19 mg/L and shows a mean of 6.92 mg/L. At the same time, rainwater samples collected from Miri city shows slight variation in concentration of parameters analysed. In Miri, pH in the rainwater samples varies in the range of 4.66 to 6.09 with a mean of 5.35. EC varies from 52.61 to 108.91 μS/cm with a mean of 83.02 μS/cm. TDS varies in the range of 36.83 to 76.23 mg/L with a mean of 58.11 mg/L. DO during the period of study range from 7.20 to 9.92 mg/L and shows a mean of 8.30 mg/L.

Rainwater collected from the Limbang city and Miri city shows slight variation in the concentration of pH, EC, TDS and DO. Considering the concentration of pH, rainwater falling in the Limbang city shows comparatively higher values than those in Miri city and is well reflected in the mean value also. The rainwater samples collected show slightly higher mean pH (6.07) in the Limbang city and this might be due to the presence of increased amount of particulate matter in the atmosphere and also due to the local influence of evaporation over rainfall characteristics (Rajeev et al., 2016; Sanhueza et al., 1989; Galloway et al., 1982). The mechanism behind this is the water soluble particulate matter and gaseous aerosols present in the atmosphere through which raindrop travel thousands of meters are responsible for the nature of rainfall i.e. increase or decrease of pH at each location. This is also highly influenced by the rainfall amounts, diluting concentration of most of the ions with higher rainfall amounts (Ramírez Lara et al., 2010; Shende et al., 2007). Further, due to the location, the dominant way of precipitation is convective in nature and always there will be a higher level moisture contribution through local evaporation (Galloway et al., 1982). Lower mean pH (5.35) observed in the Miri suggests the presence of more concentration of NOx and SO2 in the atmosphere (Charlson and Rodhe, 1982). This indicates the presence of petrochemical industries and offshore petroleum production centres in the region.

Considering the electrical conductivity (EC) and total dissolved solids (TDS), rainwater samples collected from Miri city shows slightly lower concentration than the rainwater samples collected from the Limbang city with mean

| Parameters | Limbang city | Miri city |
|------------|-------------|-----------|
|            | Min. | Max. | Mean | Min. | Max. | Mean |
| pH         | 5.330 | 7.140 | 6.070 | 4.660 | 6.090 | 5.350 |
| EC (μS/cm) | 58.170 | 117.160 | 87.560 | 52.610 | 108.910 | 83.020 |
| TDS (mg/L) | 40.720 | 82.010 | 61.290 | 36.830 | 76.230 | 58.110 |
| DO (mg/L)  | 3.400 | 9.190 | 6.920 | 7.200 | 9.920 | 8.300 |
| Co (mg/L)  | 0.008 | 0.050 | 0.025 | 0.005 | 0.045 | 0.023 |
| Ni (mg/L)  | 0.083 | 0.210 | 0.146 | 0.069 | 0.219 | 0.129 |
| Cd (mg/L)  | 0.003 | 0.0097 | 0.006 | 0.0035 | 0.011 | 0.0057 |
| Fe (mg/L)  | 0.961 | 1.609 | 1.09 | 0.715 | 1.63 | 0.975 |
| Mn (mg/L)  | 0.022 | 0.042 | 0.030 | 0.013 | 0.043 | 0.026 |
| Pb (mg/L)  | 0.012 | 0.115 | 0.035 | 0.012 | 0.088 | 0.036 |
| Zn (mg/L)  | 0.002 | 0.030 | 0.014 | 0.002 | 0.043 | 0.024 |
| Cu (mg/L)  | 0.007 | 0.026 | 0.015 | 0.005 | 0.021 | 0.012 |
Concentration of EC and TDS is directly proportional and is heavily depends on the concentration of dissolved ions, ionic strength, and temperature of water (Abulude et al., 2018; Marandi et al., 2013) showing local influence over temporal variation in concentration. Higher values of EC and TDS indicate the higher concentration of particulate matter in the atmospheric air within the study locations (Cobbina et al., 2013). Variation in concentration of DO in rainwater samples was noted with higher mean DO in rainwater samples collected from Miri city (8.30 mg/L) than the Limbang city (6.92 mg/L). Due to the specific geographical location of the study sites i.e. nearness to equator, the region experiences different weather and atmospheric processes like structure of the raincloud and height of cloud base, mechanism of raindrop formation and diffusion from the surrounding air and by aeration (rapid movement of raindrops) might have influenced the higher concentration of DO in rainwater samples collected from these locations (Williams and Tighiri, 2015; Komabayasi, 1959). Overall, higher values of physical parameters measured from these locations indicate increased presence of particulate matter, which indirectly point towards plausible concentration of trace metals in the region because particulate matters are the scavengers and carriers of trace metals (Pizarro et al., 2021; Majumda et al., 2020; Martin et al., 2017; Sjöblom et al., 2010).

3.2 Trace Metal Concentration

Concentration (minimum, maximum and mean) of trace metals in rainwater samples collected from Limbang city and Miri city are given in Table 2 and comparison of monthly concentration is shown in Fig. 4. The assessed values show slight variation in concentration between the locations. Mean concentration of trace metals also shows variation among the sampling locations. Rainwater samples collected from Limbang city shows mean concentration of Co, Ni, Cd, Fe, Mn, Pb, Zn and Cu as 0.025, 0.146, 0.006, 1.09, 0.030, 0.035, 0.014 and 0.015 mg/L respectively. Considering the mean concentration of trace metals in rainwater samples collected from Miri city, it shows slight variation than rainwater samples collected from the Limbang city. Mean trace metal concentration in rainwater samples collected from Miri city was 0.023, 0.129, 0.0057, 0.975, 0.026, 0.036, 0.024 and 0.012 mg/L for
Co, Ni, Cd, Fe, Mn, Pb, Zn and Cu respectively. While considering the relative abundance of trace metals in rainwater samples collected from the Limbang city follows the decreasing order of Fe > Ni > Pb > Mn > Co > Cu > Zn > Cd and for Miri city it shows the decreasing order of Fe > Ni > Pb > Mn > Zn > Co > Cu > Cd respectively. Though variability in concentration of trace metals in
individual rainwater sampling locations was observed in the study, it doesn’t show any particular pattern. Co, Ni, Fe, Mn, and Cu shown comparatively higher concentration in rainwater samples collected from Limbang city whereas Pb and Zn show relatively higher concentration in rainwater samples collected from Miri city. Concentration of Cd shows similar value in both locations. The major process that controls the concentration of trace metals in rainwater is the atmospheric pollutant scavenging (Hasan et al., 2017). Source contributors of trace metals in atmosphere are either natural or anthropogenic such as forest fire, dust from mining and exposed areas, petrochemical pollutions due to fossil fuel combustion for energy production and also through motor vehicles (Rivera-Rivera et al., 2020; Sabuti and Mohamed, 2016; Ghadimi et al., 2013; Vuai and Tokuyama, 2011; Farahmandkia et al., 2010). In the study locations, both have the influence of petrochemical industries including off-shore drilling, heavy industrial and transportation activities, dust derived from forest burning and logging activities. These might have contributed the trace metals in the rainwater and the variation in concentration can be by the wind reaching the region, which carries the materials originated from marine and or land sources.

3.3 Statistical Analysis of Rainwater Characteristics

3.3.1 Correlation

In order to identify the inter-relationship and dependency of parameters present in the rainwater samples, Pearson’s correlation test was performed with a significance level of 0.05. Analysis revealed varying inter-dependencies (either positive or negative) between the parameters and correlation coefficient \( r \geq \pm 0.60 \) were considered as strong correlation in the present study. Correlation test results of parameters analysed from the rainwater samples collected from the Limbang city was given in Table 3. In Limbang city rainwater, pH shows strong positive correlation with Fe and Pb \( (r = 0.79 \text{ and } 0.70) \). EC shows very strong positive correlation with TDS \( (r = 1) \) and strong negative correlation with Co, Ni and Zn \( (r = -0.83, -0.70 \text{ and } -0.75 \text{ respectively}) \). TDS showed strong negative correlation with Co, Ni and Zn \( (r = -0.83, -0.70 \text{ and } -0.75 \text{ respectively}) \). A strong positive correlation was noted between Ni with Zn and Cu \( (r = 0.73 \text{ and } 0.75) \). Fe also showed a very strong positive correlation with Pb \( (r = 0.85) \) whereas Zn showed a strong positive correlation with Cu \( (r = 0.63) \). However, rainwater samples collected from Miri city shown comparatively different correlation characteristics than those observed in Limbang city (Table 4). pH showed a strong positive correlation with EC and TDS \( (r = 0.74) \) and a strong negative correlation with Ni and Cu \( (r = -0.89 \text{ and } -0.72) \). EC shows a strong positive correlation with TDS \( (r = 1) \). EC and TDS showed a strong negative correlation between Co, Fe and Cu \( (r = -0.83, -0.89 \text{ and } -0.67 \text{ respectively}) \). A strong positive correlation was observed between DO with Co, Fe and Pb \( (r = 0.61, 0.76 \text{ and } 0.84 \text{ respectively}) \). Co showed strong positive correlation with Fe \( (r = 0.80) \). Ni showed strong positive correlation between Cu \( (r = 0.64) \). A strong positive correla-

| Variables | pH | EC (μS/cm) | TDS (mg/L) | DO (mg/L) | Co (mg/L) | Ni (mg/L) | Cd (mg/L) | Fe (mg/L) | Mn (mg/L) | Pb (mg/L) | Zn (mg/L) | Cu (mg/L) |
|-----------|----|-----------|------------|----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| pH        | 1  | -0.10     | 1          | -0.10    | 1         | 1         | 1         |           |           |           |           |           |
| EC (μS/cm)| -0.10| 1   | -0.34     | -0.34    | 0.33      | 1         |           |           |           |           |           |           |
| TDS (mg/L)| 0.49| -0.83   | -0.83     | -0.83    | 0.33      | 1         |           |           |           |           |           |           |
| DO (mg/L) | 0.28| -0.70    | -0.70     | -0.70    | 0.10      | 0.53      | 1         |           |           |           |           |           |
| Co (mg/L) | -0.41| 0.11   | 0.11      | 0.29     | -0.14     | -0.12     | 1         |           |           |           |           |           |
| Ni (mg/L) | 0.34| 0.79    | -0.24     | -0.24    | 0.43      | 0.16      | -0.40     | 0.38      | 1         |           |           |           |
| Cd (mg/L) | 0.30| -0.04   | 0.04      | 0.09     | -0.24     | -0.05     | 0.48      | 0.08      | 1         |           |           |           |
| Fe (mg/L) | 0.70| 0.07    | -0.51     | -0.51    | 0.56      | 0.30      | -0.02     | 0.47      | 0.85      | 0.26      | 1         |           |
| Mn (mg/L) | 0.30| -0.04   | 0.04      | 0.09     | -0.24     | -0.05     | 0.48      | 0.08      | 1         |           |           |           |
| Pb (mg/L) | 0.70| -0.51   | -0.51     | 0.56     | 0.30      | -0.02     | 0.47      | 0.85      | 0.26      | 1         |           |           |
| Zn (mg/L) | -0.10| 0.75   | -0.75     | 0.04     | 0.48      | 0.73      | 0.21      | 0.15      | 0.15      | 0.42      | 1         |           |
| Cu (mg/L) | -0.32| -0.44   | -0.44     | 0.24     | 0.27      | 0.75      | 0.26      | -0.26     | 0.22      | 0.14      | 0.63      | 1         |

Values in bold are different from 0 with a significance level alpha = 0.05
tion was observed between Cd and Pb \( (r = 0.67) \) and Fe and Pb \( (r = 0.85) \). These results indicate the variability of source contributors of rainfall in these two locations along with change in characteristics of local moisture sources because strong negative and low correlation suggests different source of origin whereas strong positive correlation indicates same source of origin (Silva et al., 2020; Tripathi et al., 2014; Owoade et al., 2012; Helmreich et al., 2010).

### 3.3.2 Factor Analysis

Results of the physical and trace metal analysis are used to extract the factor components through Principal component analysis (PCA) to identify and explain the source of trace metals in rainwater samples. Varimax rotation technique used to extract the factor components (Eigen value > 1), because it will aid to make results easier to interpret by minimising the variance of squared normalized factor loadings across variables for each factor (Báez et al., 2007). Factor analysis results of Limbang city and Miri city rainwater’s were shown in Table 5. Factor loading between \( \pm 0.50 \) to \( \pm 0.75 \) considered as good loading and \( \geq \pm 0.75 \) as strong loading in the present research. Considering the rainwater sample analysis results of Limbang city, factor analysis revealed three factor components which explained 80.18% of total variance. Factor 1 explained 37.18% of total variance with strong negative loadings of EC and TDS and strong positive loadings of Co, Ni, Zn and good loading of Cu. Positive loading of Co, Ni, Zn and Cu indicates the contribution of pollutants from industrial and traffic activity present in region (Ghadimi et al., 2013; Vuai and Tokuyama, 2011). Factor 2 explained 27.92% of total variance with strong positive loading of pH, Fe and Pb and good loading of DO. pH and DO are the physical parameters which determine the overall water chemistry by dissolution of other major ions and trace metals. Fe and Pb in the rainwater can be by the anthropogenic-pollution based factors such as fossil fuel burning (petrochemical industries and transportation) and forest fires in the region (Cheng et al., 2011). Factor 3 explained 15.08% of total variance with strong positive loading of Cd and good loading of Mn. The source for Cd and Mn in the rainwater samples are from anthropogenic activities (iron and steel manufacturing industry, fossil fuel combustion and mining) as well as from natural factors such as sea salt spray, forest fire, windblown dust rich in Mn (Ressler et al., 2000; Nriagu, 1989).

In the case of parameters identified in rainwater samples collected from the Miri city, factor analysis revealed four factor components which explained 93.11% of total variance. Factor 1 explained 37.49% of total variance with strong negative loadings of EC and TDS and strong positive loadings of DO, Co, Fe and Pb. Co, Fe and Pb in the rainwater can be attributed by the atmospheric pollution caused by the anthropogenic activities such as fossil fuel burning (petrochemical industries and transportation) and forest fires (biomass burning) in the region (Cheng et al., 2011). Factor 2 explained 30.21% of total variance with strong positive loading of Ni and Cu and strong negative loading of pH. Presence of Ni and Co indicates the contribution of pollutants from industrial
and traffic activity present in the region (Ghadimi et al., 2013; Vuai and Tokuyama, 2011). Factor 3 explained 15.15% of total variance with strong positive loading of Cd and Zn. This suggests the influence of anthropogenic processes such as pollution from vehicles, petro-chemical industries, forest (biomass) burning along with dust particles transported from distant palaces as well as from local areas. Factor 4 explained 10.26% of the total variance with strong positive loading of Mn and it can be from anthropogenic (iron and steel manufacturing industry, fossil fuel combustion and mining) as well as natural factors such as sea salt spray and forest fires (Moreda-Piñeiro et al., 2014). Though different loading of trace metals in various factors were observed in the rainwater samples collected from the Limbang city and Miri city, cumulatively it points towards few major sources and processes which contribute the trace metal fraction in the atmosphere of the region. These processes are dominated by anthropogenic activities like burning of fossil fuel and biomass (forest) with comparatively minor contribution from natural sources like fine soil dust particles from exposed and barren lands in the region (Williams et al., 2021; Orlović-Leko et al., 2020; Akpan et al., 2018; Gunawardena et al., 2012; Sternbeck et al., 2002; Allen et al., 2001).

### 3.3.3 Elemental Ratio (Pb/Zn ratio)

The elemental ratio of Pb/Zn examined in the present study shows varying values in monthly rainwater samples collected from the Limbang city and Miri city (Fig. 5). In both the locations, the Pb/Zn ratio varies from 0 to 1 but shows difference in mean ratio i.e. 0.28 and 0.20 for the Limbang city and Miri city respectively. Lower values (< 0.25) of Pb/Zn ratio were observed in the months of October, December, January, February, March and July in rainwater collected from the Limbang city whereas in the Miri city, rainwater samples collected during the months of December, January, February, March, April, May, August and September shows the lower ratio of Pb/Zn. Higher Pb/Zn (≥ 0.25) ratios are observed in the months of November, April, May, June, August and September in rainwater collected from the Limbang city and October, November, June and July from the Miri city. Both locations show maximum value (1) in June. The observed higher values indicate the favorable atmospheric conditions such as lower mixing layer height, boundary layer dynamics, local transport processes, and thermal inversion that resulted in the accumulation of trace metals (Rivera-Rivera et al., 2020; Serafin et al., 2018; Siudek and Frankowski, 2017). Considering the seasonality in Pb/Zn ratios, rainwater collected during the south-west monsoon season shows higher ratio in Limbang city whereas the

### Table 5. Results of factor analysis of physico-chemical parameters in rainwater samples collected from the Limbang city and Miri city.

| Parameters | Limbang city | Miri city |
|------------|-------------|-----------|
| F1 | F2 | F3 | F1 | F2 | F3 | F4 |
| pH | -0.12 | 0.92 | 0.09 | -0.22 | -0.89 | 0.01 | -0.30 |
| EC | -0.95 | -0.27 | 0.10 | -0.78 | -0.60 | -0.01 | -0.14 |
| TDS | -0.95 | -0.27 | 0.10 | -0.78 | -0.60 | -0.01 | -0.14 |
| DO | 0.27 | 0.58 | 0.17 | 0.86 | -0.41 | 0.24 | 0.10 |
| Co | 0.78 | 0.30 | -0.14 | 0.90 | 0.25 | -0.25 | 0.04 |
| Ni | 0.88 | -0.39 | 0.08 | -0.07 | 0.88 | 0.05 | 0.36 |
| Cd | -0.12 | 0.30 | 0.86 | 0.30 | -0.08 | 0.85 | 0.11 |
| Fe | -0.01 | 0.93 | 0.07 | 0.91 | 0.26 | 0.28 | 0.12 |
| Mn | 0.04 | 0.17 | 0.69 | 0.15 | 0.19 | 0.01 | 0.92 |
| Pb | 0.32 | 0.83 | 0.31 | 0.81 | -0.08 | 0.52 | -0.02 |
| Zn | 0.82 | 0.00 | 0.31 | -0.13 | 0.47 | 0.76 | -0.13 |
| Cu | 0.65 | -0.32 | 0.55 | 0.25 | 0.88 | 0.19 | -0.26 |
| Eigen value | 4.77 | 3.27 | 1.59 | 5.79 | 2.77 | 1.59 | 1.01 |
| Variability (%) explained | 37.18 | 27.92 | 15.08 | 37.49 | 30.21 | 15.15 | 10.26 |
| Cumulative (%) variability | 80.18 | 93.11 |  |  |  |  |  |

Bold italic values indicate strong loading.
Miri city does not follow any specific pattern and few higher ratios are observed in north-east and south-west monsoon seasons. High Pb/Zn ratio in the Limbang city is strongly influenced by anthropogenic activities such as off-shore petroleum exploration and natural gas combustion, local (both short range and long range) transportation processes and dust derived from interior land region which brought the trace metals into the specific locations.

3.4 Source Determination through HYSPLIT Model

To confirm the sources of chemical constituents reaching Limbang city and Miri city, backward air mass trajectory analysis for the period of rainwater sample collection was regenerated from NOAA ARL HYSPLIT transport model. In air quality and pollution movement modelling, trajectory analysis is widely used (Ma et al., 2021; Sulaymon et al., 2021; Grewling et al., 2019; Punsompong and Chantara, 2018; Tatsuta et al., 2017; Schwarz et al., 2016; Chen et al., 2013) because it helps to determine the origin and sources of pollutants in the atmosphere which reaching the surface through rainfall (Tu et al., 2019; Stein et al., 2015). In the present research, cumulative monthly representation for the whole study period (i.e. October 2016 to September 2017) was considered to build the backward air mass trajectory at a height of 1,000 m above the ground level for the individual stations.

Air mass backward trajectories reaching the Limbang rainwater sampling location is shown in Fig. 6. During 2016 October in Limbang sampling station, pollutants reaching from western and south-western sides of the study area indicate dominant marine contribution. In 2016 November, trajectories shows almost mixed crustal and marine source with dominant marine contribution and a change in the direction i.e. the pollutants reaching the sampling points from north-east direction. A drastic change in the direction of air mass reaching the sampling point was found in 2016 December. It displays a distributed nature with dominated crustal contribution with less attribute from marine sources. Crust/land dominated pattern with less contribution from marine sources of air mass reaching the Limbang was continued in January 2017 with a directional change to north-east i.e. from the Sabah landmass. In February 2017, air mass trajectory shows a mixed contribution of marine and crustal sources i.e. from the Sabah land mass in north-east and the South China Sea in north and the Sulu Sea in north-east. Air mass trajectory continued the same trend in the month of March 2017, with dominance of crustal (north-east, Sabah landmass) with minor contribution from marine (the South China Sea and Sulu Sea) sources. In April 2017, it shows a distributed (spread) nature with contribution from crustal as well as marine sources. During May 2017, air mass trajectory reaching the sampling point revealed a distributed nature with mixed contribution (crustal and marine sources) in which crustal contribution dominates. In June 2017, air mass trajectories shown a change in its direction with south-west direction dominated trend with almost equal contribution from marine (the South China Sea) and crust i.e. Borneo landmass. In July 2017, air mass
trajectory continued the same directional trend i.e. south-west with a dominant contribution from marine source. However, in August 2017, the air mass trajectories shown distributed nature indicating combined influence of marine and crustal sources with dominance of crustal contribution. In September 2017 the air mass reaching the study area showed dispersed nature as recorded in the month of August, indicating contribution from crustal as well as marine sources.

Air mass backward trajectories reaching the Miri rainwater sampling location is shown in Fig. 7. Cumulative monthly air mass trajectory analysis of October 2016 shows slope contribution from the South China Sea on north-west side of the Miri City. In November 2016, the pattern of air mass trajectories reaching the sampling point changes to a distributed or dispersed nature which shows mixed contribution from marine and crustal sources. The same dispersed pattern of air mass trajectories were observed in December 2016, with mixed marine and crustal contribution of pollutants. In January 2017, the air mass reaching the sampling point indicates both marine and crustal contribution to the chemical characteristics of rainfall and the air mass shows dominant direction of origin from north-eastern part of the study area i.e. from the Sabah landmass. The same pattern of air mass trajectories continued in the months of February 2017 and March 2017, with a mixed contribution from marine (South China Sea and Sulu Sea) and crustal (Sabah landmass) sources. In April 2017, air mass trajectories had shown a dispersed pattern with the contribution of chem-
ical constituents in rainwater from crustal as well as marine sources. During May 2017, rainwater was chemically enriched by dominant marine contribution with less input from crustal sources. The chemical characteristic of rainwater during the month was controlled by the air mass reaching from the South China Sea from north-west and north direction as observed in the backward trajectory analysis data which showed a dispersed pattern with more concentration toward marine side. In the month of June 2017, trend of air mass trajectories indicates dominant marine contribution i.e. the South China Sea from west-south-west direction as inferred from the air mass trajectory analysis. The same trend continued in July 2017 indicating foremost contribution from the South China Sea. However, a change in trajectory pattern i.e. dispersed pattern, with a combined contribution of crust and marine as the dominant source was noted in during August and September 2017. During these months, the air mass trajectories found to be originated from almost all direction with prevailing south-west direction.

Overall, the analysis of air mass backward trajectories provided information about the trace metal concentrations observed in the rainwater samples as local and transported pollutants from various sources, suggesting contribution from combined marine and land sources (Anil et al., 2019; Schneider et al., 2019; Caggiano et al., 2017; Likuku et al., 2013). As inferred from the monthly cumulative backward trajectory based on HYSPLIT model, both the sampling locations indicate major source contribution as marine and that from the southwest and west.
direction, during in the months of October (2016), June, July, August and September (2017) because the prevailing season is south-west monsoon (SWM). Similarly, during December (2016), January, February and March (2017), majority of the backward trajectories indicate contribution from dominant land (crustal) sources with very less contribution from marine with major direction from north-east and east side of the sampling locations. During these months, the dominant monsoon season is the north-east monsoon (NEM). Based on these, the dominant sources which contribute trace metals to rainwater reaching the region are petro-chemical drilling (off-shore), transportation (road and ship), biomass burning and soil dust particles derived from the non-protected areas in the land due to intense logging activities present in the region (Sabuti and Mohamed, 2016).

4. CONCLUSION

A year-long collection and analysis of rainwater samples from spatially separated coastal cities, namely Limbang city and Miri city in Malaysian Borneo focused to determine the concentration of trace metals (Cu, Pb, Fe, Mn, Zn, Cd, Ni and Co) and plausible sources of trace metal enrichment were performed during October 2016 to September 2017. Physical parameters analysed (pH, EC, TDS and DO) shows variations in values among two sampling locations with maximum values are shown in the samples collected from the Limbang city than the Miri city. Trace metal concentration also follows almost same pattern with a higher concentration of Co, Ni, Fe, Mn, and Cu in rainwater samples collected from the Limbang city and Pb and Zn in samples collected from Miri city. Statistical analysis (Pearson correlation and Factor analysis) provided insight into probable contributing sources of trace metals in rainwater samples by highlighting anthropogenic activities as the primary sources along with limited natural supply. This finding was complemented by the observed high rate of Pb/Zn ratio in rainwater samples, especially in samples collected from the Limbang city (more than 6 months). This can be attributed to the closeness of Limbang city with Brunei, where the petro-chemical drilling, burning of natural gas, shipping and water transportation activity is dominant (especially in Brunei Bay and off shore region) than Miri region. Further, this point towards the control of meteorological characteristics, particularly, local and long-range transportation of air mass in the region over the trace metal concentration in rainwater. The air mass backward trajectory analysis also provided an important insight into the dynamics of atmospheric processes took place in the study area. This indicates the combination of marine and crustal origin of trace metal and its movement over the spatial and temporal domain is heavily controlled by the prevailing monsoon characteristics of the region. The findings of the present research point towards the necessity of setting up a network of rainwater sampling locations in the region along with air quality monitors for periodical monitoring of chemistry of rainwater and air to reduce the impending health risk for the human and the total environment.

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REFERENCES

Abulude, F., Ndaitso, M.M., Abdulkadir, A. (2018) Environmental situation of an agricultural area in Akure, Nigeria, based on physico-chemical properties of rainwater. Pollution, 4(2), 317–325. https://doi.org/10.22059/poll.2017.242241.322
Akpan, A.O., Udosen, E.D., Offiong, N.A.O. (2018) Rainwater chemistry within the vicinity of Qua Iboe Estuary, Nigeria. CLEAN Soil, Air, Water, 46(3), 1700114. https://doi.org/10.1002/clen.201700114
Al-Khashman, O.A. (2009) Chemical characteristics of rainwater collected at a western site of Jordan. Atmospheric Research, 91(1), 53–61. https://doi.org/10.1016/j.atmosres.2008.05.007
Allen, A.G., Nemitz, E., Shi, J.P., Harrison, R.M., Greenwood, J.C. (2001) Size distributions of trace metals in atmospheric aerosols in the United Kingdom. Atmospheric Environment, 35(27), 4581–4591. https://doi.org/10.1016/S1352-2310(01)00190-X
Anil, I., Alagha, O., Blaisi, N.I., Mohamed, I.A., Barghouthi, M.H., Manzar, M.S. (2019) Source identification of episodic rain pollutants by new approach: combining satellite observations and backward air mass trajectories. Aerosol and Air Quality Research, 19(12), 2827–2843. https://doi.org/10.4209/aqr.2019.04.0187

APHA (American Public Health Association) (2012) Standard methods for the examination of water and waste water. 22nd ed. APHA, American Water Works Association, Water Environment Federation.

Báez, A., Belmont, R., García, R., Padilla, H., Torres, M.D.C. (2007) Chemical composition of rainwater collected at a southwest site of Mexico City, Mexico. Atmospheric Research, 86(1), 61–75. https://doi.org/10.1016/j.atmosres.2007.03.005

Bravo, H.A., Saaedra, M.I.R., Sánchez, P.A., Torres, R.J., Granada, L.M.M. (2000) Chemical composition of precipitation in a Mexican Maya region. Atmospheric Environment, 34(8), 1197–1204. https://doi.org/10.1016/S1352-2310(99)00305-2

Calvo, A.I., Olmo, F.J., Lyamani, H., Alados-Arboledas, L., Casablanca, A.R., Calamita, G., Sabia, S., Tripetta, S. (2017) Bio-monitoring of atmospheric pollution: a novel approach for the evaluation of natural and anthropogenic contribution to atmospheric aerosol particles. Environmental Science and Pollution Research, 24(9), 8578–8587. https://doi.org/10.1007/s11356-017-8534-3

Calvo, A.I., Olmo, F.J., Lyamani, H., Alados-Arboledas, L., Castro, A., Fernandez-Raga, M., Fraile, R. (2010) Chemical Composition of Wet Precipitation at the Background EMEP Station in Víznar (Granada, Spain) (2002–2006). Atmospheric Research, 96, 408–420. https://doi.org/10.1016/j.atmosres.2010.01.013

Cerqueira, M.R.F., Pinto, M.F., Derossi, I.N., Esteves, W.T., Santos, M.D.R., Matos, M.A.C., Lowinson, D., Matos, R.C. (2014) Chemical characteristics of rainwater at a southeastern site of Brazil. Atmospheric Pollution Research, 5(2), 253–261. https://doi.org/10.5094/APR.2014.031

Charlson, R., Rodhe, H. (1982) Factors controlling the acidity of natural rainwater. Nature, 295, 683–685. https://doi.org/10.1038/295683a0

Chen, B., Stein, A.F., Maldonado, P.G., de la Campa, A.M.S., Gonzalez-Castanedo, Y., Castell, N., Jesus, D. (2013) Size distribution and concentrations of heavy metals in atmospheric aerosols originating from industrial emissions as predicted by the HYSPLIT model. Atmospheric Environment, 71, 234-244. https://doi.org/10.1016/j.atmosenv.2013.02.013

Cheng, M.C., You, C.F., Lin, F.J., Huang, K.F., Chung, C.H. (2011) Sources of Cu, Zn, Cd and Pb in rainwater at a subtropical islet offshore northern Taiwan. Atmospheric Environment, 45(11), 1919–1928. https://doi.org/10.1016/j.atmosenv.2011.01.034

Cobbina, S.J., Michael, K., Salifu, L., Duwiejua, A.B. (2013) Rainwater quality assessment in the Tamale municipality. International Journal of Scientific & Technology Research, 2(5), 1–5.

Deshpande, R.D., Gupta, S.K. (2008) National programme on isotope fingerprinting of waters of India (IWIN). Glimpses of Geosciences Research in India, the Indian Report to IUGS, Indian National Science Academy (Singhvi, A.K., Bhattacharya, A. and Guha, S. Eds), INSA, New Delhi, pp. 10–16.

Farahmandkia, Z., Mehrabi, M., Sekhavatjou, M. (2011) Relationship between concentrations of heavy metals in wet precipitation and atmospheric PM10 particles in Zanjan, Iran. Journal of Environmental Health Science & Engineering, 8(1), 49–56.

Galloway, J.N., Likens, G.E., Keene, W.C., Miller, J.M. (1982) The composition of precipitation in remote areas of the world. Journal of Geophysical Research: Oceans, 87 (C11), 8771–8786. https://doi.org/10.1029/JC087iC11p08771

Ghadimi, F., Ghomi, M., Ranjbar, M., Hajati, A. (2013) Sources of contamination in rainwater by major and heavy elements in Arak, Iran. Journal of Water Sciences Research, 5(2), 67–84.

Gomyo, M., Kuraji, K., Kitayama, K., Suzuki, M. (2008) Rainfall, bulk deposition of nutrients and their seasonal variation in two tropical lowland and montane forests in Borneo. In AGU Fall Meeting Abstracts, 2008, H33F-1096.

Grewling, Ł., Bogawski, P., Kryza, M., Magyar, D., Sikoparija, B., Skjøth, C.A., Udvardy, O., Werner, M., Smith, M. (2019) Concomitant occurrence of anthropogenic air pollutants, mineral dust and fungal spores during long-distance transport of ragweed pollen. Environmental Pollution, 254, 112948. https://doi.org/10.1016/j.envpol.2019.07.116

Gunawardena, J., Egodawatta, P., Ayoko, G.A., Goonetilleke, A. (2012) Role of traffic in atmospheric accumulation of heavy metals and polycyclic aromatic hydrocarbons. Atmospheric Environment, 54, 502–510. https://doi.org/10.1016/j.atmosenv.2012.02.058

Hasan, N.Y., Driejana, D., Sulaeman, A. (2017) Composition of ions and trace metals in rainwater in Bandung City, Indonesia. IPTEK Journal of Proceedings Series, 3(6), 603–608. https://doi.org/10.12962/j23546026y2017i6.3310

Helmreich, B., Hilliges, R., Schriewer, A., Horn, H. (2010) Run-off pollutants of a highly trafficked urban road Correlation analysis and seasonal influences. Chemosphere, 80(9), 991–997. https://doi.org/10.1016/j.chemosphere.2010.05.037

IAEA (International Atomic Energy Agency) (2014) IAEA/GNEP precipitation sampling guide. Vienna: International Atomic Energy Agency.

Innocente, E., Squizzato, S., Visin, F., Facca, C., Rampazzo, G., Bertolini, V., Gandolfi, I., Franzetti, A., Ambrosini, R., Bestetti, G. (2017) Influence of seasonality, air mass origin and particulate matter chemical composition on airborne bacterial community structure in the Po Valley, Italy. Science of the Total Environment, 593, 677–687. https://doi.org/10.1016/j.scitotenv.2017.03.199

Jeelani, G., Deshpande, R.D., Galkowski, M., Rozanski, K. (2018) Isotopic composition of daily precipitation along the southern foothills of the Himalayas: impact of marine and continental sources of atmospheric moisture. Atmospheric Chemistry and Physics, 18(12), 8789–8805. https://doi.org/10.5194/acp-18-8789-2018

Khayan, K., Heru Husodo, A., Astuti, I., Sudarmadji, S., Sugandawaty Djohan, T. (2019) Rainwater as a source of drinking water in the city of Surakarta. Journal of Environmental Science, 7(2), 57–66.

Khanna, A.K. (2005) Rainfall in the Himalayas and its role in continental/sea-ice interactions over the Indian Ocean and the southern Asia. Journal of Geophysical Research: Oceans, 87 (C11), 8771–8786. https://doi.org/10.1029/JC087iC11p08771

Kulkarni, S.P., Deshpande, R.D. (2019) Flight of dust and some airborne pollutants over the Arabian Sea. Atm. Res., 169, 84–98. https://doi.org/10.1016/j.atmosres.2016.07.005

Kumar, V., Garg, M., Kumar, M., Prasad, D., Goel, R.K. (2012) Isotopic composition of precipitation at Vizag (India) and its comparison with similar precipitation sites in tropical Africa. Environmental Earth Sciences, 66(2), 369–378. https://doi.org/10.1007/s12665-011-1445-8

Kumar, V., Garg, M., Prasad, D., Goel, R.K. (2013) Isotopic composition of precipitation at Vizag (India) and its comparison with similar precipitation sites in tropical Africa. Environmental Earth Sciences, 66(2), 369–378. https://doi.org/10.1007/s12665-011-1445-8

Kumar, V., Garg, M., Prasad, D., Goel, R.K. (2013) Isotopic composition of precipitation at Vizag (India) and its comparison with similar precipitation sites in tropical Africa. Environmental Earth Sciences, 66(2), 369–378. https://doi.org/10.1007/s12665-011-1445-8

Kumar, V., Garg, M., Prasad, D., Goel, R.K. (2013) Isotopic composition of precipitation at Vizag (India) and its comparison with similar precipitation sites in tropical Africa. Environmental Earth Sciences, 66(2), 369–378. https://doi.org/10.1007/s12665-011-1445-8
water: health impacts and rainwater treatment. Journal of Environmental and Public Health, 2019, 1760950. https://doi.org/10.1155/2019/1760950

Komabayasi, M. (1959) Dissolved oxygen in rainwater and its relation to the raincloud structure (I) Precipitation from September to March. Journal of the Meteorological Society of Japan. Ser. II, 37(1), 22–34. https://doi.org/10.2151/jmsj1923.37_1_22

Kulshrestha, U.C., Kulshrestha, M.J., Sekar, R., Sastry, G.S.R., Vairamani, M. (2003) Chemical characteristics of an urban site of south-central India. Atmospheric Environment, 37(21), 3019–3026. https://doi.org/10.1016/S1352-2310(03)00266-8

Li, Z., Liu, F., Song, Y., Song, L., Tian, Q., Jia, B., Li, Y., Ma, J. (2017) Chemical characteristics of precipitation and the indicative significance for sand dust events in the northern and southern slopes of Wushaoling Mountain, northwestern China. Journal of Arid Land, 9(6), 911–923. https://doi.org/10.1007/s40333-017-0069-4

Likuku, A.S., Gboutloeloe, G.K., Mmolawa, K.B. (2013) Determination and source apportionment of selected heavy metals in aerosol samples collected from Sebele. American Journal of Environmental Science, 9(2), 188–200.

Lynam, M.M., Dvonch, J.T., Hall, N.L., Morishita, M., Barres, M.A. (2014) Spatial patterns in wet and dry deposition of atmospheric mercury and trace elements in central Illinois, USA. Environmental Science and Pollution Research, 21(6), 4032–4043. https://doi.org/10.1007/s11356-013-1911-4

Ma, Y., Wang, M., Wang, S., Wang, Y., Peng, L., Wu, K. (2021) Air pollution emission characteristics and HYSPLIT model analysis during heating period in Shenyang, China. Environmental Monitoring and Assessment, 193(1), 1–14. https://doi.org/10.1007/s10661-020-08767-4

Majumdar, A., Satpathy, J., Kayee, J., Das, R. (2020) Trace metal composition of rainwater and aerosol from Kolkata, a mega-city in eastern India. SN Applied Sciences, 2(12), 1–17. https://doi.org/10.1007/s42452-020-03933-2

Marandi, A., Polikarpus, M., Jõeleht, A. (2013) A new approach for describing the relationship between electrical conductivity and major anion concentration in natural waters. Applied Geochemistry, 38, 103–109. https://doi.org/10.1016/j.apgeochem.2013.09.003

Martin, R., Dowling, K., Pearce, D.C., Florentine, S., McKnight, S., Stelcer, E., Cohen, D.D., Stopic, A., Bennett, J.W. (2017) Trace metal content in inhalable particulate matter (PM\textsubscript{10} and PM\textsubscript{2.5}) collected from historical mine waste deposits using a laboratory-based approach. Environmental Geochemistry and Health, 39(3), 549–563. https://doi.org/10.1007/s10653-016-9833-1

MGDM (Minerals and Geoscience Department Malaysia) (2013) Geological map of Sarawak (1:1,750,000 scale). Published by Minerals and Geoscience Department, Sarawak, Malaysia.

Migliavacca, D., Teixeira, E.C., Wiegand, F., Machado, A., Sanchez, J. (2005) Atmospheric Precipitation and Chemical Composition of an Urban Site, Gaúba Hydrographic Basin, Brazil. Atmospheric Environment, 39, 1829–1844. https://doi.org/10.1016/j.atmosenv.2004.12.005

Moreda-Piñeiro, J., Alonso-Rodríguez, E., Moscoso-Pérez, C., Blanoc-Heras, G., Turns-Carou, I., López-Mahía, P., Muniategui-Lorenzo, S., Prada-Rodriguez, D. (2014) Influence of marine, terrestrial and anthropogenic sources on ionic and metallic composition of rainwater at a suburban site (north-west coast of Spain). Atmospheric Environment, 88, 30–38. https://doi.org/10.1016/j.atmosenv.2014.01.067

Nadzir, M.S.M., Lin, C.Y., Khan, M.F., Latif, M.T., Dominick, D., Hamid, H.H.A., Mohamad, N., Maulud, K.N.A., Wahab, M.I.A., Kamaludin, N.F., Lim, Y., M.A.S.M. (2017) Characterization of rainwater chemical composition after a Southeast Asia haze event: insight of transboundary pollutant transport during the northeast monsoon. Environmental Science and Pollution Research, 24(18), 15278–15290. https://doi.org/10.1007/s11356-017-9131-1

Nriagu, J.O. (1989) A global assessment of natural sources of atmospheric trace metals. Nature, 338(6210), 47–49. https://doi.org/10.1038/338047a0

Ojekunle, Z.O., Jinadu, O.O.E., Afolabi, T.A., Taiwo, A.M. (2018) Environmental pollution and related hazards at Agbara industrial area, Ogun State. Scientific Reports, 8(1), 1–8. https://doi.org/10.1038/s41598-018-24810-4

Okuda, T., Kato, J., Mori, J., Tenmoku, M., Suda, Y., Tanaka, S., He, K., Ma, Y., Yang, F., Xu, X. (2004) Daily concentrations of trace metals in aerosols in Beijing, China, determined by using inductively coupled plasma mass spectrometry equipped with laser ablation analysis, and source identification of aerosols. Science of the Total Environment, 330, 145–158. https://doi.org/10.1016/j.scitotenv.2004.04.010

Orlović-Leko, P., Vidović, K., Ciglenečki, I., Omanović, D., Sikirić, M.D., Šimunić, I. (2020) Physico-chemical characterization of an urban rainwater (Zagreb, Croatia). Atmosphere, 11(2), 144. https://doi.org/10.3390/atmos11020144

Owoade, O.K., Olise, F.S., Ogundele, L.T., Fawole, O.G., Olaniyi, H.B. (2012) Correlation between particulate matter concentrations and meteorological parameters at a site in Ile-Ife, Nigeria. Ife Journal of Science, 14(1), 83–93.

Oza, H., Padhya, V., Ganguly, A., Saikranthi, K., Rao, T.N., Pordesai, P.J., Casassa, G., Carrasco, J., Damiani, A., Llanillo, M., Lambert, F. (2021) Hydrometeorological processes in semi-arid western India: insights from long term isotope record of daily precipitation. Climate Dynamics, 54(5), 2745–2757. https://doi.org/10.1007/s00382-020-05136-2

Özsoy, T., Örneke, S. (2009) Trace elements in urban and suburban rainfall, Mersin, Northeastern Mediterranean. Atmospheric Research, 94(2), 203–219. https://doi.org/10.1016/j.atmosres.2009.05.017

Pizarro, J., Vergara, P.M., Cerda, S., Cordero, R.R., Castillo, X., Rowe, P.M., Casassa, G., Carrasco, J., Damiani, A., Llanillo, P.J., Lambert, F. (2021) Contaminant emissions as indicators of chemical elements in the snow along a latitudinal gradient in southern Andes. Scientific Reports, 11(1), 1–10. https://doi.org/10.1038/s41598-021-93895-1

Punsompong, P., Chantara, S. (2018) Identification of potential sources of PM\textsubscript{10} pollution from biomass burning in northern Thailand using statistical analysis of trajectories. Atmospheric Pollution Research, 9(6), 1038–1051. https://doi.org/10.1016/j.apr.2018.04.003
Trace Metal Concentration in Rainwater Collected from Northern Sarawak, Borneo

Radojevic, M. (2003) Chemistry of forest fires and regional haze with emphasis on Southeast Asia. Pure and Applied Geophysics, 160(1), 157–187. https://doi.org/10.1007/s00024-003-8771-x

Radojevic, M., Tan, K.S. (2000) Impacts of biomass burning and regional haze on the pH of rainwater in Brunei Darussalam. Atmospheric Environment, 34(17), 2739–2744. https://doi.org/10.1016/S1352-2310(99)00339-9

Rajeev, P., Rajput, P., Gupta, T. (2016) Chemical characteristics of aerosol and rain water during an El Niño and PDO influenced Indian summer monsoon. Atmospheric Environment, 145, 192–200. https://doi.org/10.1016/j.atmosenv.2016.09.026

Ramírez Lara, E., Miranda Guardiola, R., Gracia Vásquez, Y., Balderas Rentería, I., Bravo Álvarez, H., Sosa Echeverría, R., Sánchez Álvarez, P., Alarcón Jiménez, A., Torres, M.C., Kahlb. J. (2010) Chemical composition of rainwater in northeastern México. Atmosfera, 23(3), 213–224.

Ressler, T., Wong, J., Roos, J., Smith, I.L. (2000) Quantitative Richards, E.H. (1917) Dissolved oxygen in rain-water. The

Schneider, L., Mariani, M., Saunders, K.M., Maher, W.A., Harrison, J.J., Fletcher, M.S., Zawadzki, A., Heijnis, H., Haberle, S.G. (2019) How significant is atmospheric metal contamination from mining activity adjacent to the Tasmanian Wilderness World Heritage Area? A spatial analysis of metal concentrations using air trajectories models. Science of The Total Environment, 656, 250–260. https://doi.org/10.1016/j.scitotenv.2018.11.241

Schwarz, J., Cusack, M., Karban, J., Chalupničková, E., Havránek, V., Smolík, J., Zdímal, V. (2016) PM10, chemical composition at a rural background site in Central Europe, including correlation and air mass back trajectory analysis. Atmospheric Research, 176, 108–120. https://doi.org/10.1016/j.atmosres.2016.02.017

Serafin, S., Adler, B., Cuxart, J., De Wekker, S.F.J., Gohm, A., Grisogono, B., Kaltho, N., Kirshbaum, D.J., Rotach, M.W., Schmidli, J., Stiperski, I., Vecenaj, Ž., Zardi, D. (2018) Exchange Processes in the Atmospheric Boundary Layer Over Mountainous Terrain. Atmosphere, 9, 102. https://doi.org/10.3390/atmos9030102

Shende, R.R., Ravindran, U., Bhondave, S.D., Deshmukh, A.K. (2007). Role of aerosols in deciding pH of rainwater. Mau-sam, 58(4), 513.

Silva, G.N.D., Alves, L.D., Santos, I.E.D., Bila, D.M., Ohnuma Júnior, A.A., Corrêa, S.M. (2020) An assessment of atmospheric deposition of metals and the physico-chemical parameters of a rainwater harvesting system in Rio de Janeiro Brazil, by means of statistical multivariate analysis. Revista Ambiente & Água, 15(4). https://doi.org/10.4136/ambi-agua.2522

Siudek, P., Frankowski, M. (2017) The effect of sources and air mass transport on the variability of trace element deposition in central Poland: a cluster-based approach. Environmental Science and Pollution Research, 24(29), 23026–23038. https://doi.org/10.1007/s11356-017-9932-2

Sjöblom, Å., Håkansson, K., Allard, B. (2010) Particulate matter as scavenger and carrier of trace elements in simulated mine water. In International Mine Water Association Symposium (IMWA), Sydney, Nova Scotia, Canada, September 5–9, 2010 (403-406). Cape Breton University Press.

Sorooshian, A., Shingler, T., Harpold, A., Feagles, C.W., Meixner, T., Brooks, P.D. (2013) Aerosol and precipitation chemistry in the southwestern United States: spatiotemporal trends and interrelationships. Atmospheric Chemistry and Physics, 13(15), 7361–7379. https://doi.org/10.5194/acp-13-7361-2013

Srivastava, S., Kumar, A., Baudhdd, K., Gautam, A.S., Kumar, S. (2020) 21-Day lockdown in India dramatically reduced air pollution indices in Lucknow and New Delhi, India. Bulletin of Environmental Contamination and Toxicology, 105, 9–17. https://doi.org/10.1007/s00128-020-02895-w

Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J., Cohen, M.D., Ngar, F. (2015) NOAA’s HYSPLIT atmospheric transport and dispersion modeling system. Bulletin of the American Meteorological Society, 96(12), 2059–2077. https://doi.org/10.1175/BAMS-D-14-00110.1

Sternbeck, J., Sjödin, Å., Andréasson, K. (2002) Metal emissions from road traffic and the influence of resuspension results from two tunnel studies. Atmospheric Environment, 36(30), 4735–4744. https://doi.org/10.1016/S1352-2310
Sulaymon, I.D., Zhang, Y., Hopke, P.K., Zhang, Y., Hua, J., Mei, X. (2021) COVID-19 pandemic in Wuhan: Ambient air quality and the relationships between criteria air pollutants and meteorological variables before, during, and after lockdown. Atmospheric Research, 250, 105362. https://doi.org/10.1016/j.atmosres.2020.105362

Tatsuta, S., Shimada, K., Chan, C.K., Kim, Y.P., Lin, N.H., Takami, A., Hatakeyama, S. (2017) Contributions of long-range transported and locally emitted nitrate in size-segregated aerosols in Japan at Kyushu and Okinawa. Aerosol and Air Quality Research, 17(12), 3119–3127. https://doi.org/10.4209/aaqr.2016.12.0587

Tiwari, S., Hopke, P.K., Thimmaiah, D., Dumka, U.C., Srivastava, A.K., Bisht, D.S., Rao, P.S., Chate, D.M., Srivastava, M.K., Tripathi, S.N. (2016) Nature and sources of ionic species in precipitation across the Indo-Gangetic Plains, India. Aerosol and Air Quality Research, 16(4), 943–957. https://doi.org/10.4209/aqr.2015.06.0423

Tripathi, B., Pandey, R., Raghuvanshi, D., Singh, H., Pandey, V., Shukla, D.N. (2014) Studies on the physico-chemical parameters and correlation coefficient of the river Ganga at Holy Place Shringverpur, Allahabad. Journal of Environmental Science, Toxicology and Food Technology, 8(10), 29–36.

Tu, X., Lu, Y., Yao, R., Zhu, J. (2019) Air quality in Ningbo and transport trajectory characteristics of primary pollutants in autumn and winter. Atmosphere, 10(3), 120. https://doi.org/10.3390/atmos10030120

Vuai, S.A.H., Tokuyama, A. (2011) Trend of trace metals in precipitation around Okinawa Island, Japan. Atmospheric Research, 99(1), 80–84. https://doi.org/10.1016/j.atmosres.2010.09.010

Williams, J., Petrik, L., Wichmann, J. (2021) PM$_{2.5}$ chemical composition and geographical origin of air masses in Cape Town, South Africa. Air Quality, Atmosphere & Health, 14(3), 431–442. https://doi.org/10.1007/s11869-020-00947-y

Williams, N.E., Tighiri, H.O. (2015) Quality studies of rainwater in Uyo Municipality, Nigeria. International Journal of Scientific & Engineering Research, 6(4), 1569–1573.

Zhang, J., Chen, S.Z., Yu, Z.G., Wang, C.S., Wu, Q.M. (1999) Factors influencing changes in rainwater composition from urban versus remote regions of the Yellow Sea. Journal of Geophysical Research: Atmospheres, 104(D1), 1631–1644. https://doi.org/10.1029/1998JD100019

Zhang, S.R., Lu, X.X., Higgitt, D.L., Chen, C.T.A., Sun, H.G., Han, J.T. (2007) Water chemistry of the Zhujiang (Pearl River): natural processes and anthropogenic influences. Journal of Geophysical Research: Earth Surface, 112(F1). https://doi.org/10.1029/2006JF000493