

# Seasonal Changes and Major Cation Composition in Individual Rain Events at Ballarat, Central Victoria (Australia)

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**Abstract:** Rainfall events ( $n = 120$ , representing 702.7 mm rain) were collected at the Mount Helen site in Ballarat, Central Victoria (Australia) from November 2002 to November 2003 and analysed for rainfall amount, pH, electric conductivity (EC), temperature and major cation (Ca, Na, Mg and K) composition. The pH exhibited large variations, from 4.2 (in winter) to 7.7 (in summer) with an average value of 6.0. Twenty-one percent of the recorded rain events were acidic ( $\text{pH} < 5.5$ ) and 29% were neutral to alkaline ( $\text{pH} > 6.5$ ). The mean dissolved concentrations of major cations varied in the order:  $\text{Na} > \text{Ca} > \text{K} > \text{Mg}$ . All major cations varied significantly (Na and K >50-fold, Ca >100-fold and Mg >450-fold) in the individual rain events. While Na and Mg were the most and least abundant cations in all seasons, respectively, Ca recorded the second dominant cation in spring and winter, and K in summer and autumn seasons. About 3-fold higher K concentration in summer (average  $\sim 65 \mu\text{eq/L}$ ) as compared to the winter season (average  $\sim 21 \mu\text{eq/L}$ ), pointed to possible biogenic emissions and forest burnings in the area. The rainwater chemistry appeared to be controlled mainly by terrestrial sources; however, the higher value of Na (upto  $434.9 \mu\text{eq/L}$  in summer season) pointed to the influence of marine sources in few rain events. More than 15% of rain events recorded  $>300 \mu\text{eq/L}$  of Na, which could be significant from the agricultural and environmental point of view for both quality and quantity.

**Key words:** Rainwater, major cations, individual rain events, seasonal changes, Australia.

## Introduction

Rainfall chemistry has been studied in detail in America (USEPA, 1998; Peters et al., 2002), Europe (Horvath and Meszaros, 1984; Charron et al., 2000) and East Asia (Hara et al., 1995; Ayers and Yeung, 1996; Lee et al., 2000) due to the increasing emissions of  $\text{NO}_x$ ,  $\text{SO}_2$  and  $\text{NH}_3$  and their effects on the environment (Galloway, 1995; Sultan, 2003). Seasonal changes in rainwater chemistry can significantly affect the environment depending upon the amount and type of local emissions. Seasonal episodic input from low pH rain events can cause chronic surface water acidification (Lawrence, 2002) depending upon the acid-neutralizing capacity (ANC) of the soil-water system. The base cation status provides an index of the

capability of an environmental compartment to neutralize inputs of acidic deposition. Low base cation concentration of a storm event may cause streams and surface water bodies to be prone to acidification. Base cation status helps in the assessment of terrestrial ecosystem sensitivity to acid deposition (Cinderby et al., 1998).

During and after the gold-rushes in the 1850s, Central Victoria has been the centre of mining activities resulting in mine tailings and waste materials rich in potentially toxic metals. Surface waters have been found to be contaminated with arsenic with concentrations varying more than 28-fold seasonally associated with rainfall (Sultan, 2006). The number of rainwater chemistry studies in the Southern Hemisphere has been fewer than in the Northern Hemisphere because rainwater acidity is

not considered a problem (Bridgman, 1992). Relatively little information is available for the rainwater chemistry of Central Victoria where individual rainfall events could be important from an agricultural point of view in both quality and quantity. Event based studies of rainfall chemistry are important in the management of the environment as individual rain events could be significantly acidic. In this paper, chemical composition, especially major cations in rainwater, are investigated at a site closer to Ballarat.

## Methods

### Site Description

A rain gauge and a sample collector were installed at the University of Ballarat (~525 m ASL), located (Australian Map Grid 755310-5831526) about 8 km from the centre of the city, Ballarat (Central Victoria). Samples were collected from November 2002 to November 2003 from the self-made rain collector (~1500 cm<sup>2</sup>) on the roof of the two storey Science and Engineering building. The sample collector was about 2.0 m high attached to a 4 L bottle with no close obstructions. Rainwater sampling included summer (January to March), autumn (April to June), winter (July to September) and spring (October to December) seasons. Ballarat's climate is temperate with moderate rainfall (average annual ~705.3 mm), 59% of which is received from May to October. The mean number of rainy days in Ballarat is 169. The mean daily maximum temperature for January is 24.9°C in summer and mean daily minimum temperature for July is 3.2°C during winter. The site location represented the region's background precipitation, as no particular urban industrial emissions are present in close vicinity; however, local minor sources (e.g. residential heating, vehicle emissions and brick industry) might be present. The sampling site is surrounded by forest, agricultural land and a rural town, Buninyong. The nearest coast is located about 120 km southeast of the sampling site. Soils in the area are mildly acidic and there are many mine tailings due to the mining activities during and after the gold rushes of the 1850's. The study aims to document the major cation composition based on seasons and individual rain events in Ballarat.

### Sampling

Rain samples ( $n = 120$ ) were collected following a rain event on a daily basis or twice a day to reduce the mixing of the individual rain events as much as possible. Rainfall events represented a total rainfall amount of 702.3 mm and all four seasons of Ballarat. All rain events greater than 0.5 mm were collected during the sampling period

covering all four seasons. Rainfall amounts less than 0.5 mm were too small to be used for chemical analysis and, therefore, were neglected. The samples were collected in pre-cleaned acid washed polyethylene bottles and the collector was washed with Milli-Q deionised water ( $\leq 18 \text{ M}\Omega \text{ cm}^{-1}$ ) before the following rain event. On-site analysis included pH, Eh, electrical conductivity and temperature measurements by using standard portable meters. The samples were filtered through a 0.45  $\mu\text{m}$  Millipore membrane filter and acidified ( $\text{pH} < 2$ ) with analytical grade  $\text{HNO}_3$  for stabilisation. The samples were stored in a refrigerator at 4°C for later cation (Na, Ca, Mg and K) measurements about every fortnight.

### Analysis

The pH was measured using an Orion pH meter (model EA940) with a resolution of measurement of 0.01 pH units. The major cations (Ca, Mg, Na and K) were determined by flame atomic absorption spectrometry at University of Ballarat. Samples with low dissolved cations were pre-concentrated (20 $\times$ ) by sub-boiling evaporation in clean beakers on a laminar-flow clean bench before determination. Intermediate standards of various concentrations of elements (Ca, Mg, Na and K) were prepared from certified aqueous standards (BDH Spectrosol), Concentrations of all the cations in field blanks were found to be below the detection limit. Both analytical precision (based on replicate analysis) and accuracy (evaluated by analysing standard reference rainwater SRM 2694a) were generally better than  $\pm 10\%$  and  $\pm 7\%$ , respectively.

## Results and Discussion

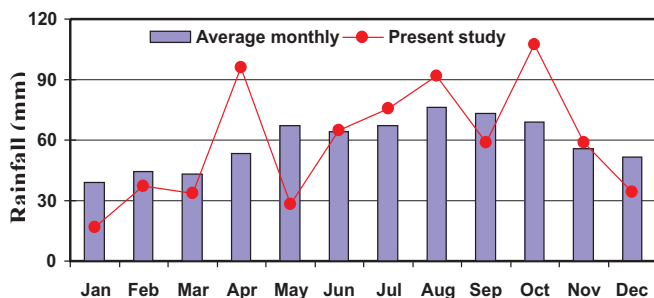
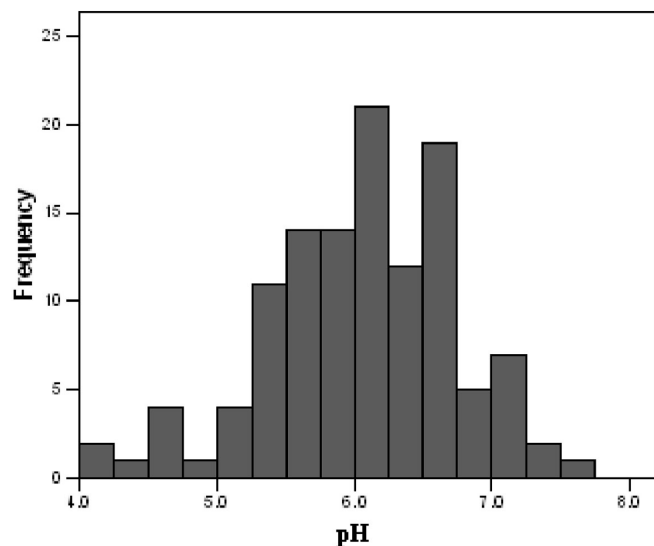
The rainwater physico-chemical data along with volume-weighted mean values are presented in Table 1. The temperature of rainwater samples ranged from 1.8 to 25.2 °C.

A total of 702.7 mm of rainfall was collected comprising 120 rain events and did not include rain events <0.5 mm. The rain measured in this study was about 1.0% lower than the annual average of the Ballarat city (Figure 1). The sampling location, Mount Helen Campus, is about 8 km from the city. While April and October registered higher rainfall, January and May recorded only half the average. About 87% of the rain events were below 10 mm and only 6% were above 20 mm of rain. Ballarat receives more of its rain as drizzly showers.

The pH varied from 4.2 to 7.7 with an average value of 6.0. Twenty-one percent of the recorded rain events

**Table 1: Chemical composition of rainwater in Ballarat (Australia)**

Component	Samples (n)	Min	Max	Mean (arithmetic)	Mean (volume-weighted)	SD
Rainfall event (mm)	120	0.5	32.6	5.86	–	–
Temperature (°C)	120	1.5	25.20	13.6	–	–
EC (µS/cm)	120	5.0	157.0	32.7	–	29.5
pH	120	4.15	7.7	6.03	5.24	0.69
Na (µeq/L)	118	8.7	434.90	136.4	90.7	113.4
K (µeq/L)	118	3.14	170.20	34.2	14.3	46.2
Ca (µeq/L)	118	3.0	470.10	54.1	30.5	74.8
Mg (µeq/L)	118	0.37	168.0	34.0	19.7	34.2

**Figure 1: Chart showing average monthly and measured rainfall data.****Figure 2: Frequency distribution of pH of rainwater (n=120).**

were acidic (pH <5.5) and about 29% were neutral to alkaline (pH >6.5). The rain events with a pH <4.5 were only 4%. The rainwater pH frequency distribution is shown in Figure 2. The pH values recorded the lowest in winter season, July (average ~5.1), and the highest in the summer season, January (average ~6.6). Neutral to

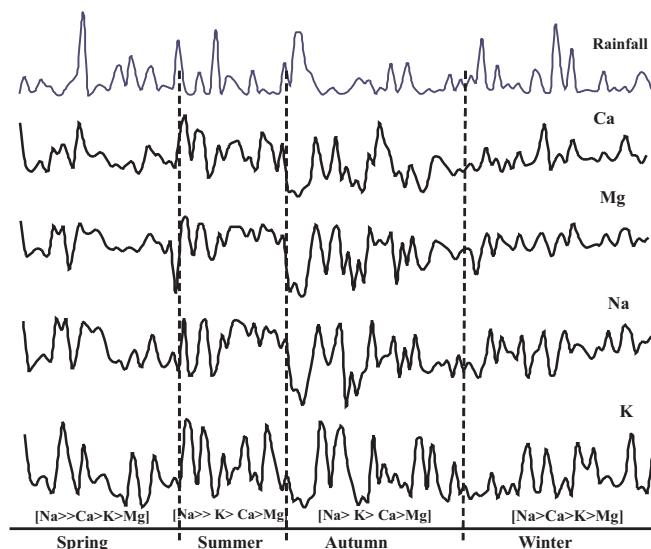
alkaline pH values (=7.0) are likely to be due to the dissolution of the alkaline dust particles of Ca and Mg in the atmosphere. Emissions of other alkaline substances such as gaseous  $\text{NH}_3$  can also neutralize some fractions of the acids. A weak positive correlation between pH, and Ca and Mg suggested the potential role of gaseous ammonia ( $\text{NH}_3$ ) in partial neutralization of acidity. Summer season (January to March) rainfall showed a large variation in pH values as compared to winter (July to September) and autumn (October to December) rainfall.

Electrical conductivity (EC) ranged from 5.0 to 157.0 µS/cm with an average value of 32.7 µS/cm. A weak negative correlation of EC with rainfall amount indicated an influence on chemical composition of rainwater due to the dilution effect. The summer season with lowest rainfall (87.4 mm) registered the highest average EC value (67.5 µS/cm). EC values showed a weak negative correlation ( $r = -0.35$ ) with rainfall amount and a positive correlation ( $r = 0.48$ ) with the rainwater temperature due to the dilution effect and dissolution of dust particles, respectively. All major cations showed good positive correlation ( $r = 0.64$ ) with EC values. The winter season recorded the highest rainfall and lowest average EC value (µS/cm). A large amount of rain tends to be dilute due to the rainout process in which rainwater takes on chemistry in the clouds during condensation. During the long rainfall events the concentration of elements decrease with time due to cleaning of the atmosphere by the progressive rain. Brief rainfall events carry a signature of the washout process where dissolution of elements occurs in the lower atmosphere with the falling rain.

Sodium concentration varied from 8.7 to 434.9 µeq/L with an average value of 136.4 µeq/L. Sodium registered the dominant cation in rainwater and varied 50-fold. An average Na concentration in summer (~221.5 µeq/L) was more than 23-fold higher than the average concentration in autumn season (~9.4 µeq/L). About 53% of rain events

recorded Na less than 100  $\mu\text{eq/L}$  and 15% rain events recorded Na greater than 300  $\mu\text{eq/L}$ . A large variation in Na concentrations are possibly due to the chemical signature of the source whether these originated from marine or terrigenous continental air masses. Higher Na concentrations are representative of marine influences; however, the study site is located about 120 km from the coast. Yoboue et al. (2005) reported an influence of marine inputs on rainwater chemistry over the Wet Savanna Ecosystem of Lamto (Africa), located more than 100 km from the nearest coast. Sea-salt correction of Na could not be applied as the Cl concentration was not measured in samples. Al-Momani et al. (1995) reported average Na concentration in rainwater from Izmir (Turkey) to be 450  $\mu\text{eq/L}$ , which is more representative of marine sources (Table 2). Mouli et al. (2005) document an average value of 33.1  $\mu\text{eq/L}$  of Na in rainwater of Tirupati (India), which is strongly influenced by terrestrial sources. Major cation concentration in rainwaters along with the order of abundance is presented in Table 2.

Potassium concentration ranged from 3.1 to 170.2  $\mu\text{eq/L}$  with an average value of 34.2  $\mu\text{eq/L}$ . Potassium recorded the third most abundant cation in rainwater and represented more of a continental origin than marine. Lower values of K concentration are indicative of rain events of marine influence and dilution effect as studied by Bravo et al. (2000) and Al-Momani et al. (1995) of rainwater with seawater as a dominant source (Table 2). Khemani et al. (1989) reported an average K concentration of 41  $\mu\text{eq/L}$  in the rainwater of Agra (India). Primary sources of K in rainwater are soil dust and biological emissions. Other sources could be from plants (as  $\text{K}_2\text{SO}_4$ ), biomass burning and fertilizers. Negrel (1998) observed K contributions of marine origin in the range of less than 1 to 41.2% in rainwater from the Massif Central (France). Potassium concentration registered highest in the summer season (average  $\sim 65 \mu\text{eq/L}$ ) and lowest in the winter season (average  $\sim 21 \mu\text{eq/L}$ ), which is possibly due to inputs from forest burning, as bushfires



**Figure 3: Seasonal variations of major cations and rainfall amount in Ballarat.**

are common in the summer season (Figure 3) but more likely is derived from local illite dominated soils.

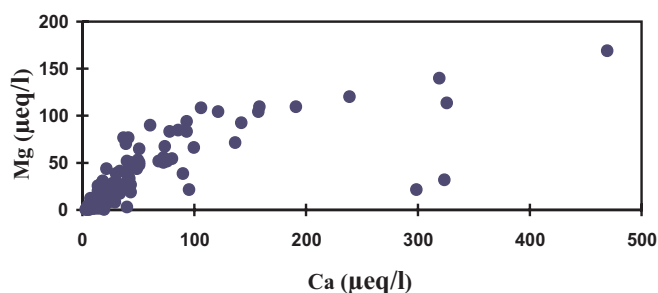
Calcium concentration varied from 3.0 to 470.1  $\mu\text{eq/L}$  with an average value of 54.1  $\mu\text{eq/L}$ . Ca content varied more than 150-fold in individual rain events. Ca concentration was the highest in summer (average  $\sim 98.5 \mu\text{eq/L}$ ) and the lowest in winter season (average  $\sim 43.5 \mu\text{eq/L}$ ). Higher Ca concentration is possibly related to eolian particles of terrestrial origin which include calcite ( $\text{CaCO}_3$ ), dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ) and gypsum ( $\text{CaSO}_4$ ). The average Ca concentration was found to be much lower as compared to Ankara and Agra, 210  $\mu\text{eq/L}$  (Tuncel and Ungor, 1996) and 237  $\mu\text{eq/L}$  (Khemani et al., 1989), respectively. The Ca contents in local soils could have neutralized the acidity, inferred by  $\text{SO}_4$  and  $\text{NO}_x$  concentrations, along the path of the air masses, although no meaningful correlation was observed between Ca concentration and rain pH ( $r = 0.46$ ) in this study.

**Table 2: Mean concentration ( $\mu\text{eq/L}$ ) of major cations and pH values at selected locations around the world**

Location	pH	Na	K	Ca	Mg	Order of abundance
Tirupati (India) <sup>a</sup>	6.8	33.1	33.9	150.7	50.5	Ca>Mg>K>Na
Izmir (Turkey) <sup>b</sup>	5.17	450	12.1	140	94	Na>Ca>Mg>K
Puerto Morelos (Mexico) <sup>c</sup>	5.35	128	5	17	28	Na>Mg>Ca>K
Athens (Greece) <sup>d</sup>	6.1	67.7	14.5	137	30	Ca>Na>Mg>K
Campinas (Brazil) <sup>e</sup>	4.5	2.7	3.4	3.7	1.3	Ca>K>Na>Mg
Beijing (China) <sup>f</sup>	6.05	29	24	148	21	Ca>Na>K>Mg

<sup>a</sup>Mouli et al. (2005); <sup>b</sup>Al-Momani et al. (1995); <sup>c</sup>Bravo et al. (2000); <sup>d</sup>Dikaiajos et al. (1990); <sup>e</sup>Lara et al. (2001); and <sup>f</sup>Fujita et al. (2000).

Magnesium concentration ranged from 0.37 to 168.0  $\mu\text{eq/L}$  with an average value of 34.0  $\mu\text{eq/L}$ . Dissolved Mg content varied significantly (>450-fold) in the individual rain events. Mg registered the least abundant major cation and showed a similarity in behaviour to Ca in the rain events (Figure 3). While summer rain events measured higher Mg concentrations, the autumn rain events recorded lower Mg concentrations. Mg concentration was better correlated with Ca ( $r = 0.79$ ,  $n = 118$ ; Figure 4) indicating the same terrestrial sources. Considering the location of the sampling area, sea salt is not an important contributor of Mg. Wind blown agricultural dust could be a possible source of Mg in rainwater.



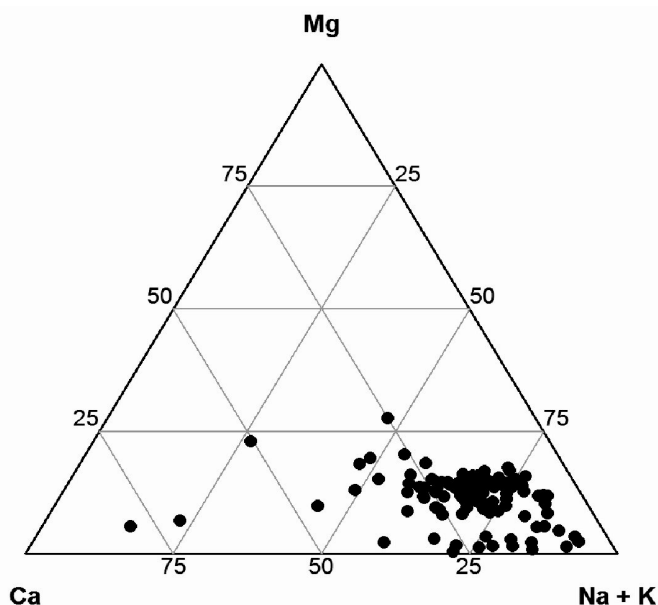
**Figure 4: Plot of Ca concentration against Mg in the Ballarat rain.**

The average rainwater pH higher than 6.0 is possibly due to the near neutral to slightly alkaline nature of the soils of the area. Generally, the pH decreased in the following rainfall in consecutive rain events due to the washing out of alkaline dust by the initial rain. During the consecutive rain events, the pH of the rain increased or decreased to attain pH 5.6. The pH of rainwater is generally about 5.6 as a result of  $\text{CO}_2$  dissolution in the atmosphere (Charlson, and Rodhe, 1982). Tuncel and Ungor (1996) reported a change of 1.1 pH units within the first twenty minutes of a rain event. Rain events with a pH lower than 5.6 indicated an addition of acid causing substances. The lower concentration of Ca also reduced the buffer capacity of the rainwater. Low pH values in winter rain events are due to the higher input of major acids ( $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$ ) from local emissions. The higher dilution by rain (~226 mm) prevents further lowering of pH in the winter season. Some of the organic acids might contribute to the acidity but generally are considered a negligible fraction as compared to the mineral acids in the urban precipitation (Likens et al., 1979). However, organic acids have been found to contribute significantly to rainwater acidity in tropical Australia (Post et al., 1991), which reduces toward the

higher latitude. Yoboue et al. (2005) calculated the contribution of organic acidity of 56% of the rainwater in equatorial Africa. Relatively low pH in the summer season is possibly due to the lesser dilution by rainfall (~87 mm).

The mean dissolved concentrations of major cations varied in the order of  $\text{Na} \gg \text{Ca} > \text{K} > \text{Mg}$ . The triangular diagram of major cations (Mg v. Ca v. Na+K; Figure 5) depicted the dominance of Na+K end member. Most of the rainwater samples are dominated by mixtures of Ca and Na+K, with very little influence from Mg.

All major cations registered higher average values in the summer season and lowest in the winter season due to the dilution effect. Higher Na concentration in the summer season with low rainfall and high evaporation could increase the salinity in the area. There is a similarity in seasonal variations of Ca with Mg. All major cations showed a higher variation in concentrations in the autumn season and lesser variations in the winter season. Na was the most dominant and Mg the least abundant cation in all seasons. While K was the second dominant cation in summer and autumn seasons, Ca was recorded as the second dominant cation in spring and winter seasons (Figure 3). Overall, the concentration of major cations was higher in summer due to a lack of dilution by rain and/or high atmospheric concentrations of soil particles. The measured K/Na (0.224), Mg/Na (0.245) and Ca/Na (0.397) ratios in rainwater were found to be considerably elevated than the seawater 0.037, 0.038 and 0.12, respectively. About 14% of the rain events registered a



**Figure 5: Triangular predominance diagram of major cations for the Ballarat rainwater.**

K/Na ratio of  $\leq 0.06$  which pointed to the possible marine influence on rainwater chemistry. Overall, the results indicate the dominance of terrestrial sources of major cations over the marine sources. However, the modification of the marine derived salts by the terrestrial dust is the most likely reason for the deviation of the ratios.

## Conclusion

Major cations in the rainwater were observed to be in the order:  $\text{Na} \gg \text{Ca} > \text{K} > \text{Mg}$ . Concentrations of major cations varied significantly (Na and K >50-fold, Ca and Mg >100-fold) in the individual rain events and registered particularly higher concentrations in the summer season. Overall, among the dissolved cations, Na is the major and Mg is the minor component in all the seasons of rainwater at Ballarat. K registered as the second most dominated cation in summer and autumn, and Ca in spring and winter season. A high average K concentration in summer and low K concentration in the winter season points to forest burnings in the area or aerosol of illitic nature. High Na concentration in rain events indicated the marine influence at a site more than 120 km away from the nearest coast; however terrestrial sources dominated the rainwater chemistry. Higher Na concentration in the summer season with low rainfall and high evaporation could increase the salinity in the area. Twenty-one percent of the recorded rain events were acidic (pH <5.5). This study indicated the background and seasonal variations in the rainwater composition in the Central Victorian region of Australia. The study showed that less urbanised inland areas of Australia also receive rain events that are quite acidic in nature.

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