



# Chemical Composition and Seasonal Variation of Acid Deposition in Chiang Mai, Thailand

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## Abstract

This study aims to determine the chemical composition and seasonal variation of atmospheric acid deposition in order to identify possible sources contributing to precipitation. Sampling and analysis of 132 wet deposition samples were carried out from January to December 2008 at Mae Hia Research Center, Chiang Mai University, Chiang Mai Province. Total precipitation was 1,286.7 mm. Mean electro-conductivity and pH values were 0.94 mS/m and 6.27, respectively. Major cations ( $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ) and major anions ( $\text{HCOO}^-$ ,  $\text{CH}_3\text{COO}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) were determined by Ion Chromatography. The relative volume weight mean concentrations of anions, in descending order, were  $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Cl}^- > \text{CH}_3\text{COO}^- > \text{HCOO}^-$  and those of cations were  $\text{NH}_4^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+ > \text{Na}^+$ . Results of a principle component analysis highlighted the influence of various possible sources of ions such as agricultural activity, fuel combustion, marine sources, soil resuspension, and biomass burning.

**Keywords:** Acid deposition, Chemical composition, Principle component analysis, Seasonal variation

## 1. Introduction

Atmospheric acid deposition is an environmental concern worldwide, and the determination of its impacts in remote areas can be problematic [1]. Acidification is primarily related to the emissions of  $\text{SO}_2$  and  $\text{NO}_x$ , since these gases are the precursors of major acids like  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$ . On the other hand, the neutralization of acidity in rainwater can be achieved either by  $\text{CaCO}_3$  in airborne dust [2, 3] and or by ammonia released from industrial, agricultural, and other natural sources [4].

The chemical composition of rainfall depends strongly on the atmosphere. The concentrations of chemical species in precipitation vary widely in relation to a multiplicity of factors, such as the type and distribution of aerosol sources, transport of aerosols, chemical species, and the scavenging processes of species. Meteorological factors are often considered to be the predominant factor [5-7] in relation to rainfall; the two main processes [8, 9] are described. The first process refers to the washout of the below-cloud atmosphere during precipitation events by raindrops which scavenge and dissolve particles and gases as they fall; the washout components are mainly of local/regional origin. The second process, called "rainout," corresponds to the condensation of water vapor on aerosol particles during the formation of cloud droplets and incorporation of gases surrounding the droplets by aqueous-phase reactions. This process cor-

responds mainly to the incorporation of long-range transported components which may be traced back to their origins by air masses trajectories.

Acid rain is primarily caused by a mixture of strong acids (e.g.  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$ ) resulting from fossil fuel combustion, therefore it has been observed in many industrial regions, particularly in North America and Central Europe. The composition of wet deposition actually reflects the composition of the atmosphere through which it falls. Determination of the composition of rain helps to evaluate the relative importance of the different sources for gases and, particulate matter. Thus the research of chemistry composition of precipitation has been a primary focus of the field of acid deposition [10-13]. In contrast to Europe and North America, emissions of air pollutants in Asia are increasing rapidly [14, 15], resulting from its large population, rapidly growing economy, and the associated systems of energy consumption and production. The potential for air pollution problems in Asia is great due to high emissions and the close proximity of many of the major industrial and urban centers (e.g. Tokyo, Seoul, Hong Kong, Bangkok, Kuala Lumpur, Singapore, and Jakarta). Biomass burning is an important source of air pollutants, especially in Southeast Asia, where the haze phenomenon is a major and recurring problem.

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## 2. Experimental Methods

### 2.1. Sampling Site

The sampling site is located at a meteorological station in the area of Mae Hia Research Center, Chiang Mai University, Muang District, Chiang Mai Province (Fig. 1). It is bounded by longitude 98° 55' 54.3" E and latitude 18° 45' 40.3" N, and is 334 m above sea level. It is surrounded by an agricultural plantation. This site was classified as a rural site based on Acid Deposition Monitoring Network in East Asia (EANET) criteria [16].

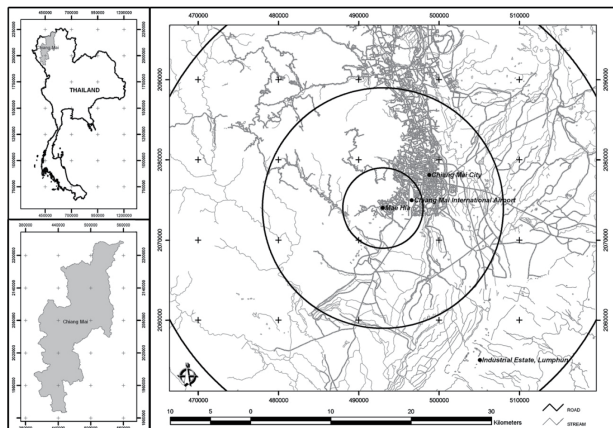


Fig. 1. Sampling site.

### 2.2. Sample Collection and Analysis

Rainwater samples were collected on a daily basis for one year (January–December 2008) using an automated wet only collector. Samples were collected at 9 AM local time and dispatched to the laboratory for analysis within a week. The sampling bucket was cleaned every day after sample collection using de-ionized water with conductivity <math><0.15\text{ mS/m}</math>.

The rainwater samples were weighed to determine the amount of water and were measured for electro-conductivity (EC) and pH under a controlled temperature of 25.0°C using conductivity and pH meters, respectively. The samples were filtered through 0.45  $\mu\text{m}$  cellulose acetate filter paper and stored in the dark at 4°C until ion analysis was performed. Inorganic cations ( $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ) and anions ( $\text{HCO}_3^-$ ,  $\text{CH}_3\text{COO}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) in the rainwater samples were analyzed by Ion Chromatograph (Metrohm, Herisau, Switzerland). Hydrogen ion ( $\text{H}^+$ ) concentrations were derived from pH values [16].

In this study, principal component analysis (PCA) was used to determine the various sources of ion composition of the wet depositions. A varimax rotation with Kaiser Normalization of Principle Components Analysis by SPSS program (version 14; SPSS Inc., Chicago, IL, USA) was used for the determination of factors underlying the inter-correlations between the measured species. Pearson correlation ( $r$ ) was used for data analysis to identify relationships between ionic species in rainwater.

## 3. Results and Discussion

### 3.1. Relationship Between pH and Rainfall Amount

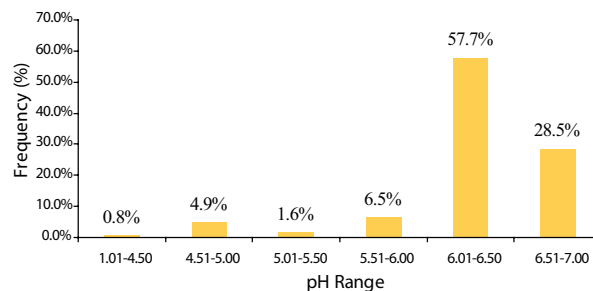


Fig. 2. Frequency distribution of rain pH.

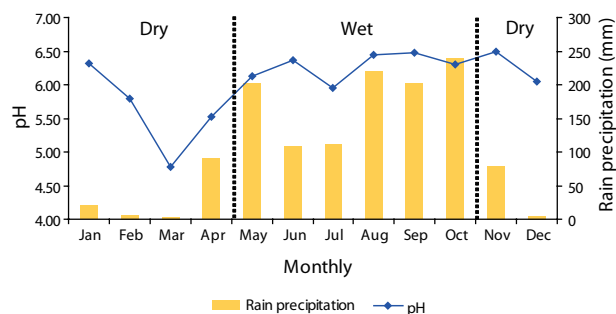


Fig. 3. Variation of monthly precipitation amounts and mean pH values.

The total number of collected samples was 132 and precipitation volume was 1,286.7 mm. The frequency distribution of the rainwater pH values ( $n = 123$  with precipitation  $>4\text{ mm}$ ) is illustrated in Fig. 2.

pH values of almost 60% of the samples were in a range of 6.01–6.50, which agreed with a previous study [17]. About 28% of samples had a higher pH (6.51–7.00). Only 7% of the samples illustrated had a pH of less than 5.60. The samples with pH values above 6.0 may suggest various inputs of alkaline species into precipitation.

Monthly mean pH values are shown in Fig. 3. Acidic precipitation ( $\text{pH} < 5.6$ ) was found during the months of March to April. Monthly mean pH values were lowest in March ( $\text{pH} = 4.78$ ) and highest in November ( $\text{pH} = 6.50$ ). In the dry season, high amounts of pollutants accumulated in the atmosphere due to lower levels of precipitation. Therefore, rain samples collected during the dry season contain high ionic concentrations, which presented low pH values.

### 3.2. Relationship Between EC and Rainfall Amount

The number of rainwater samples measured for EC was 123. EC values ranged from 0.18 – 6.82 mS/m and the volume weight mean EC value was 0.62 mS/m. The month with the highest mean EC value was March, whereas the month with the lowest value was November (Fig. 4).

The EC value is positively correlated with the level of ionic contamination in precipitation. Therefore, samples with high EC values contain high ionic concentrations. Based on the dilu-

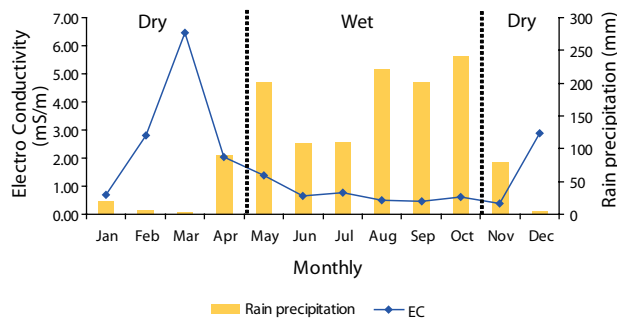


Fig. 4. Variation of monthly precipitation amount and mean electro-conductivity values.

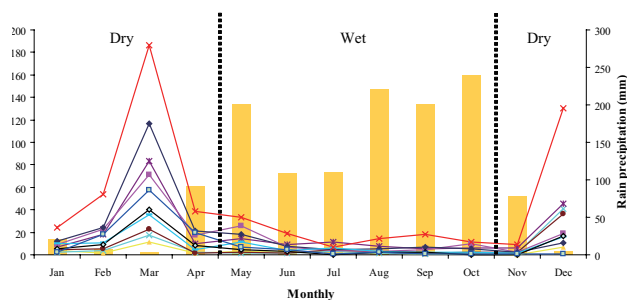


Fig. 5. Monthly average variability of anion and cation concentrations in units of  $\mu\text{eq/L}$ .

tion effect, the amount of precipitation is inversely related to the EC values.

### 3.3. Ion Concentration

The rainwater shows remarkable seasonal variations. Monthly average anion and cation concentrations, together with precipitation amount, are illustrated in Fig. 5.

The highest ion concentrations were found in March (dry season) due to the low amount of rain precipitation and long period of non-precipitation. On the other hand, low ion concentrations were found in the rainy season.

The order of anion and cation equivalents were  $\text{NH}_4^+$  (28%) >  $\text{Ca}^{2+}$  (18%) >  $\text{SO}_4^{2-}$  (14%) >  $\text{NO}_3^-$  (13%) >  $\text{Cl}^-$  (6%) >  $\text{CH}_3\text{COO}^-$  (5%) =  $\text{Mg}^{2+}$  (5%) >  $\text{K}^+$  (4%) =  $\text{Na}^+$  (4%) >  $\text{HCOO}^-$  (3%). The cation and anion species with the highest percentage of their respective categories were  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$ . The ratio between total anions and total cations was 43:57 with units of  $\mu\text{eq/L}$ .

### 3.4. Data Screening

In order to get high precision and correct data, it is necessary to do quality control. Data quality of rainwater samples was checked by ionic balance ( $R_1$ ) and conductivity balance ( $R_2$ ). The data was rejected if it did not meet the quality criteria.  $R_1$  and  $R_2$  values illustrate the accuracy of ion analysis and EC measurements, respectively. The principle of electro-neutrality in precipitation water requires that total anion equivalents be equal to total cation equivalents. Ion balance in a precipitation samples

can be calculated by the following equations [18];

$$C(\mu\text{eq/L}) = 10^{(6-\text{pH})}/1.008 + \sum C_{\text{ci}} \times V_i \quad (1)$$

Where  $C$  represents anion equivalents,  $C_{\text{ci}}$  is the concentration of  $i$ -th cation in  $\mu\text{mol/L}$ ,  $V_i$  is the valence of the given ion.

$$A(\mu\text{eq/L}) = \sum C_{\text{Ai}} \times V_i \quad (2)$$

Where  $A$  represents cation equivalent,  $C_{\text{Ai}}$  is the concentration of  $i$ -th anion in  $\mu\text{mol/L}$ .

$$R_1 = [(C - A)/(C + A)] \times 100\% \quad (3)$$

This is a simplified form of the corresponding equation used by the US Environmental Protection Agency where the denominator is the average of the two sums.

For dilution solutions (e.g. below  $10^{-3}$  M), the total conductivity can be calculated in mS/m from the molar concentrations and molar conductivity (at infinite dilution) of the individual ions. The calculation is as follows:

$$\Lambda_{\text{calc}} = \sum c_i \Lambda_i^0 \times 10^{-4} \quad (4)$$

Where  $\Lambda_{\text{calc}}$  denotes the calculated conductivity of the solution (in mS/m),  $C_i$  represents the ionic concentration of the  $i$ -th ion (in  $\mu\text{mol/L}$ ), and  $\Lambda_i^0$  represents the molar conductivity (in  $\text{S cm}^2/\text{mol}$ ) at infinite dilution and  $25.0^\circ\text{C}$ .

The calculation of conductivity values can then be compared to the observed value for precipitation samples with the relationship described in the equation below;

$$R_2 = ((\Lambda_{\text{calc}} - \Lambda_{\text{meas}}) / (\Lambda_{\text{calc}} + \Lambda_{\text{meas}})) \times 100\% \quad (5)$$

The total number of precipitation samples with complete measurements was 123 (100%). The number of samples that met the quality control criteria in terms of  $R_1$  and  $R_2$  were 62 (50.4%) and 96 (78.0%) respectively. Only 58 samples (47.2%) were qualified based on  $R_1$  and  $R_2$  combination criteria and therefore were used for statistical analysis.

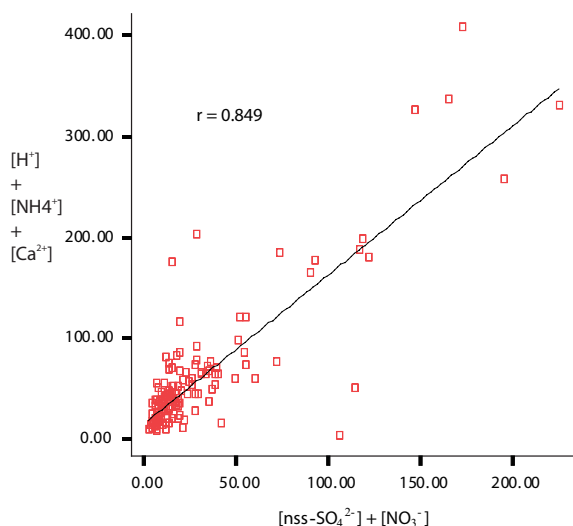
### 3.5. Acid Neutralization

Some base ions found in precipitation (e.g.,  $\text{Ca}^{2+}$  and  $\text{NH}_4^+$ ) act as buffers for the acidity of rainwater. To estimate the neutralization capacity of each alkaline compound, the neutralization factor (NF) was calculated by the following equation [19]:  $\text{NF}_{\text{xi}} = [\text{Xi}] / ([\text{SO}_4^{2-}] + [\text{NO}_3^-])$ , where  $\text{Xi}$  is the chemical component of interest, with all the ions expressed in  $\mu\text{eq/L}$ . The corresponding values for the major cations are given in Table 1.  $\text{NH}_4^+$  was found to have the highest neutralization effect, with  $\text{Ca}^{2+}$  having the second highest, while the contribution of  $\text{Na}^+$  and  $\text{Mg}^{2+}$  to the overall neutralization process was very low.

The regression coefficient of the relationship of the sum of

Table 1. Neutralization factors (NF) of the major ions in the rainwater samples

	$\text{NH}_4^+$	$\text{Ca}^{2+}$	$\text{Na}^+$	$\text{Mg}^{2+}$
NF	1.03	0.81	0.24	0.22



**Fig. 6.** Correlation of  $([H^+] + [NH_4^+] + [Ca^{2+}])$  and  $([nss-SO_4^{2-}] + [NO_3^-])$  ( $n = 132$ ).

the concentrations of the cations to the sum of the concentration of the anions was 0.849 (Fig. 6). From this relatively high correlation, we can estimate that  $nss-SO_4^{2-}$  and  $NO_3^-$  are in neutralized forms. Such neutralization is frequently reported and attributed to  $NH_4^+$  and/or  $CaCO_3$  [20]. It is expected that the sum of the concentrations of the three main cations  $([H^+] + [NH_4^+] + [Ca^{2+}])$  correlated with sum of the concentrations of the major anions  $([nss-SO_4^{2-}] + [NO_3^-])$  if the acidity of the precipitation is mainly neutralized by typical bases in the atmosphere, i.e.  $NH_3$  and soil dust.

### 3.6. Statistical Analysis

#### 3.6.1 Correlation Analysis of Ion Composition

Correlation analysis is a useful technique to characterize relationships among the ions present in rainwater. In order to find the associations among ions in precipitation as well as the pos-

sible sources of pollutants, correlations among ions in precipitation are calculated and listed in Table 2. A correlation was seen between  $SO_4^{2-}$  and  $NO_3^-$  ( $R^2 = 0.70$ ) indicating they originated from similar sources. This was due to the similarity in their behavior in precipitation and the result of the emissions of their precursors,  $SO_2$  and  $NO_x$ . Similarly, a strong correlation was seen between  $Ca^{2+}$  and  $Mg^{2+}$  ( $R^2 = 0.82$ ), suggesting the common source of these ions was a natural source (e.g. crustal origin). Other relatively good correlations were observed between  $NH_4^+$  and  $NO_3^-$ ,  $Mg^{2+}$  and  $Cl^-$ ,  $Ca^{2+}$  and  $NO_3^-$ , and  $NH_4^+$  and  $SO_4^{2-}$ .

Most of these well-correlated pairs have common occurrences in precipitation as a result of atmospheric chemical reactions likely from the reaction of the acid in the atmosphere, such as  $HNO_3$  and  $H_2SO_4$ , with alkaline compounds rich in carbonate materials, which were carried into the atmosphere by wind-blown dust. This shows that the wind-carried dust and soil play important roles in the chemistry of rainwater. Ammonium was correlated with  $NO_3^-$  ( $R^2 = 0.88$ ). Other relatively good correlations were observed between  $Mg^{2+}$  and  $Cl^-$ , between  $Ca^{2+}$  and  $NO_3^-$ , and between  $NH_4^+$  and  $SO_4^{2-}$ , with correlation values of  $R^2 = 0.73, 0.72,$  and  $0.68$ , respectively. The ammonium compounds applied to soil can escape into the atmosphere by means of gaseous  $NH_3$  or as  $NH_4NO_3$  and  $(NH_4)_2SO_4$  particles. When  $NH_4NO_3$  and  $(NH_4)_2SO_4$  particles are incorporated into rain, they change the  $NO_3^-$  and  $SO_4^{2-}$  concentrations, but do not affect the pH. However, when ammonium is incorporated into rain, it can neutralize the acidity of rainwater [3].

#### 3.6.2. Source Analysis of Chemical Components

Statistical analysis was performed using SPSS for Windows version 14.0 (SPSS Inc.). Data was log-transformed to achieve a normal distribution. Table 3 shows the varimax rotated principal component patterns for individual precipitation events. Three factors contributed to wet deposition during the sampling. Components 1, 2, and 3 were contributed 64.6%, 12.9 and 7.5%, respectively. Based on observations, only factor loads of higher than 0.5 have been deemed to be statistically significant [21]. The first principal component consists of high loads of  $SO_4^{2-}$ ,  $NO_3^-$  and  $NH_4^+$ .  $SO_4^{2-}$  and  $NO_3^-$  were derived from fuel combustion [22] and  $NH_4^+$  mainly originated from anthropogenic activities such

**Table 2.** Binary correlation coefficient between ions

Ion species	$SO_4^{2-}$	$NO_3^-$	$Cl^-$	$HCOO^-$	$CH_3COO^-$	$NH_4^+$	$Na^+$	$K^+$	$Ca^{2+}$	$Mg^{2+}$
$SO_4^{2-}$	1.00	<b>0.70</b>	0.55	0.56	0.57	<b>0.68</b>	0.15	0.23	0.53	0.43
$NO_3^-$		1.00	0.66	0.45	0.52	<b>0.88</b>	0.10	0.30	<b>0.72</b>	0.63
$Cl^-$			1.00	0.23	0.23	0.57	0.40	0.47	0.69	<b>0.73</b>
$HCOO^-$				1.00	0.89	0.62	-0.01	0.16	0.33	0.23
$CH_3COO^-$					1.00	0.63	-0.01	0.12	0.32	0.18
$NH_4^+$						1.00	0.01	0.33	0.66	0.60
$Na^+$							1.00	0.80	0.51	0.62
$K^+$								1.00	0.63	0.82
$Ca^{2+}$									1.00	<b>0.82</b>
$Mg^{2+}$										1.00

All values in  $\mu eq/L$ .

**Table 3.** Principal components analysis of wet deposition

Parameters	Component		
	1	2	3
SO <sub>4</sub> <sup>2-</sup>	0.793	0.454	0.295
NO <sub>3</sub> <sup>-</sup>	0.821	0.402	0.295
Cl <sup>-</sup>	0.598	0.405	0.414
NH <sub>4</sub> <sup>+</sup>	0.965	0.092	0.096
Na <sup>+</sup>	0.245	0.693	0.388
K <sup>+</sup>	0.234	0.260	0.907
Ca <sup>2+</sup>	0.514	0.558	0.384
Mg <sup>2+</sup>	0.224	0.891	0.129
Eigenvalue	9.197	1.841	1.070
%Variance	64.582	12.928	7.512
Source	fuel combustion and agricultural activity	soil resuspension and marine sources	biomass burning

as agricultural activity [23]. The second component shows high loads of Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>, which indicated that they originated from soil resuspension (Mg<sup>2+</sup> and Ca<sup>2+</sup>) and marine sources (Na<sup>+</sup>) [24]. The last principal component had a high load of K<sup>+</sup> came from biomass burning [25].

## 4. Conclusions

This study of the chemical composition of rainwater was carried out in the northern part of Thailand from January to December 2008. NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> were the dominant cation and anion found in the wet deposition in this area. Ion concentrations were high in the dry season and low in the rainy season.

The PCA illuminated the sources of the pollutants detected in precipitation, including fuel combustion (SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>) and agricultural activity (NH<sub>4</sub><sup>+</sup>) in the first component. The second component suggested the contribution of soil resuspension (Mg<sup>2+</sup> and Ca<sup>2+</sup>) and marine sources (Na<sup>+</sup>). The third component was associated with biomass burning (K<sup>+</sup>).

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