

Analysis of Precipitation Chemistry at Rural Site in the Eastern Coast, Korea

Gongunn Kang*, Dae-Ywen Shin¹⁾, and Hui-Kang Kim²⁾

Department of Environmental Science, Wonkwang Health Science College

¹⁾*Department of Environmental Engineering, Chosun University*

²⁾*Department of Environmental Engineering, Konkuk University*

(Received 28 December 2002; accepted 28 March 2003)

Abstract

The 10-day interval basis measurements of precipitation samples at Yangyang, the rural and coastal area on the eastern coast of the Korea peninsula were accomplished for understanding the precipitation chemistry and the temporal variations of major ions September 1991 to February 1997. The precipitation was slightly acidic, and 37% of the samples in winter were pH less than 4.5. The concentrations of cations were found on the order $\text{Na}^+ > \text{NH}_4^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ and those of anions followed the pattern $\text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^-$. Neglecting sea salt components, the major ions controlling precipitation chemistry were nss-SO_4^{2-} and NO_3^- in anion and NH_4^+ and nss-Ca^{2+} in cation. Concentrations of these ions were lower than those measured at urban sites in Korea, but were higher than those measured in Japan. Most of nss-SO_4^{2-} and NO_3^- were neutralized by ammonia and calcium species, especially alkaline soil particles in spring and ammonia gas in other seasons. Considering also the annual value of $[\text{nss-SO}_4^{2-}]/[\text{NO}_3^-]$ ratio of 2.62 and the neutralizing factors, ammonium sulphate compounds were dominant. Annual mean concentrations of these ions showed relatively small fluctuations, while larger seasonal variations were observed with higher levels in spring and winter. Precipitation amount, influence extent of acidic gases and alkaline particles long-range transported from China continent, and energy consumption pattern in each season might be able to explain this seasonal trend.

Key words : Precipitation chemistry, Sulfate, Nitrate, Ammonium sulphate, Coastal area

1. INTRODUCTION

Huge amount of SO_2 and NO_x is observed in the East Asian region, including Korea (Akimoto and Narita, 1994; Fujita *et al.*, 1991). The emitted SO_2 and NO_x are oxidized to sulfate and nitrate aerosols and have been transported from the Asian continent to other

downwind regions or countries through the Asian monsoon system before dry or wet deposition (Ishikawa *et al.*, 1998). In general, these precursor gases result in acid precipitation, which will have a harmful impact on aquatic and terrestrial ecosystems. The acid precipitation in the East Asia region has been recognized as one of the most important consequences of long-range transport of these acidic pollutants (Galloway *et al.*, 1987). Under this situation, the Korea Ministry of Environment has been operating a national acid rain

* Corresponding author.

Tel : 063-840-1274, E-mail : gukang@wkhc.ac.kr

monitoring network to measure the pH of precipitation since 1983 (Kim, 1997), even though the concentrations of major soluble ions in precipitation samples had not been analyzed until the late 1990s. Various studies on precipitation chemistry have also been carried out at several universities and government institutions in Korea. However, most of the studies made in 1980s and early 1990s had been performed with not enough periods to estimate the precipitation chemistry and mainly at large cities, including the capital of South Korea, Seoul (Na and Chung, 1997; Jung *et al.*, 1995; Koo and Park, 1993; Shin *et al.*, 1986; Son and Yang, 1985). In addition, the enough data set on precipitation chemistry at unpolluted region in Korea during the first half of 1990s that would be very useful in assessing the atmospheric pollution level at specific urban and polluted region had not been almost obtained.

In this paper, we present the observations of precipitation pH and concentrations of ion components at Yangyang considered as the unpolluted region, an rural area on the eastern coast of Korean peninsula, during the sampling period of five and half years. Precipitation samples were collected from September 1991 to February 1997. The precipitation acidity in conjunction with pH frequency and temporal variations of seasonal and annual pH were analyzed. To understand the source of ions measured in precipitation, we conducted a factor analysis and enrichment factor computation, and then examined the annual and seasonal concentration variations of major ions, controlling the precipitation chemistry at rural site on the eastern coast.

2. EXPERIMENT

2.1 Sampling site

Precipitation samples were collected at Yangyang area (38° 05' 57" N, 128° 25' 24" E) on the eastern coast of Korean Peninsula, which considered as clean area and a famous tourist resort in Korea. Precipitation samples were collected at 10-day intervals (on the 1, 11, 21 of each month, in principle) using a wet-only sam-

pler (M1-014, Metic Inc.) with an aperture area of 190 cm². The precipitation sensor responds to rain drops with a diameter larger than 0.5 mm and opens the lid of the wet side of the sampler, in which there is a five-liter polyethylene bottle. When rainfall stops, the lid of the wet side closes. The sampler was installed on the rooftop of the elementary school building at 3m above ground level and located about 5km away from the shoreline of the East Sea to the west. There are 20 commoner's houses around this sampling site and no industrial facilities with the exception of some livestock farming in this neighborhood. To prevent snow accumulation and freezing of samples in the inlet of the sampler, the driving part and funnel surface of the collector were heated automatically when the temperature fell below 0°C. The amount of precipitation was measured using a tipping-bucket rain gauge.

2.2 Sample analysis

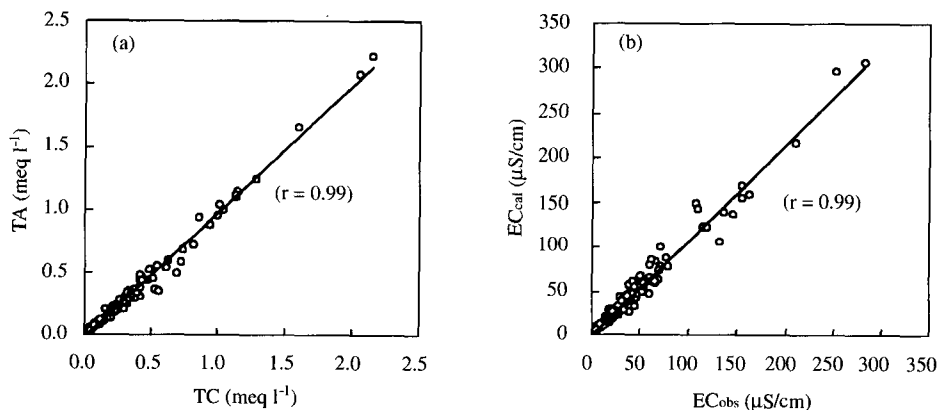
After sample recovery, the five-liter polyethylene sample bottle was sealed and placed in wooden box and then transported by post post to the chemical laboratory in the Department of Environmental Engineering of Konkuk University in Seoul, Korea. The analytical parameters of samples included pH, electric conductivity, Na⁺, K⁺, Ca²⁺, Mg²⁺, NH₄⁺, SO₄²⁻, NO₃⁻ and Cl⁻. As soon as the precipitation sample was received in the laboratory, pH and electric conductivity of the sample were measured by pH meter with glass electrode (pH/ion analyser-250, Corning Inc.) and electric conductivity by conductivity meter (Model SC-17A, Suntex Inc.) without any pretreatment. Before pH measurement, the electrode was calibrated by standard buffer solutions of pH 4.00 and 7.00 respectively. For the measurement of other parameters, samples were filtered through a Millipore filter with a pore size of 0.45 μm, and then these samples were stored in pre-cleaned polyethylene bottles at 4°C in the refrigerator until the concentrations of ion components were analyzed.

During the beginning of this monitoring, its concentrations were measured by the following analytical

Table 1. Analytical methods and equipment used in precipitation sample analysis.

Analytical parameter	Methods	Equipment
pH	Glass electrode	pH/Ion analyzer-250, Corning Inc.
Electric conductivity	Conductivity meter	SC-17A, Suntex Inc.
Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻	Ion chromatography	ICA-3000, TOA Inc. TOA PCI-201S column with 2.5 mM phthalic acid + 2.4 mM trishydroxy methyl aminomethane pH 4.0 as eluent
Na ⁺ , K ⁺ , Ca ²⁺ , Mg ²⁺ , NH ₄ ⁺	Ion chromatography	Dionex 100, Dionex Inc. CG 12 and CS 12 with 20mM methanesulfonic acid as eluent
	AAS*	AA 6500, Shimadzu Inc.

*Atomic absorption spectrophotometry

**Fig. 1. Scatter plots of cation sum vs. cation sum (a) and observed conductivity vs. calculated conductivity (b) for precipitation samples. The values in parentheses represent correlation coefficient.**

method. SO₄²⁻ was analyzed by the glycerol alcohol acid method, NO₃⁻ by the salicylic acid method, Cl⁻ by the mercury (II) thiocyanate method, NH₄⁺ by the indophenol method (UV-240, Shimadzu Inc.) and Na⁺, K⁺, Ca²⁺ and Mg²⁺ by atomic absorption spectrophotometer (GBC 901, GBC Inc.). After 1992 June, ion concentrations were analyzed by IC and a new atomic absorption spectrophotometer. Table 1 shows the analytical methods and equipment used in the analysis of the precipitation samples collected from 1992 June to 1997 February. Precision of the analytical measurement was determined by repeated analysis of standard solutions at different concentrations. The results for all ions were within 6%.

2.3 Data quality

Collection of precipitation samples began in February 1991. The first several months were used to establish the suitability of the sampling, sample transport, and analysis procedures. Precipitation samples included in this paper were between September 1991 and February 1997. During this monitoring period of 66 months, 137 samples were collected and 10-day periods with no precipitation or insufficient volume for chemical analysis during 10-day intervals were 49. 8 samples were also not measured due to sampling problems, analytical errors, or sample losses from the bottle during transport from the sampling site of Yangyang to the chemical laboratory in Seoul.

Table 2. Summary of annual mean composition data (September 1991 ~ February 1997).

	pH	AP ^a mm	NH ₄ ⁺	Ca ²⁺	K ⁺	Mg ²⁺	Na ⁺	NO ₃ ⁻	SO ₄ ²⁻	Cl ⁻
			μeq/L							
VWM ^b	5.05	1364	41.4	23.0	7.4	16.6	62.8	19.1	57.6	73.4
STD ^c	0.12	149	2.5	2.9	0.7	1.6	7.6	1.2	2.4	9.5
Max	5.57	1515	58.3	33.3	8.7	27.8	108.9	23.8	67.1	120.8
Min	4.77	1144	32.8	14.6	4.2	12.4	36.5	10.9	38.1	48.2

^aAP: Annual precipitation amount.

^bVWM: Volume-weighted mean of annual data of each analytical parameter.

^cSTD: Volume-weighted standard deviation.

As shown in Figure 1, the quality of the analytical data was checked by a cation-anion balance and by comparison of measured conductivity with the conductivity calculated from all measured ion concentrations and their specific conductivities, according to the EMEP manual (EMEP, 1996). The observed difference in cations and anions felt within the acceptable range (Ayers and Yeung, 1996). The measured conductivities were also in good agreement with the calculated conductivities for precipitation samples.

3. RESULTS AND DISCUSSION

Table 2 summarizes annual composition data for precipitation samples during the sampling period of 66 months at Yangyang. Also included in Table 2 are annual minimum and maximum values and standard deviations. These values were calculated from each annual mean, computed in monthly mean data set of precipitation samples. The monthly data of each analytical parameter were also calculated from the analysis result of each sample collected at 10-day intervals. All mean values for the chemical variables were computed in volume-weighted mean by considering the precipitation amount, and those of pH were replaced by the volume-weighted mean concentrations of hydrogen ion calculated from the values of each sample pH.

To study seasonal variations of precipitation composition, the samples were separated into four groups: spring (March-May), summer (June-August), fall (September-November) and winter (December-February).

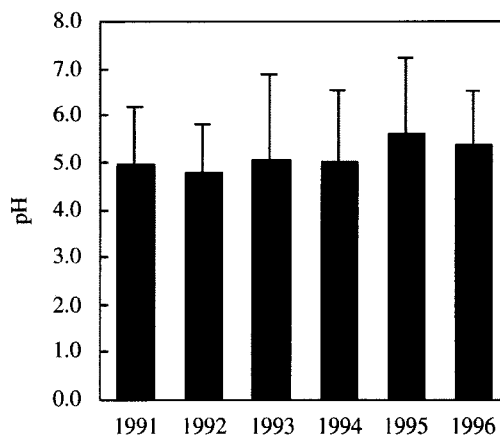


Fig. 2. Annual variation of pH. Error bars mean maximum and minimum of monthly pH.

ruary).

3.1 Precipitation acidity

The annual mean pH was 5.1 with ranging from 4.8 ~ 5.6 (Fig. 2). On the basis of the pH values, precipitations measured at rural site of Yangyang were found to be in an acidic nature as compared to the reference level of 5.6 (Charlson and Rodhe, 1982). This annual mean pH was higher than those in other sites. Kang *et al.* (1999) studied precipitation in a small city in southwest Korea from March 1995 to February 1997 and reported the annual mean pH to be 4.8. Lee *et al.* (2000) studied precipitation composition at nine locations on the Korean peninsula and found that the annual mean pH ranged from 4.6 to 4.8 during the period between

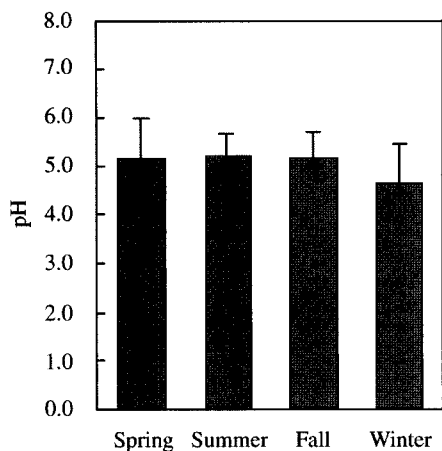


Fig. 3. Seasonal variation of pH. Error bars mean maximum and minimum of seasonal pH.

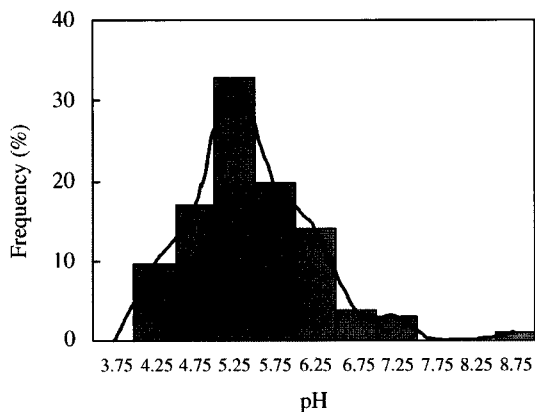


Fig. 4. Frequency distribution of pH.

May 1996 and April 1998. The pH value in this study was similar to those of precipitation in Japan (Seto *et al.*, 2000; Hara, 1993; CRIEPI, 1992) and in southern China (Wang and Wang, 1996), but much lower than those of precipitation in northern China (Wang and Wang, 1996).

The seasonal pH, as shown in Fig. 3, was found to be the similar value of 5.1~5.2 for all season except winter showing pH 4.6. Frequency distribution of pH during the whole sampling period and each season are depicted Fig. 4 and Fig. 5, respectively. The most fre-

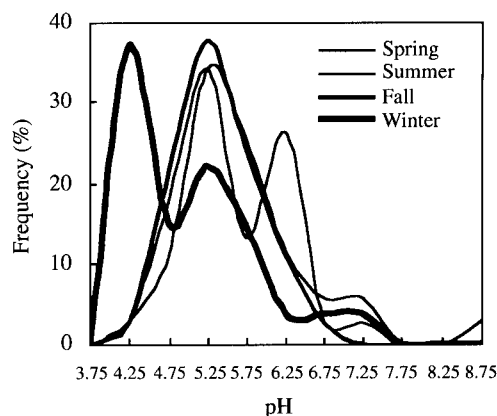


Fig. 5. Seasonal variations of pH frequency.

quently observed pH in Fig. 4 was about 5.3 ranging with 5.0~5.5 and for seasonal pH distribution its mode also seemed to be similar with that in previous figure except winter having 4.3 of pH mode. In a clean atmosphere, the pH was supposed to be between 5.0 and 5.6 due to dissolution of CO₂ in rain and cloud droplets and existence of background SO₂. Considering this criterion, most of precipitation samples were considered as not very acidic at this sampling site, but very acidic precipitation below pH 4.5 was present in 37%.

3. 2 Source analysis of major ion components

Considering the sampling site is very closely located on the seashore of the East Sea, sea salt is abundant in the chemical composition of precipitation samples (Crawley and Sievering, 1986). To understand the long-range transport of air pollutants emitted from other regions and the input and influence of acidic pollutants for the precipitation acidity, we need to consider the contribution of seasalt ions originated in the sea to the concentration of each ion component in precipitation and analyze the chemical characteristics of non-seasalt ions exclusive seasalt fraction in precipitation. So, the source characteristics of each ion component in precipitation were examined through factor analysis. This statistical analysis was conducted by principal component analysis with varimax rotation to a set of orthogo-

Table 3. Results of factor analysis with varimax rotation for the analytical data of 10-day event precipitation at Yangyang.

	Factor 1	Factor 2
NH ₄ ⁺	0.720	0.489
Ca ²⁺	0.925	0.058
K ⁺	0.574	0.671
Mg ²⁺	0.359	0.908
Na ⁺	0.182	0.968
NO ₃ ⁻	0.959	0.229
SO ₄ ²⁻	0.947	0.287
Cl ⁻	0.094	0.981
Variance explained	46.2%	44.4%

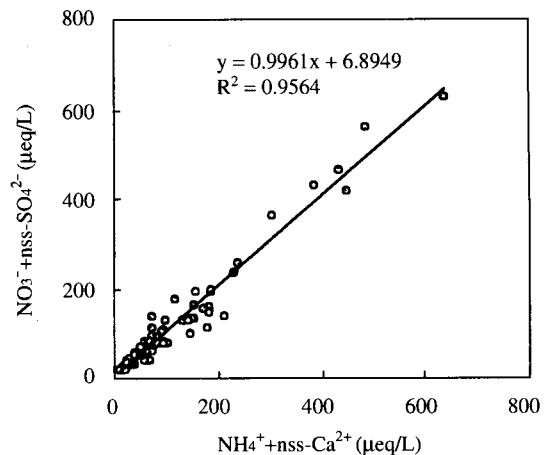
Factor loading above 0.7 are shown in bold type

Table 4. Enrichment factors and sea salt fractions of ion components.

	Ca ²⁺	K ⁺	Na ⁺	Mg ²⁺	SO ₄ ²⁻	Cl ⁻
EF	9.3	6.0	1.0	1.1	7.7	1.0
SSF (%)	11	17	100	91	13	100

nal axes for the analytical data of 10-day event precipitation samples. Table 3 contains the result of factor analysis. Two factors explaining 91% of data set variance were extracted by the statistical procedure. Factor 1 appears to represent clearly sea salt source, as it shows high loadings on sea salt constituents such as Na⁺, Mg²⁺, and Cl⁻. Factor 2 is considered as the contributions of non-sea salt source because of high loading on NH₄⁺, Ca²⁺, NO₃⁻, and SO₄²⁻.

To interpret the source analysis better and to quantify the extent of sea salt contribution to the concentrations of ion components with the possibility of sea salt origin in the precipitation, enrichment factor (EF) and seasalt fraction (SSF) by assuming that all the sodium in the precipitation is derived from the sea were calculated (Duce *et al.*, 1983). Table 4 shows enrichment factors and sea salt fractions. It can be inferred that if its EF value approaches unity, seasalt is the dominant source of element X. The enrichment factors and sea salt fractions calculated in Table 4 showed that the seasalt was the dominant source for Na⁺, Mg²⁺ and Cl⁻ in the precipitation. It is also considered that SO₄²⁻ and Ca²⁺ might be originated from sea salt. So, we calcu-

**Fig. 6. Acid-base balance between anion and cation with factor loading above 0.7 in Factor 2.**

lated the concentrations of nss-SO₄²⁻ and nss-Ca²⁺ by subtracting the fractions derived in the sea spray from the original concentration of these ions measured in the precipitation samples (Riley, 1975).

Annual mean concentrations of nss-SO₄²⁻ and nss-Ca²⁺, calculated from the above equations, were 50.0 µeq/L and 20.5 µeq/L, respectively. Neglecting sea salt components, the major ion components in the precipitation at Yangyang, controlling the precipitation chemistry, were found to be NO₃⁻ and nss-SO₄²⁻ in anion and NH₄⁺ and nss-Ca²⁺ in cation, even if the dominant components measured in the precipitation samples were Na⁺ and Cl⁻. The observed annual mean concentration of nss-SO₄²⁻ was lower than that (67.9 µeq/L) in precipitation measured at Iksan (Kang *et al.*, 1999) and that (87.4 µeq/L) in precipitation at Seoul (Kang *et al.*, 1996). However, the observed nss-SO₄²⁻ concentration was higher than that (40.2 µeq/L) in precipitation measured at Shobara, the rural area in Japan (Sinya *et al.*, 2000).

These cations and anions, known as the major components and Factor 1, are plotted in Fig. 6, showed a very well defined acid-base balance. This fact suggests that even though NO₃⁻ and nss-SO₄²⁻ in the precipitation appear to be the major acidifying species, these

are neutralized by the cation such as NH_4^+ and nss-Ca^{2+} originated from ammonia gases and alkaline particles, incorporated into cloud droplets or falling precipitation, and are present in the forms of neutralized salts rather than as sulfuric and nitric acids. In addition, the annual $[\text{H}^+]/([\text{nss-SO}_4^{2-}] + [\text{NO}_3^-])$ ratio, fractional acidity defined by Daum *et al.* (1984), was 0.13. This ratio also indicates that nearly 87% of the two acids were neutralized by ammonia and calcium species. Tsuruta (1989) has pointed out that ammonia and calcium species partly neutralize the acidic components in precipitation in Japan. The annual value of fractional acidity in this study is almost similar to that in precipitation at a rural site in Hiroshima Prefecture, Japan (Sinya *et al.*, 2000).

Assuming that precipitation acidity originates primarily from sulfuric and nitric acids, the equivalent ratio of $[\text{nss-SO}_4^{2-}]/[\text{NO}_3^-]$ would display the relative importance of their contributions to precipitation acidity (Chan *et al.*, 1987; Summers and Barrie, 1986). The annual $[\text{nss-SO}_4^{2-}]/[\text{NO}_3^-]$ ratio was 2.62. This ratio suggests that the fractions of H_2SO_4 and HNO_3 to the inorganic precipitation acidity were 72.4% and 27.6%, respectively.

Neutralization factors were calculated to evaluate the relative neutralization of precipitation by the crustal components and ammonia. Neutralization factors (NF) were calculated as

$$\text{NF}_{\text{NH}_4^+} = \frac{[\text{NH}_4^+]}{[\text{nss-SO}_4^{2-}] + [\text{NO}_3^-]} \quad (\text{Possanzini } et al., 1988) \quad (1)$$

$$\text{NF}_{\text{nss-Ca}^{2+}} = \frac{[\text{nss-Ca}^{2+}]}{[\text{nss-SO}_4^{2-}] + [\text{NO}_3^-]} \quad (\text{Kang } et al., 1999) \quad (2)$$

where $[\text{NH}_4^+]$, $[\text{nss-SO}_4^{2-}]$, $[\text{NO}_3^-]$ and $[\text{nss-Ca}^{2+}]$ denote concentrations in $\mu\text{eq/L}$. The $\text{NF}_{\text{NH}_4^+}$ and $\text{NF}_{\text{nss-Ca}^{2+}}$ were 0.60 and 0.30, respectively. This fact suggests that H_2SO_4 and HNO_3 in the precipitation are neutralized more easily by ammonia gas or ammonium containing aerosols than by calcium compounds. The

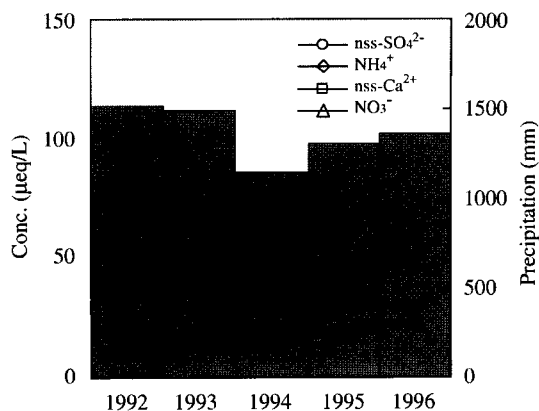


Fig. 7. Annual variation of the conc. of major ions and the precipitation amount.

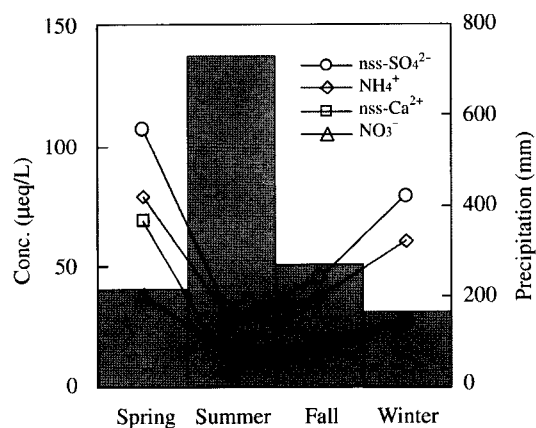


Fig. 8. Seasonal variation of the conc. of major ions and the precipitation amount.

high value of $\text{NF}_{\text{NH}_4^+}$ might be attributed to local sources such as livestock and application of fertilizers as well as wet deposition of ammonia gas or ammonium containing aerosols transported from outside natural and anthropogenic emission sources. On the basis of the high $\text{NF}_{\text{NH}_4^+}$ values and the $[\text{nss-SO}_4^{2-}]/[\text{NO}_3^-]$ ratio it may be said that NH_4^+ in precipitation is mainly present in the form of $(\text{NH}_4)_2\text{SO}_4$ rather than NH_4HSO_4 , NH_4NO_3 . Among the compounds of ammonium, ammonium sulphate compounds are known to predominate

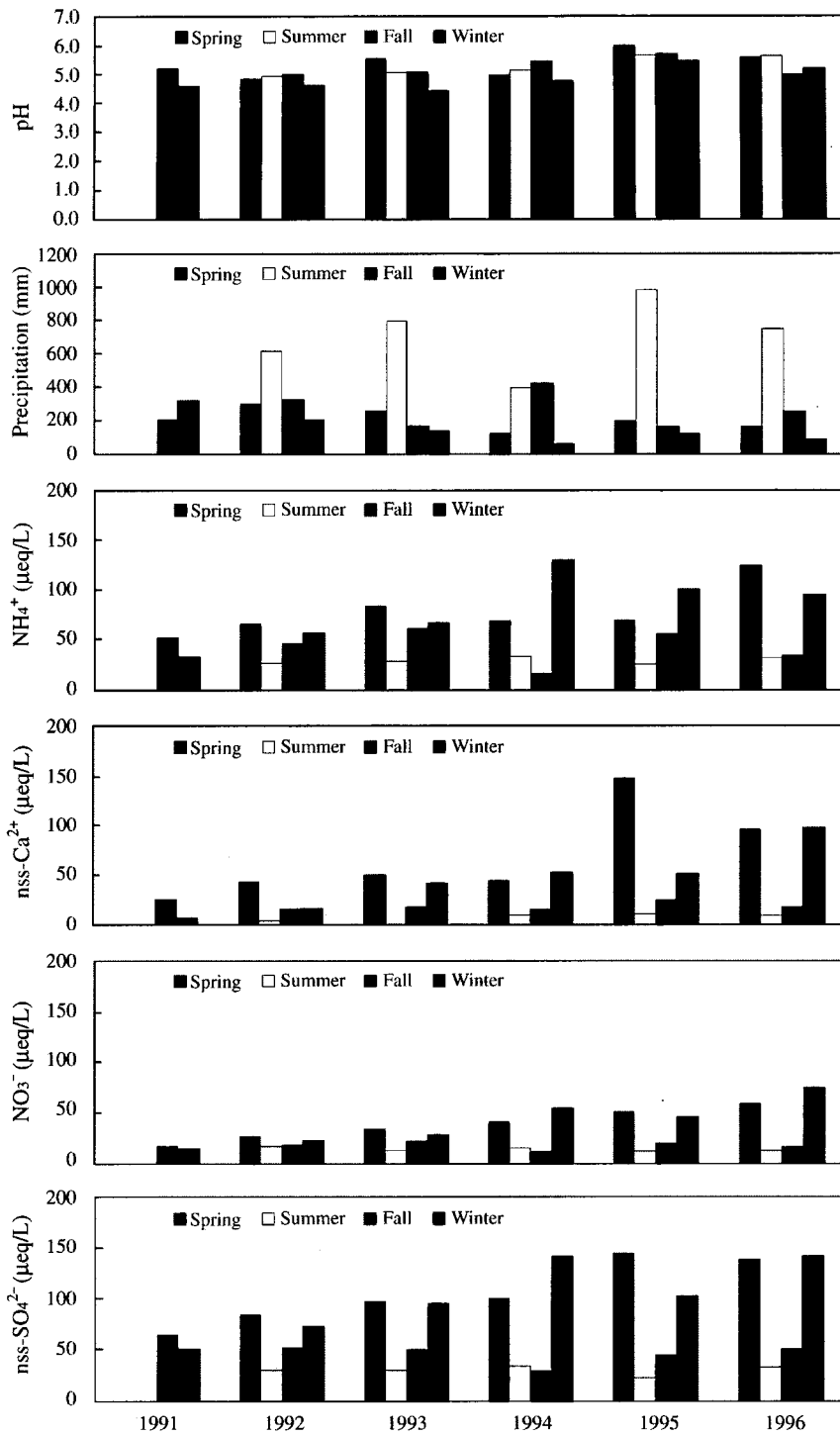


Fig. 9. Annual variations of seasonal pH, precipitation amount and the concentrations of major ions in precipitation samples.

in the atmosphere (Seinfeld, 1986).

3.3 Seasonal and annual variations of major ion components

The annual and seasonal variations of major ion components in the precipitation are presented in Fig. 7 and Fig. 8, respectively. The concentrations of major ion components are known to be relatively small differences between the annual mean values, while those of major ion components between the seasonal mean levels are shown to be very large differences, having the decreasing order of spring, winter, fall and summer. The concentrations of nss-SO_4^{2-} in anion and NH_4^+ in cation are dominant. The concentrations of NO_3^- and nss-Ca^{2+} , showing similar levels each other in annual variations except of those in 1995 under the strong influence of Yellow Sand long-range transported from northern Chinese continent. The elevated levels of major ions in spring, especially nss-Ca^{2+} , may be due to the influence of Yellow Sand transport, which has been mainly happened during this season, and light precipitation amount. In winter, high energy consumption rate for heating and short precipitation amount could be explanation for high levels in all major ions. Summer is the wettest season in Korea and this also contributes to the summer minimum. More detailed seasonal variations of these major ion concentrations, in addition to pH and precipitation amount during this study, are presented in Figure 9. Relatively big seasonal fluctuations for these major ion concentrations and precipitation amount were also observed. As a result of this, the precipitation chemistry at this site is considered to depend the seasonal emission characteristics of air pollutants through natural and anthropogenic activities and seasonal meteorological conditions such as wind system and precipitation amount.

4. CONCLUSIONS

The 10-day interval basis measurements of precipitation chemistry were conducted at Yangyang located

at a rural and coastal area of the Eastern Coast in Korea peninsula for five and half years early in 1990s.

The annual mean pH was and 5.1 with ranging from 4.8~5.6, which means that precipitation was slightly acidic. Strong acid precipitation below pH 4.5 was 37% of the samples in winter. Factor analysis was performed on 10-day interval data of ion components. Varimax rotated two factors accounting for approximately 91% of the total variance on the data set, were interpreted as non-sea salt and sea salt. The communalities for all ions were greater than 0.8, which suggests that their variability could be successfully explained by two factors. On the basis of enrichment factors and factor analysis, most of Na^+ , Mg^{2+} and Cl^- in precipitation were originated from seawater. Even though Na^+ and Cl^- were in the highest dominance in precipitation, the major acidifying components appeared to be nss-SO_4^{2-} and NO_3^- , and the main bases responsible for the neutralization of precipitation acidity were nss-Ca^{2+} and NH_4^+ . The annual mean concentrations of nss-SO_4^{2-} , NO_3^- , NH_4^+ and nss-Ca^{2+} were 50.0, 19.1, 41.4 and 20.5 $\mu\text{eq/L}$, respectively. Considering the acid-base balance of these ions and $[\text{H}^+]/([\text{nss-SO}_4^{2-}] + [\text{NO}_3^-])$ ratio of 0.13, nearly most of these two acids were found to be neutralized by ammonia and calcium species, especially ammonium sulphate compounds. The annual $[\text{nss-SO}_4^{2-}]/[\text{NO}_3^-]$ ratio was 2.62. The concentrations of nss-SO_4^{2-} , NO_3^- , NH_4^+ and nss-Ca^{2+} showed pronounced seasonal variations with lower concentrations in summer, when precipitation amounts are greater, and higher concentrations in winter and spring, when rainfall is reduced and energy consumption amount is increased for heating.

ACKNOWLEDGMENT

We are grateful to two anonymous reviewers for their comments. This study was supported by Wonkwang Health Science College as a part of research supporting program.

REFERENCES

- Akimoto, H. and H. Narita (1994) Distribution of SO₂, NO_x and CO₂ emissions from fuel combustion and industrial activities in Asia with 1° × 1° resolution, *Atmospheric Environment*, 28, 213–225.
- Ayers, G.P. and K.K. Yeung (1996) Acid deposition in Hong Kong, *Atmospheric Environment*, 30, 1581–1587.
- Chan, W.H., A.J.S. Tang, D.H.S. Chung, and N.W. Reid (1987) An analysis of precipitation chemistry measurements on Ontario, *Environ. Sci. Technol.*, 21, 1219–1224.
- Charlson, R.J. and H. Rodhe (1982) Factors controlling the acidity of natural rainwater, *Nature*, 295, 683–685.
- Crawley, J. and H. Sievering (1986) Factor analysis of the Map3s/rain precipitation chemistry network: 1976 ~ 1980, *Atmospheric Environment*, 20, 1001–1013.
- CRIEPI (1992) Acidic deposition in Japan, CRIEPI Report, 1–36.
- Daum, P.H., T.J. Kelly, S.E., and L. Newman (1984) Measurements of the chemical composition of strati-form clouds, *Atmosphere Environment*, 18, 2671–2684.
- EMEP (1996) EMEP manual for sampling and chemical analysis, EMEP/CCC-Report 1/95, Kjeller, Norway.
- Fujita, S., Y. Ichikawa, R.K. Kawaratani and Y. Tonooka (1991) Preliminary inventory of sulfur dioxide emissions in East Asia, *Atmospheric Environment*, 25A, 1408–1409.
- Fujita, S.I., A. Takahashi, J.H. Weng, L.F. Huang, H.K. Kim, C.K. Li, T.C. Frank, Huang, and F.T. Jeng (2000) Precipitation chemistry in East Asia. *Atmospheric Environment*, 34, 525–537.
- Galloway, J.N., D. Zhao, J. Xing, and G.E. Liknes (1987) Acid rain: China, United States, and remote area, *Science*, 236, 1559–1562.
- Hara, H. (1993) Precipitation chemistry on Japan, International Workshop on Acidic Deposition in East Asia, 24 ~ 26 November 1993, Hakone, Japan, pp. 9–12.
- Ishikawa, Y., K. Yoshimura, A. Mori, and H. Hara (1998) High sulfate and nitrate concentrations in precipitation at Nagasaki impacted by long-distance and local sources, *Atmospheric Environment*, 32(17), 1352–2310.
- Jung, S.H., J.C. Nam, S.K. Lee, and J.C. Choi (1995) Distribution pattern and chemical properties of acid precipitation in central region of Korean peninsula, *Journal of Korea Meteorology Society*, 31, 117–128.
- Kang, G.U. (1997) Wet deposition characteristics of acid precipitation, Dissertation of Konkuk University in Korea, 183–184.
- Kang, G.U., J.H. Lee, and H.K. Kim (1996) Interpretation of analytical data of ion components in precipitation, *Seoul, J. KAPRA*, 12(3), 323–332.
- Kang, G.U., I.K. Oh, and H.K. Kim (1999) Seasonal variations of acidity and chemistry of precipitation in Iksan area, *Journal of Korean Society for Atmospheric Environment*, 15(4), 393–402.
- Kang, G.U., D.Y. Shin, and H.K. Kim (1999) Precipitation chemistry and neutralizing capacity during spring at Iksan area, *Journal of Korean Society of Environmental Engineers*, 21(1), 197–206.
- Kim, H.K. (1997) Air pollution research and monitoring of wet deposition in Korea. Proceedings of the CRIEPI International Seminar on Transport and Effects of Acidic Substances, 28 ~ 29 November 1996, CRIEPI, Tokyo, Japan, pp. 29–38.
- Koo, J.K. and K.L. Park (1993) A study on the chemical characteristics of acid rain in Taejon city, *Journal of Korea Air Pollution Research Association*, 9(2), 147–153.
- Lee, B.K., S.H. Hong, and D.S. Lee (2000) Chemical composition of precipitation and wet deposition of major ions on the Korean peninsula, *Atmospheric Environment*, 34, 563–575.
- Na, C.K. and J.I. Chung (1997) Chemical composition of rainwater in Chonju-city, *Journal of Korea Air Pollution Research Association*, 13, 371–381.
- Possanzini, M., P. Buttini, and V. Dipalo (1988) Characterization of a rural area in terms of dry and wet deposition, *Science Total Environment*, 74, 111–120.
- Seinfeld, J.H. (1986) *Atmospheric chemistry and physics of air pollution*, Wiley/Interscience, New York.
- Seto, S., M. Oohara, and Y. Ikeda (2000) Analysis of precipitation chemistry at rural site in Hiroshima Prefecture, Japan, *Atmospheric Environment*, 34, 621–628.
- Shin, E.B., S.K. Lee, and K.H. Ahn (1986) Investigation on source strength to acid rain in Seoul area, *Journal of Korea Air Pollution Research Association*, 1, 66–74.

- Sinya Seto, Mayumi Oohara, and Yukoh Ikeda (2000) Analysis of precipitation chemistry at a rural site in Hiroshima Prefecture, Japan, *Atmospheric Environment*, 34, 621–628.
- Son, D.H. and S.C. Yang (1985) Studies on acid precipitation in Seoul, *Journal of Korean Air Pollution Research Association*, 1, 33–41.
- Summers, P.W. and L.A. Barrie (1986) The spatial and temporal variation of the sulfate to nitrate ratio in precipitation in eastern North America, *Wat. Air Soil Pollut.*, 30, 275–283.
- Tsuruta, H. (1989) Acid precipitation in Eastern Asia, *Kagaku*, 59, 305–315.
- Wang, W. and T. Wang (1996) On acid rain formation in China, *Atmospheric Environment*, 30, 4091–4093.