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Chemical characteristics of wet precipitation in urban and mountainous sites of Jeju Island

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Abstract: Wet precipitation samples were collected in Jeju City and Mt. Halla-1100 site (a site at an altitude of 1100 m on Mt. Halla) during 2011–2013, and their major ionic species were analyzed to examine the chemical composition and characteristics. A comparison of ion balance, electric conductivity, and acid fraction of precipitation revealed correlation coefficients in the range of r = 0.950~0.991, thereby implying the high quality of analytical data. Volume-weighted mean pH and electric conductivity corresponded to 4.86 and 25.5 μS/cm for Jeju City, and 4.98 and 15.1 μS/cm for Mt. Halla-1100 site, respectively. Ionic strengths of the wet precipitation in Jeju City and Mt. Halla-1100 site corresponded 0.3 ± 0.5 and 0.2 ± 0.2 mM, respectively, thereby indicating that more than 30 % of total precipitation was within a pure precipitation criteria. The precipitation with a pH range of 4.5 – 5.0 corresponded to 40.8 % in Jeju City, while the precipitation with a pH range of 5.0 – 5.5 corresponded to 56.9 % in Mt. Halla-1100 site, thereby indicating slightly more weak acidity than that in Jeju city. The volume-weighted mean concentration (μeq/L) of ionic species was in the order of Na^+ > Cl^- > nss-SO_4^{2-} > NO_3^- > Mg^{2+} > NH_4^+ > H^+ > nss-Ca^{2+} > PO_4^{3-} > K^+ > CH_3COO^- > $HCOO^-$ > NO_2^- > F^- > HCO_3^- > $CH_3SO_3^-$ at Jeju City area, while it corresponded to Na^+ > Cl^- > nss-SO_4^{2-} > NO_3^- > NH_4^+ > H^+ > Mg^{2+} > nss-Ca^{2+} > PO_4^{3-} > CH_3COO^- > K^+ > $HCOO^-$ > NO_2^- > F^- > HCO_3^- > $CH_3SO_3^-$ at Mt. Halla-1100 site. The compositions of sea salts (Na^+, Cl^-, Mg^{2+}) and secondary pollutants (NH_4^+, nss-SO_4^{2-}, NO_3^-) corresponded to 66.1 % and 21.8 %, respectively, in Jeju City and, 49.9 % and 31.5 %, respectively, in Mt. Halla-1100 site. The acidity contributions in Jeju City and Mt. Halla-1100 site by inorganic acids, i.e., sulfuric acid and nitric acid, corresponded to 93.9 % and 91.4 %, respectively, and the acidity contributions by organic acids corresponded to 6.1 % and 8.6 %, respectively. The neutralization factors in Jeju City and Mt. Halla-1100 site by ammonia corresponded to 29.8 % and 30.1 %, respectively, whereas the neutralization factors by calcium carbonate corresponded to 20.5 % and 25.2 %, respectively. From the clustered back trajectory analysis, the concentrations of most ionic components were higher when the airflow pathways were moved from the continent to Jeju area.

Key words: precipitation, Jeju Island, Mt. Halla-1100 site, acidity contribution, neutralization factor, clustered back trajectory

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1. Introduction

Wet precipitation plays an important role in the washing and elimination of air pollutants from the atmosphere.[1-2] Therefore, the chemical compositions of precipitation mainly depends on the major pollutants in ambient air.[3] Removal of air pollutants by wet precipitation mainly occurs in two scavenging mechanisms. The first mechanism is termed as in-cloud scavenging (rainout), which mostly absorbs or condenses gaseous pollutants and fine particles around the cloud droplet. The other mechanism is termed as below-cloud scavenging (washout), which is the most efficient pathway to remove fine or coarse particles.[1,3]

The atmospheric pollutants removed by the processes affect the acidification of precipitation. Specifically, the acidification of precipitation is closely related to the increase in the use of fossil fuels, and it is recognized as a result of rapid industrialization and industrial economy.[4] Increases in the use of fossil fuels leads to various atmospheric pollutants including sulfur oxides (SO_X) and nitrogen oxides (NO_X), which is reported to intensify the acidification of precipitation.[4] Specifically, SO_X and NO_X, correspond to typical pollutants that cause the acidification of precipitation and, are converted into strong acidic substances by ozone, hydrocarbons, hydrogen peroxide, and peroxides (e.g. HCOO•, RCOO•).[5]

The acidification of precipitation leads to destruction of aquatic ecosystems, degradation of crop productivity, destruction of forests, and erosion of buildings. It is typically affected by local and long-range transported pollutants. Therefore, the transboundary of acidic air pollutants including sulfur oxides and nitrogen oxides constitutes a significant environmental controversy between neighboring countries. North America and Northwest Europe have already suffered from serious acid rain and are faced with environmental controversies between neighboring countries. Furthermore, the countries are continuing to examine policy formulation and research to reduce acid rain.[6-7]

In East Asia, since the mid-1990s, the need for international cooperative research on monitoring of acid rain increased via ongoing consultation with the experts of International Conference on Acid Rain. Subsequently, Acid Deposition Monitoring Network in East Asia (EANET) is in operation since 2001. In Korea, since 1983, the ministry of environment has operated an acid rain measuring network in major cities including Seoul and industrial complexes.[7]

Jeju Island is a typical background site in Korea with few industrial facilities, low population density, and low pollution sources. Thus, it constitutes an advantageous location to monitor long-range transported air pollutants from the continent of Northeast Asia.[4,7] In the study, the ionic components of precipitation samples collected at Jeju City area and Mt. Halla-1100 site which are typical urban and mountainous sites in Jeju Island, were analyzed to investigate chemical and pollution characteristics with respect to altitude and airflow transport pathways.

2. Experimental

2.1. Collection of precipitation samples

A total of 147 precipitation samples were collected in the Jeju City area (Jeju National University; 33.26°N, 126.33°E) and 58 precipitation samples were collected in Mt. Halla-1100 site (33.21°N, 126.27°E) during 2011-2013. Automated rain samplers (model SL-4-001, Shinil Science Co., Gyeonggi, Korea) equipped with a rain sensor and stainless steel bucket (253 mm diameter) were used to collect precipitation samples. The samplers were installed on the roof of a building located in Jeju National University campus and the top of a trailer in Mt. Halla-1100 site. The HDPE bottles and sampling bucket were washed frequently before and after precipitation events with distilled water.

The precipitation samples were transferred to the laboratory, and a few of the samples were used to measure pH and electric conductivity. The remaining samples were partitioned into separate aliquots. An aliquot was preserved in a freezer at -20 °C without pretreatment for the analysis of inorganic ions and an other aliquot was stored at the same temperature for the analysis of organic acids after adding a drop of

chloroform.[4,7-8]

2.2. Chemical analysis

The pH value of each precipitation sample was measured via a pH meter (Orion 720A, 81-02 electrode; Thermo Fisher Scientific, Waltham, MA, USA). Two low ionic strength buffer solutions of pH 4.10 and 6.97 at 25 ºC were used for calibration prior to the measurement. Electric conductivity was measured via a conductivity meter (Orion 3-Star, 013005MD electrode, Thermo Electron, Waltham, MA, USA) at the same temperature.

Ion concentrations of the precipitation samples were analyzed for cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) and anions (SO_4^{2-}, NO_3^-, Cl^-, PO_4^{3-}, NO_2^-, F^-, $CH_3SO_3^-$, $HCOO^-$, CH_3COO^-) via ion chromatography (Metrohm Modula IC). With respect to the analysis of ionic species, the conditions of ion chromatography (IC) are shown in *Table* 1. The instrument detection limit (IDL) and coefficient of variation (CV) of the ion chromatography analyses are listed in *Table* 2.

2.3. Back trajectory analysis

Airflow pathway patterns were derived from a five day back trajectory analyses during the study periods. They were calculated via the PC version of HYSPLIT v. 4.0 (HYbrid Single-Particle Lagrangian Integrated Trajectory) developed by the National Oceanic and Atmospheric Administration/Air Resources Laboratory (NOAA/ARL).[9] Specifically, HYSPLIT was used to calculate back trajectories using meteorological data with a resolution of 1 × 1° that was generated by via the Global Data Assimilation System (GDAS) model from the National Weathers Service's (NWS) National Centers for Environmental Prediction (NCEP) (ftp://arlftp.arlhq.noaa.gov/archives/gdas1).[7] The total run time corresponded to 120 h, and the starting heights of the trajectories corresponded to 320 m in Jeju City and 1100 m in the Mt. Halla-1100 site.

3. Results and Discussion

3.1. Quality control of analytical data

It is necessary for the precipitation analysis to measure trace level concentrations of soluble ionic species. Therefore, the analytical data can vary based on the sample treatment and analytical process.[7] In precipitation analysis, the comparison methods of ion balance, electric conductivity and acid fraction are effectively applicable to improve the accuracy of the analyzed data. If the analysis of the precipitation components is performed quantitatively, then the ion balance between the sums of cations and anions should be correlated well. Furthermore, the correlation

Table 1. Instrumental conditions for ion chromatography analysis

Instrument	Cation	Anion & Organic Acid
IC	Metrohm Modula IC	Metrohm Modula IC
Column	Metrosep Cation C6	Metrosep A-SUPP-16
Eluent	3.0 mM HNO_3	7.5 mM Na_2CO_3
Suppressor	-	Metrohm 753 (200 mM H_2SO_4)
Flow rate	0.9 mL/min	0.8 mL/min
Injection volume	25 μL	100 μL

Table 2. Instrument detection limit (IDL) and coefficient of variation (CV) for the analysis of ionic components (n=7)

Components	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_2^-
IDL (μg/L)	2.2~18.7	1.5~9.6	4.4~17.9	2.4~17.7	2.6~14.8	1.2~8.0	11.6~12.0
CV (%)	0.8~4.9	0.4~2.1	0.8~5.5	0.2~7.7	0.8~4.4	0.2~2.0	3.1~4.3
Components	NO_3^-	SO_4^{2-}	PO_4^{3-}	F^-	$HCOO^-$	CH_3COO^-	$CH_3SO_3^-$
IDL (μg/L)	1.2~11.3	1.5~25.3	21.8~25.1	1.4~1.9	0.7~1.9	0.9~2.0	1.7~1.8
CV (%)	0.1~3.2	0.1~7.3	6.5~8.3	0.6~1.3	0.9~1.2	0.4~1.2	1.1~1.2

between the electric conductivity calculated by the ion concentrations and the electric conductivity measured by the conductivity meter should be high to significantly improve the accuracy of analytical data.[7,10] Additionally, the accuracy of analytical data can be inferred via calculating the acid fractions and their correlations.[11]

In the study, the ion balances, i.e. the correlation coefficient between the sums of the cation and anion equivalent concentrations corresponded to 0.980 in Jeju City, whereas it was 0.991 in Mt. Halla-1100 site. Additionally, the comparison between the calculated and measured electric conductivities exhibited high correlation coefficients of 0.986 and 0.971 in Jeju City and Mt. Halla-1100 site, respectively. Furthermore, the correlation coefficients between the acid fraction calculated from ionic concentrations and that from pH and conductivity corresponded to 0.962 and 0.950 in Jeju City and Mt. Halla-1100 site, respectively (*Fig.* 1). The results from three comparison methods confirmed that the quality of analytical data is good and available.

The percent imbalance (%) between the sums of cationic and anionic equivalent concentrations (Σ[Cation] and Σ[Anion]) was calculated based on the equation (Σ[Cation] − Σ[Anion])/(Σ[Cation] + Σ[Anion]) × 100 to examine the variance of the analytical data (*Fig.* 2). The results indicated that five (3.4 %) out of the total 147 precipitation samples exhibited imbalances exceeding 25 % in Jeju City. Conversely, the results revealed that six (10.3 %) out of the total 58 precipitation samples exhibited imbalances exceeding 25 % in Mt. Halla-1100 site. The results satisfy a common criterion of relative error, which is typically required for the trace level analysis in experiments.[7]

3.2. Concentrations of precipitation components

The volume-weighted mean pH and electric conductivity of the precipitation measured at Jeju City corresponded to 4.86 and 25.5 μS/cm, respectively. Conversely, the values at Mt. Halla-1100 site corresponded to 4.98 and 15.1 μS/cm, respectively. The ionic strength of precipitation corresponded to

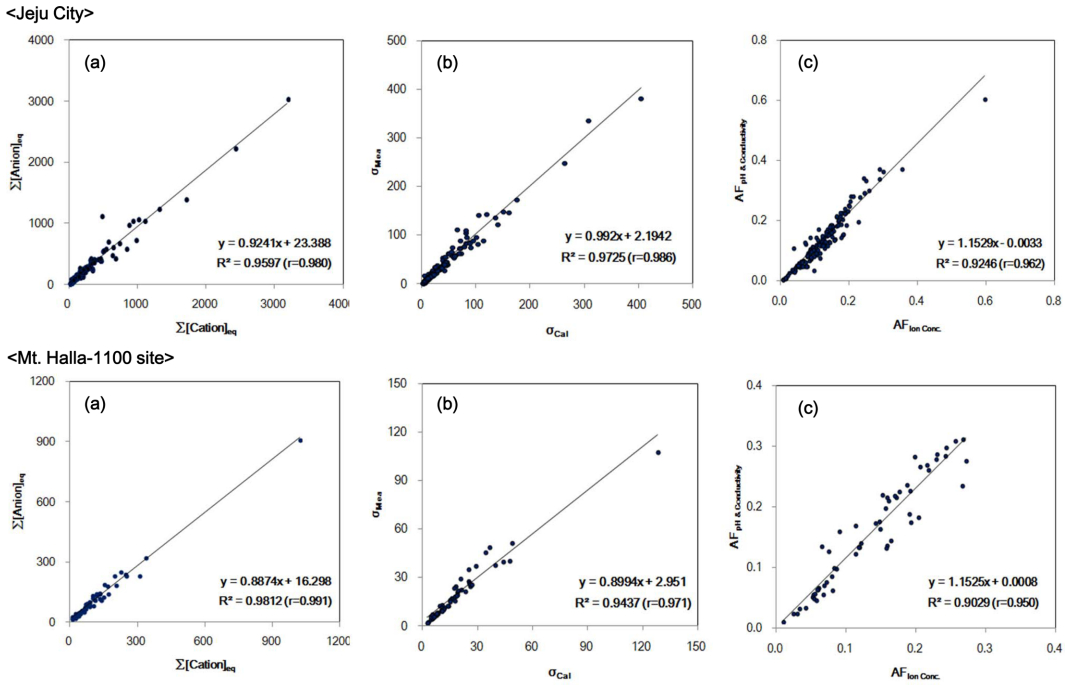

Fig. 1. Evaluation of analytical data by the comparison of ion balance (a), electric conductivity (b), and acid fraction (c).

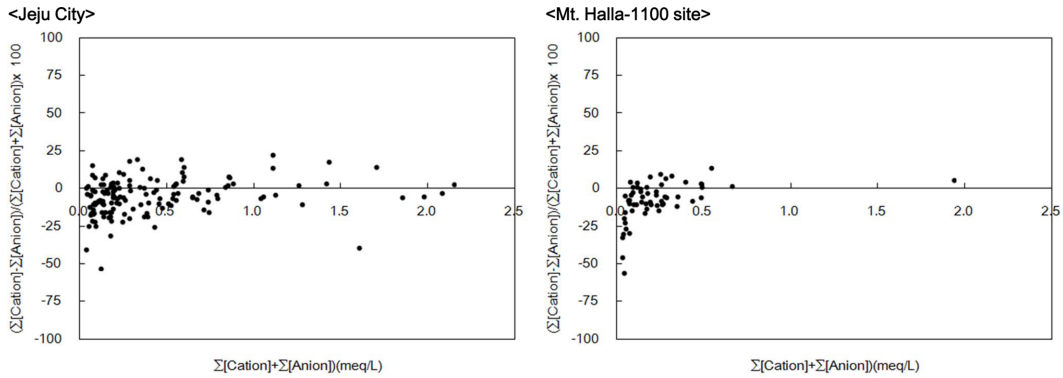

Fig. 2. Scattergram of (Σ[Cation] − Σ[Anion])/(Σ[Cation] + Σ[Anion])×100 vs. (Σ[Cation] + Σ[Anion]).

0.3 ± 0.5 mM, thereby indicating that 31.3 % of the total precipitation was within the pure precipitation criteria in Jeju City, whereas the ionic strength corresponded to 0.2 ± 0.2 mM in Mt. Halla-1100 site and 46.6 % of the total precipitation samples corresponded to the pure precipitation.[10]

Frequency of the precipitation with a pH range of 4.5 – 5.0 corresponded to 40.8 % in Jeju City, and this is the most proportional among the total precipitation samples. Conversely, in Mt. Halla-1100 site, the frequency with a pH range of 5.0 – 5.5 corresponded to 56.9 %, thereby indicating a little more basic than in Jeju city. The results suggested that the precipitation acidity can be more affected by altitude and air pollution in Jeju city. Based on the report of the Global Atmosphere Watch (GAW, 2013) on the acidity of precipitation, the pH range of precipitation between 1997 and 2013 corresponded to 4.68 – 4.88, thereby indicating the absence of regional differences or temporal variations.[12] The results of the study at two sites were similar to those in previous studies in Jeju Island.

The volume-weighted mean concentration (μeq/L) of the ionic species was in the order of $Na^+ > Cl^- > nss\text{-}SO_4^{2-} > NO_3^- > Mg^{2+} > NH_4^+ > H^+ > nss\text{-}Ca^{2+} > PO_4^{3-} > K^+ > CH_3COO^- > HCOO^- > NO_2^- > F^- > HCO_3^- > CH_3SO_3^-$ at Jeju City area, while it was in the order of $Na^+ > Cl^- > nss\text{-}SO_4^{2-} > NO_3^- > NH_4^+ > H^+ > Mg^{2+} > nss\text{-}Ca^{2+} > PO_4^{3-} > CH_3COO^- > K^+ > HCOO^- > NO_2^- > F^- > HCO_3^- > CH_3SO_3^-$ at Mt.

Table 3. Volume-weighted mean ionic concentrations (μeq/L) of precipitation in Jeju Island and other sites

Components	Jeju City	Mt. Halla-1100	Gosan[12]	Anmyeun[12]	Okinawa[13]	Shenzhen[14]	Shanghai[15]
NH_4^+	16.9	12.4	26.8	45.6	9.5	33.5	80.7
Na^+	98.2	37.4	106.7	57.9	308.0	11.2	50.1
K^+	3.0	1.6	3.6	3.0	9.4	1.8	14.9
Mg^{2+}	18.7	10.7	25.9	15.5	63.9	3.3	29.6
Ca^{2+}	13.9	10.6	19.0	21.8	25.2	35.4	204.0
Cl^-	86.0	34.3	123.4	68.7	351.0	20.6	58.3
NO_3^-	20.5	13.2	28.0	35.2	7.0	21.9	49.8
SO_4^{2-}	41.3	40.0	50.8	58.0	53.9	64.7	199.6
PO_4^{3-}	3.9	3.6	-	-	-	-	0.3
NO_2^-	1.2	1.1	-	-	-	-	0.2
F^-	0.7	0.7	1.1	1.8	-	1.7	11.0
$HCOO^-$	2.0	1.4	-	-	-	0.1	0.1
CH_3COO^-	2.7	2.3	-	-	-	0.0_3	0.4
$CH_3SO_3^-$	0.0_2	0.0_5	-	-	-	-	0.0_{03}

Halla-1100 site (*Table* 3). The concentration of non-sea salt sulfate (nss-SO_4^{2-}) and non-sea salt calcium (nss-Ca^{2+}) are obtained by the equation corresponding to '[nss-SO_4^{2-}] = [SO_4^{2-}] – 0.251×[Na^+]' and '[nss-Ca^{2+}] = [Ca^{2+}] – 0.044×[Na^+]', respectively.

Overall, the concentrations of sea salt species exceeded those of other species, and next by the secondary pollutant species in two sites in Jeju area. Based on the concentrations of precipitation components, the sea salts (Cl^-, Na^+, Mg^{2+}) and secondary pollutants (NH_4^+, nss-SO_4^{2-}, NO_3^-) contributed to the total composition of ionic precipitation species by 66.1 % and 21.8 %, respectively, in Jeju City. The composition of sea salt and secondary pollutants corresponded to 49.9 % and 31.5 %, respectively, in Mt. Halla-1100 site, thereby suggesting that the effect of sea salts is relatively high in low altitude areas (*Fig.* 3).

Table 3 shows the ionic concentrations of precipitation obtained from Jeju and other domestic/foreign sites. The results indicate that, the concentrations of sea salt components are higher in island areas, such as Jeju City, Mt. Halla-1100 site, and Okinawa, than in urban areas such as Shenzhen and Shanghai. Conversely, secondary pollutants, such as ammonium, nitrate, and sulfate, exhibit higher concentrations in urban areas than in Jeju City and Mt. Halla-1100 site.[12-15]

3.3. Acidification and neutralization characteristics

The acidification of precipitation is generally affected by the organic acids (RCOOH) and inorganic acids (H_2SO_4, HNO_3, HCl). Therefore, their acidification contribution rate can be deduced via a comparison of acidic anions, such as SO_4^{2-}, NO_3^-, Cl^-, and $RCOO^-$ concentrations.[7-8] However, Cl^- is considered to be mainly from the sea salt in Jeju Island, and thus the effect of Cl^- on the pH of precipitation is negligible.[16-17] In the study, the acidification contribution (AC) was calculated via Eq. (1) to confirm the effects of acidic substances on the acidification of precipitation.[8]

$$AC(\%) = \frac{[X]}{[nss-SO_4^{2-}]+[NO_3^-]+[HCOO^-]+[CH_3COO^-]} \times 100 \quad (1)$$

X = concentration of acidic substance

The acidification contributions of inorganic and organic acids are calculated in the equation, and the results are listed in *Table* 4. During the study period, the acidification contribution by inorganic acids in Jeju City and Mt. Halla-1100 site corresponded to 93.9 % and 91.4 %, respectively. Conversely, the acidification contribution by inorganic acids at two sites corresponded to 6.1 % and 8.6 %, respectively. The results confirmed that the acidification of precipitation mainly occurred via inorganic acids in Jeju Island.

Table 4. Acidification contributions (%) of acidic anions in precipitation

Site	nss-SO_4^{2-}	NO_3^-	$HCOO^-$	CH_3COO^-
Jeju City	56.7	37.2	3.3	2.8
Mt. Halla-1100 site	58.8	32.6	3.5	5.2

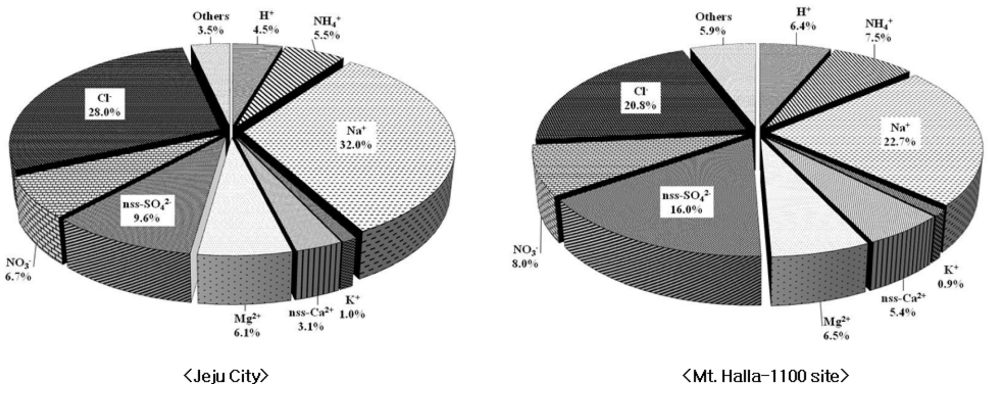

Fig. 3. Chemical composition ratios of precipitation components.

Table 5. Neutralization factors (NF) for ammonia and calcium carbonate

Site	Ammonia	Calcium carbonate
Jeju City	0.298	0.205
Mt. Halla-1100 site	0.301	0.252

The neutralization contribution is calculated by the neutralization factors (NFs) from Eqs. (2) and (3).[7,18-19] The results are listed in *Table* 5.

$$NF_{NH_4^+} = \frac{[NH_4^+]}{[nss-SO_4^{2-}]+[NO_3^-]+[HCOO^-]+[CH_3COO^-]} \quad (2)$$

$$NF_{nss-Ca^{2+}} = \frac{[nss-Ca^{2+}]}{[nss-SO_4^{2-}]+[NO_3^-]+[HCOO^-]+[CH_3COO^-]} \quad (3)$$

Acidic substances are mainly neutralized by basic substances, such as NH_3, $MgCO_3$, and $CaCO_3$, and neutralization is estimated by measuring NH_4^+, Mg^{2+}, and Ca^{2+} concentrations, which correspond to basic cations of precipitation. Specifically, NH_3 and $CaCO_3$ are known to mainly contribute to neutralization reaction of acidis substances.[16-17] The neutralization factors of the precipitation in Jeju City by ammonia and calcium carbonate corresponded to 0.30 and 0.21, respectively. Similarly, the values for the Mt. Halla-1100 site corresponded to 0.30 and 0.25, respectively. With respect to Jeju Island, the sums of the neutralization factors by ammonia and calcium carbonate approximately corresponded to 51~55 %, and two components were observed to mainly affect the neutralization reaction.

3.4. Effect of airflow transport pathways

Northeast Asia is characterized by rapid economic growth, and the long-range transport of air pollutants is consistently examined since the mid-1980s by organizations such as EANET and LTP.[8,20] The long-range transport of air pollutants is very sensitive and constitutes a significant issue between neighboring

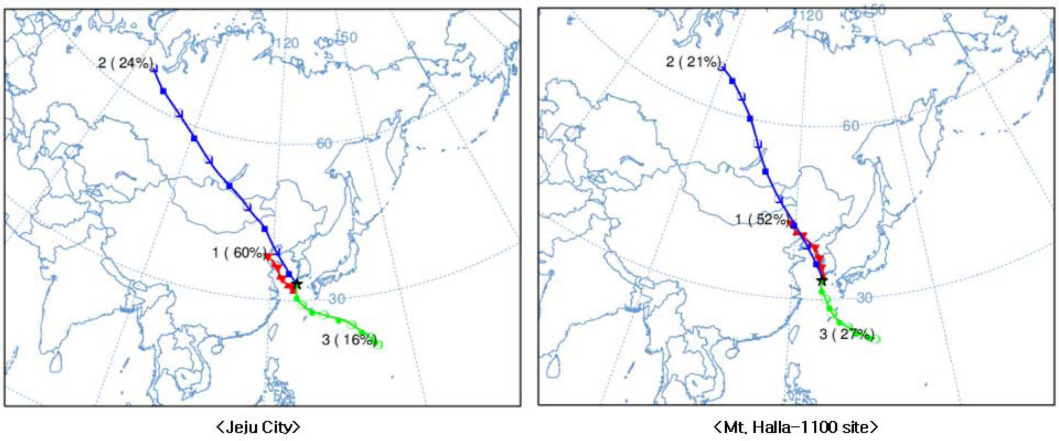

Fig. 4. Clustered 5-day back trajectories of air masses corresponding to precipitation sampling date.

Table 6. Ionic concentrations (μeq/L) of precipitation in accordance with airflow transport pathways by cluster back trajectory

Components	Jeju City			Mt. Halla-1100 site		
	Cluster 1 (n = 88)	Cluster 2 (n = 35)	Cluster 3 (n = 24)	Cluster 1 (n = 30)	Cluster 2 (n = 12)	Cluster3 (n = 16)
nss-SO_4^{2-}	65.66	84.18	34.55	31.59	49.51	39.11
NO_3^-	42.21	56.43	23.48	16.51	27.88	24.91
NH_4^+	31.52	44.39	23.89	14.49	23.98	28.89
nss-Ca^{2+}	24.69	30.94	10.22	13.37	27.32	7.87

countries in this area, and thus it is necessary to understand and clarify their transboundary transport mechanism.[7] Based on the sampling days at two sampling sites (147 days and 58 days), clustered back trajectory analysis was performed using NOAA's HYSPLIT 4 model and GDAS meteorological data to confirm transport pathways of air masses (*Fig*. 4).[21]

In the cluster analysis at Jeju City, 147 trajectories were classified into three clusters. Similarly, three clusters were identified at the Mt. Halla-1100 site. Thus, the clustered back trajectories were grouped into three pathways in Jeju City and Mt. Halla-1100 site as follows: In Jeju City, Cluster 1 (Eastern Coast of China & Korean Peninsula) corresponded to 60 %, Cluster 2 (Siberia, Mongolia & China Continent) corresponded to 24 %, and Cluster 3 (East sea & The North Pacific Ocean) corresponded to 16 %. In Mt. Halla-1100 site, Cluster 1 corresponded to 52 %, Cluster 2 corresponded to 21 %, and Cluster 3 corresponded to 27 %. We compare the concentration of ionic components, such as nss-SO_4^{2-}. NO_3^-, NH_4^+, and nss-Ca^{2+} by airflow transport pathway, and their components exhibited high concentrations when the airflow moved to Jeju from Siberia, Mongolia, and China continent (Cluster 2) (*Table* 6). Additionally, the concentrations of ionic components in Cluster 1 and Cluster 2 were high in Jeju City, which exhibited a relatively low altitude, and the concentrations of ionic components were low in Cluster 3. However, in high altitudes, such as the Mt. Halla-1100 site, the concentrations of ionic components in Cluster 3 exhibited high concentrations than that in the case of Cluster 1. The results indicated that the concentrations of components were different based on the airflow transport pathway and altitude.

4. Conclusions

In the study, the chemical compositions of precipitation collected in urban and mountainous sites of Jeju Island were compared to understand the differences each order. Additionally, their pollution characteristics were investigated relative to the altitude and airflow transport pathways.

The volume-weighted mean pH of precipitation in Jeju City and Mt. Halla-1100 site was weak acidic and corresponded to 4.86 and 4.98, respectively, and the electric conductivity corresponded to 25.5 μS/cm and 15.1 μS/cm, respectively. The precipitation with a pH range of 4.5–5.0 corresponded to 40.8 % in Jeju City, and the precipitation with a pH range of pH 5.0–5.5 corresponded to 56.9 % in Mt. Halla-1100 site, thereby indicating a little slightly weaker acidity than in Jeju city. Furthermore, the ionic strengths of precipitation at two sites corresponded to 0.3 ± 0.5 mM and 0.2 ± 0.2 mM, respectively, thereby indicating the pure precipitation criteria corresponded to 31.3 % and 46.6 %.

The acidification contributions by inorganic acid in Jeju City and Mt. Halla-1100 site corresponded to 93.9 % and 91.4 %, respectively, and the neutralization factors by ammonia and calcium carbonate approximately corresponded to 51~55 %.

The results of clustered back trajectory analysis indicated, that the concentrations of most ionic components were higher when the airflow pathways moved from the Asian continent to Jeju area.

Acknowledgements

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