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Monthly Characteristics of Rainwater Chemistry at a Coastal Site in Southwestern Japan

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ABSTRACT

Monthly characteristics of rainwater chemistry at a coastal site in southwestern Japan were examined based on an eight year record. In the period November-May when rain was mainly caused by cyclones, the monthly mean concentrations of nss-SO_4^{2-}, NO_3^-, NH_4^+, nss-Ca^{2+}, Na^+ and Cl^- over the eight years were 25.1-57.8, 9.9-25.0, 11.3-31.4, 5.5-18.7, 24.2-154.9 and 30.0-178.5 μeq L^{-1}, respectively. In June and July when rain was mainly caused by stationary fronts, i.e. Meiyu fronts, the concentrations were 14.4-20.7, 7.2-9.5, 7.7-12.9, 4.1-6.8, 21.7-33.6 and 26.4-40.5 μeq L^{-1}, respectively. In August and September when typhoons contributed substantial rainfall, the respective concentrations of Na^+ and Cl^- were as high as 97.7-105.3 and 116.8-122.9 μeq L^{-1}, while the concentrations of other ions were low. These results indicate a large variation of monthly rainwater chemistry, which is basically dependent on the synoptic weather patterns causing rain. From later autumn to early spring, rain contains ions in high concentration and large variation ranges. In the Meiyu season, rain contains less ions which vary in a range much smaller than that in later autumn-early spring. In summer and autumn, the concentrations are low, except Na^+ and Cl^- which can be large due to typhoons' contribution.

Key words: Ions, Cyclones, Stationary fronts, Typhoons, Synoptic weather

1. INTRODUCTION

Rain plays a crucial role in the chemical transformation and the removal of airborne species in the atmosphere and links various variations and evolutions on the Earth (Seinfeld and Pandis, 2006). The presence of ions such as sulfate and nitrate in rainwater and cloud water is a consequence of multiple processes and factors, including the emission and dispersal of air pollutants, chemical conversions in the air, thermodynamic properties of air parcels, and microphysical variations of cloud and rain droplets (Iribarne and Cho, 1989; Easter and Luecken, 1988). The input of ion species to ocean area via wet deposition is one of the key processes to drive the evolution and conservation of the marine ecosystems in the open ocean (Duce *et al.*, 2008).

On synoptic scales, rain over the East China Sea is usually caused by cyclones, Meiyu fronts (i.e. stationary fronts) and typhoons. Thermodynamic properties of air parcels in these weather patterns are different and have distinctive seasonal characteristics. Cyclones are low-pressure systems generated initially by the southward intrusion of cold polar air in the Euro-Asian continent and move eastward in the mid-latitude westerly winds (Barry and Chorley, 2003). A cyclone usually has a cold front, and sometimes also has a warm front after moving away from the Asian continent. Cyclone-associated rain, which is from clouds along fronts, can be observed in every month throughout a year. Meiyu fronts (also called Baiu fronts in Japanese) are stationary fronts. They are the boundaries between warm and humid tropical Pacific air in the south and cold and dry air in the north, when the tropical air expands northward under the prevention of the cold air. The fronts bring long-term and large rainfall in East Asia, usually in June and July, i.e., late spring to early summer (Zhou *et al.*, 2004). Typhoons are strong low-pressure systems in northwestern Pacific. They are initially generated in the tropical ocean areas and move to the middle latitude areas (Su *et al.*, 2012). Some typhoons arrive at and pass the East China Sea, causing short-term and extremely heavy rain in July-September, i.e., summer to early fall.

Rainwater chemistry is expected to be different according to the rain types because the thermodynamic mechanisms of the cloud formation on synoptic scales are different. Seasonal variations of the rainwater chemistry over the East China Sea should be closely dependent on the predominant rain types in different months or seasons. In particular, typhoons and Meiyu fronts do not occur in every season or month.

Monthly or seasonal variation of ions in rainwater, and wet deposition fluxes at land and ocean areas have been well studied with long-term records. The studies are usually discussed with an integrated base of records while seasonal characteristics have rarely been carefully examined according to rain types in a quantitative manner (e.g. Pan *et al*., 2013; Yang *et al*., 2012). Toyonaga and Zhang (2016) concentrated on the wet deposition fluxes of ions to the coastal areas of the East China Sea area, and reported that the annual variation of the fluxes were closely associated with dominance of the rain types. However, seasonal or monthly variations of rainwater chemistry according to the variation of the predominant rain types, and the relation between emission and rainwater chemistry at the coastal areas of the East China Sea have not been carefully examined.

In this study, seasonal characteristics of the concentrations of four terrigenous-origin ions (SO_4^{2-}, NO_3^-, NH_4^+ and Ca^{2+}) and two marine-origin ions (Na^+ and Cl^-) in the rainwater at a coastal site of southwestern Japan are examined. The records of 348 rain episodes over eight years from 1996 to 2003, which was the same dataset as used by Toyonaga and Zhang (2016), are applied. The major purposes are to demonstrate the association of the monthly variation of the rainwater chemistry with rain types causing the rain, and to provide accurate information for the inter-comparisons and the elucidation of rainwater chemistry.

2. MATERIALS AND METHODS

The concentrations of SO_4^{2-}, NO_3^-, NH_4^+, Ca^{2+}, Na^+ and Cl^- in the rainwater collected at the Reihoku-shiki Observatory (32°30′N, 130°03′E; 5 m asl; Fig. 1) from each rain event in the period from January 1996 to December 2003 are used. The observatory is located at a coastal area of Amakusa Island in southwestern Japan, with the East China Sea approximately 400 m to its west and 1.7 km to its north. Rain at the site is usually caused by weather changes on a synoptic scale. Rain events on a small scale, such as thunderstorms, rarely occur.

The collection of rainwater and the measurements of ions in the rainwater were carried out by the Kumamoto Prefectural Institute for Public-Health and Environmental Science, as a part of the acid rain monitoring project of Kumamoto prefectural government (Ogata *et al*., 2004). Rain events were classified into 4 types: cyclones (Cy), stationary fronts (SF), typhoons (Ty), and "Other", according to the weather information and surface weather charts issued by the Japan Meteorological Agency. Some rain events could not be categorized into the types of Cy, SF or Ty, because of the multiple or complex weather patterns causing the rain. They were grouped as "Other". The details of the procedure of rainwater collection, preservation and analyses, data quality control, and rain type classification are described by Toyonaga and Zhang (2016).

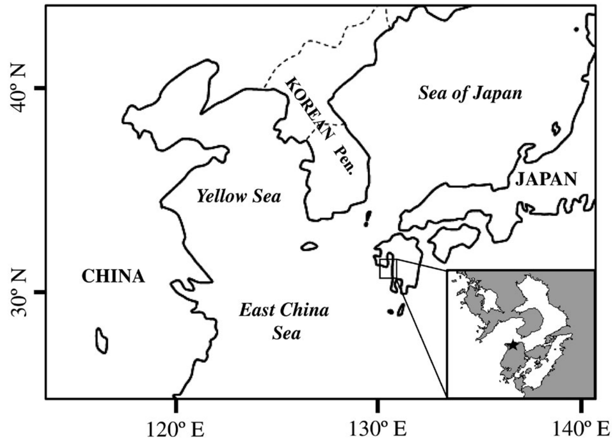

Fig. 1. Location of the rainwater collection site, Reihoku-shiki Observatory (marked with a star).

Monthly mean concentration of ions in rainwater was compiled into the "volume-weighted mean" concentration (Galy-Lacaux *et al*., 2009) of each month over the eight years. If there were less than three samples for a rain type in a certain month over the eight years, the monthly mean concentration was treated as "not available" (N.A.). The relative contribution of each rain type to the ions in the total rainwater in a certain period was evaluated according to ion deposition fluxes to the surface. The fluxes were calculated with the concentration of one or multiple ions in the rainwater multiplied by the amount of the rainfall. The monthly flux of an ion of a rain type was the summation of the fluxes in all episodes of the rain type in a month, and the mean flux was the mean of the eight one-month integrated values. Non-sea salt fractions of SO_4^{2-} (nss-SO_4^{2-}) and Ca^{2+} (nss-Ca^{2+}) were estimated with Na^+ as the conservative tracer for sea salt (Keene *et al*., 1986). The concentration and the flux of sea salt fractions of SO_4^{2-} and Ca^{2+} will not be described or discussed in this paper, because of their small contributions.

3. RESULTS

Monthly mean concentrations of nss-SO_4^{2-}, NO_3^-, NH_4^+, nss-Ca^{2+}, Na^+ and Cl^- over the eight-years in total rainwater were 14.4-57.8, 7.2-25.0, 7.7-31.4, 2.3-18.7, 21.7-154.9 and 26.4-178.5 μeq L^{-1}, respectively

Table 1. Monthly mean concentrations (in $\mu eq\,L^{-1}$) of ions in the rainwater at the study site and at remote sites in Japan.

Site name	Period	Ion species ($\mu eq\,L^{-1}$)	Rain type	Month											
				Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Reihoku-shiki[a]	1996-2003	nss-SO_4^{2-}	Total	45.8	57.8	37.4	25.1	26.3	14.4	20.7	20.6	21.4	20.7	29.1	43.4
			Cy	46.7	55.8	37.3	24.9	27.1	25.4	23.0	15.4	–	20.6	38.6	39.9
			SF	–	–	–	–	29.1	12.3	21.4	20.2	23.0	–	–	–
			Ty	–	–	–	–	–	–	19.3	5.7	15.2	–	–	–
		NO_3^-	Total	19.3	25.0	18.7	10.3	9.9	7.2	9.5	8.3	8.2	8.2	14.6	20.7
			Cy	19.8	24.3	18.7	10.2	10.1	9.0	10.3	8.7	–	8.5	19.3	18.1
			SF	–	–	–	–	10.6	5.7	9.8	7.9	6.6	–	–	–
			Ty	–	–	–	–	–	–	8.8	1.3	13.8	–	–	–
		NH_4^+	Total	17.6	31.4	21.2	13.5	12.5	7.7	12.9	10.1	10.4	8.6	11.3	16.4
			Cy	18.0	30.5	21.1	13.3	12.4	13.9	13.0	9.2	–	8.3	15.5	15.4
			SF	–	–	–	–	14.2	7.1	14.2	8.3	10.2	–	–	–
			Ty	–	–	–	–	–	–	9.9	3.4	20.7	–	–	–
		nss-Ca^{2+}	Total	10.0	18.7	11.4	8.6	5.5	6.8	4.1	2.3	6.4	4.8	11.8	11.0
			Cy	10.4	16.7	11.4	8.5	6.2	3.0	2.0	1.0	–	2.6	14.2	8.7
			SF	–	–	–	–	4.8	1.9	5.3	3.7	1.5	–	–	–
			Ty	–	–	–	–	–	–	2.7	3.7	15.1	–	–	–
		Na^+	Total	154.9	123.4	58.1	29.0	24.2	21.7	33.6	97.7	105.3	75.2	70.2	135.4
			Cy	159.6	122.3	58.1	29.3	24.2	16.0	13.9	37.1	–	31.6	126.4	115.8
			SF	–	–	–	–	16.4	19.5	34.7	40.8	23.2	–	–	–
			Ty	–	–	–	–	–	–	54.3	423.3	1004.2	–	–	–
		Cl^-	Total	178.5	148.1	72.5	34.7	30.0	26.4	40.5	116.8	122.9	85.5	85.5	152.3
			Cy	184.2	147.9	72.6	35.0	31.3	20.5	16.9	38.0	–	35.9	151.5	129.2
			SF	–	–	–	–	16.8	23.5	41.3	49.3	27.8	–	–	–
			Ty	–	–	–	–	–	–	67.5	513.5	1195.7	–	–	–
Rishiri[b]	2000-2007	nss-SO_4^{2-}	Total	31.5	67.3	55.9	40.0	44.6	33.8	34.6	14.4	6.1	13.0	30.6	32.9
		Na^+	Total	238.1	394.4	194.4	104.6	28.5	75.5	18.6	22.3	39.2	241.2	515.4	425.5
Hedo[b]	2000-2007	nss-SO_4^{2-}	Total	17.8	28.9	37.1	19.8	14.0	10.8	12.2	4.6	3.3	15.3	12.4	18.1
		Na^+	Total	352.1	233.9	219.9	67.8	64.5	71.7	423.4	2218.3	297.1	347.3	163.6	329.5

[a]This study, [b]Calculated from the data available from EANET homepage (Network Center for EANET, 2014).
–: Not available (N.A.).

(Table 1). The concentrations of nss-SO_4^{2-}, NO_3^- and NH_4^+ were the highest in February and the lowest in June. The concentration of nss-Ca^{2+} was the highest in February and the lowest in August. The concentrations of Na^+ and Cl^- were the highest in January, the lowest in June, and relatively high in August and September.

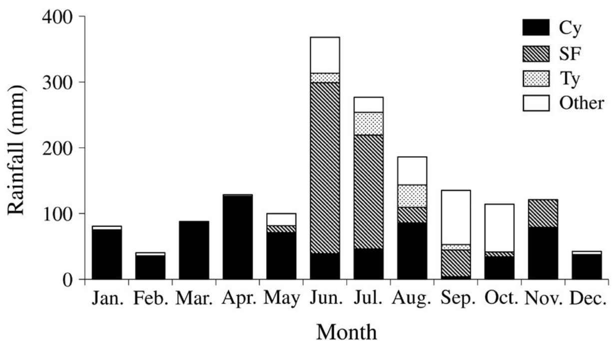

Fig. 2. Monthly mean rainfall by Cy, SF, Ty and Other over the eight years.

These trends were closely associated with the dominant rain types, which will be discussed in details.

In November-May, the monthly mean concentrations of terrigenous ions, i.e., nss-SO_4^{2-}, NO_3^-, NH_4^+ and nss-Ca^{2+}, in total rainwater were 25.1-57.8, 9.9-25.0, 11.3-31.4 and 5.5-18.7 μeq L^{-1}, respectively. They were relatively higher than the concentrations in other months. The concentrations of marine-origin ions, Na^+ and Cl^-, were 24.2-154.9 and 30.0-178.5 μeq L^{-1}. The concentrations of all these ions were higher in November-May than the concentrations in other months, except for in August and September, and were actually very close to the concentrations in Cy rainwater. Monthly mean rainfall by Cy was more than 30 mm, which occupied 85% of total rainfall in November-May (Fig. 2). Cy rain contributed 84-92% to the total fluxes of the six ions in these months (Fig. 3). Therefore, the ions in the total rainwater in November-May were dominated by Cy rain.

In June and July, the monthly mean concentrations of nss-SO_4^{2-}, NO_3^-, NH_4^+, nss-Ca^{2+}, Na^+ and Cl^- in

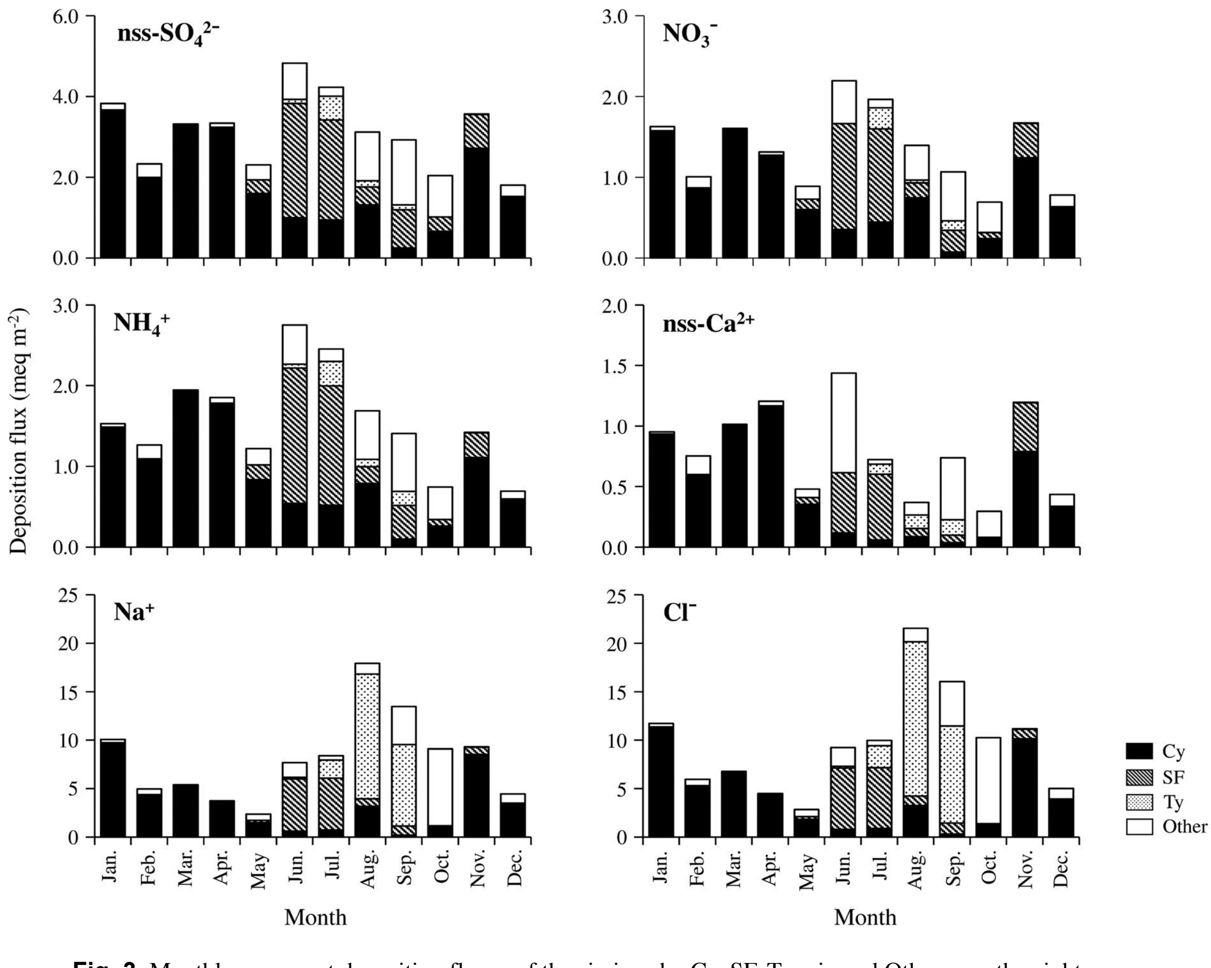

Fig. 3. Monthly mean wet deposition fluxes of the six ions by Cy, SF, Ty rain and Other over the eight years.

total rainwater were 14.4-20.7, 7.2-9.5, 7.7-12.9, 4.1-6.8, 21.7-33.6 and 26.4-40.5 μeq L^{-1}, respectively. The concentrations were relatively lower than those in November-May (Table 1). This result was mainly due to the large rainfall by SF in June and July. The rainfall by SF was 260 mm in June and 173 mm in July. Each of these constituted more than 60% of the total rainfall in the months (Fig. 2). For this reason, SF rain contributed 35-75% of the fluxes of the six ions in the two months (Fig. 3). In addition, the concentrations of the six ions in the Cy rainwater in the two months were lower than those in November-May, and were relatively close to the concentrations in the SF rainwater (Table 1).

In August and September, Ty rain substantially contributed to marine-origin ions Na^+ and Cl^- in total rainwater. It was estimated that more than 60% in the fluxes of the two ions in the two months were from Ty rain (Fig. 3). This was due to the extreme abundance of the two ions in Ty rainwater. It is found that the concentrations of the two ions in Ty rainwater were 10-43 times the concentrations in Cy and SF rainwater (Table 1). The concentrations were large enough to significantly influence the rainwater chemistry in the two months although the rainfall caused by Ty was 8-34 mm $month^{-1}$ and occupied only 6-18% of the total rainfall (Fig. 2). Ty rain also caused rainfall of 35 mm in July, which occupying 13% of the total rainfall in the month. However, the relative contribution of Ty rain to the fluxes of Na^+ and Cl^- was low in July, because of the dominant contribution by SF rain. In October, the rainfall and the ions were dominated by the rain type of Other. These cases were frequently associated with multiple rain types, because the month is a transition period from fall to winter.

4. DISCUSSION

The above results indicate the remarkable seasonal characteristics of rainwater chemistry due to changes of the predominant rain types in different months or seasons. This is naturally acceptable because of the distinctive nature of rainwater chemistry in different rain types (Toyonaga and Zhang, 2016). Besides the changes of the predominant rain types and the respective frequency of different type, the variation of ion concentrations in Cy rainwater was also an important factor influencing the monthly variation of rainwater chemistry in the total rainwater. Terrigenous ions SO_4^{2-}, NO_3^-, NH_4^+ and Ca^{2+} in Cy rainwater were closely associated with rainfall and emission amount of precursors (Vet *et al.*, 2014; EANET, 2006).

To further evaluate the effect of the rainfall and the precursor emission on the ion concentrations, a simple regression analysis was conducted for the monthly rainfall and the fluxes by Cy in four seasons, i.e. winter (Dec.-Feb.), spring (Mar.-May), summer (Jun.-Aug.), fall (Sep.-Nov.). The regression equation is

$$Y = aX + b$$

where Y is monthly wet deposition fluxes and X is the total rainfall in a month during a season, a is the slope, and b is the intercept.

There were strong correlations ($R > 0.6$) in all seasons between the monthly fluxes of anthropogenic ions (nss-SO_4^{2-}, NO_3^- and NH_4^+) and rainfall by Cy, indicating the rainfall is a factor influencing the fluxes of the ions (Table 2). Slopes, a, in the case of nss-SO_4^{2-} and NO_3^- were the highest in winter and the lowest in summer. The slopes corresponded to the loading of ions per unit rainfall, which could be interpreted as the strength of the emissions influence. Cy rain was mainly affected by continental air masses. The seasonal trend of the slopes should be corresponded to precursor gases and particulate matter emissions at Asian continent (Seto and Hara, 2006; Fujita *et al.*, 2000). The seasonal characteristics of nss-SO_4^{2-} and NO_3^- variations were consistent with the concentration of

Table 2. Coefficients of the regression analysis between the monthly wet deposition fluxes and rainfall caused by Cy.

	Slope	Intercept	Correlation coefficients
	a	b	R
nss-SO_4^{2-}			
Winter	0.0324	0.8636	0.78
Spring	0.0218	0.6412	0.73
Summer	0.0167	0.4571	0.89
Fall	0.0223	0.5062	0.74
NO_3^-			
Winter	0.0124	0.4521	0.73
Spring	0.0104	0.1676	0.72
Summer	0.0080	0.0717	0.96
Fall	0.0082	0.3008	0.62
NH_4^+			
Winter	0.0116	0.5295	0.65
Spring	0.0126	0.3218	0.71
Summer	0.0094	0.2148	0.84
Fall	0.0089	0.2184	0.67
nss-Ca^{2+}			
Winter	0.0043	0.4510	0.40
Spring	0.0052	0.3551	0.44
Summer	0.0008	0.1038	0.37
Fall	0.0029	0.2790	0.30

Regression equation: $Y = aX + b$.
Y: monthly deposition fluxes (meq m^{-2}), X: monthly rainfall (mm).

precursor gases (SO_2 and NO_X) and particulate SO_4^{2-} and NO_3^- in the air in China, which is high in winter and low in summer (Pan *et al*., 2013, 2012; Meng *et al*., 2009).

The slope in the case of NH_4^+ was the highest in spring and the lowest in fall. The precursor gas of NH_3 in China is high in summer and low in fall or winter (Meng *et al*., 2010). However, particulate NH_4^+ is higher in winter or spring than in other seasons in China (Meng *et al*., 2014; Cao *et al*., 2009). Therefore, NH_4^+ in the Cy rainwater might be more closely associated with particulate NH_4^+ from China than the precursor gas NH_3.

There was no clear correlation between the flux of nss-Ca^{2+} and the rainfall in any season. This was mainly attributed to the fact that the rainfall of Cy rain with extreme high concentration of nss-Ca^{2+} was usually small. It is likely such Cy rain are the results of the influence by Asian dust (Toyonaga and Zhang, 2016; Seto *et al*., 2007). Asian dust-related rain is frequently observed in spring (Kawamura and Hara, 2006; Seto *et al*., 2004). We found that the flux of nss-Ca^{2+} by Cy rain in spring was higher than in other seasons (Fig. 3), which is consistent with the expectation from results of the previous studies.

The annual fluxes by Cy rain showed a year-by-year variation which is characterized by the influence of the anthropogenic emissions from the Asian continent because this type of rain is more significantly influenced by continental air than other types of rain (Toyonaga and Zhang, 2016). These results indicate that, as expected, the emission trend of anthropogenic pollutants in China is also an important factor to influence the seasonal and annual variation of the chemistry in Cy rainwater.

The seasonal variation of rainwater chemistry at other sites could also be interpreted as the consequence of seasonal changes of dominant rain types. For example, the variation of monthly mean concentration of Na^+ and nss-SO_4^{2-} in the rainwater collected at Rishiri, the island located in the northern part of the Sea of Japan, was similar to the variation in the Cy rainwater at the site of this study (Table 1). This is consistent with the fact that the major type of rain at Rishiri is Cy rain, and the fact that Rishiri is rarely affected by SF and Ty rain (Berry *et al*., 2011; Kimura, 1970).

In contrast, the variation of monthly mean concentrations of the ions at Hedo, the north cape of Okinawa located approximately 1,300 km to the south from the site of this study, was similar to the variation in the total rainwater at the site of this study (Table 1). Moreover, in July-September, the concentration of Na^+ was much larger than that at the site of this study (Table 1). This was due to the more frequent typhoon at Okinawa than at the site of this study, besides Cy and SF rain (Sakihama and Tokuyama, 2005).

Therefore, quantifying ions in rainwater according to rain types is important for a meaningful inter-comparison of the seasonal variations at different geographic regions, and is essential for accurate understandings of the correlation between rain type and rainwater chemistry.

5. SUMMARY

Monthly variation of rainwater chemistry of four terrigenous-origin ions (nss-SO_4^{2-}, NO_3^-, NH_4^+ and nss-Ca^{2+}) and two marine-origin ions (Na^+ and Cl^-) at a coastal site in southwestern Japan was investigated with the records of 348 rain episodes over eight years according to the rain types of Cy, SF and Ty. Monthly mean concentrations of ions in November-May, when the rain was mainly caused by Cy, were higher than the concentrations in June and July, when the rain was mainly caused by SF. In August and September, the concentrations of marine-origin ions were relatively high due to the contribution by Ty rain. Further analysis suggested that the monthly variation of the chemistry in Cy rainwater was associated with the variation of the emission of air pollutants in China, in addition to the variation of rainfall. Comparison analysis with other sites suggested that the difference of seasonal variations of ion concentrations at the present site from those at Hedo of Okinawa and Rishiri of the Sea of Japan were due to the different seasonal dominance of the rain types depending on the regions. These results indicate that the monthly variation of the rainwater chemistry was closely associated with the variation of contribution by the rain types.

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