

Chemical characteristics of cloudwater over Maoer Mountain district

Yao Rongkui, Bai Yuhua, Chen Shufen, Shao Min,
Tang Xiaoyan

Environmental Chemistry Division, Department of Technical
Physics, Peking University, Beijing 100871, China

Abstract Samples of cloudwater, rainwater and ambient aerosol were collected over Maoer Mountain in the northeast of Guangxi Province in March 1988. The pH value of cloudwater ranged from 3.37 to 6.20 with a mean value of 4.34. SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{2+} and H^+ were the principal ionic species of cloudwater. The advance of cold fronts into Maoer Mountain appeared to lead to higher major ionic concentration and acidity than that of warm fronts. The relative acidity and concentration of NO_3^- of cloudwater were much greater than that of aerosol samples. With the exception of NO_3^- and H^+ , most of the concentration of SO_4^{2-} and NH_4^+ in cloudwater came from the nucleation scavenging of aerosol. Gaseous nitric acid and organic acid from local may be one of the important source of cloudwater acidity. Compared with other sites, Maoer Mountain can be as a clean contrast station for studying acidic precipitation in Guangdong and Guangxi provinces.

Keywords: acid precipitation; cloudwater; rainwater; aerosol; clean site.

1 Introduction

A growing interest can be seen in the research of acid precipitation in China since 1980s. More and more studies have shown that there is serious acid rain pollution in the region of southern Yangtze River, especially in southern and western China (Zhou, 1988). Most previous field work in China has based on the sampling and analyzing of rainwater, only a few reports are related to the cloudwater (Deng, 1987; Zhang, 1988, Ding, 1989). Some researchers have suggested that clouds play an important role in the transformation and deposition of acidic materials. So it is an imminent work for us to collect cloudwater over mountainous districts for a better understanding of the formation and source of acid precipitation in southern and western China.

This paper presents the results that we collected the data of cloudwater and rainwater from remote Maoer Mountain in Guangxi Province during March 14-24, 1988, which may be used as the data of clean site to compare with the urban areas

such as Guilin, Liuzhou and other mountainous districts.

2 Sampling site

Maoer Mountain is located at the northeast of Guangxi Province, the north end of Hunan-Guangxi corridor and about 65 km to the north of Guilin City. Its elevation of 2141 m and is known as "the first mountain in south China" (Fig. 1). Several factors were considered in selecting this site: first, the mountain is high enough; second, there are no important pollutant source; third, it is one of the very few locations in both Guangdong and Guangxi provinces which provides open exposure to impacting clouds; and fourth, it is located not very far from Guilin City. It is very convenient to be a contrast point for Guilin to study the action of below-cloud washout, especially during the northern air flow coming.

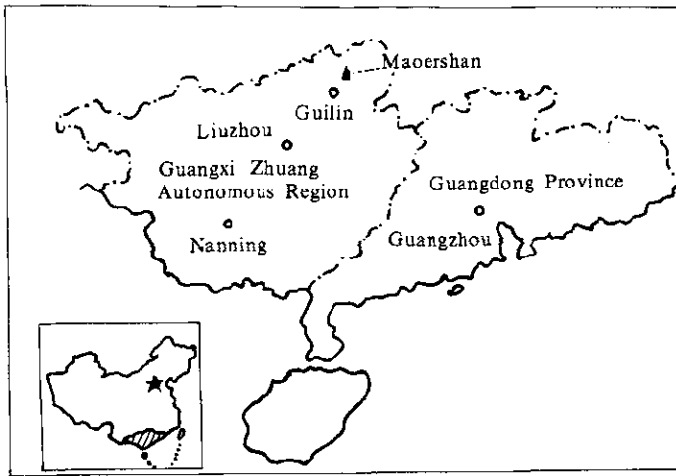


Fig. 1 Sketch map of Maoer Mountain site used for cloudwater sampling
The inset shows the location of the mountain relative to Guilin City and the rest of Guangxi and Guangdong provinces

3 Measurement techniques

Cloudwater samples were collected with the full-plastic cloudwater collector produced in the United States as depicted in Fig. 2. rainwater were collected as

whole and segmental samples by using plastic tank with 27 cm diameter and polyethylene segmental collector respectively during rain events. After collection, the volume of liquid samples were scaled using cylinder and measured the pH value as well as conductivity immediately and then maintained at a temperature of 4 °C after filtration by micropore filter (0.45 μm pore size).

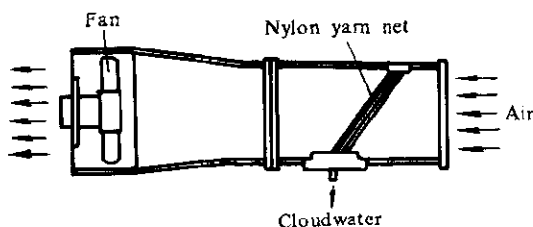


Fig. 2 The full-plastic collector of cloud-fog water produced in the United States

After filtering with 0.2 μm filter, the eluted solution were diluted to 25.00 ml in volumetric flask. This is just the aerosol solution which was ready to be analyzed as did as the cloudwater.

The pH measurement was made with a conventional electrode and Orion research pH meter (model 211). Conductivity was measured with model DDS-11 conductivity meter made in Second Analytical Instrument Plant in Shanghai.

The major ions: SO_4^{2-} , NO_3^- , NH_4^+ , Na^+ and K^+ as well as organic acid (formic acid and acetic acid) were measured in our laboratory using Dionex 2010 Ion-Chromatograph, Ca^{2+} and Mg^{2+} concentration were determined by Hitachi 180-8 Atomic Absorption Spectrometer.

4 Results

A total 24 cloudwater samples were collected during 8 cloud impaction events. Some of sampling events were associated with cold fronts approaching from the north or northwest. Twenty-five rainwater samples were collected during 9 rain events. Three aerosol samples were also collected in only three sunny days.

The results of the analysis of cloudwater and aerosol composition are given in Table 1 and Table 2, respectively. It can be seen that the major cations of cloudwater over Maoer Mountain district were Ca^{2+} , NH_4^+ and H^+ , the major anions were

Aerosol samples were collected on a Pallflex Qao 2500 quartz filter using KB-120 model air sampler pump with a flow rate of 100 L/min for 2–4 h. All samples were stored at ambient temperature.

The aerosol samples on filter were eluted with distilled-deionized water by about 20 minutes in a ultrasonic cleaner (model HQQ 005 produced in Wuxi Ultrasonic Electronic Equipment Plant). After

SO_4^{2-} and NO_3^- . The concentrations of NH_4^+ , Ca^{2+} and SO_4^{2-} and NO_3^- in cloudwater accounted for around 43.7%, 24.6% and 56.1%, 29.3% of the concentration of total cations and anions, respectively. SO_4^{2-} and NH_4^+ were the principal ions.

Table 1 The chemical composition of cloudwater over Maoer Mountain, $\mu\text{eq/L}$

Event	Date	Time	Air flow	Formic acid	Acetic acid	Sum (+)	Sum (-)
1	3/15	1620-1800	S N			590.1	427.8
		1840-1945	S N	78.2	27.3	510.2	351.8
2	3/16	1610-2030	N	92.8	43.7	1551	1621
3	3/17	1050-1240		38.0	21.8	457.2	359.3
		1245-1430	N	26.1	12.8	341.5	178.8
		1440-1640		18.2	10.9	160.2	75.6
		1640-1990		14.2	10.9	126	56.8
4	3/18	0700-1100	N(NE)			191.4	119.6
5	3/20	0000-0800				642.1	413.9
		0820-1045	S	26.1	20.0	214.8	60.2
		1045-1245		11.2	18.2	84.3	27.4
		1245-1510		21.1	18.2	216.9	64.2
		1510-1730				110.8	44.8
		1730-2100				79.8	36.1
6	3/21	0000-0715	S			111.8	33.8
		0905-1940	SW			134.9	73.1
7	21-22	1940-0800				133	84.6
8	3/23	0920-1025		40.9	21.8	462.7	347.9
		1030-1220		32.6	20.0	304.6	163.3
		1220-1410	N	17.2	14.6	152.0	57.4
		1410-1530		12.2	10.9	114.3	26.9
		1530-1830		21.1	10.9	123.3	50.8
	23-24	1830-0840		13.2	5.5	89.7	61.8
9	3/24	0840-1630	N			189.9	82.6

Table 1 (continued)

pH	H ⁺	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Ca ²⁺	Mg ²⁺
4.23	58.9	18.8	50.4	116.7	241.9	49.2	286.6	33	135.7	26.7
4.07	93.3	6.3	43.6	91.7	210.2	38.7	216.2	25.1	112.3	24.6
3.37	426.36	36	79.8	497.2	1008	51.3	639.7	73.2	290.4	70.2
4.00	100	12.2	22.7	106.4	218	16.1	214.5	25.1	81.3	20.2
4.06	87.1	0.053	12.4	51.8	114.6	5.6	159.6	20.2	58.9	10.1
4.27	53.7	0.0053	6.0	22.0	47.6	5.2	58.8	8.7	28.4	5.43
4.79	16.2	0.05	8.1	14.2	34.4	4.4	66.0	5.6	29.4	4.36
4.36	43.6	7.2	15	27	70.4	17.4	51.6	16.6	51.9	10.3
5.06	8.71	10.6	33.4	155.1	214.8	32.6	262.2	49.6	251.5	37.5
5.42	3.80	0.05	10.6	17.5	32.0	28.3	84.8	14.8	74.9	8.2
6.20	0.63	0.05	3.2	7.6	16.6	2.2	38.8	4.1	35.9	2.7
5.08	7.41	3.96	11.2	17.7	31.3	14.8	92.0	11.3	48.4	4.3
5.47	3.4	9.4	8.7	9.6	17.1	8.3	38.8	10.2	45.9	4.2
5.40	3.3	9.0	5.9	8.8	12.4	0.9	20.0	3.8	47.4	4.4
5.62	2.4	5.4	6.8	10.2	18.1	5.6	51.6	8.7	39.9	3.6
6.2	0.6	21.6	25.5	8.4	17.6	30	14.4	14.3	66.4	9.2
5.82	1.5	34	14.4	16.4	19.8	8.7	66.0	9.2	41.4	6.2
4.20	63.1	23.2	19.9	111.3	193.5	10.0	124.8	28.6	40.9	28.6
4.30	50.1	14.1	10.6	47.0	91.6	3.48	44.4	6.3	32.6	6.3
5.04	9.1	7.4	10.2	16.6	23.2	10.0	40.4	5.4	17.2	5.4
4.73	18.6	0.43	11.3	6.7	8.5	13.5	24.5	2.8	12.2	2.8
4.73	18.6	10.0	6.9	14.4	19.5	9.1	23.0	2.8	21.1	2.8
5.26	5.5	0.05	14.9	12.0	34.8	11.3	21.5	3.7	13.2	3.7
4.62	24.0	3.3	9.0	30.9	39.4	20.9	103.7	15.4	21.5	4.4

The pH values of cloudwater ranged from 3.37 to 6.20 with a mean value of 4.34 and a median of 4.74. The acidic frequency of cloud water is about 84% (defined as the percentage of samples with pH < 5.60 divided by the total samples). The largest frequency of pH occurred in the range of 4.0 to 4.5, which accounted for around 34% of the total samples. Fig. 3 shows the frequency distribution of pH of cloudwater. It can be seen from Fig. 3a that most of the cloudwater over Maoer Mountain district were already acidified.

Table 2 The ions composition of aerosol in different sites, neq/m³

Name of mountain	Date	Sampling time	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NH ₄ ⁺	K ⁺	Ca ²⁺	Mg ²⁺	H ⁺	Total
Maoer shan	3/16	1220-1610	22	8.09	13.5	119.8	269.8	22.2	57.1	14.3	34.7	
	3/19	920-1730	9.73	-	11.55	73.4	106.5	12.9	62.1	6.76	3.51	
	3/22	1300-1930	4.27	2.96	4.4	95.7	121.2	15.4	30.6	8.63	8.59	
Maoershan	1988.3		10.5	3.4	9.5	87.5	146	15.7	50.3	9.0		351.1
Hengshan	1988.3		1.1	21.3	69.9	163.4	141.7	34.6	85.6	5.7		558.1
Shizishan	1988.3		1.2	9.9	19.1	162.4	122.7	26.9	91.4	32		486.1

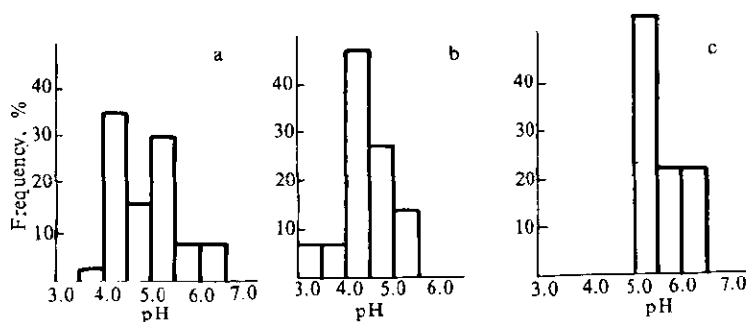


Fig. 3 Acidic frequency distribution of cloudwater

- a. total 24 samples; b. the samples collected during cold fronts;
c. the samples collected during warm fronts

The concentration of chemical composition and acidity of cloudwater were obviously different in different air flow. Fig. 4 shows the diurnal mean concentrations of major cations and anions of cloudwater and the wind direction during sampling in 1988. March 16-19 and 23-24 were controlled by north air flow, March 20-22 were did by the south air flow. Fifteen samples collected during north air flow were all acidic, the pH value of them ranged from 3.37 to 5.26 with a mean value of 4.15, the acidic frequency was about 100%; for the other 9 samples collected during south air flow, the pH value ranged from 5.06 to 6.20 with a mean value of 5.45 and the acidic frequency was only 56% (Fig. 3b and c). On the other word, the concentration of H⁺ during north air flow was 20 times as high as the [H⁺] during

south air flow. The variation of concentration of SO_4^{2-} , NO_3^- and NH_4^+ , except Ca^{2+} , have similar tendency as H^+ . It can be seen from Fig. 4 that there were consequently valley points on the curves on March 21, which was exactly during the south air flow.

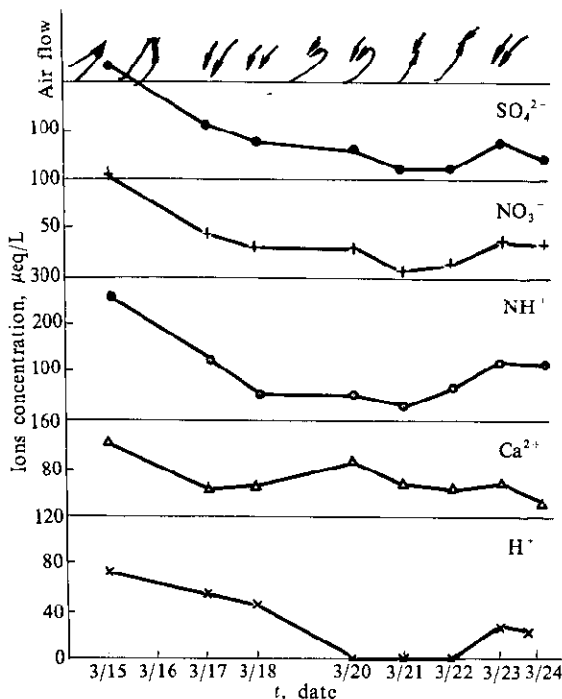


Fig. 4 Variation of diurnal mean concentration of major ions of cloudwater and the air flow direction each day

Low molecular weight organic acid (formic acid and acetic acid) appear to be important contributors to the overall chemical composition of cloudwater over Maer Mountain. The research has shown that the dissociated parts of the organic acid in cloudwater accounted for around 5.5% and 12.6% of the total ion concentration and the total anion concentration respectively. The ratio of sum (-)/sum(+) % for the samples will be enhanced from 68.1% to 78.1%, if organic acid were considered. It suggested that if the constituent of organic acid were included, the ion balance of the samples were getting much better (Fig. 5).

The pH value and chemical composition of cloudwater has been found to be a function of time (Seinfeld, 1986). Fig. 6 shows the variability of it on March 23,

1988. It can be seen that at the beginning of the cloud impaction event, the concentrations of its major chemical composition, typically sulfate, nitrate, ammonium, calcium and hydrogen ion, were much higher than the latter. Fig. 6 has described the overall process of formation of cloudwater. The similar situations can be found for other cloudwater events during sampling (Table 1).

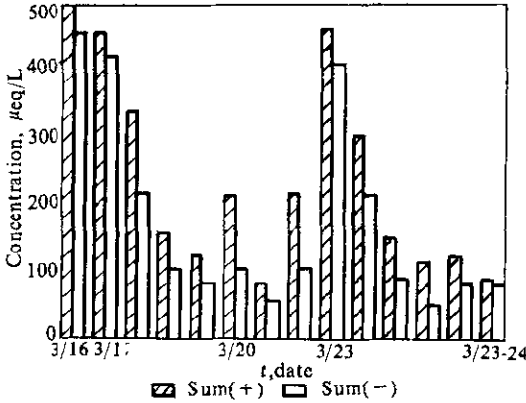


Fig. 5 The comparison of ion balance of cloudwater
 Sum(+): total organic acid concentrations
 Sum(-): total anion concentrations including the dissociated parts of the organic acid

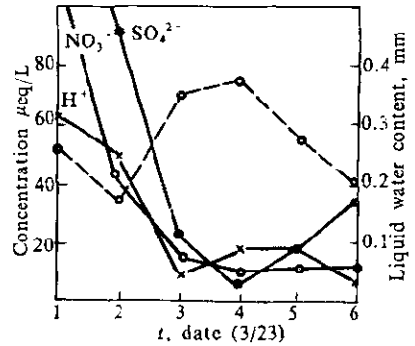


Fig. 6 Diurnal variation of sulfate, nitrate and H⁺ concentration relative to the liquid content of cloud water

Dotted line: liquid water content
 1. 09:20 - 10:25 4. 14:10 - 15:30
 2. 10:30 - 12:20 5. 15:30 - 18:30
 3. 12:20 - 14:10 6. 18:30 - 08:40

5 Discussion

5.1 The sources of major chemical composition and acidity in cloudwater over Maoer Mountain district

The major chemical composition of cloudwater over Maoer Mountain district, as shown in Table 1, were SO₄²⁻, NO₃⁻, NH₄⁺ and Ca²⁺. It is necessary to get a clear understanding for the source of those ions in cloudwater.

It has been known that sulfate and nitrate can be incorporated into the precipitation by three major pathways: nucleation scavenging of sulfate and nitrate-containing aerosol particles during cloud formation; aqueous-phase formation from absorbed SO₂ and gaseous nitric acid in cloud droplets and below-cloud scavenging of aerosol sulfate and nitrate and gaseous SO₂ by rain drops (Seinfeld, 1986). It is interesting for us to know which pathway was the prime during the formation of cloudwater

over Maoer Mountain district. Because the cloud are formed by condensation of water vapor on aerosol particles and much of the aerosol mass is incorporated into cloud droplets (Daum, 1984). The key to the question is to compare the chemical composition and acidity between cloudwater and ambient aerosol. If both composition are the same, then aerosol scavenging cannot be ruled out as a prime mode of incorporation. If, on the other hand, the composition differ obviously, a process other than direct aerosol scavenging is indicated (Zhang, 1988).

During sampling, three aerosol samples were got, the compositions of cloudwater compared with the aerosol samples are to use the data of diurnal mean concentration of same days. Because the absolute concentrations both the aerosol and cloudwater samples were quite variable, comparisons are made in terms of the concentration ratio of main constitutes (Table 3). Because the resultant of $[K^+]/[Mg^{2+}]$ ratio was not markedly different between aerosol and cloudwater, we chose it as the starting point for addressing this question. Which suggested that most of the concentration of Mg^{2+} and K^+ in cloudwater were perhaps derived by dissolution of preclude aerosol. So it is convenient and confident to use each one of them to be a standard to measure other concentration ratio such as $[NO_3^-]/[K^+]$, $[SO_4^{2-}]/[K^+]$ and so on. The ratio values of the major chemical compositions are shown in Table 3. It can be seen from these data: there are 95% confidence to say that the ratio of $[SO_4^{2-}]/[K^+]$ and $[NH_4^+]/[K^+]$ belong to the same population. In other word, most of sulfate and ammonium in cloudwater came from aerosol. For the ratio of $[NO_3^-]/[K^+]$, however, it is clear from these data that the compositions of the two kind of samples are obvious different: the ratios for aerosol are substantially less than 1 (mean 0.6), whereas the ratios for the cloudwater samples are always greater than 1 (mean 2.45). Similar

Table 3 Ratio of the major ions in cloudwater and aerosol

Date	K^+/mg^{2+}		NH_4^+/K^+		NO_3^-/K^+		SO_4^{2-}/K^+		H^+/NH_4^+	
	Aerosol	Cloud	Aerosol	Cloud	Aerosol	Cloud	Aerosol	Cloud	Aerosol	Cloud
3/16-3/17	1.55	1.48	12.2	8.4	0.61	3.26	5.4	6.96	0.13	0.67
3/19-3/20	1.91	1.53	8.3	5.7	0.9	2.3	5.1	3.45	0.03	0.05
3/22-3/23	1.78	1.49	7.9	6.8	0.29	1.78	6.2	4.1	0.07	0.265
Avg.	1.75	1.35	9.5	7.0	0.6	2.45	5.57	4.84	0.078	0.33
STD	0.18	0.27	2.38	1.36	0.3	0.75	0.57	1.87	0.049	0.31

conclusion can be derived by comparing the ratio of $[H^+]/[NH_4^+]$ (Fig. 7), it is about 4 times for the cloudwater as high as that in aerosol samples. These results indicated that the enhanced acidity of cloudwater over that of ambient aerosol is attributed to the absorption of gaseous nitric acid by cloud droplets. The NO_x or gaseous nitric acid may be one of the important sources of acidity and NO_3^- of cloudwater over Maoer Mountain district.

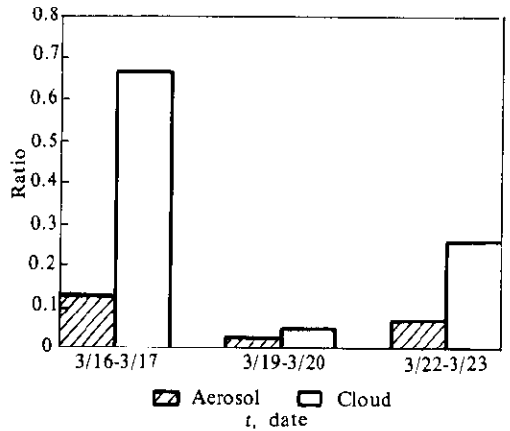


Fig. 7 Ratio of $[H^+]/[NH_4^+]$ in aerosol and cloudwater

In order to understand better the relation between aerosol and cloudwater, the method of enrichment factors (Lautzy, 1979) was introduced by using the equation:

$$EF = \frac{(\text{conc. of ion}/\text{conc. } K^+)_{\text{cloudwater}}}{(\text{conc. of ion}/\text{conc. } K^+)_{\text{aerosol}}}$$

In Table 4 the EF value of major composition of cloudwater are listed. K^+ ion is used for the normalization. It can be found that the EF value of SO_4^{2-} , NH_4^+ and Mg^{2+} are close to one (in the range of 1.00 ± 0.34), EF value of NO_3^- range from 2.55 to 5.86 with a mean value of 4.58. These results are consistent with above conclusion: the most of SO_4^{2-} , NH_4^+ and Mg^{2+} ions in cloudwater came from aerosol, the NO_3^- in cloudwater should be have another source with the exception of aerosol.

Moreover, the organic acid emitted by natural plants may be another important source of acidity of cloudwater over Maoer Mountain. If the organic acids of incorporation into cloudwater were all in the forms of acid rather than the others, the most contribution of it to the acidity of cloudwater was about 57%. Therefore the organic acids should not be ignored when investigating the source of the acidity of cloudwater over Maoer Mountain district.

Table 4 Enrichment factors for some ions in cloudwater over Maoer Mountain district

Ion	Date	Mg ²⁺	SO ₄ ²⁻	NH ₄ ⁺	NO ₃ ⁻
EF	3/16	1.05	1.29	0.69	5.34
	3/19	1.25	0.68	0.69	2.55
	3/22	1.19	0.66	0.86	5.86
AVG		1.16	0.88	0.75	4.58
STD		0.08	0.29	0.08	1.45

5.2 Maoer Mountain is a natural protective district of China, it is a rather clean site in comparing the acid precipitation with other mountains and cities in Guangdong and Guangxi provinces. Table 5 shows the analytical results of cloudwater at different sites. It was found that not only the major chemical composition but also the ratio of SO₄²⁻/NO₃⁻ of cloudwater in Maoer Mountain were lower than that over other mountains. The ratio of SO₄²⁻/NO₃⁻ was lower than 2 over Maoer Mountain and at other sites were usually higher than 4. This may be due to the fact that the concentration of gas phase SO₂ in ambient air of Maoer Mountain was very low, usually, it was lower than 0.5 µg/m³, which is the limit of detection of colorimetric method. The concentration of nitrate for Maoer Mountain and most other sites, however, was almost at the same level, ranging from 34–90 µeq/L.

Table 5 Chemical composition of cloudwater in different sites, µeq/L

Name of mountain	Altitude, m	Date	NO ₃ ⁻	H ⁺	NH ₄ ⁺	Ca ²⁺	NO ₃ ⁻	SO ₄ ²⁻	SO ₄ ²⁻ /NO ₃ ⁻	Sum
Maoershan	2141	1988.3	24	45.8	129	72.5	59	114	1.9	496.6
Shizishan	1050	1988.3	24	42.4	199.7	61.5	49.4	228.9	4.6	668.4
Fanlingshan		1986.5–6	19	28.5	234	171	34.4	256	7.4	
Lushan		1987.5		4.4	46.5	199.6	33.8	159	4.7	508.6
Hengshan	1200	1988.3	31	83.4	333.2	101.1	89.2	362.9	4.1	1115.3
Baiyushan	380	1988.3		22.9	2642	1302	707	3776	5.3	11515

On the other hand, total suspended particulate concentration ranged from 0.027–0.067 mg/m³ and airborne particles ($D < 10\mu\text{m}$) were just in the range of 0.022–0.052 mg/m³, which were much lower than the primary air quality standard of China. The ion concentrations of aerosol over Maoer Mountain were also lower than that at other sites (Table 2).

The rainwater over Maoer Mountain was also rather clean. Fig. 8 shows the situation of the major chemical compositions in segmental sample of rainwater over there on March 17, 1988. It is seen that the concentration of major chemical composition do not decrease obviously with increase in number of grade. This results have implied that the rainwater was clean, because the action of washout below-cloud there was not important.

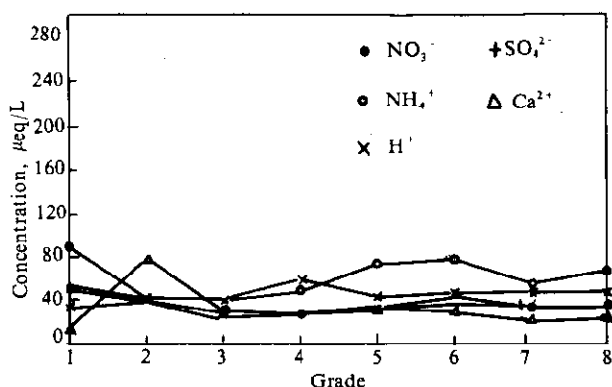


Fig. 8 The variation of major ion concentrations of rainwater in segmental samples over Maoer Mountain

In a few words, Maoer Mountain is clean enough as a clean contrast station for studying the acidic precipitation in Guangdong and Guangxi provinces.

6 Summary and conclusion

Cloudwater was sampled during the spring of 1988 in Maoer Mountain in the northeast of Guangxi Province. The major chemical compositions of cloudwater were SO_4^{2-} , NO_3^- and NH_4^+ , Ca^{2+} , which accounted for around 56.1%, 29.3%, and 43.7%, 24.6% of total anions and cations respectively. The pH values ranged from 3.37 to 6.20 with a mean value of 4.34. Concentration of the major cloudwater ions were found to vary within a large range, some of these difference were found to be associated with the air flow direction: the samples collected during warm fronts are generally much less acidity and ionic concentrations than that during cold fronts.

The nucleation scavenging in rainout of sulfate-containing aerosol was the most important source for sulfate and ammonium of cloudwater, by comparing the concentration ratio of the major ions in aerosol and cloudwater collected on the

correspondent time. The two important exceptions to this were acidity and nitrate, cloudwater was found to be significantly more acidic and had more NO_3^- than aerosol analyzing in terms of the ratio $[\text{H}^+] / [\text{NH}_4^+]$, $[\text{NO}_3^-] / [\text{K}^+]$ and value of *EF*, this excess acidity is consistent with the hypothesis of absorption of gaseous nitric acid by cloudwater and the contribution of organic acid emitted from natural plants.

Compared the concentration of total ions and major chemical compositions of cloudwater as well as aerosol in Maoer Mountain with other sites (mountains and cities), it can be found that Maoer Mountain is a much clean site for the acid precipitation. As a clean contrast station for studying acid rain in Guangdong and Guangxi provinces of China.

Acknowledgements— We thank Prof. Li Jinlong of our division for valuable discussion, thank him and Dr. Zhang Yanhang for the helping us to treat the data by computer and Assoc. Prof. Li Xin to work a lot in analyzing the samples. We thank Prof. Mao Jietai *et al.* of the department of Geophysics for the closely cooperation in Maoer Mountain, thank Environmental Monitoring Station of Guilin City and Maoer Mountain Microwave Station of Guangxi Province for their supporting and the closely cooperation.

References

- Deng Jiaquan, Mo Tianlin. *Environmental Science*, 1987; 6(1)
- Ding Guoan, Ji Xiangming, Fang Qiumei, Fu Jimeng, Fan Wu, Yu Xianren. Characteristics of cloud-fog water in Lushan Mountain in spring, *Proceedings of the International Conference on Global and Regional Environmental Atmospheric Chemistry*. Beijing, China, 1989
- Daum PH, Schwartz SE, Newman L. *Journal of Geophysical Research*, 1984; 89(D1):1447
- Lautzy RT, Maikenzie FT. *Geochim Cosmochim Acta*. 1979; 43:511
- Seinfeld JH. *Atmospheric chemistry and physics of air pollution*. New York: Willey, 1986
- Zhang Tao, Zeng Huafan, Ji Xiangxiong. *Environmental Chemistry*, 1988; 7(1)
- Zhao Dianwu, Xiong Jiling, Walter HC. *Atmospheric Environment*, 1988; 22(2):349

(Received October 8, 1991)