Rapid changes of precipitation pH in Qinghai Province, the northeastern Tibetan Plateau

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Abstract
Acid rain monitoring programmes have been carried out in two industrial cities of China's Qinghai Province in the some periods of the1980s and 1990s. The results indicate that precipitation in this area originally is alkaline. Compared with the monitoring from the late 1980s, the pH values in middle 1990s have declined greatly from around 8 of the 1980s to below 7. Such rapid change was caused by fast growth of local industrial plants that released a large amount of pollutants. However, tough control on pollutant emission has led to the pH value back up above 7 in the late 1990s. The pH and rainfall chemical analyses indicate that alkaline rain in this region is caused by airborne dusts, which originate from local alkaline soils. With decrease of pH value, the total ionic concentration of rainwater is increased.

Keywords: precipitation pH, precipitation chemistry, alkaline rain, air pollutant, air dust

Introduction

Qinghai Province is located on the northeastern part of the Tibetan Plateau. It has a total area of over 721,000 km^2, stretching east-west for about 1200 km and spanning north-south a width of nearly 800 km. The province, extensive yet sparsely populated, has a total population about 4 millions. It has an average elevation of 3000-4000 m.a.s.l. with mountain range and vast basins (Fig. 1). Across extensive areas on the plateau surface, average annual temperature ranges from –5.7^oC to 8.6^oC and average annual precipitation from 17 to 750 mm. Precipitation falls in the form of rain, snow and hail, more than 80% of which occurs from June to September, which is basically controlled by the Indian monsoon [Tibetan Expedition Team of Chinese Academy, 1983]. Low relative humidity (30-50%), high intensity of total annual solar radiation (586-741 kcal/cm^2) and

high potential evaporation (1500-2500 mm) are additional climatic characteristics of the plateau. All of its lands are arid and semiarid areas with frequently windy days. Desertification and soil erosion problems are very serious in the province. During the last decade, Qinghai has experienced rapid economic development. Germu and Xining are the two largest and most industrialized cities in the Qinghai Province with one third of its total population (Fig. 1). Germu is an important logistic station to Tibet, where the railway to Tibet ends. Its population has tripled from 1988 to 1999, from 0.1 to 0.3 million because of exploration of the oil field in the Qaidam Basin and development of Tibet. Many heavy industrial plants have been built around this city during the last 15 years, including oil refinery, metal smelters and chemical industries. Xining, the capital city of Qinghai Province, has had a fast pace of industrialization as well. The city's population has doubled during the last 10 years, up to the current 1.6 million.

Rapid economic development in the province must have brought air pollution problems because a lot of newly established industries are of primary processing plants. In the remote countryside, the grassland degradation is also a serious problem, which ahs been considered a result of overgrazing and climatic warming. However, acid rain can also cause the degradation of grassland. Therefore, it is necessary to investigate the extent to which recent rapid development has led to change of rainfall pH. Because of its remoteness and difficult accessibility, the measurement of precipitation pH was only initiated in 1986 by the Environmental Monitoring Station of Qinghai in Xining and Germu. Some reports have been published in a local journal without detailed analysis (Hu and Wu, 1989; Ni and Gao, 1989; Wang 1993). The regular measurement of pH has been carried out by the Qinghai Meteorological Station since 1991 in Xining and 1994 in Germu. The first author started the rainwater measurements, including pH and chemistry of rainwater, during 1987-1988 with the support from the Royal Geographical Society and Manchester University. The study locations were focused on the sites along the expedition routes. The measurements of rainwater pH and conductivity were done in the field and the samples were taken to the University of Manchester for

chemical analysis. A more recent research project has been carried out from 1997, with the support of local scientists and meteorological stations. The project used the pH data collected by the meteorological stations in Germu (2,807 m.a.s.l.) and Xining (2,295 m.a.s.l.) and made its own measurements on rainwater chemistry and atmospheric pCO_2. The aims of this investigation are to examine the precipitation pH and chemical characteristics at such high altitude, arid and cold conditions, to evaluate the rainfall pH and chemical changes caused by the fast industrialization associated with heavy pollutant loading and the recent measures to abate air pollution.

Methodology

A precipitation monitoring network have been established by the General Meteorological Bureau of China since 1991 and well-trained technicians to operate various stations by following the guidelines that are based the WMO recommendations for background networks. Our research grants contributed towards the operation of the stations in Xining and Germu. The stations run on a 24-hour basis and the bulk precipitation samples were measured in laboratory after each precipitation event.

Field measurements

In the 1987-1988 and 1998-1999 fieldwork periods the pH, conductivity and other parameters of bulk precipitation were collected and measured in field during the field trip seasons by the author because of insufficient laboratory facilities on the plateau. For each precipitation event the following parameters were recorded and analysed in the field and field laboratories following Standard Methods (APHA, 1980), plus some additional items:

a. precipitation---- mm (volumetric)
b. pH--- pH units at 20^oC (glass electrode)
c. CO_2---- atm partial pressure (air quality monitor)
d. TSP---- mg/m^3 (airborne particulates monitor)
e. conductivity---- $\mu S\ cm^{-1}$ at 25^oC (conductivity cell)

f. temperature (air and water)----^{o}C (thermometer)

Every sample has three readings on pH, CO_2, TSP and conductivity. Other meteorological parameters were collected. Besides, air-borne dusts were collected by plastic sheets laid on the surface. The meteorological stations measured pH and conductivity constantly from 1991 in Xining and 1994 in Germu, which provided a long-term record of rainfall pH and other parameters. The pH values and conductivities of the rainfall events measured by us in field seasons of 1998 and 1999 were compared with those acquired by the meteorological stations. The largest differences in pH and conductivity are 0.4 unit of pH and 17 $\mu S\ cm^{-1}$ in conductivity in these two years.

Laboratory analysis

Bulk precipitation samples were collected manually with a precipitation collector, which was washed every day, and stored in a refrigerator. The samples were well sealed in the local laboratories in order to keep the plateau's CO_2 partial pressure and brought to the University of Manchester, Chinese State Key Laboratory of Environmental Geochemistry and the University of Hong Kong for chemical analysis because of insufficient laboratory facilities on the plateau. The measured parameters were:

a. K^+, Na^+, Ca^{2+}, Mg^{2+}, Fe, and Mn ---- mg/L (Inductively Coupled Plasma, ZEISS PLASMAQUANT110 and Flame Atomic Emission Spectroscopy)
b. F-, Cl^-, NO^-_2, NO^-_3, SO^{-2}_4 Br^- and HPO^{-2}_4 ----mg/L (ion chromatography, HP LC 1100)
c. NH^+_4----mg/L (colorimetric and ICP)
d. HCO^-_3---mg/L (titration)

The pH and conductivity values were re-examined in the laboratories. The measurement started immediately after the sample bottles were unsealed, due to the need to consider the partial pressure difference of CO_2 and other gases between high and low altitudes. However, the concentration of HCO^-_3 has to be determined by titration because there is no available instrumental technique.

Results

pH values

The results of pH measurement for rainfall in 1987-1988 field seasons indicate that the precipitation in Xining and Germun was alkaline (Table 1), with a volume-weighted average of 8.15 and 8.23. This result is similar with those measured by Hu and Wu (1989) and Ni and Gao (1989) (Table 1). In 1990, Wang (1993) measured pH and rainfall chemistry at 8 stations in Xining City, and a significant drop of pH value was observed, from the averages of 7.96 and 7.83 in 1986 and 1987-88 to 7.14 in 1990 (Table 1). Since 1991 and 1994, the precipitation pH in Xining and Germu has continuously declined with some fluctuations. However, the pH of Xining returned to 7 since 1995, while the pH of Germu kept in the range of 6 in during the 1994-1998 seasons until the 1999. From the continuous records on the meteorological stations in Xining and Germu, a series of statistical analyses were conducted between rainwater pH and climatic factors. There was no large difference between annual precipitation amounts in these years. Therefore, the difference of annual rainfall amounts could not explain such a wide variation and changing pattern on pH values during the periods. There is no obvious correlation between pH and conductivity (Xining: r= 0.19; Germu: r=0.021), nor between pH value and rainfall amount (Xining: r= 0.04; Germu:r= -0.198), between pH and rainwater temperature (Xining: r=-0.12; Germu:r= -0.006), and between pH and wind speed during the period of precipitation (Xining: r=0.06; Germu: r= 0.034). However, some weak correlation appears between rainfall amount and conductivity of the rainwater (Xining: r= -0.30; Germu:r= -0.27).

We also studied the relationship between the pH value changes with seasonality. According to classification of the wet season (May-September) and dry season (October-April), there is no regularity in pH value change. If we divide the year into the windy season (December to May), which has 75% of annual gale days, and the calm season (June to November), pH value and conductivity changes follow the seasonal changes in Xining and Germu (Table 2). In windy seasons, pH values and conductivity of rainwater are generally higher than those in the calm seasons. The

conductivity of rainfalls of Germu was exceptionally high in 1995 because of oil field fires in the year.

Table 1. Annual weighted mean rainfall pH values of Germu and Xining

Location	Elevation(m) /precipitation (mm)	1986	1987-88	1990	1992	1993	1994	1995	1996	1997	1998	1999
Germu	2807/50	**8.03**#	**8.23(9)**	-	-	-	**7.10**	**6.88**	**6.91**	**6.59**	**6.46**	**7.28**
Xining	2295/350	7.96*	**7.83(11)**	7.1@	**6.30**	**6.70**	**6.55**	**7.32**	**7.09**	**7.09**	**6.65**	**6.91**

Hu and Wu 1989; * Ni and Gao 1989; @ Wang 1993; – No examination. Numbers in brackets indicate numbers of samples field seasonal measurements. Others are volume weighted mean of all rainfall events in the year.

Xining Germu

Year	Season	pH	Conductivity
1992	Windy	6.70	98.33
	Calm	6.37	65.19
1993	W	7.38	84.08
	C	6.47	58.79
1994	W	6.97	76.78
	C	6.71	39.59
1995	W	7.59	86.31
	C	7.19	66.86
1996	W	7.31	93.87
	C	7.42	67.11
1997	W	7.30	62.06
	C	6.83	55.33
1998	W	7.02	61.06
	C	6.55	55.10
1999	W	7.29	92.11
	C	6.97	64.28

Year	Season	pH	Conductivity
1994	Windy	7.70	115.9
	Calm	6.97	99.67
1995	W	6.95	238.8
	C	6.85	896.73
1996	W	7.32	162.3
	C	7.13	137.9
1997	W	6.56	147.8
	C	6.65	110.0
1998	W	6.87	104.2
	C	6.61	108.8
1999	W	6.89	58.30
	C	7.06	24.3

Table 2 Volume-weighted means of pH and conductivity in wind and calm seasons of Xining and Germu

Chemical composition

Table 3 Volume-weighted mean of chemical concentration (μeq L^{-1}) of precipitation of Qinghai Province

Location	Year/ Sample number	K^+	Na^+	Ca^{2+}	Mg^{2+}	NH^+_4	Cl^-	NO^-_2	NO^-_3	SO^{-2}_4	HCO^-_3	F^-

Germu +Xining	1988/7	36.15	23.88	149.25	30.37	11.62	11.62	*	8.16	7.92	263.40	*
Germu + Xining	1999-2000/11	69.17	96.56	314.31	37.90	160.61	48.77	2.41	48.08	84.01	534.29	46.62

*No examination

The rainfall chemical analysis demonstrates the changes of atmospheric chemistry as well (Table 3). Fe, Mn, HPO^{-2}_4 and Br^- present in trace quantities in the samples, within the range of a few μg L^{-1} or undetectable, hence their contents have not been reported here. There were a little SO^{-2}_4 and NO^-_3 in the samples of the 1988 rainfalls of Xining and and Germu and their contents are similar to those measured in world's remote islands (Galloway et al., 1982). In the 1998-99 period, a large increase occurred on almost all elements. It is noted that the total ionic concentration of the rainwater in the 1999-2000 is 160% higher than that of the 1988. The samples of 1999-2000 from Xining and Germu were seriously polluted. The SO^{-2}_4, NH^+_4 and NO^-_3 in the rainwater increased greatly in the 1999-2000 samples compared to those of 1988, and high concentrations of F^- has been found in the 1999-2000 samples, which may be related to metal smelting and brick plants in the city. Because of increase in acid content, the concentrations of all cations in the samples have been increased dramatically.

Basically, Ca^{2+} content is the dominant cation species and HCO^-_3 is the major anion species in all samples. The major anions and cations can be ordered as Ca^{2+}, K^+ , Mg^{2+} and Na^+ in 1988 and Ca^{2+}, NH^+_4, Na^+ K^+ and Mg^{2+} in 1999-2000. For the anions the reference is HCO^-_3 , Cl^-, NO^-_3 and SO^{-2}_4 in 1988 and HCO^-_3 , SO^{-2}_4 Cl^-, and NO^-_3 in 1999-2000.

Discussion

The causes of the rapid change of pH

Measurements in the two industrial cities in Qinghai Province exhibit fast decline of pH value in 1990s, with average values having dropped from around 8 in 1980s to below 7 in 1992 and 1995 respectively. The decrease in pH values from the 1987-88 period to the 1990s is indicated by its

population and industrial growth and the changes of pH followed the development and pollution control measures closely. With economic development of the two cities, the emission of air pollutants had gradually increased in early 1990s. For example, the SO_2 emission increased from 22,554 tons in 1991 to 33,009 tons in 1995 for the whole province (Table 4) as many heavy industrial plants were built. During this period, a lot of small ferrosilicate smelting plants were also built along Xining river valleys and the NOx content in air reached up to 0.067 mg/m^3, SO^{-2}_4 up to 0.115 mg/m^3 and TSP to 1.036 mg/m^3 during the winter period [Wang, 1995]. The average pH of rainfall in Xining declined in tandem and rather drastically from 7.83 in 1987-88 to 7.1 (1990), 6.64 (1992), 6.86 (1993) and 6.77 (1994). During the 1995-96 period, the Qinghai Government enforced environmental regulations and shut down many seriously polluting plants, and SO_2 emissions have reduced by nearly a half of those in the previous year. The rainfall pH in Xining swiftly increased from 6.77 in 1994 to alkaline (7.58 and 7.39 in 1995 and 1996). However, fast economic development and many newly built factories in this city caused pH to decline again until 1999 when the tougher environmental control measures have been carried out and many factories and domestic heating plants have been forced to use oil instead of coal. The case of Germu is different from Xining in terms of its economic growth period and geographical location. Germu is a remote city with faster growth in late 1990s. The development of the Qaidam Basin oil field nearby has created a lot oil and chemical industrial plant in late 1990s. The reinforcement of environmental control policies in this remote city was slower than the provincial capital. Therefore the rainfall pH was not lowered below 7 until 1995 and went back above 7 in 1999. Such experience also occurred in semiarid areas, such as Chembur, a highly industrialized area in the Bombay region. Its rainfall was reported as being acidic (4.8) from 1974 to 1980, and turned to 6-7 in response to effective pollution control measures after 1980 (Khemani, 1993).

The change of pH also was reflected in rainwater chemistry. It may be found that there were little pollutants, such SO^{-2}_4 , NH^{+}_4 and NO^{-}_3 , in bulk deposition of 1988. However, they have increased greatly in the period 199-2000, compared to the early monitoring (Table 3). Increase of

the acids could dissolve more dusts in air and lead to increase of total concentration of rainwater in the late 1990s.

Although a large change has occurred on pH values and the pollutants emission in Xining is higher than many south China industrial cities with serious acid rain problem, the rainfall in this area is still far from the acid rain. We

Table 4 SO_2 emission (Tons) of Qinghai Province

Year	1991	1992	1993	1994	1995	1996
SO_2 emission	22554	21306	24272	-	33009	17343

* Environmental Protection Bureau of Qinghai Province 1997. The Environmental Statement of Qinghai Province, 1991-1996.

The causes of alkaline rain

General scientific knowledge lends support that natural rain is weakly acidic [Howells, 1990; Meszaros, 1992; Miller, 1998. Merritts et al., 1998]. However, our measurements indicate that the less polluted atmosphere in this rather arid province can produce alkaline rain. Such long-term record of alkaline rain has rarely been recorded in the literature. The general knowledge about natural rain acidity is based on the measurements on remote oceanic islands and theoretical calculation of potential acidity. The original acidity of natural rain is calculated by CO_2 partial pressure in the atmosphere at sea-level or by CO_2 plus other natural acids, which is around 5.6 [Radojevic and Bashkin, 1999] or 5.0 [Galloway, et al., 1982] respectively. There are five potential causes to increase pH value of rainfall and lead to the occurrence of alkaline rain: volcanic ash, local industrial alkaline dusts, sea salts, low atmospheric pCO_2 and continental soil dusts. There was no volcanic eruption in and around the study areas during the alkaline rain period. The scale of local industrial mining around these two cities was too small and did not generate very large amount of dusts. Therefore, the first two potential causes can be ignored. From the chemical analysis of rainwater, the ratios of different elements have been calculated (Table 5). The result indicates that the rainwater on this part of the plateau is typical continental rainfall, not dominated

by sea salts. As the high altitude of this place can lower atmospheric pCO_2, which may increase pH values, we calculated the rainfall acidity according to the CO_2 partial pressure measurements in the province. The lowest value of pCO_2, which is about a half of that at sea-level and at the height of 5,600 m.a.s.l., may reduce rainwater pH values up to only 0.1 unit based on the attainment of chemical equilibrium between rainwater and atmospheric CO_2 partial pressure. Such a small drop of pH value implies that the lower pCO_2 on the plateau is an insiginificant contributor of the high pH value of rainfall. The only potential cause left is the continental soil dust. From the statistical analysis, it has been found that the rainfall in windy seasons generally has higher pH and conductivity. Local measurement on soil exhibits that its pH ranges from 6-10 [Land and Resource Bureau of Tibetan Government, 1994]. Our measurement on pH of dusts also shows that all TSP samples are alkaline (Table 6). The arid and windy climate and our analysis in rainfall chemistry, therefore, explain that the alkaline rain in these two cities is most possibly caused by continental dusts with little pollutants with scant contribution from marine aerosols. It must be noted that the arid soils commonly contain base-forming cations, CO^{-2}_3 , HCO^{-}_3, SO^{-2}_4 and free soluble salts, which could contribute alkaline particulates to the atmosphere. In addition, calcification is the major soil formation process in semiarid and arid regions, which produces abundant of calcite ($CaCO_3$). The arid soils tend to have accumulation of alkaline salt on the soil surface due to capillary rise of soil moisture and its evaporation. Soil dusts are mobilized from the surface soil with an elevated content of alkaline materials. The wind-blown dusts mobilzed from these soils are usually >2.5 μm in diameter, that is mainly of the coarse aerosol type that can be effectively and rapidly scavenged by the rainfall washout process. Between rainfall episodes, the atmospheric loading of soil-borne particulates could be quickly replenished by winds. Such continental soil dusts could buffer weakly acidic precipitation leading to pH values higher than 5.6 in many places, especially in arid and semi-arid areas where there are plenty of air-borne dusts [Berner and Berner, 1987; Sequeira 1993]. Hedin and Likens [1996] have pointed out the importance of atmospheric dusts in neutralizing the acidic air pollutants and the reduction of atmospheric dust release has

aggravated the acid rain problem. Evidence from arid areas, such as Israel, India, arid and semi-arid areas of the USA, Africa, Spain and other sites along the world's desert belts, also demonstrates that continental dusts can generate alkaline rain, although the records of the alkaline rains in these areas re short-term or occasional. The alkaline rain events have been associated with African trajectories and local dusts. [Subramanian and Saxena, 1980; Felly and Liljestrand, 1983; Khemani *et al*., 1985; Mamane *et al*.,1987; Avila and Alarcon 1999; Goni *et al*., 2001]. The fact that these areas do not have long-term and constant alkaline rain records is probably because that the measurements were usually carried out in cities and after industrialization when the acid rain problem had already entrenched itself, or because of the taking of only short-term measurements due to financial constrains or the absence of serious acid rain problem.

Table 5. Ratios of Na^+ to Cl^-, SO^{-2}_4, K^+, Ca^{2+} and Mg^{2+} of the Qinghai rainwater

	Cl^-/Na^+	SO^{-2}_4/Na^+	K^+/Na^+	Ca^{2+}/Na^+	Mg^{2+}/Na^+
Germu 1988	0.61	0	3.45	15.58	2.46
Xining 1988	0.69	0.03	0.18	6.43	0.83
Xining 1999	0.58	3.10	0.04	3.48	0.52

Conclusion

The long-term monitoring of rainfall pH in two industrial cities of Qinghai Province indicates that the rainfall is this region is naturally alkaline and moderately mineralized. Chemical composition of rainwater is dominated by Ca^{2+} and HCO^-_3. Fast changes of precipitation pH and rainfall chemical concentration in Qinghai have demonstrated that human beings can change the nature of rainfall chemistry in a very short period. Emission of pollutants without control can rapidly induce rainwater acidity. Meanwhile, with tough environmental control measurements, reduction of the emission and change of energy-use pattern could also rapidly shift rainfall pH back close to its natural state.

From our research results, the following implications can be drawn: a. pH value cannot be used as an indicator of air pollution in arid and semiarid areas and as a sole indicator of air pollution in

other areas because air-borne dusts can buffer acids in cloud water; b. HCO^-_3 is a very important anion in precipitation, it should be recommended to the rainfall monitoring guidelines as a standard measurement for all precipitation; c. environmental consequences that were caused by rapid change of pH in fragile arid and semi-arid regions need to be investigated as soon as possible because the species used to live in alkaline environment may have been damaged under acidic condition; d. the anthropogenic acidification can increase the total concentration of precipitation because of abundant of airborne dusts in arid regions.

Acknowledgement

The authors acknowledge the assistance of the Meteorological Bureau of Qinghai, and of the Chinese State Key Laboratory of Geochemistry and Ms Liu of The University of Hong Kong for chemical analysis. Dr. Radojevic and Dr. Chen provided very stimulating technical advices and discussions. This work is supported by the Seed Funding Grant of The University of Hong Kong, Nanjing University, University of Manchester and Royal Geographical Society.

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Captions:

Fig. 1. Location of the rainwater monitoring stations on the northeastern Tibetan Plateau.

Table 1. Annual weighted mean rainfall pH values of Germu and Xining

Table 2 Volume-weighted means of pH and conductivity in wind and calm seasons of Xining and Germu

Table 3 Volume-weighted mean of chemical concentration (μeq L^{-1}) of precipitation of Qinghai Province

Table 4. SO_2 emission (Tones) of Qinghai Province

Table 5 Ratios of Na^+ to Cl^-, SO^{-2}_4, K^+, Ca^{2+} and Mg^{2+} of the Qinghai rainwater

Table 6. pH values of dust deposits in Qinghai

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Location	*Season*	*PH*
Xining	Windy	8.1
	Calm	7.6
Germu	Windy	8.4
	Calm	7.9