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VARIATION OF PH IN ATMOSPHERIC PRECIPITATION IN CITIES OF JILIN PROVINCE

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ABSTRACT: Through statistical analysis on data of atmospheric precipitation samples of 8 cities from 1992 to 1997, this paper discussed temporal and spatial variations of pH in atmospheric precipitation in Jilin Province, China. Monitored pH value in the 8 representative cities was converted into hydrogen ion consistency and average consistency was calculated by weighting precipitation, then mean pH value was calculated according to average hydrogen ion consistency. Results show that atmospheric precipitation of cities in Jilin Province is basically neutral, but in Tumem and Hunchun cities atmospheric precipitation is seriously acid. The average pH value of atmospheric precipitation of the eastern cities of Tumen and Hunchun in many years is 5. 12 and 5. 38 respectively, and the atmospheric precipitation acidity in the two cities is quite serious with the extreme low pH is 3. 91 and 4. 28 respectively. Most of the acid precipitation occurred in summer and autumn seasons in the 8 monitored cities, so that the low pH value of atmospheric precipitation occurred in summer and autumn seasons and high pH value occurred in winter and spring seasons. The temporal and spatial variations of pH in atmospheric precipitation in Jilin Province are attributed to both local sources and peripheral sources, and are related to both natural and artificial factors.

KEY WORDS: atmospheric precipitation; pH; acid rain; Jilin Province

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Acid rain has been a global environmental problem. South China and the southwest part are the two main acid rain areas in China with the range of pH variation from 4. 5 to 4. 0, and scientific studies on acid rain focus on these areas(WANG *et al.*, 1997). In recent years, however, in the north part of China acid rain occurred not unusually and even more seriously such as in cities of Tumen and Hunchun in Jilin Province. It is essential to delve into the temporal and spatial variations of atmospheric precipitation acidity and the variations causes in these areas to provide scientific basis for management and further research.

1 CITY SELECTION AND STATISTICAL METHODS

Jillin Province is located in the east of Eurasia and the center of northeast China, from 121°38′ to 131°19′ E and 40°52′ to 46°18′N. Representative cities from the west to the east in the province are selected as followings: Changchun, Siping, Dunhua, Tonghua, Yanji, Tumen and Hunchun.

pH value is an important parameter to measure the acidity of atmospheric precipitation. Both the minimum pH value of natural rain saturated with CO_2 and the minimum pH value of glacier are about 5. 5 to 5. 6. So we classify precipitation with its pH value below 5. 6 as acid rain, while categorize precipitation with its pH value above 8. 5 as alkaline rain. On the basis of atmospheric precipitation data from 1992 to 1997 provided by local environmental monitoring departments, pH value was converted into hydrogen ion consistency and the average consistency was calculated by weighting

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of precipitation. Mean pH value was calculated according to average hydrogen ion consistency. The statistical methods are given bellow:

$$pH_i = -\log [H^+]_i$$

$$[H^+]_{ave} = \sum [H^+]_i P_i / \sum P_i$$

$$pH_{ave} = -\log [H^+]_{ave}$$

where pH_i and P_i are the monitored pH value and precipitation(mm), $[H^+]_i$ means converted hydrogen ion consistency in each precipitation, $[H^+]_{ave}$ represents the average hydrogen ion consistency, pH_{ave} is the average pH value.

2 TEMPORAL AND SPATIAL VARIATIONS IN pH OF ATMOSPHERIC PRECIPITATION

2.1 Temporal pH Variations

2.1.1 Seasonal variations

The calculated results of seasonal average pH value of precipitation in 8 cities are shown in Table 1.

All the seasonal average pH value in Siping, Tonghua, Dunhua and Yanji varies from 5.95 to 7.17, and the atmospheric precipitation in these cities is generally neutral. Except for winter season all the seasonal average pH value in Tumen and Hunchun is bellow 5.6. In Tumen City in autumn the average pH value of atmospheric precipitation is 4.99, and the extreme low value is 3.91(November 3rd, 1997, rain, 1.8 mm), while in Hunchun city the lowest value is 4.28(September 24th, 1996, rain, 9.8 mm). Acidity of precipitation in the above two cities is quite serious comparing with South China and the southwest where precipitation acidity is nearly the most serious in the world.

According to acid rain samples and all the atmospheric precipitation samples in representative cities from 1992 to 1997, occurrence frequency of acid rain can be calculated. The results show that in the cities with low seasonal average pH value of precipitation the acid rain occurrence frequency is high, while in the cities with high seasonal average pH value of precipitation the acid rain occurring frequency is low. There were no acid precipitation through the monitored years in Jilin, Siping, Tonghua and Yanji cities, so the acid rain occurrence frequency is 0. In Dunhua City there were only one acid rain during 1992 to 1997(June 13rd, 1996, rain, 15.3 mm, pH = 3.97). Situation in the other three cities are shown in Fig 1.

Table 1 Mean pH value of atmospheric precipitation in representative cities in Jilin Province

	Changchun	Jilin	Siping	Tonghua	Dunhua	Yanji	Hunchun	Tumen
Spring(April – May)	6.66	7.26	6.86	6.85	6.83	6.63	5.33	5.14
Summer(June – August)	5.92	6.79	6.99	6.96	6.29	6.84	5.34	5.13
Autumn(September – October)	6.09	7.29	7.17	6.51	6.84	6.61	5.46	4.99
Winter(November – March)	6.75	7.79	7.17	5.95	6.70	6.57	5.87	5.80
Annual mean	6.01	6.87	7.02	6.74	6.39	6.74	5.38	5.12

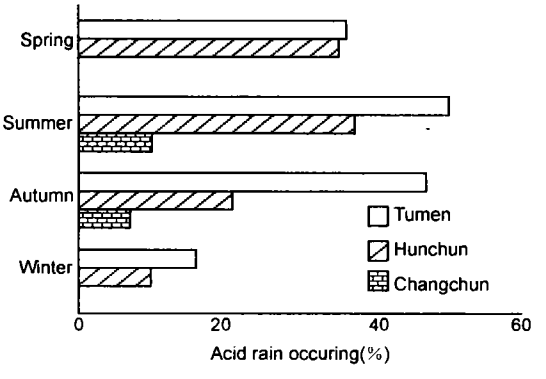

Fig. 1 Occurrence frequency of acid rain in representative cities in Jilin Province

From Fig. 1 it is clear that Tumen and Hunchun are the two cities which have the highest acid rain occurrence frequency in Jilin Province. Acid rain occurrence frequency in summer and autumn is higher than that in spring and winter. Low pH value of atmospheric precipitation occurred in summer and autumn seasons and high pH value occurred in winter and spring seasons.

Alkaline rain once occurred in Jilin Province but with less frequency than acid rain. Table 2 shows all of the alkaline rain in Jilin Province from 1992 to 1997. Owing to its limited times there is no obvious temporal occurrence law but in Jilin City most of the alkaline rain concentrated in winter in the form of snow.

Table 2 All of the saline rain in Jilin Province from 1992 to 1997

City	Date	Type	Precipitation(mm)	pH
Jilin	1992-02-18	snow	3.0	8.93
	1992-04-21	snow		10.00
	1992-11-19	snow	0.6	11.25
	1992-12-21	snow	0.5	11.21
	1994-06-12	snow	7.9	9.00
	1995-06-12	snow	0.8	10.98
	1995-12-27	snow	1.8	10.48
	1997-06-10	rain	0.5	9.42
	1997-11-17	snow	1.5	9.00
Siping	1993-06-19	rain	9.0	8.59
Tonghua	1995-08-07	rain	156.3	8.62
Hunchun	1996-04-17	rain	3.0	8.80

2. 1. 2 Yearly variations

In the 8 representative cities, atmospheric precipitation in Jilin, Siping, Tonghua, Dunhua and Yanji is generally neutral and the yearly variations of pH value is not intensive. Yearly pH variations in Tumen and Hunchun are shown in Fig. 2.

Before the year 1993 mean yearly pH values of atmospheric precipitation in the two cities were both above 5.6, while since the year 1994, mean pH values were both below 5.6. Mean yearly pH value in the two cities tends to descend and acid rain situation becomes serious.

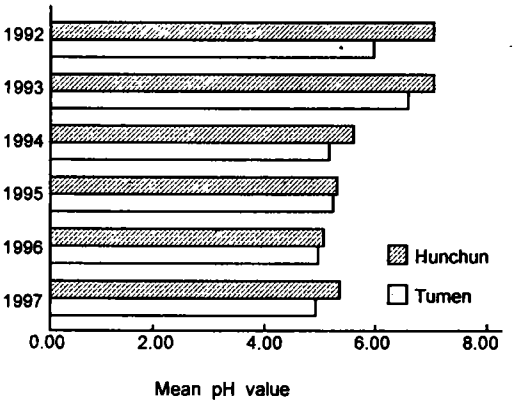

Fig. 2 Mean yearly pH value variation of atmospheric precipitation in Tumen and Hunchun

2.2 Spatial pH Variations

Jilin Province is topographically made up of two main landforms: the eastern mountainous region and the central-western plains. Among the 8 representative cities Tumen and Hunchun lie in the Tumen River Basin in the eastern part of the province. In all of the central-western cities the precipitation acidity can be neglected with an exception of Changchun. Acid rain mainly occurred in Tumen and Hunchun cities (Table 3).

Acid rain samples and the sum of acid precipitation in Tumen and Hunchun cities contribute more than 90% to the province respectively. Considering Changchun City acid rain samples and the sum of acid precipitation in the three cities contribute 99.5% and 99.43% respectively. The other 5 cities contribute very little precipitation acidity to the province. Acid rain concentrated in Tumen, Hunchun and Changchun especially in Tumen and Hunchun the precipitation acidity is very severe(Table 4).

Alkaline rain occurs mainly in Jilin City especially in Hadawan industrial area where in winter and spring the occurrence frequency is 46.67% and 33.33% separately. In the other 7 cities alkaline rain seldom occurs.

3 CAUSES OF THE TEMPORAL AND SPATIAL VARIATIONS OF PH

The pH value of atmospheric precipitation is determined by mutual balance of acid materials and alkaline materials in the precipitation process, and also related to the factors including water vapor sources, transmission distances, and passed underlying surface .

Table 3 Contribution of Tumen and Hunchun cities to acid rain in the 8 representative cities in Jilin Province

	Acid rain samples	Contribution(%)	Precipitation(mm)	Contribution(%)
Tumen	122	50.41	1226.1	45.91
Hunchun	105	43.39	1274.1	47.71
Total	227	93.80	2500.2	93.63

Table 4 Statistical parameters of pH value in Tumen and Hunchun from 1992 to 1997

	N	Difference	Median	Mode	Coefficient	Standard deviation
Tumen	284	3.79	5.81	6.00	0.70	4.97E-0.2
Hunchun	346	4.52	6.69	6.90	0.74	4.62E-0.2

N: number of samples; Difference: the difference between the maximum and the minimum

So when causes of pH variation of atmospheric precipitation are investigated, we must study relationships between pH variations and acid-causing materials, as well as the mitigation function of alkaline microsomes, and at the same time the influence of peripheral sources should be considered.

3.1 The Balance of Acid Materials and Alkaline Materials

Chemical composition of atmospheric precipitation in representative cities in Jilin Province is that $SO_4^{2-} > Cl^- > NO_3^-$. The main acid causing materials are SO_2 and NO_x. Seasonal mean concentration of SO_2 and NO_x in Tumen City is lower than that in other cities(Table 5), however, precipitation acidity in Tumen is the most serious, so that acid-causing materials are not the leading factor in causing pH value variation of atmospheric precipitation.

Table 5 presents that seasonal mean concentration of TSP in Tumen City is much lower than that in the other three cities. TSP is microsomes in the atmosphere with complex chemical composition and particle radii assembly. TSP in the cities in the northern part of China is alkaline. Alkaline TSP can buffer and neutralize the acid rain forming process and improve the pH value of the atmospheric precipitation. Acidity from SO_2 can be neutralized half by TSP(ZHAO, 1987). Pearson correlation between pH value and TSP concentration is 0.78, and the correlation is significant at the 0.01 level. Relations between pH and these factors can be expressed as:

$$pH = 4.740 + 3.481[TSP] - 3.139[SO_2] + 4.720[NO_x]$$

where [TSP], $[SO_2]$ and $[NO_x]$ represents the consistency of TSP, SO_2 and NO_x respectively. Since the TSP concentration in Tumen is lower than that in other cities, thus pH value of atmospheric precipitation in this city is correspondingly lower than that in other cities.

In Tumen City, the SO_2 and NO_x content in summer and autumn seasons are lower than those in winter and spring seasons, but the TSP content in atmosphere is opposite, so that precipitation pH value in summer and autumn is higher than that in winter and spring. The TSP content in atmosphere is the leading factor to the temporal and spatial variations of pH value of atmospheric precipitation.

3.2 Influence of Topography and Meteorology

Wind speed in the eastern mountainous area of Jilin Province is slowed down below 3 m/s by topography screen function. This makes the acid-causing materials not easy to diffuse, thus improving the precipitation acidity. While in the central-western plain areas, the lane formed by the Da Hinggan Mountains and the Changbai Mountains, is the center of high wind speed in northeast China with the average level at 5 m/s. High wind speed enhances diffusion of acid materials in the atmosphere, so pH value of city precipitation in the central-western part of the province is higher than that in the eastern part.

In summer and autumn seasons, sunshine time is

Table 5 Daily mean concentration of SO_2, NO_x and TSP in cities of Changchun, Jilin, Tumen and Yanji (mg/m³)

City	Items	Spring	Summer	Autumn	Winter
Changchun	SO_2	0.087	0.013	0.011	0.094
	NO_x	0.072	0.036	0.044	0.096
	TSP	0.453	0.350	0.308	0.369
Jilin	SO_2	0.156	0.047	0.026	0.105
	NO_x	0.083	0.040	0.044	0.078
	TSP	0.772	0.679	0.556	0.731
Tumen	SO_2	0.038	0.006	0.011	0.049
	NO_x	0.043	0.017	0.025	0.056
	TSP	0.302	0.148	0.240	0.334
Yanji	SO_2	0.102	0.016	0.014	0.121
	NO_x	0.118	0.032	0.039	0.118
	TSP	0.561	0.228	0.492	0.601

Resources: Report on Environmental Quality in Jilin Province (1991 - 1996)

long and temperature is relatively high, and rich precipitation makes humidity high. All this meteorologic factors are beneficial for SO_2 to transform into SO_4^{2-}, so precipitation acidity in summer and autumn exceeds that in winter and spring and causes seasonal precipitation pH variations.

3.3 Influence of Peripheral Region

Acid rain in Jilin Province belongs to sulfuric acid type. The more the peripheral region contributes to the sulfur content subsidence, the more the region influences the precipitation acidity of cities in Jilin Province. Sulfur content subsidence in Jilin Province is mainly contributed by Jilin Province, northeast China and North China(Table 6).

Northern peripheral areas contribute less sulfur content subsidence but more alkaline TSP to Jilin Province than the southern peripheral areas do. For instance, sulfur content subsidence contribution from Heilongjiang Province is only 1/4 to 1/2 of the counterpart from Liaoning Province. Impacted by east Asian monsoon climate, wind directions in Jilin Province have seasonal variations, northwest wind prevails in winter and spring seasons and southeast wind prevails in summer and autumn seasons. Northwest wind brings less sulfur content and more alkaline TSP from northern peripheral areas to Jilin Province than southwest wind does from southern peripheral areas. It is easy to conclude that pH value of atmospheric precipitation in winter and spring seasons with the northeast wind prevails are higher than that in summer and autumn season

Table 6 Contribution of sulphur from peripheral provinces to Jilin Province (YANG *et al.*, 1998)

	Contribution(%)						
	50 - 40	40 - 30	30 - 20	20 - 10	10 - 5	5 - 1	1 - 0
Overhead souces				Jilin	Liaoning	Hebei, Inner Mongolia, Shandong, Henan, Hebei, Heilongjiang	Shanxi, Jiangsu Zhejiang, Anhui, Henan, Hubei, Sichuan, Shaanxi, Ningxia, Beijing, Tianjin, Shanghai
Low sources		Jilin		Liaoning		Shanxi, Inner Mongolia, Shandong, Tianjin, Heilongjiang	Jiangsu, Zhejiang, Anhui, Jiangxi, Henan, Hubei, Hunan, Sichuan Guizhou, Shaanxi, Gansu, Xinjiang, Beijing, Shanghai
Overhead and low sources	Jilin		Liaoning		Heilongjiang	Hebei, Shanxi, Inner Mongolia, Shandong, Henan, Beijing, Tianjin	Jiangsu, Zhejiang, Anhui, Jiangxi, Hubei, Hunan, Sichuan, Guizhou, Shaanxi, Gansu, Ningxia, Xinjiang, Shanghai

with wind from the direction of southern peripheral areas.

Precipitation acidity in Tumen and Hunchun is closely related to peripheral region. Through chemical properties analysis of precipitation in the two cities near the Sea of Japan and atmospheric airflow trace retreating analysis, the study by WANG Wen-xing *et al.*, (1997) reveals that acid precipitation airflow comes from Japan especially Osaka industrial areas. In fact, not only Tumen and Hunchun but also cities in Liaodong Peninsula and Shandong Peninsula is influenced by airflow from Japan and South Korea. It is necessary to cooperate with other provinces, neighbouring countries (Japan and South Korea) in controlling acid rain problems, and regional relations should be studied to take common corresponding measures to limit the total amount of acid materials in the atmosphere.

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