

Chemical and Mineral Characteristics of Melted Snow-Water in the Jilin Province, North-East China

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Abstract

The significant investigation was performed to establish a chemical baseline for snowfall in Northeast of China since there was no such record. The chemical and mineral composition of snowfall samples were evaluated in Jilin Province, Northeast China. The specific purposes of the present study are to characterize the chemical and mineral composition of melted snow-water samples and to identify the sources of atmospheric pollutants, as well as to investigate the spatial variability of melted snow-water chemistry in this region. Snowfall and aerosol samples were collected from Changchun city, Baicheng city, and Yanji city during November 2015 to March 2016. All the filtered (0.45 m) snow-water was analyzed for conductivity, pH, major cations (Na^+ , K^+ , NH_4^+ , Ca^{2+} , Mg^{2+}) and anions (F^- , SO_4^{2-} , Cl^- , NO_3^- , HCO_3^-). The pH of these samples ranged from 6.01 to 7.10, with an average value of 6.44. The conductivity ranged between $17.50 \mu\text{S cm}^{-1}$ and $86.00 \mu\text{S cm}^{-1}$ with an average value of $55.0 \mu\text{S cm}^{-1}$. These parameters indicated high chemical concentrations in the snow-water. It was observed that Ca^{2+} and NH_4^+ were the major cations which accounted the concentration for 5.05% to 18.71% and 3.47% to 15.54%. HCO_3^- and SO_4^{2-} were major anions which accounted the concentration for 23.41% to 38.49% and 21.10% to 35.24%, respectively. $(\text{SO}_4^{2-} + \text{NO}_3^-) / (\text{Ca}^{2+} + \text{NH}_4^+)$ was 1.38. The organic carbon of particles in the snowfall was higher and the average content was 52.85 g kg^{-1} . The size of the snow-water particles in the three Cities was mainly based on clay complexes ($<2 \mu\text{m}$). The order of average content was: ($<2 \mu\text{m}$) $>$ ($>200 \mu\text{m}$) $>$ ($20\text{-}200 \mu\text{m}$) $>$ ($2\text{-}20 \mu\text{m}$). The dominant mineral of particles in the snow-water was quartz and feldspar, while clay mineral was composed of montmorillonite, illite, kaolinite and vermiculite. According to the above analyses, the possible sources of the particles in the snowfall should be soil and ground dust and coal-burning.

Keywords: Chemical ions; Melted-snow water; Mineral composition; Particles; Source analysis

Introduction

Atmospheric, as an important part of the ecosystem, has an important influence on the environment and ecology. Air pollution had become one of the environmental issues concerned widespread [1-8]. The ecological environment had been affected by the particles in the atmosphere [9]. Furthermore, particles were also harmful to the environment, cloud formation, atmosphere visibility, climate, and human health [10-12]. Due to the relative large size, coarse particles generally precipitated to the ground through wet deposition. While fine particles still remained in the atmosphere for several weeks and could be transported through atmospheric circulation. Therefore, fine particles greatly impacted the environment [13]. Understanding the physical and chemical properties of the particles in the snowfall is the most important prerequisite to investigate the impact of winter snowfall on the environment [14]. Saxena and Ruggiero [15] found Na^+ , Cl^- , SO_4^{2-} and NO_3^- to be the predominant ions in precipitation at McMurdo. Delmas [16] reported sulfate, sodium, chloride, carbonaceous species and trace amounts of NH_4^+ and NO_3^- in precipitation on the Antarctic Plateau. Warburton and Linkletter [17] found Na^+ , Mg^{2+} , and K^+ to be the predominant ions collected in snow on the Ross Ice Shelf. Mulvaney and Wolff [18] found low nitrate and sulfate, and high chloride concentrations in January ice core samples from the Weddell Sea sector of Antarctica.

The different particle fractions could have substantially different sources [19]. Coarse particles were mainly derived from mechanical processes related to road transport [20,21]. Fine particles primarily originated from combustion processes and from atmospheric, gas-to-particle conversion processes [22]. The study about characteristics of wet deposition in atmospheric from some cities in Northern China have been conducted. Most previous studies were concentrated on

ionic components, while the researches about the particle compositions and mineral compositions were less. Most previous studies in China focused on big cities such as Beijing, Shanghai, and the Pearl River Delta region [23]. The report about Northern China was less. However, the increasing winter heating in Northern China made the air quality much worse compared to the other seasons. Due to such characteristics, the research on the particles in the snowfall can be helpful to investigate the sources of the air pollution. To investigate the sources of the contaminant in the atmospheric environment, physical characteristics and chemical composition of the particles in the snowfall must be understood [24]. Further research was done about the chemical properties and mineral composition of particles in winter snowfall in Northeast China by XRD to analyze the chemical composition of its soluble substances. Studies on particles chemistry are important for evaluating air quality and understanding anthropogenic contributions to atmospheric environment. The main subject of this study was to play an active part in investigating the possible sources of the particles of winter snowfall in the Northeast China. The results from this study will provide the theoretical foundation for the government to put forward scientific and rational measures to prevent and control environmental pollution.

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Received July 19, 2016; Accepted August 01, 2016; Published August 03, 2016

Citation: Zhao X, Guo X, Mao Y, Yan H, Li M, et al. (2016) Chemical and Mineral Characteristics of Melted Snow-Water in the Jilin Province, North-East China. J Civil Environ Eng 6: 248. doi: [10.4172/2165-784X.1000248](https://doi.org/10.4172/2165-784X.1000248)

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Materials and Methods

The study area

Figure 1 showed the situation of study area within province and China. The first sampling site was Changchun City, and located in Jilin Agricultural University campus (43° 81'N, 125° 41'E). The second one was Baicheng city, and located in the testing station of Jilin Agricultural Sciences (45° 62N, 122° 81'E). The last one was Yanji City, and the specific location was in the farmland from College of Agriculture in Yanbian University (42° 91'N, 129° 81'E). Seven snowfall samples were collected in Changchun City. The snowfall in Baicheng City and Yanji City were mixed samples without human interference. Changchun is the capital of Jilin Province, and this city contains a bigger population and energy consumption than that of Baicheng city and Yanji city. Therefore, only one sample from Baicheng city and Yanji city was collected as comparison research. The snowfall samples were stored for analysis after melting. Basic information about the samples was shown in Table 1.

Chemical analysis about the melted snow-water and the particles

The values of pH and conductivity were measured by Crison-Basic-20 pH and DDS-11A produced in Spain. After filtering, the snow-water without the particle was obtained and ion chromatography

(ICS3000) was used to determine the anion concentrations of SO_4^{2-} , HCO_3^- , Cl^- , F^- and NO_3^- . The concentration of K^+ , Na^+ was measured by flame photometer and Ca^{2+} , Mg^{2+} , NH_4^+ was measured by atomic absorption spectrometer (ZEEnit700).

Analysis of the particles composition in the melted snow

The pipette method and sieving method were used to determine the composition of the particles from snowfall [25]. First of all, hydrogen peroxide was used to remove the organic matter in the particle samples, and then dematerialized with dilute hydrochloric acid. According to the Secretary Alex Benedict law, the clay complexes ($<2 \mu\text{m}$) and the silt complexes ($2\sim 20 \mu\text{m}$) were separated with the siphon after ultrasonic dispersion, respectively. The fine complex ($2\sim 200 \mu\text{m}$) and the coarse sand complex ($>200 \mu\text{m}$) would be got by sieving method. After sedimentation, centrifugation, drying and weighting, the percentage of each particle components would be calculated.

X-ray diffraction (XRD) analysis of the particles from the melted snow

The method of dithionite-sodium citrate-bicarbonate (DCB) was employed to remove the iron from the clay complexes $<2 \mu\text{m}$ and then made them saturated by using potassium and magnesium specimens. The saturated specimens were dried naturally after directional specimen. The X-ray diffractometer (Shimadzu 7000) was used to determine

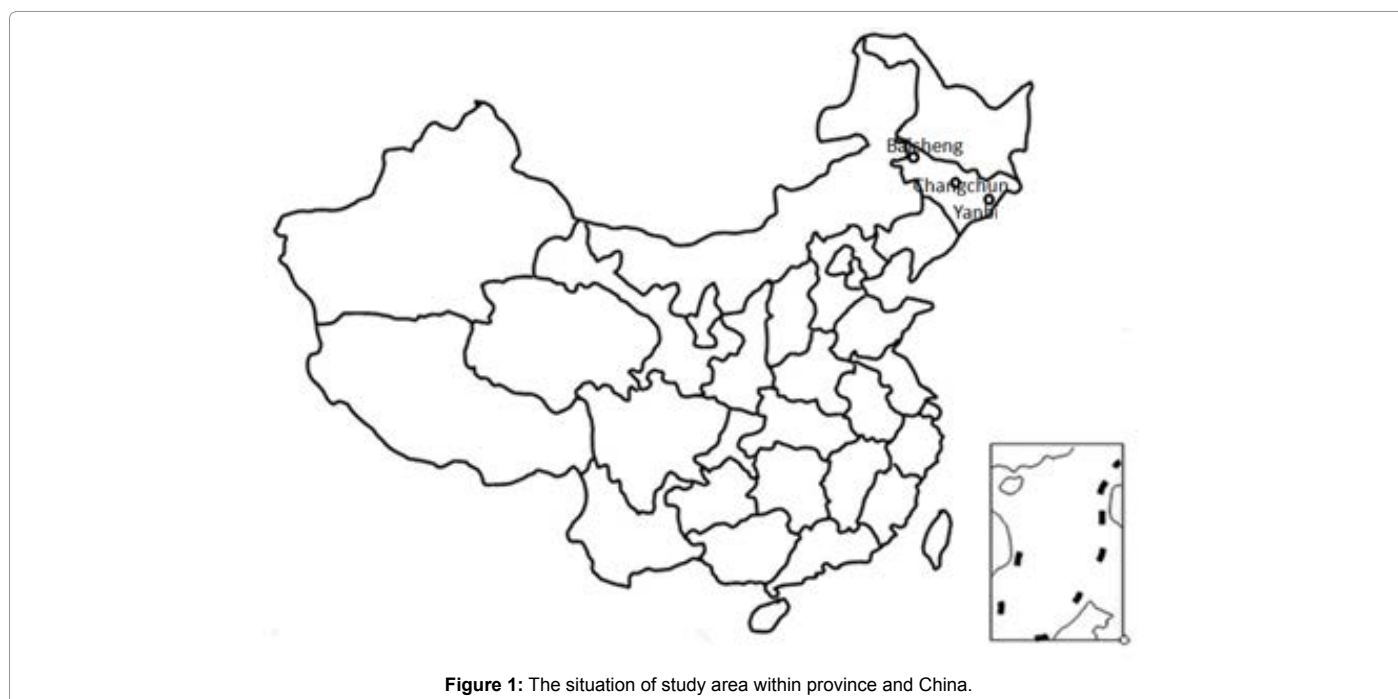


Figure 1: The situation of study area within province and China.

Collection sites	Number	Snowfall Date	Snowfall Height/mm	Wind direction
Changchun city (N43°81' E125°41')	C1	2015.11.26	61	North
	C2	2015.12.04	33	North
	C3	2015.12.25	17	Northeast
	C4	2015.12.28	80	Northeast
	C5	2016.01.27	150	North
	C6	2016.03.14	143	North
	C7	2016.03.22	7	Northwest
Baicheng city (N45°62' E122°81')	B1	2016.01.12	130	Northwest
Yanji city (N42°91' E129°81')	Y1	2016.01.18	142	West

Table 1: Basic information about the sampling sites.

mineral composition under the condition of CuK α radiation, Ni filter, 40.0 kV, 30.0 mA and each step size was 0.04 $^\circ$. Then, the potassium saturated specimens were heated in muffle furnace for 2 h at 300 $^\circ$ C and 550 $^\circ$ C, respectively. The magnesium saturated specimens should be saturated by using glycerin before determination. A non-directional test for the silt complexes (2~20 μ m) was conducted. The data of clay minerals identification was analyzed by MDI Jade 5.0.

Results and Discussion

Chemical properties of melted snow-water

It can be seen from Figure 2 that the average pH of the snowfall in Jilin province was 6.44 in 2015-2016, ranging from 6.01 to 7.10, which did not belong to acid precipitation. So there was no impact on the environmental acidification. The conductivity values were medium, ranging from 17.50 to 86.00 μ s cm $^{-1}$ and the mean value was 55.0 μ s cm $^{-1}$ (Figure 3). This value was approximately 37 times the value of the

clean rainfall of the atmospheric baseline observatory in Waliguan, China [26]. This indicated that a certain degree of the snowfall had been polluted by the chemical ions with higher content. The ion concentrations of the snow-water were shown in Figure 4. It can be seen that the concentrations of positive Ca $^{2+}$ and NH $_4^+$ were higher than that of other positive ions. Ca $^{2+}$ was considered to be the symbolic elements of city dust and soil dust [27] and the relatively high concentration of Ca $^{2+}$ could be the evidence that the sources of the particles from snow-water in Northeast China was mainly city dust and soil dust. The concentration of the SO $_4^{2-}$ was relatively higher than that of the NO $_3^-$, indicating that the main acid-causing substance was SO $_4^{2-}$. While SO $_4^{2-}$ was formed by the oxidation of SO $_2$ produced by coal burning, which indicated that coal-fired pollution from industrial production was the mainly factor. Since Changchun City is located in the north temperate latitudes in the Northern Hemisphere. Spring is dry and windy, winter is cold and long which also leads to the winter-heating season much longer. Meanwhile the winter-heating was mainly based on coal

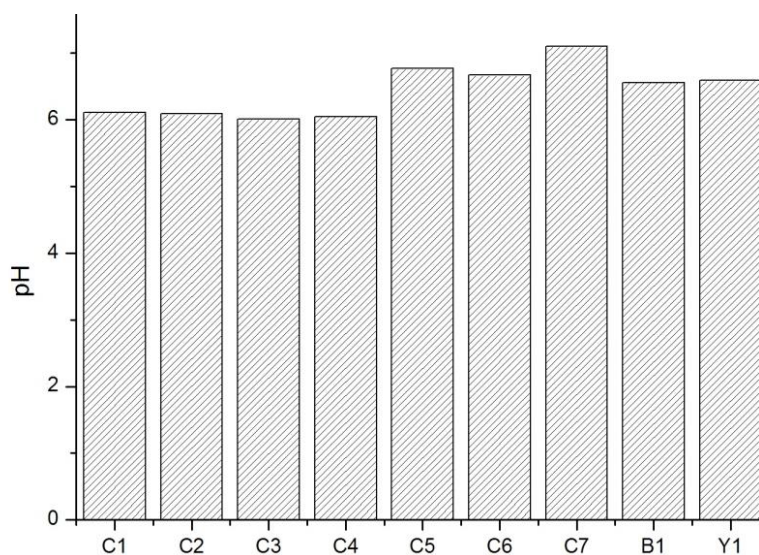


Figure 2: pH value of the nine melted-snow water samples.

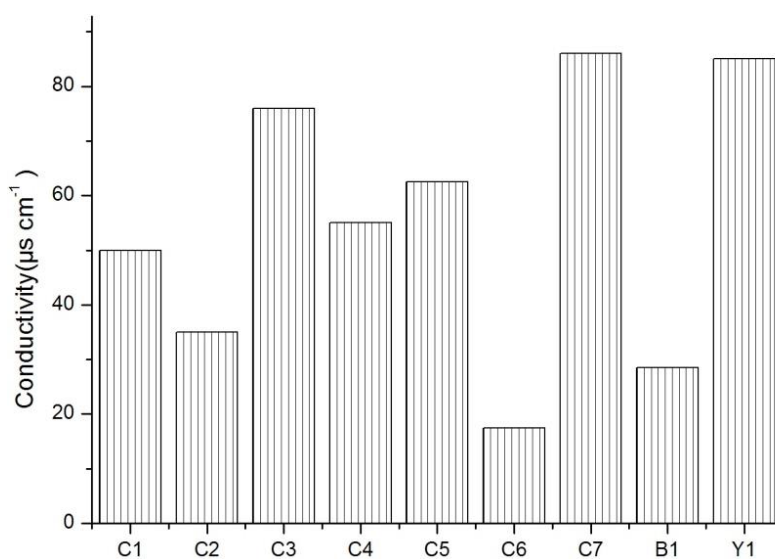


Figure 3: Conductivity of the nine melted-snow water samples.

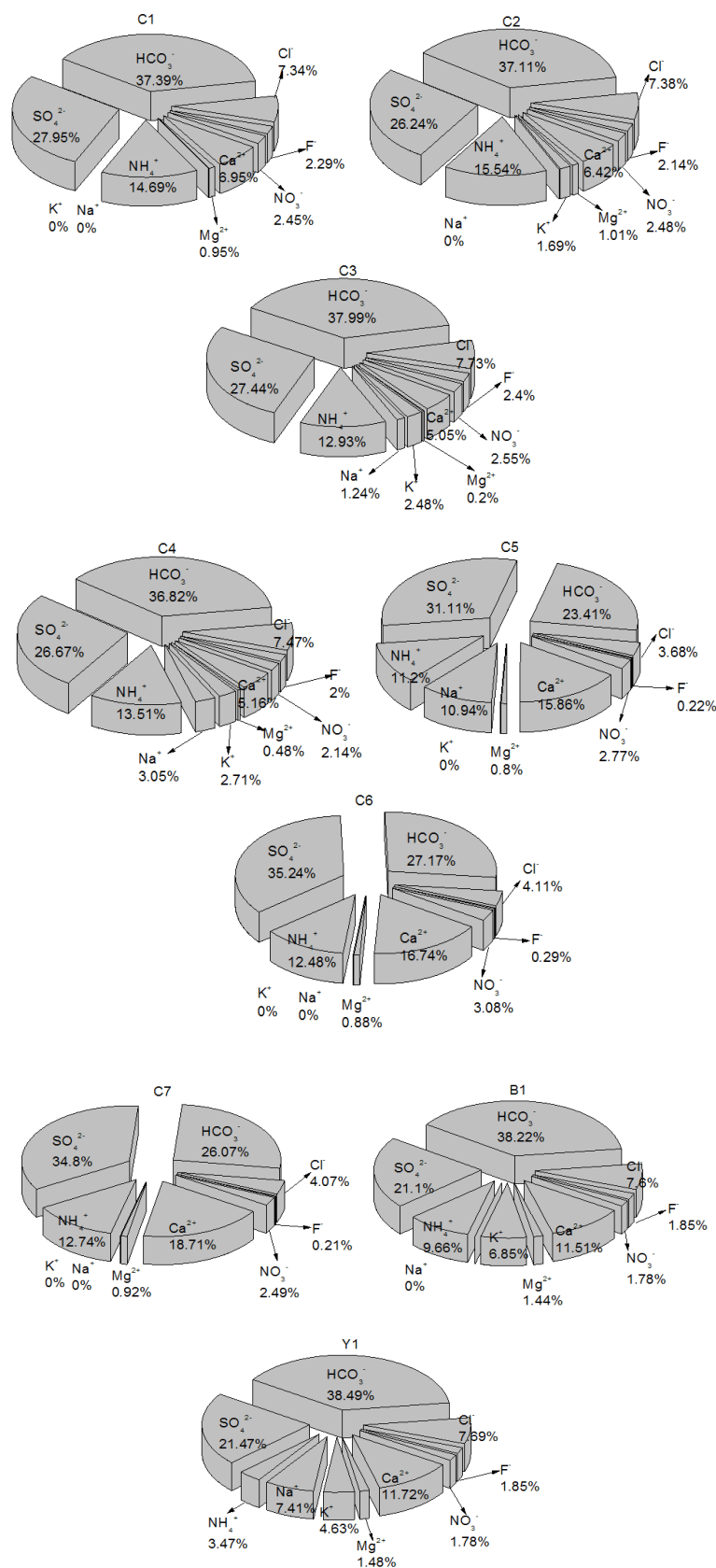


Figure 4: Chemical composition of melted-snow water in the three cities of Jilin Province, North-east China.

burning, which produced a large amount of fine particulate matter. This part of fine particulate matter makes a great contribution to the air pollution. HCO_3^- and SO_4^{2-} were major anions which accounted the concentration for 23.41%-38.49% and 21.10%-35.24%, respectively. Acidic ion was mainly based on SO_4^{2-} , and the concentration of NO_3^- was very low. Alkaline ions were based on NH_4^+ , Ca^{2+} and HCO_3^- [28]. Although the contents of acidic ions in snow-water were relatively high, winter snowfall was not acidic under the neutralization of alkaline ions [29]. The higher ratio of $(\text{SO}_4^{2-} + \text{NO}_3^-) / (\text{Ca}^{2+} + \text{NH}_4^+)$, the higher acid precipitation intensity, that was to say the lower pH [30]. The value of $(\text{SO}_4^{2-} + \text{NO}_3^-) / (\text{Ca}^{2+} + \text{NH}_4^+)$ in the precipitation of Jilin province was 1.38, which indicated a strong neutralizing capacity of Jilin atmosphere. For the three cities, HCO_3^- and SO_4^{2-} were all the dominant water soluble ions. The third ion was NH_4^+ for Changchun city, while for Baicheng city and Yanji city the third one was Ca^{2+} . There was still difference between the different sampling months in Changchun city. The percentage of the HCO_3^- for sampling points C1, C2, C3 and C4 was all over 30%. But for sampling points C5, C6 and C7, the percentage

was all under 30%, SO_4^{2-} had the opposite law. Figure 5 showed a linear regression analysis of neutralization ($\text{Ca}^{2+} + \text{NH}_4^+$) and acidification potential ($\text{SO}_4^{2-} + \text{NO}_3^-$) for snowfall samples in Northeast China. The correlation coefficient between neutralization and acidification ions in the snowfall was 0.86 in Northeast China. The contribution of other ion species in the acidification and neutralization processes in the snowfall samples of the Northeast region was not significant [28]. Data analyses showed that SO_4^{2-} , Cl^- , NO_3^- were the major indicators of the coal and automobile exhaust. SO_4^{2-} was formed by the oxidation of SO_2 produced by coal burning. Among them, the mass ratio of the NO_x/SO_x exhausted by the vehicle emissions was about 8~13, while the ratio by the emissions from coal-fired boilers was about 1~2. So the mass ratio of $\text{NO}_3^-/\text{SO}_4^{2-}$ was commonly used to analyze the source of the city pollution (fixed source pollution or mobile source pollution) [31]. The mass ratio of the $\text{NO}_3^-/\text{SO}_4^{2-}$ was negligibly small in this study. It was obviously that the source of the city pollution in northeast was mainly by fixed source pollution (such as fire coal).

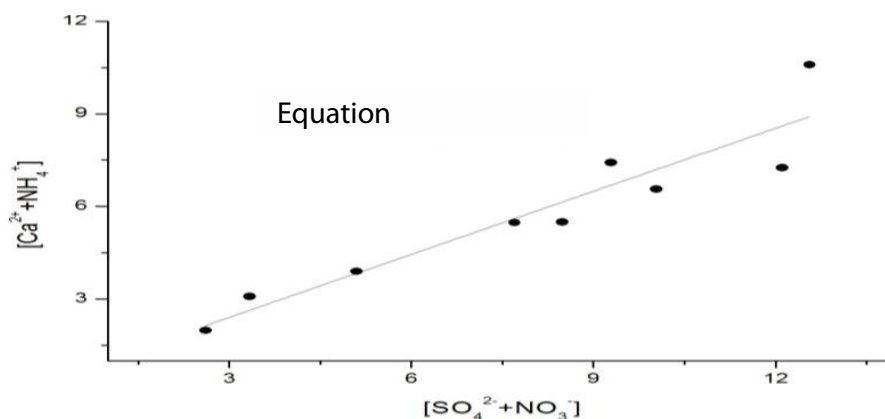


Figure 5: Correlation between $([\text{Ca}^{2+} + \text{NH}_4^+])$ and $([\text{SO}_4^{2-} + \text{NO}_3^-])$ in the melted-snow water.

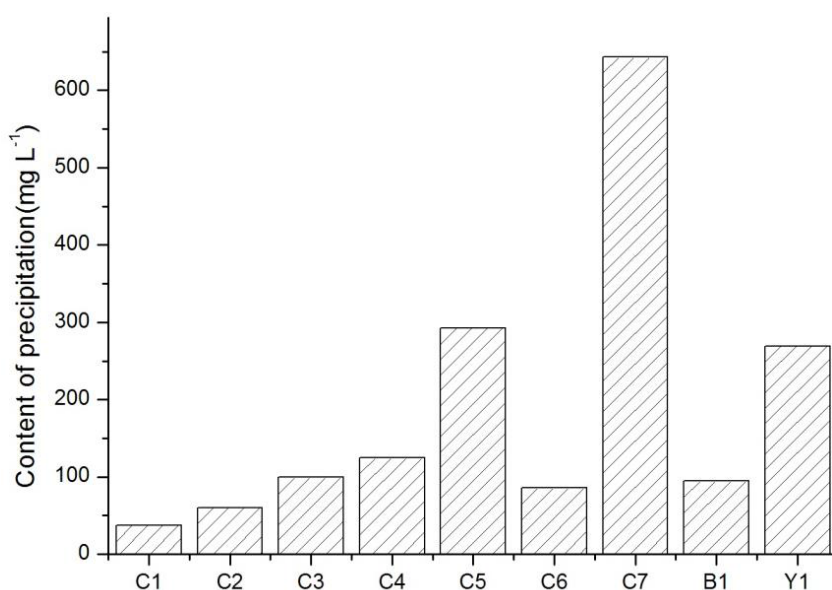


Figure 6: The quality content of particles in the unit volume of melted-snow water.

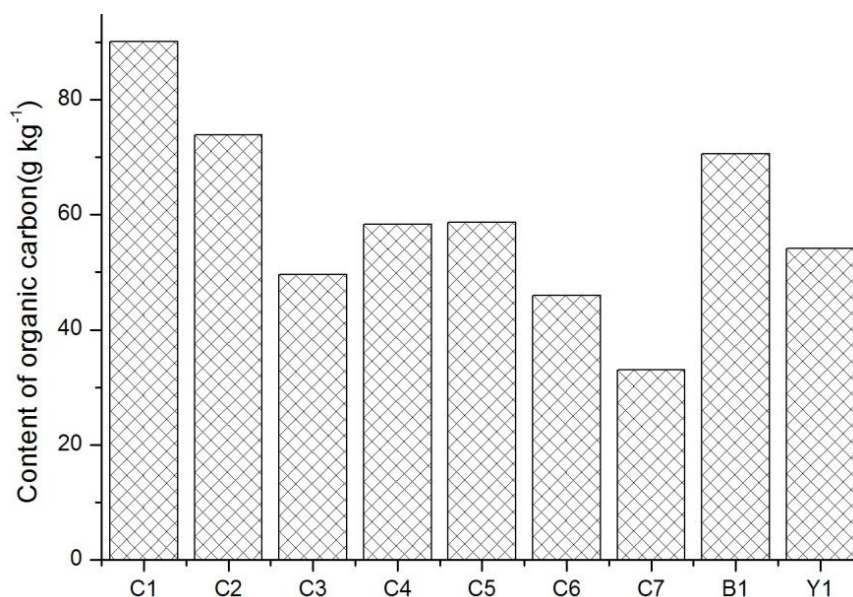


Figure 7: The organic carbon content in the particles from melted-snow water.

Number	Composition of particles (%)			
	<2 μm	2-20 μm	20-200 μm	>200 μm
C1	51.23	4.76	27.99	16.02
C2	83.48	0.19	9.53	6.80
C3	74.76	0.39	22.63	2.22
C4	70.02	1.38	20.84	7.76
C5	70.88	4.56	20.05	4.51
C6	96.17	0.19	0.47	3.17
C7	46.15	1.79	47.45	4.61
B1	77.04	0.59	22.23	0.14
Y1	62.03	13.10	24.42	0.45

Table 2: The size composition of particles in the snowfall.

The content of organic carbon and the composition of the particles from the melted snow

As it could be seen from Figure 6 that the mean content value of the particles in melted snow was 189.71 mg L⁻¹, ranging from 37.47 to 643.06 mg L⁻¹. More atmospheric particulate matter fell to the ground by snow falling, indicating that snowfall played a role in purifying the air. As shown in Figure 7, the organic carbon content of the particles in Changchun City snowfall was higher and the mean value of organic carbon was 52.85 g kg⁻¹, ranging from 33.05 to 90.07g kg⁻¹. Sample C1 collected from Changchun in November had the highest concentration of organic carbon among the nine samples. After that, the concentration of other months had a tendency to decrease. The content of organic carbon existed obvious difference, which may be related to the different ratio of each pollutant and the meteorological conditions [32,33]. In northeast China, the burning of coal in winter and exhaust emissions made the organic carbon content of the particles in the snowfall higher.

Table 2 showed the particle size of snowfall. the particle size in the snowfall of Changchun City, Baicheng city and Yanji city was mainly based on clay complexes (<2 μm). Clay complexes (<2 μm) was primarily derived from soil surface weathering and did not belong to the native minerals by Niu et al. While the silt complexes (2~20 μm) belonged to the native minerals.

Mineral composition of the particles from the melted snow

Silt complexes (2~20 μm) were used for non-directional X-ray diffraction test. Figure 8 showed that the primary mineral of the particles in the snowfall of Jilin Province was mainly of quartz, feldspar. Only the particles in the snowfall of Changchun City (43° 81'N 125° 41'E) contained hematite. The primary mineral of the particles in Baicheng and Yanji city had no hematite. Diffraction peaks of quartz were seen at 0.42 nm and 0.35 nm. The diffraction peak at 0.35 nm was intensity while the other one at 0.42 nm was slightly weaker. Feldspar diffraction peaks could be seen at 0.4 nm, 0.37 nm, 0.36 nm and 0.31 nm. Meanwhile it was a strong and intensity one at 0.31 nm. Hematite diffraction peaks was seen at 0.27 nm but the intensity of it was weaker [34].

Figure 9 (a, b and c) showed the X-ray diffraction curves of the clay complexes (<2 μm) of the particles in the snowfall on November 26, 2015 of Changchun city, on January 12, 2016 of Baicheng city and on January 18, 2016 of Yanbian city. For the three cities, all the five diffraction curves were the similar, which meant the particles from the three different cities had almost the same mineral compositions. the magnesium dried curve peaks appeared at 1.42 nm, 1.0 nm, 0.71 nm, 0.50 nm, 0.47 nm, 0.42 nm, 0.35 nm and 0.33 nm. It could be seen from the saturated and 550°C heating potassium curve that diffraction peaks disappeared at 0.71 nm, indicating the kaolinite was contained

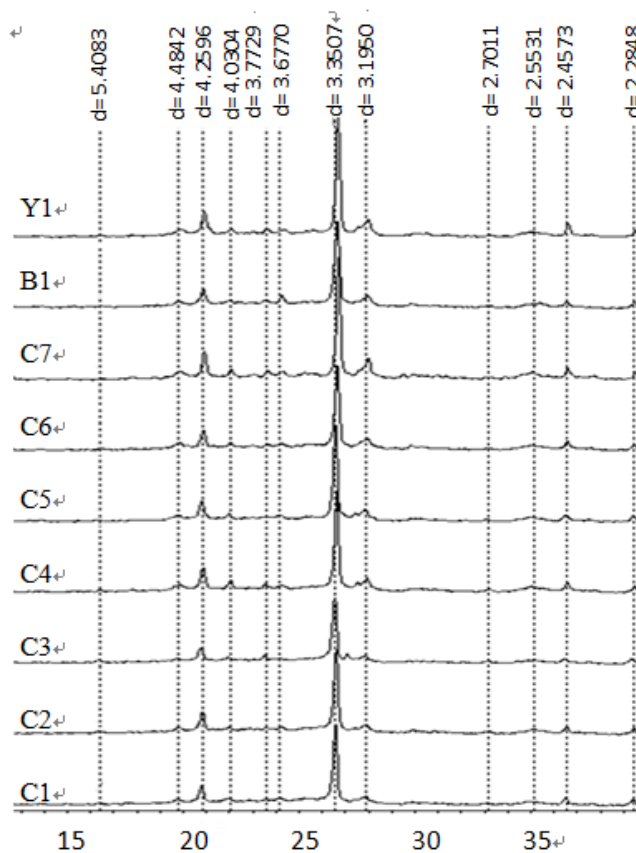


Figure 8: Non-directional X-ray diffraction of filtered particles from melted-snow water.

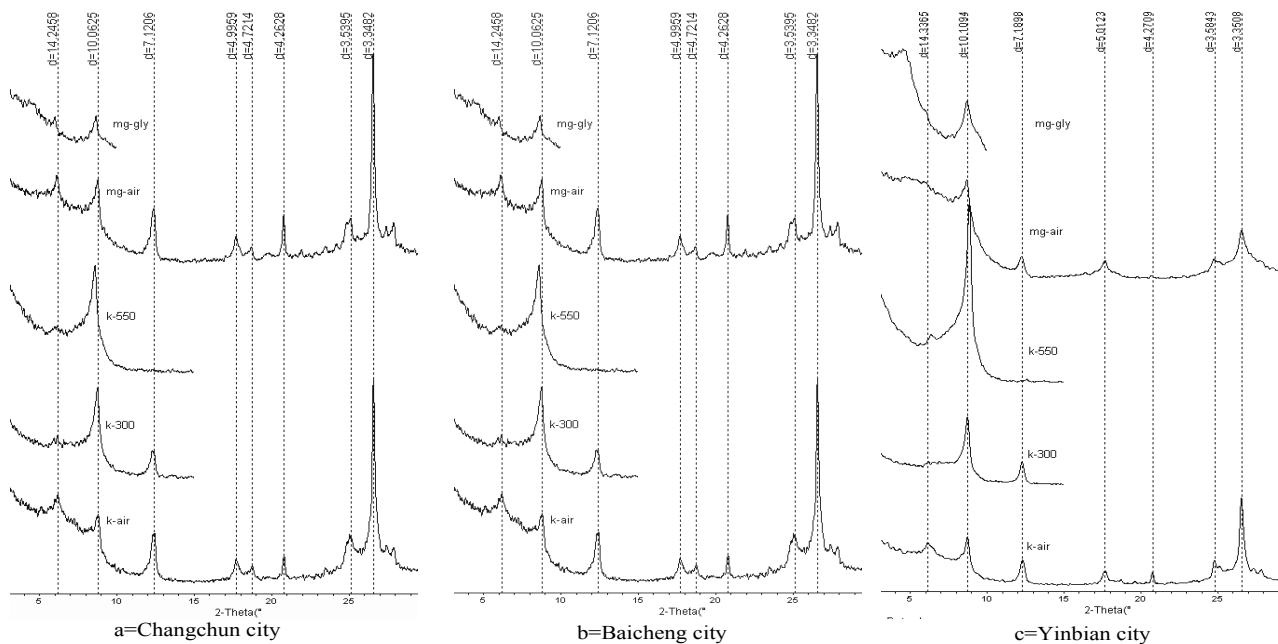


Figure 9: (a, b, c) The X-ray diffraction curves of the clay complexes (<2 μm) in the snowfall particles in the three cities.

in the particles. While at 1.42 nm the intensity of diffraction peak was decreased, indicating that a small quantity of vermiculite was contained. Only for Changchun city, the intensity of the diffraction peak at 1.42 nm was slightly changed, showing the presence of chlorite. Because the diffraction peak position at 1.0 nm was not significantly changed after glycerol and potassium saturation and heating treatment, a lot of illite were contained. After potassium saturation and glycerol treatment, the diffraction peak on the left of the low-angle at 1.42 nm had a contraction phenomenon, showing the presence of montmorillonite. According to the above analysis, it could be determined that the clay minerals in the three snowfall particles contained montmorillonite, illite, vermiculite and kaolinite. Only the particles from Changchun city contained chlorite [35-37].

Conclusion

The concentrations of positive Ca^{2+} and NH_4^+ were higher than that of other positive ions, and HCO_3^- and SO_4^{2-} were major anions. The chemical ion content of the snowfall in Jilin Province was higher. The relatively high concentration of Ca^{2+} could be the evidence that the sources of the city pollution in Northeast China was mainly from city dust and soil dust. The concentration of the SO_4^{2-} was relatively higher than that of the NO_3^- , indicating that coal-fired pollution from industrial production was still the mainly factor. The mean value of the organic carbon content of the particles in the melted snow was 52.85 g kg^{-1} . For varieties of industrial raw materials (such as coal), quartz was seldom in it. Therefore the maximum possible source of the quartz was from the ground dust. Illite and chlorite in the particles were present as a sign of sources from ground dust. The source of the Kaolinite and montmorillonite were from the soil dust. Overall, the possible source of the particles in the snowfall could be the city dust, soil dust, ground dust and coal-burning.

Acknowledgments

This research work was funded by the National Natural Science Foundation of China (No. 41403077) and Special Grand National Science-Technology Project (No. 2014ZX07201-011). The author would like to acknowledge, Key Laboratory of Soil Resource Sustainable Utilization for Jilin Province Commodity Grain Bases for technical and financial support during sampling and analysis.

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