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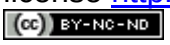

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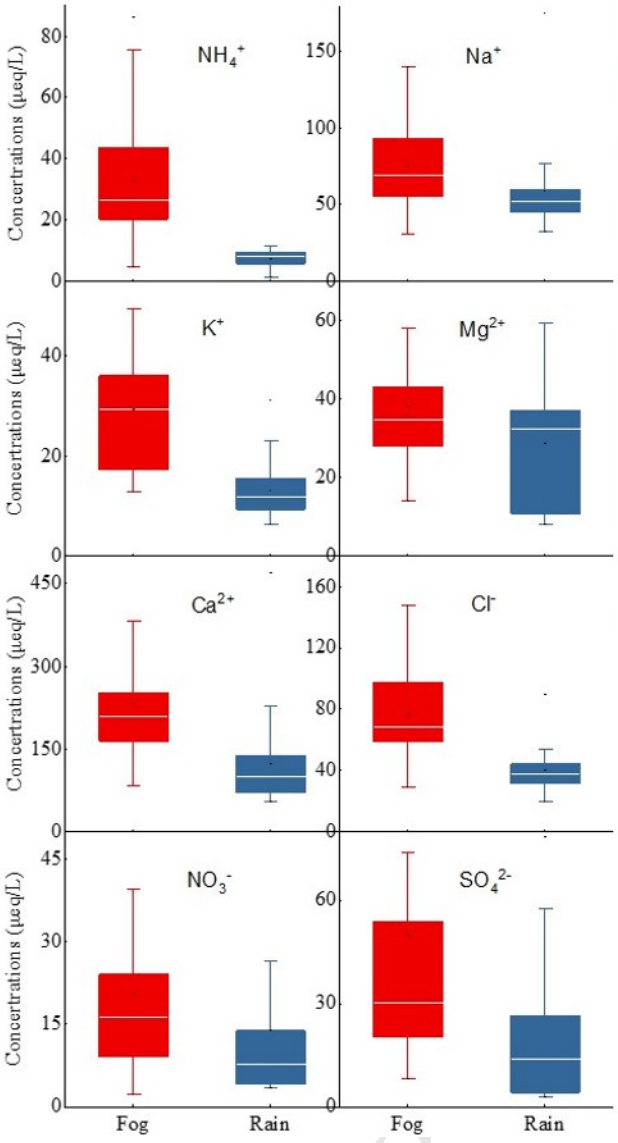
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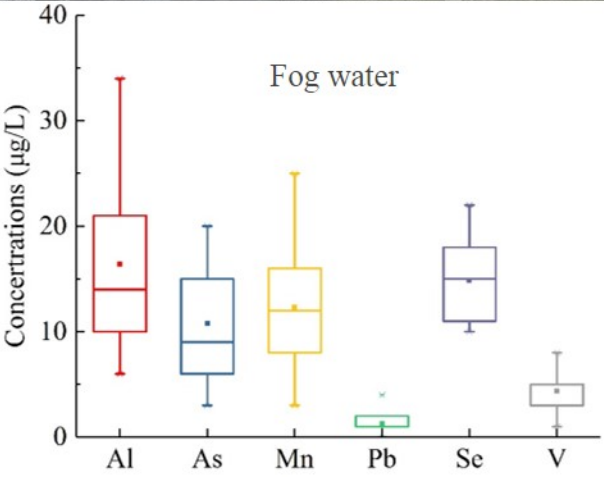
Na+
K+
Mg2+
Ca2+
Cl-
NO3-
SO4 2-
Concentrations (μeq/L)
Fog
Rain


Fog water
Concentrations (μg/L)
Al
As
Mn
Pb
Se
V

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Chemical compositions of fog and precipitation at Sejila Mountain in the southeast Tibetan Plateau, China

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Abstract:

Chemical compositions of fog and rain water were measured between July 2017 and September 2018 at Sejila Mountain, southeast Tibet, where fog events frequently occurred in original fir forests at altitude 3950 m. Fog water samples were collected using a Caltech Active Strand Cloud Collector (CASCC), and rain samples were collected using a precipitation gauge. Differences were observed between fog water and rain composition for most analyzed ions. Ion abundance in fog water was Ca^{2+} >

Cl^- > Na^+ > SO_4^{2-} > Mg^{2+} > NH_4^+ >K^+ > NO_3^- whereas an order of Ca^{2+} > Na^+ > Cl^- > Mg^{2+} > SO_4^{2-} > NO_3^- > K^+ > NH_4^+ was observed for rain water. All ion concentrations were higher in fog water than in rain water. Additionally, Ca^{2+} was the dominant cation in both fog and rain samples, accounting for more than half of all measured cations. NH_4^+ and SO_4^{2-} concentrations were notable for being higher in fog than rain water when compared with other ions. For trace elements, Al, As, Mn and Se were the most abundant elements in fog water; only Al and As were detected in rain water. Seventy-two hour back-trajectory analysis showed that air masses during fog and/or rain events mainly came from the south of Sejila Mountain. Spearman correlation analysis and source contribution calculations indicated that both marine and terrestrial sources contributed to the observed ion concentrations. Considering the higher concentrations of NH_4^+ and higher ratio of NH_4^+/NO_3^- measured in fog than in rain, we suggest that quantification of fog nitrogen deposition and its ecological effect in this area should be given more attention.

Keywords: Fog water; Ion concentration; Trace elements; Emission source; Southeast Tibet

Capsule: Comparison of water-soluble ions in fog and rainwater in southeast Tibet.

1. Introduction

Clouds and fogs play an important role in processing pollutants and other trace chemical species in the atmosphere. Scavenging of particles and soluble trace gases by cloud and fog droplets, followed by direct droplet deposition to the ground or incorporation into precipitation, represent important pathways for pollutant removal from the atmosphere (Liu et al., 2004). In order to assess the impacts of cloud and fog on various ecological environments, numerous studies of cloud and fog water chemical composition have been conducted in Europe (e.g., Collett et al., 1993a; Blas et al., 2010; Blas et al., 2008; Malcolm et al., 2003; Giulianelli et al., 2014), North America (Waldman et al., 1985; Weathers et al., 1988; Collett et al., 1990, 1991a; Saxena and Lin, 1990; Straub et al., 2012, Herckes et al., 2015, Templer et al., 2015, Straub, 2017), South America (Weathers et al., 2000; Strater et al. 2010), and Asia

(Aikawa et al., 2005; Kim et al., 2006; Beiderwieden et al., 2007; Sheu and Lin, 2011; Wang et al., 2011; Shen et al., 2012). In many cases, clouds and fogs have high ion concentrations and can be 5–20 times more concentrated than rain water (Anderson et al., 1999; Beiderwieden et al., 2007). Although the direct hydrologic input from fogs and intercepted clouds is typically much lower than from rain and snow, the higher solute concentrations mean that cloud and fog deposition should not be ignored when considering nutrient and pollutant input, especially for high elevation ecosystems (Collett et al., 1990, 1993a; Aleksic et al., 2009).

Montane cloud forests are defined as forests that are frequently covered in cloud or mist (Hamilton et al., 1995). In the forest ecosystems of higher mountains, fog deposition has been recognized as an important component of hydrological and nutrient cycling (Dawson et al., 1998; Elias et al., 1995; Weathers et al., 2000). For example, Lovett (1982) found that the input of NH_4^+ and NO_3^- through fog deposition was 4 times that by rainfall for a fir forest ecosystem in New Hampshire. In contrast, the water input via fog deposition was only 18–23% of the corresponding precipitation amount (Yamaguchi et al., 2015). Although the chemical characteristics of fog water were highly variable in different regions, the contribution of fog water was found to be crucial in the water and nutrient cycle of the forest ecosystem (Fuzzi et al., 1988; Basset et al., 1991; Mueller et al., 1991a; 1991b; Berresheim et al., 1993; Chang et al., 2006; Klemm et al., 2007; Novak et al., 2015). Moreover, leaves could gather water and ions by direct contact with fog, and even become the main source of plant nutrients (Hutley et al., 1997; DeFelice, 2002; Liu et al., 2004). Additionally, the chemistry of fog and rain were usually different even at the same area, because precipitation incorporates cloud condensation nuclei (aerosol particles) when cloud droplets are formed at higher altitudes but often experiences dilution by significant water vapor condensation (e.g., Collett et al., 1991b) whereas fog droplets at the surface condense in lower boundary layer air (Templer et al., 2015; Novak et al., 2015). The pH value of fog water was often lower than rain water, and acid damage to vegetation by fog water (Waldman et al., 1982) can be considerable because plant leaves are immersed in fog for long periods of time (Waldman et al., 1982; Charlson

et al., 1982; Wrzesinsky et al., 2000; Adzuhata et al., 2001). As a result, the chemical characteristics of fog water at montane forests have been highlighted by researchers in several regions (Aleksic et al., 2009; Polkowska et al., 2014; Michna et al., 2015; Wang et al., 2015). Köhler et al. (2015) reported that atmospheric water and element inputs increased with the rise of elevation in temperate mountain forests, attributable mainly to fog and cloud deposition.

The above-ground carbon reserves of forests in Tibet can exceed 250 t hm^{-2}. Moreover, the forest with the highest monthly average net productivity is the dark coniferous forest near an altitude of 4000 m (a.s.l.) in Nyingchi city (He, 2008). The chemical composition of rainwater and fogwater in this region has not previously been reported; consequently, potential impacts of pollutant and nutrient deposition on these forests are unquantified. Considering the unique geographical location of the Qinghai-Tibetan plateau and the relatively low anthropogenic disturbance in this region, understanding the chemical composition of cloud/fog water and rain water in this region is not only significant to the nutrient circulation of the local forest ecosystem, but also could provide useful insight into long range transport of air pollutants. Here we select a high elevation native fir forest as a representative region to investigate the fog chemistry at Sejila mountain, with the objectives of 1) understanding fog and rainfall chemical characteristics in a southeast Tibet forest ecosystem; 2) clarifying the possible sources of inorganic ions in fog and rainfall in the area. We hypothesize that fog interception of the forest will be frequent and that concentrations of key solutes will be higher in fog water than in rainfall, pointing to a need to further explore potential ecosystem impacts of fog deposition.

2. Materials and methods

2.1. Site description

The sampling site was established at the National Field Scientific Observation Station of the Alpine Forest Ecosystem (29.65° N, 94.72° E 3950 m a.s.l.) on Sejila Mountain. Sejila Mountain is a nationally protected area because of its primeval fir forest with a variety plants and animals. The climate is dominated by warm air currents from the Indian Ocean, with air masses arriving mainly from a southerly

direction during April to October, when fog events are likely to occur. The annual average temperature was -0.73 ^{o}C and the annual average precipitation was approximately 1000 mm. This station was surrounded by forest and there were no nearby agricultural and industrial pollution sources except a state road (#318). Fog events occurred mainly in the evening, lasting sometimes till the following morning. Unfortunately, fog events were often coupled with rain events in the night, however, our experiment condition was limited, even continuous power supply was not guaranteed in our research station, and we were unable to identify fog events during nighttime. In order to avoid contamination of fog samples by rainfall, only samples collected in the daytime were included in this study. Location and sampling environment are shown in **Fig. 1**.

2.2. Sample collection

Fog samples were collected from July 2017 to September 2018. No fog events were observed in any part of the research area in July 2017 or between November 2017 and March 2018, although frost or rime could be seen. Fog was observed almost every day at the mountain hillside forest area during the experimental period. However, due to experimental constraints, 35 fog water samples were obtained from 35 individual fog events during the sampling period, with most of those samples collected in July, August and September. A Caltech Active Strand Cloud Collector (CASCC) was used to collect fog samples (Demoz et al., 1996). The CASCC family of fog collectors has been widely used for fog sampling (e.g., Collett et al., 1990, 2002; Bator and Collett, 1997; van Pinxteren et al., 2016; Li et al., 2011; Guo et al., 2012). Air was drawn by a fan into the CASCC at 24.5 m^3 min^{-1}. Six rows of 508 μm Teflon strands were used to capture droplets by inertial impaction; the 50% lower size cut for the collector corresponded to a drop diameter of 3.5 μm. Collected droplets were drawn down the strands by aerodynamic drag, accumulated in a Teflon collection trough, and flowed through a Teflon tube to a pre-cleaned sample bottle. Fog samples were collected with the CASCC when fog interception events were observed at the site and continued until the fog disappeared. After sampling, all collected fog water samples were mixed as one sample for each individual fog event,

154 then transferred to clean polyethylene bottles (50 ml) for storage. The CASCC was
155 cleaned in late afternoon (e.g. 6 p.m.) in order to collect fog which mainly occurred in
156 the evening and morning. In addition, the national road No. 318 in Sejila Mountain
157 section was often closed to vehicles between 12:00 p.m. (midnight) and 7:00 a.m.
158 (morning) next day. Fog collection was not carried out during or after precipitation
159 events to avoid contamination by rain water.

160 Rain samples were collected during the same study period (from July 2017 to
161 September 2018 with the exception of the period between November 2017 and March
162 2018) by continuously open rain gauges (Fuyuanming Inc., Tianjin, China). All parts
163 are made of stainless steel except the glass water collection bottle. Precipitation (snow
164 and rain) samples were collected by the rain gauges during the monitoring period.
165 Since the collector remained open, rainwater concentrations measured in this study
166 reflect additional inputs from dry deposition of aerosols and trace gases to the
167 collector surface. After each precipitation event, the samples were thoroughly stirred
168 and immediately transferred to clean polyethylene bottles (50 ml) for storage. The
169 rainwater collection bottle was rinsed with deionized water to eliminate cross-event
170 contamination. Further details of measuring methods and collection are given in our
171 previous studies (Xu et al., 2015, 2018, 2019a)

172 *2.3. Sample analysis and quality control*

173 After each sample collection, fog water or rain samples were immediately taken
174 into the laboratory and filtered with a 0.45 μm pore size cellulose acetate filter
175 (Tengda Inc., Tianjin, China), then divided into three equal parts that were used to
176 measure sample pH and concentrations of water-soluble ions (Na^+, NH_4^+, K^+, Mg^{2+},
177 Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) and trace elements (Mn, V, Cr, As, Se, Pb, Cd, Al). The
178 pH was measured by a pH meter (SG3, Mettler Toledo Company) as soon as possible
179 after filtration (pH meter calibrated against pH 4.00 and 6.86 buffers). The other two
180 sample portions were stored in a refrigerator at 4°C prior to ion and element analysis;
181 all samples were analyzed within 48 hours.

182 Water-soluble ions were analyzed by ion chromatography (IC, Dionex
183 Corporation). Anions were analyzed by an ICS-2100 ion chromatograph equipped

with a Dionex Ionpac AG11/AS11 guard/separation column pair using a potassium hydroxide eluent. Cations were analyzed on a DX-600 ion chromatograph equipped with a Dionex AG12A/CS12A guard/separation column pair using a methanesulfonic acid eluent; the detection limit was 0.02 μeq L^{-1} for Na^+ and K^+, 0.03 μeq L^{-1} for Mg^{2+} and Ca^{2+}, 0.06 μeq L^{-1} for NH_4^+, and 0.01 μeq L^{-1} for Cl^-, NO_3^-, and SO_4^{2-}. No rain or fog samples were below detection limits. Trace elements were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES, JOBIN-YVON), the detection limit was 0.001 mg L^{-1}. H^+ concentrations were calculated from the measured pH values.

2.4. Backward trajectory analysis

Air mass back-trajectory analysis was performed using the Hybrid-Single Particle Integrated Trajectory Model (HYSPLIT 4) (http://ready.arl.noaa.gov/HYSPLIT.php), provided by the Air Resource Laboratory of the National Oceanic and Atmospheric Administration (NOAA). Meteorological data were input from the Global Data Assimilation System (GDAS1 global, 2006 to present-ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/) with a model resolution of 1°×1°. Three-day (72 h) backward trajectories were calculated at 6 h intervals (06:00, 12:00, 18:00, 24:00 UTC) on sampling days, with an arrival height of 500 m (above ground level) for rain events and of 100 m for fog events. Cluster analysis was performed using the trajectories based on the total spatial variance (TSV) method (Draxler et al., 2012).

2.5. Statistical analysis

In this study, the Spearman correlation values between the sum of total cations ($Na^+ + NH_4^+ + K^+ + Mg^{2+} + Ca^{2+}$) and sum of total anions ($Cl^- + NO_3^- + SO_4^{2-}$) were 0.874 and 0.964 for fog and rainwater, respectively, suggesting that the measured ion balance in this experiment was credible. Additionally, the ratios of total cations to anions in fog and rain water were 3.1 and 2.8, respectively, suggesting that one or more major anions were missing in our determination. It has been reported that HCO_3^- and CO_3^{2-} were the dominant anions in precipitations and major rivers in Qinghai-Tibet plateau (Li et al., 2007; Xiang et al., 2009; Yang et al., 2012). The pH

value of atmospheric water is 5.6 for equilibrium with atmospheric carbon dioxide (Charlson and Rodhe, 1982). The amount of HCO_3^- was estimated using the following equation $HCO_3^-=10^{(pH-5.05)}$ (Xing et al., 2017), yielding estimated HCO_3^- concentrations on average of 24.2 and 16.9 μeq L^{-1} for rain and fog samples, respectively. These concentrations are not sufficient to explain the missing anions. Similar results were also reported for Qinghai-Tibet plateau in previous study (Li et al., 2007; Liu et al., 2015; Xing et al., 2017). Therefore, the relatively low total anion concentration was attributed to unmeasured organic acids (Migliavacca et al., 2005) and other unknown components. At the high pH values of these rain and fog samples, gas phase organic acids such as formic and acetic acid are highly soluble.

The Enrichment Factor (EF) has been widely used to examine the source contributions of major ions in both fog and rain water (Hissler and Probst 2006; Cong et al., 2010; Liu et al., 2015; Xu et al., 2017). Generally speaking, Na^+ can be considered as a typical marine source tracer while Ca^{2+} derives mainly from the continental crust. In this study, Na^+ and Ca^{2+} were therefore used as background tracer elements for marine and continental sources, respectively. The background of sea source elements is described in Keene et al., (1986) and continental source elements in Taylor (1964).

The EF of an ion concentration in fog (rain) relative to the concentration in sea was estimated as follow (Liu et al., 2015):

$$EF_{sea} = \frac{[X/Na^+]_{sample}}{[X/Na^+]_{sea}}$$

Similarly the EF of an ion concentration in fog (rain) relative to the ion in the continental crust was estimated as follow (Liu et al., 2015):

$$EF_{sea} = \frac{[X/Ca^{2+}]_{sample}}{[X/Ca^{2+}]_{soil}}$$

The sources of ions can be divided into ocean, land and human activities. In this study, the follow equations were used to calculate the ion sources in fog and rain water (Liu et al., 2015):

$$\%SSF = \frac{[X/Na^+]_{sea}}{[X/Na^+]_{sample}}$$

$$\%CF = \frac{[X/Ca^{2+}]_{soil}}{[X/Ca^{2+}]_{sample}}$$

$$AF = 100\%-\%SSF-\%CF$$

SSF refers to the sea salt fraction, CF to the crustal fraction and AF to the anthropogenic fraction; it should be noted that, if the SSF (CF) ratio was greater than 1, SSF (CF) was calculated as 1 minus CF (SSF).

Spearman correlation and principal component analyses have been widely used to determine natural and anthropogenic source contributions to chemical composition of precipitation (Cao et al., 2009; Sheu and Lin, 2011; Yue et al., 2014). In this study, Spearman correlation and principal component analysis were conducted using the SPSS software package, version 20.0 (SPSS Inc., Chicago, IL).

3. Results and discussion

3.1. Fog and rain chemistry characterization

As shown in **Table 1** and **Fig. 2**, the relative composition of fog water and rain water was similar, while all mean ion concentrations observed were higher in fog water than in rainwater. The concentration abundance order of ion concentrations in fog water was $Ca^{2+} > Cl^- > Na^+ > SO_4^{2-} > Mg^{2+} > NH_4^+ > K^+ > NO_3^-$ and an order of $Ca^{2+} > Na^+ > Cl^- > Mg^{2+} > SO_4^{2-} > NO_3^- > K^+ > NH_4^+$ was observed in rain water. Additionally, average total cations summed to 0.23 and 0.41 meq L^{-1} while total anions summed to 0.07 and 0.14 meq L^{-1} in rainwater and fog samples, respectively. The mean concentrations of NH_4^+, SO_4^{2-}, K^+, Ca^{2+}, Cl^-, NO_3^-, Mg^{2+}, Na^+ in fog were 4.6, 2.6, 2.3, 1.9, 1.9, 1.5, 1.3 and 1.3 times those in rainwater, respectively. The percentages of Ca^{2+}, NH_4^+ and K^+ in total cations were higher in fog than in rainwater. NH_4^+ increased from 3% in rain to 8% in fog water. The percentage of SO_4^{2-} was also higher in fog than in rainwater. By contrast, opposite behaviors were observed for Cl^- and NO_3^- (**Fig. 2a and b**).

In high-elevation environments, cloud and fog water have generally been recognized as being more acidic than rain (Anderson et al., 1999; Collett et al., 2002; Weathers et al., 1988; Herckes et al., 2002). Characterizing the acidity of fog and rain water is crucial to better understanding inputs of acidic deposition at high-elevation

locales. Higher acidity was observed in these Tibetan fog samples than in collected precipitation samples; but the difference was relatively small (6.4 versus 6.2). One possible explanation is that pH values in fog and rain water were strongly influenced by high Ca^{2+} concentrations.

The effect of cloud-precipitation concentration differences has previously been highlighted by other studies (Lovett et al., 1982; Collett et al., 1990, 1991a). Generally speaking, ion concentrations in fog water have been found to be higher than those in rain water; however, the differences have varied largely across regions(Chang et al., 2006; Aleksic et al., 2009). For instance, large variations for all ion concentrations between fog and rain water were found in NE Taiwan (Chang et al., 2006; Beiderwieden et al., 2007), the concentrations of Na^+, NH_4^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} in fog water were 53, 13, 9.4, 5.7, 15, 15 and 14 times higher than those in rain water, respectively. In contrast, minor differences of ion concentrations between fog and rain water were reported at Mangdang Mountain, China (Huo et al., 2010), the concentrations of Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-} in fog water were 5.0, 1.6, 1.7, 2.4, 3.7, 2.0, 3.5 and 1.6 times higher than those in rain water for that study, respectively. Compared to variations of ion conentrations between fog and rain water in other regions (Bridges et al., 2002; Aleksic et al., 2009; Straub et al., 2012), the differences of ion concentrations in Sejila were minor. One reason might be that ion concentrations in fog water were low due to a lack of major nearby emission sources from human activity (**Table 2**). A second factor is that precipitation samples were collected by continuously open rain gauges, resulting in some dry deposition being mixed in those samples, such that the measured ion concentrations in rain samples should be higher than in the local precipitation.

Compared with previous studies (**Table 2**), high Ca^{2+} concentrations were found in fog water samples. High Ca^{2+} concentrations are expected to be found in the Qinghai-Tibet plateau ambient environment, as with aerosol (Cong et al., 2007), rivers (Huang et al., 2008; Xiang et al., 2009), and soil (Li et al., 2009). Most areas of the Qinghai-Tibetan plateau are above 4,000 m altitude, with sparse vegetation, bare land and relatively high winds, and the weathering of the rocks on top of the mountain is

298 severe. Thus, concentrations of particulate Ca^{2+} in the atmosphere are relatively high
299 (Zhang et al., 2001) and expected to contribute to high Ca^{2+} concentrations in local
300 fog water. Moreover, the high Ca^{2+} concentrations reflect abundant atmospheric dust
301 particles that can be efficiently incorporated into precipitation though in-cloud and
302 below-cloud scavenging processes (Huang et al., 2014). As a result observed Ca^{2+}
303 concentrations in precipitation in the Tibetan plateau have been extremely high (Li et
304 al., 2007; Yang et al., 2012; Li et al., 2015; Liu et al., 2015). HCO_3^- was reported as
305 the main anion in precipitation, accounting for as much as 62% of the total anion
306 concentration. HCO_3^- was not measured in our work, one factor contributing to total
307 measured anion concentrations much lower than total cation concentrations in both
308 fog and rain samples (**Table 1**).

309 NH_4^+ concentrations in fog water averaged 4.6 times higher than in rain water
310 (**Table 1**). Often, rain and fog have different origins, with precipitation falling from
311 clouds formed at higher altitudes and fog forming in lower boundary layer air. High
312 NH_4^+ concentrations in fog water might reflect high atmospheric NH_3 concentrations
313 from local agricultural activity at our study site. Wang et al. (2019) has reported that
314 concentrations of NO_3^--N and NH_4^+-N in precipitation were similar in southeast Tibet,
315 but atmospheric NH_4^+-N (sum of gaseous NH_3 and particulate NH_4^+) concentrations
316 were much higher than NO_3^--N (sum of gaseous NO_2, HNO_3 and particulate NO_3^-)
317 concentrations. It is worth noting that NH_4^+ concentrations measured in this study
318 were relatively low compared with those in other areas (**Table 2**). One possible
319 explanation is that NH_3 volatilization from soil is limited by low temperature at the
320 altitude of 4000 m. SO_4^{2-} and NO_3^- mainly come from oxidation of sulfur dioxide and
321 nitrogen oxides, emitted by a wide range of anthropogenic sources, such as coal and
322 fuel combustion (Yang et al., 2012; Xu et al., 2019b). High concentrations of SO_4^{2-}
323 and NO_3^- have, not surprisingly been reported in areas near industrial districts
324 (Bridges et al., 2002; Strater et al., 2010). In this study, both SO_4^{2-} and NO_3^-
325 concentrations were relatively low (**Table 2**), consistent with a lack of substantial
326 anthropogenic activities in the study area.

327 *3.2. Concentration of trace elements in fog and rain water*

328 Although trace element concentrations are much lower than the major ion
329 concentrations, they can still play an important role in aqueous reactions in clouds
330 (e.g., Mancinelli et al., 2005; Harris et al., 2013). The composition of trace elements
331 in the atmosphere is mainly attributed to anthropogenic emissions, biogenic emissions,
332 biomass buring, and soil dust (Viana et al., 2009). Trace element concentrations
333 measured in fog water in this study are shown in **Table 3**, where concentrations of Al,
334 As, Mn and Se are seen to be higher than those of Pb and V. Cd and Cr were also
335 measured in the study, but concentrations were below our detection limit. Mn and Se,
336 essential micronutrient for plants, averaged 13.8 and 16.4 μg L^{-1}. Mn is also an
337 efficient catalyst for in-cloud reactions such as the oxidation of dissolved SO_2 to
338 sulfate (Rao and Collett, 1998; Harris et al., 2013). Al and As, considered as toxic
339 metals, had mean concentrations of 19.5 and 10.8 μg L^{-1}, respectively. All trace
340 elements in rain water were below the detection limit except Al and As, with mean
341 average concentrations of 2 and 6 μg L^{-1}, respectively.

342 Compared with more economically developed areas in China, such as Mt. Lu and
343 Mt. Tai (**Table 3**), all Sejila mountain trace element concentrations were lower except
344 for Se. Al, Ca, and Mn all can be derived from terrestrial sources (Cong et al., 2010).
345 In this study, the correlation (p<0.05) between Al and Ca was 0.469, while the
346 correlations between Al and Mn, and Ca and Mn reached 0.784 and 0.797 (both
347 p<0.01), respectively. These results suggest that those elements came mainly from
348 terrestrial sources in our research area. Pb and Se are considered primarily as
349 anthropogenic source elements. Considering the low anthropogenic activities in our
350 research area, concentrations of trace elements should be lower than in highly
351 populated regions. Interestingly, high concentraitons of As and Se were found in this
352 study; one explanation could be that those elements mainly derived from local crustal
353 material. Similar results were found in Qinghai-Tibetan plateau observations in other
354 previous studies (Cong et al., 2007; Wang et al., 2012). Vehicle exhaust might also be
355 a factor contributing to local atmospheric heavy metal concentrations because our
356 monitoring station was located close to the national road 318# (Tinus et al., 2010;
357 Srimuruganandam et al., 2011).

3.3. Source assessment of ions in fog and rain water

Air mass back-trajectories are frequently used to trace regional transport of atmospheric pollutants (Sun et al., 2015; Yue et al., 2014). In this study, 72 hour back-trajectories for each fog event (rainfall event) from August 2018 to September were computed. As shown in **Fig. S1a**, almost all air masses arrived from a southerly direction when fog events occurred. Additionally, all air masses from different directions were influnced by surface sources. As shown in **Fig. S1b**. major air mass transport patterns for rainfall events could be divided into 4 categories, the percentage of south, southwest, west and northwest source regions were 43%, 37%, 7% and 13% respectively, similar to findings in previous studies (Liu et al., 2015; Wang et al., 2018). These results reflect that southeast Tibet is influenced by the southwest monsoon and air mass transport from the Indian Ocean (Yang et al., 2012). Not surprisingly, therefore, high Na^+ concentrations were found in both rainfall and fog samples (**Table 1**).

Concentrations of water soluble inorganic ions have been extensively studied because they often dominate fog composition in areas with a continental background and high air pollution (Collett et al., 2002; Marinoni et al., 2004). Spearman correlation analyses have been widely used to examine the relationships among various water-soluble inorganic ions. The correlations between different ions in fog water were all significant (**Table S1**), perhaps reflecting simultaneous dilution (concentration) of all fog solutes with increasing (decreasing) fog liquid water content. In contrast, the correlation between more than half of the ions was not significant in rain water (**Table S2**).

Principal component analysis (PCA) as a multivariate statistical method is commonly used to find a small number of factors from a data set of many correlated variables (Brereton, 2003). The original data matrix is decomposed into the product of a matrix of factor loadings and a matrix of factor scores plus a residual matrix. The residual matrix contains the part of variance of the data set that cannot be explained by common factors (e.g. analytical uncertainties). On the basis of the correlation matrix, orthogonal factors, sorted by descending order (Marengo et al., 1995) are

388 extracted solving an eigenvalue problem. In general, the number of extracted factors
389 is lower than the number of measured features. After rotation of the factor loading
390 matrix, the factors can often be interpreted, for example, as origins or common
391 sources of pollutants. Based on the varimax rotated PCA, we characterized fog
392 samples using three factors which collectively explain 90.0% of the total variance
393 (**Table 4**). In fog water, PC1 is dominated by Mg^{2+}, Ca^{2+}, NO_3^- and SO_4^{2-}, and thus
394 can be regarded as crustal/soil dusts combined with transported anthropogenic
395 emissions. PC2 features high fractions of Na^+ and Cl^-, indicating a marine source. PC3
396 contained high levels of NH_4^+ and K^+, which could be explained by local source
397 emissions from agriculture and residential biomass burning which is commonly used
398 for local cooking and domestic heating. For rain water, three factors were identified
399 with the combined explained variance more than 89%. PC1 had high loading for Mg^{2+},
400 Ca^{2+}, Na^+, NO_3^- and SO_4^{2-}, and may be a combination of dust, sea salt, and long range
401 transported anthropogenic emissions. PC2 shows high loadings for K^+ and Cl^-,
402 suggesting a biomass burning source. The third component was NH_4^+, suggesting
403 contributions from an agricultural source, perhaps dominated by local emissions as
404 described above.

405 Recognizing that Na^+ is typically associated with a marine source and Ca^{2+} with
406 a terrestrial source, EF is frequently used to evaluate ion enrichments in precipitation.
407 If the EF value of an ion is higher (lower) than 1, this ion could be seen as enriched
408 (diluted) in the environment relative to the reference source composition. In this study,
409 NH_4^+, NO_3^- and SO_4^{2-} were all enriched for marine sources and terrestrial sources
410 (**Table S3**), suggesting that concentrations of those ions are influenced by human
411 activities. NO_3^- and SO_4^{2-} might come from India (**Fig. S1**) where anthropogenic
412 emissions of NO_x and SO_2 to the atmosphere significantly increased from 2001 to
413 2015 (Paulot et al., 2018). NH_4^+ might result from high ambient particulate NH_4^+
414 concentrations in summer in southeast Tibet (Wang et al., 2019) as well as from
415 scavenging of local agricultural NH_3 emissions. Both rain and fog showed similar
416 patterns for ion sources: Na^+, Mg^{2+} and Cl^- were mainly from a marine source, K^+ and

417 Ca^{2+} were mainly from a terrestrial source, and NH_4^+, NO_3^- and SO_4^{2-} were dominated
418 by anthropogenic emission **(Table 5**).

419 **4. Summary and conclusions**

420 This study analyzed the chemical characteristics of fog water and precipitation in
421 the southeast Tibet and examined potential sources of different ions. Air masses
422 associated with both fog and precipitation events arrived mainly from a southerly
423 direction, suggesting potential impacts of species transported over long distances from
424 India or the Indian Ocean. Concentrations of all measured ions were enriched in fog
425 water over rain, but both were dominated by Ca^{2+}. The major ion sources for fog and
426 precipitation showed a similar pattern. Na^+ and Mg^{2+} were mainly from marine
427 sources, Ca^{2+} was contributed mainly from abundant crustal dust aerosols, and SO_4^{2-}
428 and NO_3^- were dominated by transported anthropogenic emissions. Enriched NH_4^+
429 concentrations in fog water appeared to reflect local agricultural emissions of NH_3
430 while K^+ and Cl^- appeared to come from local residential biomass burning sources.
431 The most abundant trace elements in fog water were Al, As, Mn and Se,whereas only
432 Al and As were detected in rain water.

433 Because these are the first measurements of this kind in the region and because
434 the dataset is still relatively limited in scope, there remains a need for further research,
435 including better characterization of fog occurrence, liquid water content, and
436 composition over longer time periods. The enrichment of pollutant and nutrient
437 species in fog over rain and the frequent interception of fogs by the local forest
438 certainly points to a need for better examining potential contributions of occult (fog)
439 deposition to nutrient and pollutant deposition budgets for the local ecosystem.

440

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Author contributions

Xuejun Liu designed the research. Wei Wang and Wen Xu conducted the research (collected the data and performed the measurements) and wrote the manuscript. All authors were involved in the discussion and interpretation of the data as well as the revision of the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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839 **Figure captions**

840 Figure 1. Location and sampling environment of the study site at Sejila Mountain,
841 southeast Tibet

842 Figure 2. Comparison of the percentage of cations (a) and anions (b) based on
843 equivalents in fog and precipitation samples

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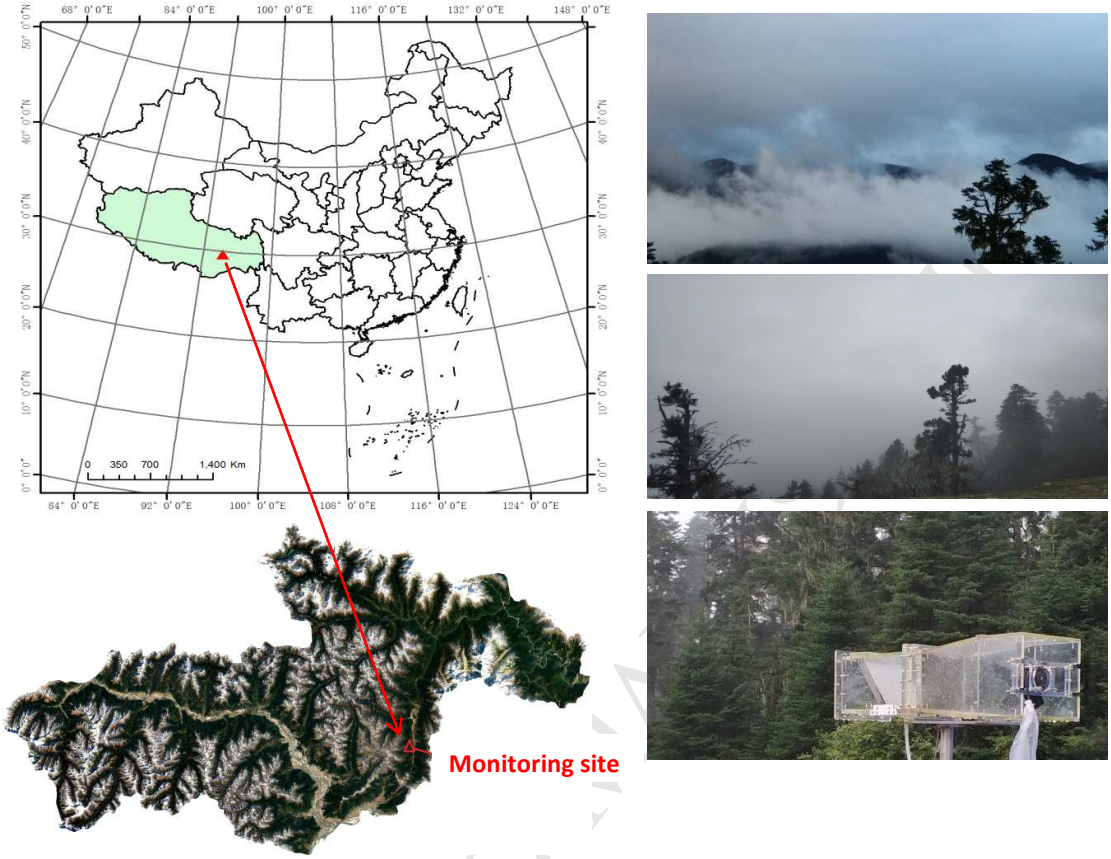
Figure 1



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881 **Figure 2**

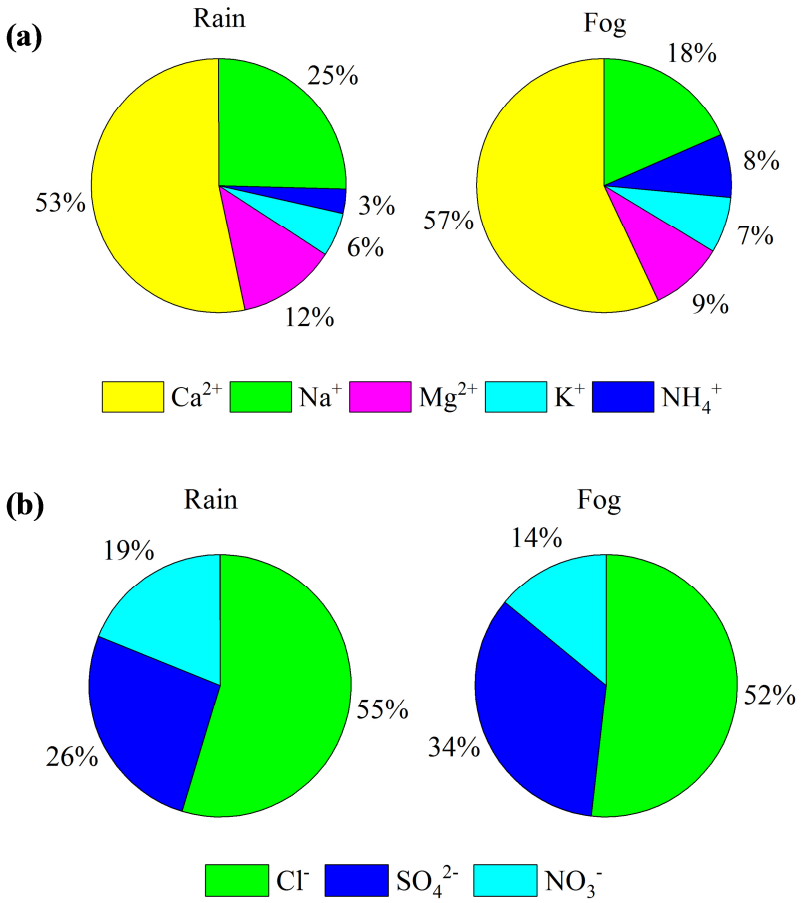

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Table 1. Concentrations (mean ± standard deviation, μeq L^{-1}) of water-soluble ions in rain and fog water at Sejila mountain.

	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	H^+	Total cations	Total anions
Rain	58.5±26.9	7.13±2.93	13.1±5.2	28.7±17.3	123±83	40.1±13.9	13.9±20.2	19.4±17.5	0.47±0.25	230	73
Fog	75.2±26.6	32.9±20.3	29.4±13.9	38.0±15.0	233±97	75.8±29.0	20.6±16.1	50.0±48.0	0.72±0.59	408	146

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915 **Table 2.** Comparison of concentrations (μeq L^{-1}) of chemical composition in fog water in previous studies.

Monitoring site	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	pH	Year	Reference
Northern Poland	19.4	16.5	5.9	1.5	8.7	18.1	14.7	17.8	6.1	2010	Polkowska et al. (2014)
Northeastern Taiwan	29.5	22.2	9.00	12.9	30.0	34.3	27.9	108	4.3	2000-2001	Chang et al. (2002)
Norway Sundsbø	69.7	3.90	1.90	11.7		102	5.3	13.1	5.0	2011	Wang et al. (2015)
Norway Bakka	75.1	10.3	2.60	14.2	5.5	106	<1	14.8	5.0		
Norway Hakadal	73.3	87.1	13.5	11.9	31.4	47	61.7	42.8	4.7		
whiteface mountain	3.10	117	1.60	5.00	17.8	5.60	79.9	200	-	1994-2006	Aleksic and Dukett, (2010)
Mangdang Mountain	25.0	63.0	5.00	12.0	92.0	12.0	105	63.0	4.8-6.0	2009	Huo et al. (2010)
Germany	35.0	216	6.10	5.10	9.80	30.0	164	43.0	4.3	2010	van Pinxteren et al. (2016)
South Korea	69.0	173	10.1	22.2	59.8	74.9	93.0	176	4.4	2002-2003	Kim et al. (2006)
Southern China	21.9	276	14.6	9.90	62.2	27.9	182.8	341	3.8	2011-2012	Sun et al. (2015)
NE taiwan	58.6	235	10.3	18.8	34.1	42.8	179	401	3.6	2006	Beiderwieden et al. (2007)
Swiss 2330 m a.s.l	43.0	143	5.00	12.6	46.8	10.6	87.0	72.3	6.4	2006-2007	Michna et al. (2015)
Swiss 1650 m a.s.l	44.3	249	10.3	13.9	35.4	20.5	200	105	6.6		
Swiss 682 m a.s.l.	78.0	1030	96.1	139	117	44.0	347	334	7.3		
Swiss (inside forest canopy)	38.2	845	72.0	54.1	32.8	32.8	294	297	7.2		
Czech	64.4	203	19.4	404	136	155	726	1250	3.0	1995-1996	Bridges et al.(2002)
Poland	130	230	50.0	31.0	100	140	170	220	4.1-5.1	2005-2006	Blas et al. (2010)
Ailaoshan Mountain	6.09	727	25.3	7.41	27.5	24.3	69.2	185	4.05	2015	Nieberding et al., 2018
Cervenohorske sedlo	26.1	44.3	7.7	<8.2	25	<19.7	<29	127	5.2	2002	Zapletal,et al.,2007

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Table 3. Comparison of concentrations (μg L-1) of trace elements in fog water at Sejila Mountain with other areas

Sites	Al	As	Mn	Pb	Se	V	Cd	Cr	Year	References
Lu Mountain	111	20.4	16.4	54.4	7.40	5.90	1.66	8.37	2011-2012	Sun et al. 2015
Tai Mountain	157	13.7	42.8	46.2	n.a.	n.a.	3.08	0.93	2007	Liu et al. 2012
Yangtze River	391	3.38	40.0	5.83	2.10	3.65	1.37	4.96	2015	Xu et al. 2017
Northern Poland	9.50	n.a.	1.86	1.06	n.a.	n.a.	7.78	n.a.	2010	Polkowska et al. 2014
Northern Chile	n.a.	9.10	<200	<10	<5	n.a.	<0.5	<2.5	2008	Sträter et al. 2010
Elden Mountain	16.6	0.80	34.0	0.40	2.00	3.20	n.a.	1.70	2005-2007	Hutchings et al. 2009
Sejila Mountain	19.5	10.8	13.8	1.23	16.4	4.38	n.a.	n.a.	2017-2018	This study

Note: n.a. denotes not available.

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Table 4. Varimax-rotated principal component analysis of major ions of major ions in fog and rain at Sejila Mountain.

		Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Variance (%)	Cumulative (%)
Fog	PC1	0.329	0.515	0.129	**0.731**	**0.685**	0.272	**0.879**	**0.941**	71.1	71.1
	PC2	**0.888**	0.105	0.584	0.473	0.414	**0.874**	0.254	0.252	12.5	83.6
	PC3	0.230	**0.683**	**0.749**	0.443	0.497	0.193	0.302	0.110	6.6	90.0
	CT	0.951	0.743	0.919	0.955	0.887	0.875	0.928	0.960		
Rain	PC1	**0.656**	0.072	-0.062	**0.957**	**0.927**	0.567	**0.857**	**0.970**	59.8	59.8
	PC2	0.518	0.182	**0.897**	-0.028	0.229	**0.700**	0.212	0.075	20.2	80.0
	PC3	0.334	**0.938**	0.290	-0.169	0.182	-0.092	0.294	0.004	9.4	89.3
	CT	0.811	0.918	0.893	0.945	0.945	0.820	0.865	0.947		

Note: PC1, PC2 and PC3 indicate the first, second and third component, respectively. CT means communality.

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Table 5. Source contributions (%) for major ions in fog and precipitation at Sejila Mountain.

		Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Fog	SSF	100	2.0	3.3	84.2	1.7	77.1	2.2	8.7
	CF		0.5	96.7	15.8	98.3	0.5	0.8	3.6
	AF		97.5				22.4	97.0	87.7
Rain	SSF	100	7.3	5.8	86.7	2.5	99.5	2.6	17.4
	CF		1.2	94.2	13.3	97.5	0.5	0.6	5.0
	AF		91.5					96.8	77.6

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Highlights

1. Characteristics of ions and trace elements in fog water in the Sejila mountain of southeast Tibet were reported.
2. Ion and trace element concentrations were higher in fog water than in rain water but pH values were on the contrary.
3. Ratios of NH_4^+-N and NO_3^--N were much higher in fog water than in rain water.
4. Local source contributed mainly to atmospheric ions in the Sejila mountain.

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Competing interests

The authors declare that they have no conflict of interest.

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