Dear Dr. Zhang,

Thanks for your work on our manuscript. We greatly thank two anonymous referees for providing constructive comments. The comments are very important for improving our manuscript. We evaluated all comments, and revised the manuscript carefully. Please find the point-by-point response to the referees as below (reviewer's comments in black, authors' responses in blue). The marked-up manuscript version was combined at the end of the response letter. Please note that the line numbers in the following text indicate the line numbers in the revised version of the manuscript, but NOT the marked-up manuscript version.

To address concerns raised by the referees, major revisions were made mainly as follows.

**Title**: The title was changed to "Wet deposition of atmospheric inorganic nitrogen at five remote sites on the Tibetan Plateau" (L1-2).

**Abstract**: Seasonal variation and regional differences of inorganic N wet deposition at the five sites on the TP were added in the abstract (L22-29).

**Materials and Methods**: We explained the resource of $Na^+$ and $Ca^{2+}$ in precipitation on the TP more clearly in the Materials and Methods section (L155-180). To examine the resource of inorganic N wet deposition on the TP more clearly, principal component analysis and backward trajectory analysis were added in the revised manuscript (L202-L222).

**Results**: We added 2 sections: "3.3 Seasonal dynamics of inorganic N wet deposition" (L253-258) and "3.4 Sources of inorganic N wet deposition" (L259-300) in the revised manuscript. Section 3.4 include three parts: 3.4.1 Enrichment factors (L260-274), (2) principal component analysis (L275-289) and (3) backward trajectories analysis results (L290-300).

**Discussion**: Section 4.2 was almost rewrote in the revised manuscript. In this section, we discussed the resource of inorganic N wet deposition on the TP, not only using enrichment factor method, but also using principal component analysis (L334-355) and backward trajectory analysis (L372-388). The discussion of the uncertainty of our short-term in situ measurements was strengthened in section 4.4 (L469-474).

**Tables**: We added a new table to show the results of principal component analysis of major ions in precipitation at five remote sites on the TP (Table 6).

**Figures**: We added a new figure to show the results of seven-day backward trajectories at five remote sites on the TP (Fig. 5).

**References**: Several references were added in the revised manuscript. Please see L554-556, L564-567, L570-572, L573-575, L586-588, L589-591, L609-611, L612-615, L637-639, L640-642, L652-654, L678-680, L681-683, L695-696, L726-728, L735-737, L741-743, L744-746, L797-798, and L811-814.

**Supplementary material**: We added a new table to show the results of Pearson correlations matrix of ionic concentrations in precipitation at five remote sites on the TP (Table S1). The spatial distribution data of inorganic N wet deposition on the Tibetan Plateau (Figure S1) was provided as a NetCDF file in the supplementary material in the revised manuscript.

Please contact us if any further information is requested. In advance, thank you for your consideration.

Sincerely yours,

Yongwen Liu
Xu-Ri
Yuesi Wang
Yuepeng Pan
Shilong Piao

**Responses to Anonymous Referee #1**

*RC* – Reviewer's Comments, *AC* – Authors' Comments.

*AC*: We greatly thank Anonymous Referee #1 for providing constructive comments, which are important for improving our manuscript. The comments were carefully evaluated. Based on the comments, we have revised the manuscript. The detailed responses to the comments are shown as below (reviewer's comments in black, authors' responses in blue).

*RC*: The authors present atmospheric nitrogen deposition based on a one-year field observation in a typical alpine ecosystem - Tibetan Plateau, China, which is an important topic and falls within the scope of the journal. While I appreciate the nature of the data set and agree that the regional data is necessary and valuable, the authors maybe neglect its reliability. Meteorological conditions are not only complex but also highly variable. Therefore, with only one-year data, it is difficult to get a factual picture of nitrogen deposition at a location. And there is lack of comparability at the five sites for the different experiment years. As it stands the current manuscript is not suitable for publication. Thus I suggest the MS need to be revised and then be reviewed.

*AC*: Thanks for the reviewer's appreciation on our work. As the reviewer has pointed out, our regional assessment of inorganic nitrogen (N) deposition based on *in situ* measurements in the Tibetan Plateau (TP) is necessary and valuable. The TP covers about 2.57 million $km^2$, occupying approximately 1/4 of the land area of China. Alpine ecosystems are widely distributed on the TP, which are sensitive to elevated N deposition. Ice core records show that the inorganic N deposition on the TP has increased during recent decades. Clarification of N wet deposition on the TP not only is conducive to accurately estimating N wet deposition for the entire nation of China, but also can provide background information for the studies focusing the alpine ecological effects of elevated N deposition. However, inorganic N deposition on the TP remains unclear, due to scarcely N deposition monitoring, especially in the central and western TP.

In this study, we preformed short-term *in situ* measurements of inorganic N wet deposition at 5 remote sites in various typical alpine ecosystems mainly on the central and western TP. Combing our *in situ* measurements and those in previous studies, we assessed the amount of inorganic N wet deposition for the entire TP. Results reveal that previous interpolation and simulation studies highly overestimated the inorganic N wet deposition for the entire TP. Thus, compared to previous estimation of inorganic N wet deposition on the entire TP, either based on interpolation at national scale or model simulation at global scale, we provided a clearer picture of inorganic N wet deposition on the entire TP. Indeed, meteorological conditions are not only complex but also highly variable. Precipitation is the meteorological condition most closely related to inorganic N wet deposition. Herein, the datasets of precipitation during recent years at the remotes sites in this study were collected and analyzed to examine the reliability of our short-term *in situ* measurements. Continuous precipitation data for at least 3 years was available at four sites: Southeast Tibet Station (2007-2012), Nam Co Station (2007-2013), Qomolangma Station (2010-2012), and Ngari Station (2010-2013). Fig. R1 shows the inter-annual variability of precipitation and mean annual precipitation at the four sites. Although with only one-year, the annual precipitation in this study is comparable with the mean annual precipitation at each site (Fig. R1). Across the four sites, the difference of annual precipitation in this study is also comparable with the difference of mean annual precipitation, although our short-term *in situ* measurements at the four sites were not conducted at an identical period (Fig. R1). These results are likely expected, because the observation sites in our study are located in different climate zones from southeastern TP to western TP. Although meteorological conditions are complex and highly variable, the climatic characteristics and the difference among climates are likely relatively stable during consecutive several years.

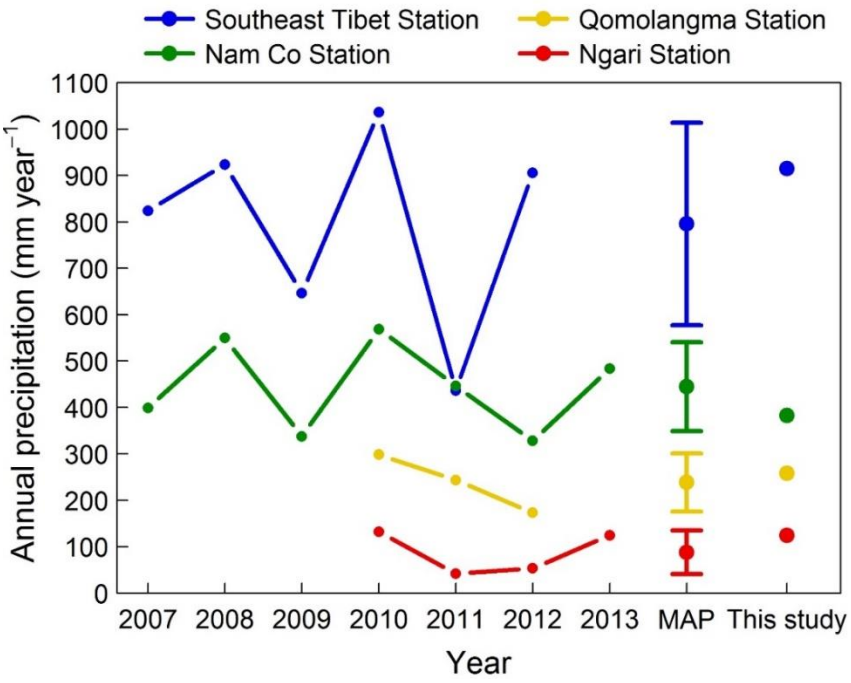


Figure R1. Annual precipitation at four sites on the TP. MAP indicates mean annual precipitation (mean±1 standard deviation).

Overall, our estimation of the inorganic N wet deposition at the remote sites on the TP is probably reliable, although with only one-year *in situ* measurements. Meanwhile, we acknowledge that our short-term *in situ*

measurements can't clarify the long-term trend and inter-annual variability of inorganic N wet deposition on the TP. Moreover, this study focused the wet deposition of inorganic N on the TP, yet with the dry deposition of N on the TP remaining unclear. We therefore recommend conducting long-term monitoring of both wet and dry deposition of N to more clearly clarify N deposition on the TP in the future work.

*RC*: Specific comments:

*RC*: Title: "Wet deposition of atmospheric inorganic nitrogen at five remote stations on the Tibetan Plateau". The MS research only focuses on the western (and central) Tibetan Plateau. Thus, it is better to be more specific.

*AC*: In this study, we preformed short-term *in situ* measurements of inorganic N wet deposition at 5 remote sites mainly on the central and western TP. Based on *in situ* measurements in this and previous studies, we assessed the inorganic N wet deposition for the entire TP. Nevertheless, our manuscript was entitled "Wet deposition of atmospheric inorganic nitrogen at five remote stations on the Tibetan Plateau". This title has been specified to site-scale ("…at five remote stations…"), rather than regional-scale (e.g. the entire TP). In the revised manuscript, we entitled the manuscript at site-level more clearly as "***Wet deposition of atmospheric inorganic nitrogen at five remote sites on the Tibetan Plateau***" (L1-2). Certainly, when more suitable title is available, we are open to alteration.

*RC*: Abstract: The part is not focused well enough. It should state the characteristics of wet deposition including its flux and seasonal variation, its regional differences including the typical factor and the important suggestions for regional development and the protection of alpine ecosystem. However, it only focused on wet deposition flux and the comparison with previous study in other regions in Tibetan Plateau.

*AC*: We agree with the reviewer that the characteristics of inorganic N wet deposition should be stated more clearly. The flux of inorganic N wet deposition has been included in the manuscript. In the revised manuscript, seasonal variation and regional differences of inorganic N wet deposition at the five sites on the TP were added in the abstract as "***The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all stations. Both $NH_4^+$-N and $NO_3^-$-N wet deposition on the TP were mainly influenced by anthropogenic activities. Backward trajectory analysis showed that the inorganic N deposition at Muztagh Ata Station was mainly transported from Central Asia and East Asia through westerlies. At Southeast Tibet Station, Nam Co Station, Qomolangma Station and Ngari Station, the inorganic N deposition was mainly contributed by anthropogenic sources in South Asia, and was mainly transported by Indian monsoon.***" (L22-29)

We agree that it is important to examine the possible responses of alpine ecosystem in the TP to elevated N deposition, and then provide suggestions to regional development and the protection of alpine ecosystem. However, this is another topic, which is difficult to be clarified by the *in situ* measurements of inorganic N wet deposition at 5 remote sites on the TP in this study. Actually, we have performed a multi-level N fertilization experiment in an alpine steppe on the TP to examine the ecological effects of simulated N addition (Liu et al., 2013). Nevertheless, direct evidences are still needed to clarify the comprehensive effect of current elevated inorganic N deposition to the regional development and alpine ecosystems in the TP. Hence, in the abstract

of the manuscript focusing on the amount of inorganic N wet deposition on the TP, we yet did not provide suggestions for regional development and the protection of alpine ecosystem, but recommended conducting long-term monitoring of both wet and dry deposition of N in various climate zones to clarify N deposition on the TP more clearly.

In addition, in the abstract, we provided field observation results of inorganic N wet deposition at 5 remote sites on the TP, and assessed the inorganic N wet deposition for the entire TP based on our and previous site-scale *in situ* measurements. Then, this regional-scale assessment based on *in situ* measurements, was NOT compared with the previous site-scale *in situ* measurements in other regions in the TP, but with previous regional-scale estimation of inorganic N wet deposition for the entire TP based on interpolations or model simulations. This point was expressed more clearly in the abstract of the revised manuscript.

*RC*: Results: It is better to introduce in this part the characteristics of wet inorganic nitrogen deposition just as the "3.2 wet deposition of atmospheric inorganic N section". Thus, this part may include three sections: 1) flux; 2) seasonal variation; 3) sources. In addition, more information is needed for the discussion of nitrogen deposition at each site, such as the wind direction, air temperature, and local human activities, and so on.

*AC*: We agree. The flux of wet inorganic N deposition has been included in the section of "3.2 Wet deposition of atmospheric inorganic N". To adequately explain the characteristics of the inorganic N wet deposition at the 5 sites, we added 2 sections: "3.3 Seasonal dynamics of inorganic N wet deposition" and "3.4 Sources of inorganic N wet deposition" in the revised manuscript. Indeed, more in-depth analysis is needed for the discussion of N deposition at each site. In the revised manuscript, we added results of (1) principal component analysis (Table 6) and (2) backward trajectories (Fig. 5) in the results section, and strengthened the discussion of N deposition at each site in the discussion section based on those added analysis. The detailed modifications were shown in the results section of the revised manuscript as follows (L253-300):

> ***3.3 Seasonal dynamics of inorganic N wet deposition***
>
> ***The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all sites (Fig. 4). Both concentrations of $NH_4^+$ and $NO_3^-$ did not exhibited any clear seasonal pattern (Fig. 2). The seasonal dynamics inorganic N wet deposition at most stations appeared the shape of single peak type (Fig. 4). The seasonal pattern of inorganic N wet deposition were similar to the seasonal pattern of precipitation, rather than that of $NH_4^+$ or $NO_3^-$ concentration (Fig. 2).***
>
> ***3.4 Source assessment of wet deposition of inorganic N and other ions***
>
> ***3.4.1 Enrichment factors***
>
> ***Table 4 shows the EFs of precipitation constituents at the five sites relative to seawater and soil. If the EF value of an ion in precipitation is much higher (lower) than 1, the ion is considered to be enriched (diluted) relative to the reference source. Among the five sites, $Cl^-$ had a relatively lower $EF_{sea}$ value, ranging from 0.50 (Nam Co Station) to 0.90 (Qomolangma***

*Station), but a relatively higher $EF_{soil}$ value, ranging from 42.4 (Muztagh Ata Station) to 286 (Qomolangma Station). Different from $Cl^-$, $NH_4^+$ in precipitation was enriched relative to both marine origin and soil reference source at all sites, because its $EF_{sea}$ values ranged from 11629 to 80684, and its $EF_{soil}$ from 350 to 2378. Similar to $NH_4^+$, $NO_3^-$ also had a relatively high value of both $EF_{sea}$ and $EF_{soil}$ at all five sites.*

*Table 5 shows the source contributions for major ions in precipitation of the five remote sites in this study. Almost all $Cl^-$ and $Na^+$ in precipitation on the TP appeared to be of marine origin, with SSF value above 95% at the five sites. Nearly all $Ca^{2+}$ in precipitation came from crust at the five sites, with the CF value being above 90%. Among the five sites, anthropogenic sources contributed at least 99% of $NH_4^+$ in precipitation, and $NO_3^-$ in precipitation was also mainly influenced by anthropogenic activities, with AF values ranging from 95.3% to 99.9%.*

### *3.4.2 Principal component analysis*

*Table 6 shows the 1st, 2nd and 3rd component of principal component analysis, which account for at least 85% of the total variance across the five sites. $Na^+$ and $Cl^-$ were mainly explained by the same component at all sites. Principal component analysis shows that the variances of $Ca^{2+}$ and $Na^+$ were represented by different components at four of five sites (except Southeast Tibet Station) (Table 6). The common variance of $Ca^{2+}$, $Mg^{2+}$ and $SO_4^{2-}$ as 1st component represents the largest proportion of the total specie variation at the 3 sites (Nam Co Station, Ngari Station and Muztagh Ata Station) in the central and western TP (Table 6). At Qomolangma Station, $Na^+$, $Cl^-$, $K^+$ and $NH_4^+$ as 1st component represents the largest proportion of the total specie variation (Table 6). Except for Qomolangma Station, at the other four sites, the variances of $NH_4^+$ were mainly represented by 3rd component (Table 6). At Southeast Tibet Station, both $Ca^{2+}$ and $Na^+$ variances were mostly represented by the 1st component, but $NO_3^-$ variances were mainly represented by the 2nd component (Table 6). At Nam Co Station, Qomolangma Station and Ngari Station, $NO_3^-$ variances were mainly represented by the 3nd component, which were different with that of both $Ca^{2+}$ and $Na^+$ (Table 6). However, $NO_3^-$ variances were mainly represented by the 1st component at Muztagh Ata Station (Table 6).*

### *3.4.3 Backward trajectory analysis*

*Fig. 5 shows the seven-day backward trajectories of air mass arriving at the five remote sites at the sampling days. The transport pathways of air masses were various with the different sites (Fig. 5). The cluster trajectory results showed that at Muztagh Ata Station, nearly all air masses at sampling days were transported from Central Asia and Middle East (Fig. 5a). Different to Muztagh Ata Station, almost all air masses at Nam Co Station were transported from South Asia (Fig. 5d). For Ngari Station, Qomolangma Station, and Southeast Tibet Station, the air masses at sampling days were mainly transported from South Asia, with the proportion of 90%, 79.8% and 90.6%, respectively (Figs. 5b-5e). Besides South Asia, Central Asia, Qaidam Basin and Middle East was the second source of air masses at sampling days*

***for Ngari Station, Qomolangma Station, and Southeast Tibet Station, respectively (Figs. 5b-5e).***

*RC*: Discussion: the authors concluded that the inorganic N wet deposition for the entire Tibetan Plateau in previous studies (either through atmospheric chemistry transport model simulations or interpolations based on limited observations) was highly overestimated, whereas they estimated N wet deposition in Tibetan Plateau by combining their field observations with previous studies. In addition, the inorganic N wet deposition on the Tibetan Plateau has increased during recent decades according ice core records. Accordingly, uncertainty analysis should be strengthened.

*AC*: Yes, we concluded that previous regional-scale assessment of inorganic N wet deposition for the entire TP, either based on interpolation at national scale or model simulation at global scale, was highly overestimated. However, when estimating the inorganic N wet deposition for the entire TP in this study, we did NOT use these previous regional-scale assessments, but comprehensively used our site-scale *in situ* measurements at the 5 sites and previous site-scale *in situ* measurements at the other sites on the TP. This point was expressed more clearly in the discussion section of the revised manuscript.

Indeed, the inorganic N deposition on the TP has shown increasing trend during recent decades according records of both ice cores and sediment cores in alpine lakes. We acknowledge that the long-term trend of inorganic N deposition can't be quantified by our short-term *in situ* measurements. In order to quantify the long-term trend of inorganic N deposition on the TP, long-term *in situ* monitoring is still necessary. Therefore, we recommend conducting long-term monitoring of both wet and dry deposition of N in various climate zones on the TP in the future work. The discussion of the uncertainty of our short-term *in situ* measurements was strengthened in the revised manuscript (L469-474).

**Reference**

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**Responses to Anonymous Referee #2**

*RC* – Reviewer's Comments, *AC* – Authors' Comments.

*AC*: We appreciate the insightful comments from Anonymous Referee #2. The comments are thought-provoking, and help us deepen our understanding of the source assessment of ions wet deposition on the Tibetan Plateau (TP). Our manuscript was revised according to the comments. The detailed responses to the comments are shown as below (reviewer's comments in black, authors' responses in blue).

*RC*: The manuscript is well writen and should be accepted for publishing in ACP. This reviewer has a minor comment for the authors considering. Biomass burning is an important air pollution source in the Tibetan Plateau and can also be a significant source of Na in wet deposition. The use of Na as the tracer of sea-salt

does not sound scientific. Construction can be a source of Ca, but this reviewer is not sure the importance in the Tibetan Plateau. The authors are encouraged to clarify this. Overall, Section 4.2 does not make any sense to this reviewer and needs revision.

*AC*: We agree that biomass burning is an important air pollution source in the TP. In 2010, 6300 Gg of dry biomass are estimated to be subjected to field burning of crop residue in India, resulting in 350 GgN emissions (Sahai et al., 2011). Various trace gases and aerosols can be emitted due to biomass burning (Andreae and Merlet, 2001). For instance, in the year 2000, biomass burning in Asia led to the emission of 34.3 Tg $SO_2$, 26.8 Tg $NO_x$, 9870 Tg $CO_2$, 279 Tg CO, 107 Tg $CH_4$, 52.2 Tg non-methane volatile organic compounds, 2.54 Tg black carbon, 10.4 Tg organic carbon, and 27.5 Tg $NH_3$ (Streets et al., 2003). However, as far as we know, water-soluble Na was not major component of total ions in trace gases or aerosols emitted by biomass burning in Asia. For example, biomass burning in Nepal and northern India is an important source of the carbonaceous aerosol at Qomolangma Station on the northern slope of the Himalayas (Cong et al., 2015). However, water-soluble $Na^+$ and $Cl^-$ consisted of a very minor portion of total ions in aerosols at Qomolangma Station (Cong et al., 2015).

Multiple lines of evidence demonstrate that $Na^+$ in precipitation on the TP mainly comes from oceans. Balestrini et al. (2014) monitored the chemical and isotopic compositions of precipitations at the Pyramid International Laboratory (5050m a.s.l.) on the southern slope of the Himalayas, and data analysis suggested that $Na^+$ and $Cl^-$ were derived from the long-range transport of marine aerosols. Ice records in the central Himalayas show that $Cl^-/Na^+$ was positively related with the monsoon rainfall in northeast India, and there was a teleconnection between the $Na^+$ and $Cl^-$ concentrations and the North Atlantic Oscillation, indicating $Na^+$ in the ice core mainly came from oceans (Wang et al., 2002). To identify potential precursors of major ions wet depositions at the five remote sites in our study, we performed a Varimax-rotated principal component analysis. Table 6 shows the 1st, 2nd and 3rd component, which account for at least 85% of the total variance across the five sites. $Na^+$ and $Cl^-$ were mainly explained by the same component at all sites. This indicates that $Na^+$ and $Cl^-$ were likely contributed by the same source: sea-salt. This assumption was also confirmed by the relatively high Pearson correlation between $Na^+$ and $Cl^-$ at all five sites (Table S1). In addition, $Na^+$ has been used as a marine tracer when analyses the source contributions of ions wet deposition on the northeastern TP (Li et al., 2015), the southeastern TP (Liu et al., 2013) and the southern slope of central Himalayas (Tripathee et al., 2014). Therefore, based on our analysis and previous studies, we suggested that $Na^+$ wet deposition on the TP was mainly contributed by sea-salt, rather than biomass burning.

On the TP, sandy desertification land covers about $3.1\times10^5$ $km^2$, accounting for 14% of the whole plateau, of which moderate sandy desertification land occupies 55.44% (Liu et al., 2005). The TP was regarded as an important dust source region (Fang et al., 2004;Han et al., 2009;Han et al., 2008). The TP dust sources contribute 69% of dust at the surface and 40% of dust in the lower troposphere on the TP (Mao et al., 2013). Moreover, arid regions are widely distributed surrounding the TP, e.g. central Asia, deserts of western China. The dust on the TP partly comes from the adjacent dust source regions, e.g. Taklimakan Desert in western China (Huang et al., 2007;Xia et al., 2008). Atmospheric dust aerosols over the TP are strongly impacted by local sources and enriched with Ca (Zhang et al., 2001). These dust aerosols in the atmosphere can interact with clouds and precipitation (Huang et al., 2014), and deposit on the surface with precipitation. Thus, $Ca^{2+}$ is commonly used as a proxy of dust for ice core studies in the TP (Kang et al., 2002;Kang et al., 2010;Kaspari

et al., 2007;Wang et al., 2008). As a dust proxy, $Ca^{2+}$ record in an ice core from the central TP even was significantly related regional zonal wind (westerlies) trends, and reflected the long-term control of regional atmospheric circulation strength over atmospheric dust concentrations (Grigholm et al., 2015). In the revised manuscript, backward trajectories analysis was added to examine the transport pathways of air masses arriving at the five remote sites at the sampling days. Fig. 5 shows the seven-day backward trajectories at five remote sites on the TP. The cluster trajectory results showed that at Muztagh Ata Station, nearly all air masses at sampling days were transported from Central Asia and Middle East (Fig. 5). At the other four sites, the air masses at sampling days were mainly transported from South Asia (Fig. 5). As expected, the precipitation at Muztagh Ata Station has the highest $Ca^{2+}$ concentrations of 119.4 μeq $L^{-1}$, which is much higher than the $Ca^{2+}$ concentrations in precipitation at the other four sites (7.9~53.4μeq $L^{-1}$). In addition, Ca also has been used as a reference element for continental crust when assess source of ion wet deposition in precipitation on the northern TP (Li et al., 2015).

In our study, $Ca^{2+}$ is a major ion in the precipitation at all five sites, representing 26.1%-54.6% of total measured specie burdens. Principal component analysis shows that variances of $Ca^{2+}$ and $Na^{+}$ were represented by different components at four of five sites (except Southeast Tibet Station), indicating different source of $Ca^{2+}$ and $Na^{+}$ in precipitation on the TP (Table 6). The common variance of $Ca^{2+}$, $Mg^{2+}$ and $SO_4^{2-}$ as 1rd component represents the largest proportion of the total specie variation at the 3 sites (Nam Co Station, Ngari Station and Muztagh Ata Station) in the central and western TP (Table 6). This is probably because these 3 sites are located relatively closer with central Asia and deserts of western China, and are more severely influenced by westerlies than the other 2 sites (Qomolangma Station and Southeast Tibet Station) (Table 6). At Qomolangma Station, $Na^{+}$, $Cl^{-}$, $K^{+}$ and $NH_4^{+}$ as 1st component represents the largest proportion of the total specie variation, indicating more severely influence of Indian Monsoon in monsoon period. However, at Southeast Tibet Station, both $Ca^{2+}$ and $Na^{+}$ variances were mostly represented by the 1st component (Table 6). This probably because South Asia is an important source of dust aerosols in the Southeastern TP during the during the monsoon period (Zhao et al., 2013). Overall, $Ca^{2+}$ in precipitation on the TP is mainly contributed by continental source, rather than marine and anthropogenic source. Therefore, it is reasonable to $Ca^{2+}$ as a reference element for continental crust when assess source of ion wet deposition in precipitation on the TP.

In the revised manuscript, we explained the resource of $Na^{+}$ and $Ca^{2+}$ in precipitation on the TP more clearly in the Materials and Methods section (L155-180). Moreover, we discussed the resource of inorganic N wet deposition on the TP, not only using enrichment factor method, but also using principal component analysis (L334-355) and backward trajectory analysis (L372-388). Section 4.2 was almost rewrote in the revised manuscript.

**Reference**

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# Wet deposition of atmospheric inorganic nitrogen at five remote ~~stations~~ sites on the Tibetan Plateau

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## Abstract

Since the mid-20th century, nitrogen (N) deposition has shown an increasing trend on the Tibetan Plateau (TP), where ~~A~~alpine ecosystems ~~on the Tibetan Plateau are~~ are sensitive to elevated ~~nitrogen (~~N~~)~~ deposition~~, and N wet deposition in this region has shown an increasing trend since the mid-20th century~~. However, the quantitative characterization ~~amount~~ of N ~~wet~~ deposition on the ~~Tibetan~~ TP remains unclear, due in most part to the lack of *in situ* measurement~~direct observations~~. Using the Tibetan Observation and Research Platform network, we ~~investigated~~ conducted short-term *in situ* measurements of ~~wet deposition of the~~ major ions ($NO_3^-$, $Cl^-$, $SO_4^{2-}$, $NH_4^+$, $Na^+$, $K^+$, $Ca^{2+}$ and $Mg^{2+}$) wet deposition ~~–~~at five remote ~~stations~~sites on the TP during 2011-2013. At Southeast Tibet Station,

Nam Co Station, Qomolangma Station, Ngari Station, and Muztagh Ata Station, the $NH_4^+$-N wet deposition was 0.63, 0.~~91~~68, ~~1.61~~0.92, 0.36 and 1.25 kg N $ha^{-1}$ $yr^{-1}$, respectively; the $NO_3^-$-N wet deposition was 0.28, 0.~~35~~24, 0.~~04~~03, 0.08 and 0.30 kg N $ha^{-1}$ $yr^{-1}$, respectively; and the inorganic N deposition was 0.91, ~~1.26~~0.92, ~~1.64~~0.94, 0.44 and 1.55 kg N $ha^{-1}$ $yr^{-1}$, respectively. The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all stations. Results of enrichment factor analysis and principal component analysis demonstrated that both $NH_4^+$-N and $NO_3^-$-N wet deposition on the TP were mainly influenced by anthropogenic activities. Backward trajectory analysis showed that the inorganic N deposition at Muztagh Ata Station was mainly transported from Central Asia and Middle East through westerlies. At Southeast Tibet Station, Nam Co Station, Qomolangma Station and Ngari Station, the inorganic N deposition was mainly contributed by anthropogenic sources in South Asia, and was mainly transported by Indian monsoon. Combining site-scale *in situ* measurements of inorganic N wet deposition in this and ~~our field observations with~~ previous studies, the average wet deposition of atmospheric $NH_4^+$-N, $NO_3^-$-N, and inorganic N on the ~~Tibetan Plateau~~TP was estimated to be 1.~~17~~09, 0.~~58~~ 57 and 1.~~75~~ 66 kg N $ha^{-1}$ $yr^{-1}$, respectively. The average ~~estimated~~ $NH_4^+$-N:$NO_3^-$-N ratio in precipitation on the ~~Tibetan Plateau~~TP was 2:1. Compared to the present study, previous regional-scale estimation of inorganic N wet deposition ~~the inorganic N wet deposition~~ for the entire ~~Tibetan Plateau~~TP ~~in previous studies~~, either through atmospheric chemistry transport model simulations or interpolations based on limited field observations, has been highly overestimated. To clarify the total N deposition on the ~~Tibetan Plateau~~TP more clearly, it is essential ~~necessary~~ to conduct long-term ~~and large-scale~~ monitoring of both wet and dry deposition of atmospheric N in various climate zones on the TP in the future.

~~**Keywords**: nitrogen deposition, alpine ecosystem, precipitation chemistry, ion concentration, enrichment factor~~

## 1 Introduction

The global nitrogen (N) cycle has been disturbed by elevated reactive N emissions from anthropogenic activities since the mid-19th century (Canfield et al., 2010; Galloway et al., 2008; Gruber and Galloway, 2008). Accumulated reactive N in the environment has led to a series of effects on climate change and ecosystems, e.g. air pollution, stratospheric ozone depletion, the potential alteration of global temperature, drinking water contamination, freshwater eutrophication, biodiversity loss, grassland seed bank depletion, and dead zones in coastal ecosystems (Basto et al., 2015; Erisman et al., 2011; Erisman et al., 2013; Lan et al., 2015; Pinder et al., 2012; Shi et al., 2015; Zaehle et al., 2010). To examine the actual amount of N inputted into ecosystems, several monitoring networks have been established at national or continent scales, e.g. the National Atmospheric Deposition Program National Trends Network (NADP/NTN, United States) (Lehmann et al., 2005), the Canadian Air and Precipitation Monitoring Network (CAPMoN, Canada) (Zbieranowski and Aherne, 2011), the European Monitoring and Evaluation Programme (EMEP, Europe) (Fagerli and Aas, 2008), the Austrian Precipitation Sampling Network (Austria) (Puxbaum et al., 2002), and the Japanese Acid Deposition Survey (JADS, Japan) (Morino et al., 2011).

Besides Europe and North America, East Asia has become another high N deposition region, due to rapid economic growth in recent decades (Dentener et al., 2006). Across China, inorganic N wet deposition has increased since the mid-20th century, albeit with inconsistent estimations of the change: 8 kg N $ha^{-1}$ $yr^{-1}$ (from 13.2 kg N $ha^{-1}$ $yr^{-1}$ in the 1980s to 21.1 kg N $ha^{-1}$ $yr^{-1}$ in the 2000s) (X. J. Liu

et al., 2013); 2.8 kg N $ha^{-1}$ $yr^{-1}$ (from 11.11 kg N $ha^{-1}$ $yr^{-1}$ in the 1980s to 13.87 kg N $ha^{-1}$ $yr^{-1}$ in the 2000s) (Jia et al., 2014); and 7.4 kg N $ha^{-1}$ $yr^{-1}$ (from 12.64 kg N $ha^{-1}$ $yr^{-1}$ in the 1960s to 20.07 kg N $ha^{-1}$ $yr^{-1}$ in the 2000s) (Lu and Tian, 2014). Enhanced N deposition has changed the structure and function of terrestrial, aquatic and coastal ecosystems in China (Liu et al., 2011). To accurately estimate the N deposition in China, several monitoring networks have been established at the regional scale, e.g. in northern China (Pan et al., 2012), in forest ecosystems along the North–South Transect of Eastern China (NSTEC; based on the ChinaFLUX network) (Sheng et al., 2013), and in subtropical forest ecosystems in South China (Chen and Mulder, 2007). However, there are few observation sites distributed in western China, particularly on the Tibetan Plateau (TP), resulting in uncertainty regarding the N deposition for China as a whole (Jia et al., 2014; X. J. Liu et al., 2013; Lu and Tian, 2014).

The ~~Tibetan Plateau~~TP~~, whose alpine ecosystems are sensitive to elevated N deposition,~~ covers an area of about 2.57 million $km^2$, occupying approximately 1/4 of the land area of China (Zhang et al., 2002). On the TP, alpine ecosystems are widely distributed and are sensitive to elevated N deposition. Multi-level N fertilization experiments have shown that alpine grassland ecosystems are N limited and have potential capacity to absorb increased N deposition (Y. W. Liu et al., 2013; Xu et al., 2014). However, long-term N addition can decrease the species richness of both vegetation and soil seed banks in alpine meadow ecosystems on the ~~Tibetan Plateau~~TP (Ma et al., 2014). Ice core records show that the inorganic N ~~wet~~ deposition on the ~~Tibetan Plateau~~TP has increased during recent decades (Hou et al., 2003; Kang et al., 2002a, b; Thompson et al., 2000; Zhao et al., 2011; Zheng et al., 2010), and this trend is also apparent in sediment cores of alpine lakes in the western and southeastern ~~Tibetan Plateau~~TP (Choudhary et al., 2013; Hu et al., 2014). To recognize the characteristics of ion deposition

on the ~~Tibetan Plateau~~TP, a number of observations of precipitation chemistry have been carried out in the eastern ~~Tibetan Plateau~~TP in recent years (Jia, 2008; Tang et al., 2000; Zhang et al., 2003; N. N. Zhang et al., 2012). Nevertheless, in the western ~~Tibetan Plateau~~TP, observation sites are scarce, indicating that the situation in terms of N deposition across the entire ~~Tibetan Plateau~~TP remains unclear.

To quantitatively estimate the inorganic N wet deposition on the ~~Tibetan Plateau~~TP, we investigated the precipitation chemistry characteristics at five remote ~~stations~~sites, situated mainly on the central and western ~~Tibetan Plateau~~TP. The ~~stations~~ sites are part of the Tibetan Observation and Research Platform (TORP) network (Ma et al., 2008). Specifically, our aims were to (1) clarify the amount of inorganic N wet deposition on the central and western ~~Tibetan Plateau~~TP, and (2) assess the amount of inorganic N wet deposition for the entire ~~Tibetan Plateau~~TP by combining ~~our~~ site-scale *in situ* measurements ~~observations~~ with those ~~of~~ in previous studies.

## 2 Materials and methods

### 2.1 Precipitation sampling and chemical analysis

Using the Tibetan Observation and Research Platform (TORP) network (Ma et al., 2008), precipitation chemistry observations were conducted at five sampling sites: Southeast Tibet Station, Nam Co Station, Qomolangma Station, Ngari Station, and Muztagh Ata Station (Fig. 1), situated from the eastern to western ~~Tibetan Plateau~~TP and covering various climatic zones and vegetation types. A brief description of the five ~~stations~~ sites is shown in Table1.

During 2011–2013, we collected precipitation samples at each site, lasting at least one year. Precipitation samples were collected following each precipitation event, using an inner removable high-density polyethylene (HDPE) plastic bag in a pre-cleaned HDPE bucket. The HDPE bucket was placed 1.5 m above the ground. We opened the plastic bag at the beginning of the precipitation event, and collected precipitation samples at the end of the precipitation process. Then, the samples were transferred into pre-cleaned HDPE bottles (50 mL). Snowfall samples were melted at room temperature before being transferred into the HDPE bottles. All samples were kept frozen at the station and during transport until analysis in the laboratory. A total of 259 precipitation samples were collected, among which eight samples were abandoned due to breakage during transportation or the samples volume being less than 10 mL.

We analyzed the chemical composition of all precipitation samplings at the State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Analyzed ions included $NO_3^-$, $Cl^-$, $SO_4^{2-}$, $NH_4^+$, $Na^+$, $K^+$, $Ca^{2+}$ and $Mg^{2+}$. All ions were analyzed by the Ion Chromatography of Dionex-ICS2100. Samples for cation analysis were eluted on a Dionex 4-mm CS12A separatory column using 20 mM Methanesulfoni acid solution for an eluent pumped with a flow rate of 1.0 mL $min^{-1}$. Suppression was provided by a Dionex CSRS300 suppressor in recycle mode. For anion analysis, an IonPac AS19-HC column, 25 mM NaOH eluent and ASRS300 suppresser were used. The analytical detection limit was 2 ng $g^{-1}$ for all ions.

## 2.2 Data quality control

Previously documented methods (Rodhe and Granat, 1984;Safai et al., 2004) were used for quality assurance and quality control purposes, resulting in six (2.4%) samples, which fell outside the range

($m-3\delta$, $m+3\delta$), being excluded. Here, $m$ is the mean value and δ is the standard deviation. The Pearson correlation between $\Sigma_{anions}$ and $\Sigma_{cations}$ was 0.82 ($p < 0.001$), suggesting credible data quality. The ratio of total anions to total cations was calculated following Eq. (1),

$$\frac{\sum \text{anions}}{\sum \text{cations}} = \frac{\sum_{k=1}^{n}(NO_3^- + Cl^- + SO_4^{2-})}{\sum_{k=1}^{n}(NH_4^+ + Na^+ + K^+ + Ca^{2+} + Mg^{2+})}, \quad (1)$$

where $n$ is the number of samples. The ratio of ($\Sigma_{anions}/\Sigma_{cations}$) was 0.26, indicating that at least one major anion was not measured (C. Li et al., 2007). Considering that pH is alkaline in both precipitation and the surface soil layer (Ding et al., 2004; Y. H. Yang et al., 2012), the unmeasured anion was likely $HCO_3^-$ (C. Li et al., 2007).

## 2.3 Statistical Analysis

For each ~~station~~site, consecutive samples in one year were selected to analyze the annual mean values of ions. The sampling times of the samples used at the five sites were as follows: Southeast Tibet Station, November 2011 to October 2012; Nam Co Station, August 2011 to July 2012; Qomolangma Station, April 2011 to March 2012; Ngari Station, January 2013 to December 2013; Muztagh Ata Station, January 2011 to December 2011. A total of ~~186~~ 168 precipitation samples were selected, among which the number of samples for Southeast Tibet Station, Nam Co Station, Qomolangma Station, Ngari Station and Muztagh Ata Station was 53, ~~29~~27, ~~46~~30, 39 and 19, respectively.

The annual average ion concentration was calculated as the volume-weighted mean (VWM) following Eq. (2),

$$C = \frac{\sum_{i=1}^{n}(C_i \times P_i)}{\sum_{i=1}^{n} P_i}, \quad (2)$$

where $C$ is the annual average ion concentration (μeq $L^{-1}$), $C_i$ is the ion concentration of an individual sample $i$ (μeq $L^{-1}$), and $P_i$ is the precipitation amount corresponding to the sample $i$ (mm).

Wet deposition of atmospheric N was calculated following Eq. (3),

$$N_{\text{wet}} = 0.00014 \times C_{\text{N}} \times P_{\text{annual}}, \quad (3)$$

where $N_{\text{wet}}$ is the annual wet deposition of atmospheric inorganic N ($NH_4^+$-N or $NO_3^-$-N, kg N $ha^{-1}$ $yr^{-1}$), $C_N$ is the annual average equivalent concentration of N ($NH_4^+$-N or $NO_3^-$-N, μeq $L^{-1}$), $P_{\text{annual}}$ is annual precipitation (mm $yr^{-1}$), and 0.00014 is the shift coefficient for the unit of μeq $L^{-1}$ × mm $yr^{-1}$ to the unit of kg N $ha^{-1}$ $yr^{-1}$. Here, 1 μeq $NH_4^+$-N or $NO_3^-$-N contains 1 μmol N, and the weight of 1 μmol N is $14 \times 10^{-9}$ kg. Thus, μeq $L^{-1}$ × mm $yr^{-1}$ = $14 \times 10^{-9}$ kg N × $10^3$ $m^{-3}$ × $10^{-3}$ m $yr^{-1}$ = $14 \times 10^{-9}$ kg N × $10^4$ $ha^{-1}$ $yr^{-1}$ = 0.00014 kg N $ha^{-1}$ $yr^{-1}$.

## 2.4 Source assessment of ion wet deposition

### 2.4.1 Enrichment factor

~~We applied e~~Enrichment factor~~s~~ (EF~~s~~) has been widely used to examine the source contributions of major ions wet deposition ~~on the Tibetan Plateau, following~~ in ~~several~~ previous studies (Cao et al., 2009; Chabas and Lefevre, 2000; Kulshrestha et al., 1996; Lu et al., 2011; Okay et al., 2002; Shen et al., 2013; Xiao et al., 2013; Zhang et al., 2007). Commonly, Na is considered as the best reference element for seawater, due to its almost purely marine origin (Keene et al., 1986; Kulshrestha et al., 2003). Another element, ~~Ca, is a typical lithophile element, and its composition in soil barely changes. Therefore,~~ Ca is normally used as a reference element for continental crust, because Ca is a typical

lithophile element and its composition in soil barely changes (Zhang et al., 2007). In this study, Na and Ca were also used as reference element for seawater and continental crust, respectively.

On the TP, multiple lines of evidence demonstrate that $Na^+$ in precipitation on the TP mainly comes from oceans. Balestrini et al. (2014) monitored the chemical and isotopic compositions of precipitations at the Pyramid International Laboratory (5050m a.s.l.) on the southern slope of the Himalayas, and data analysis suggested that $Na^+$ and $Cl^-$ were derived from the long-range transport of marine aerosols. Ice records in the central Himalayas show that $Cl^-/Na^+$ was positively related with the monsoon rainfall in northeast India, and there was a teleconnection between the $Na^+$ and $Cl^-$ concentrations and the North Atlantic Oscillation, indicating $Na^+$ in the ice core mainly came from oceans (Wang et al., 2002). $Na^+$ has been used as a marine tracer when analyze the source contributions of ions wet deposition on the northeastern TP (Li et al., 2015), the southeastern TP (B. Liu et al., 2013) and the southern slope of central Himalayas (Tripathee et al., 2014).

On the TP, sandy desertification land covers about $3.1\times10^5$ $km^2$, accounting for 14% of the whole plateau, of which moderate sandy desertification land occupies 55.44% (Liu et al., 2005). The TP was regarded as an important dust source region (Fang et al., 2004; Han et al., 2009; Han et al., 2008). The TP dust sources contribute 69% of dust at the surface and 40% of dust in the lower troposphere on the TP (Mao et al., 2013). Moreover, arid regions are widely distributed surrounding the TP, e.g. central Asia, deserts of western China. The dust on the TP partly comes from the adjacent dust source regions, e.g. Taklimakan Desert in western China (Huang et al., 2007; Xia et al., 2008). Atmospheric dust aerosols over the TP are strongly impacted by local sources and enriched with Ca (Zhang et al., 2001). These dust aerosols in the atmosphere can interact with clouds and precipitation (Huang et al., 2014), and deposit on the surface with precipitation. Thus, $Ca^{2+}$ is commonly used as a proxy of dust for ice

core studies in the TP (Kang et al., 2002a; Kang et al., 2010; Kaspari et al., 2007; Wang et al., 2008). As a dust proxy, $Ca^{2+}$ record in an ice core from the central TP even was significantly related regional zonal wind (westerlies) trends, and reflected the long-term control of regional atmospheric circulation strength over atmospheric dust concentrations (Grigholm et al., 2015). In addition, Ca also has been used as a reference element for continental crust when assess source of ion wet deposition in precipitation on the northern TP (Li et al., 2015).

In this study, ~~T~~the EF of an ion in precipitation relative to the ion in sea was estimated using Na as a reference element following Eq. (4),

$$EF_{sea} = \frac{[X/Na^+]_{rain}}{[X/Na^+]_{sea}}, \quad (4)$$

where $EF_{sea}$ is the EF of an ion in precipitation relative to the ion in sea, $X$ is an ion in precipitation, $[X/Na^+]_{rain}$ is the ratio from precipitation composition (μeq $X$ / μeq $Na^+$), and $[X/Na^+]_{sea}$ is the ratio from sea composition (Keene et al., 1986; Turekian, 1968) (μeq $X$ / μeq $Na^+$).

The EF of an element in precipitation relative to the element in soil was estimated using Ca as a reference element ~~for~~ following Eq. (5),

$$EF_{soil} = \frac{[X/Ca^{2+}]_{rain}}{[X/Ca^{2+}]_{soil}}, \quad (5)$$

where $EF_{soil}$ is the EF of an element in precipitation relative to the element in soil, $X$ is an ion in precipitation, $[X/Ca^{2+}]_{soil}$ is the ratio from precipitation composition (μg $X$ / μg $Ca^{2+}$), and $[X/Ca^{2+}]_{soil}$ is the ratio from soil composition (Taylor, 1964) (μg $X$ / μg $Ca^{2+}$).

To estimate fractions of marine, crustal and anthropogenic contributed to ions in precipitation, we calculated the sources of ionic components in precipitation using equations from previous studies (Cao et al., 2009; Lu et al., 2011; Zhang et al., 2007) as follows:

$$\mathrm{SSF}\ (\%) = \frac{[X/\mathrm{Na}^+]_{\mathrm{sea}}}{[X/\mathrm{Na}^+]_{\mathrm{rain}}} \times 100, \quad (6)$$

$$\mathrm{CF}\ (\%) = \frac{[X/\mathrm{Ca}^{2+}]_{\mathrm{soil}}}{[X/\mathrm{Ca}^{2+}]_{\mathrm{rain}}} \times 100, \quad (7)$$

$$\mathrm{AF}\ (\%) = 100 - \mathrm{SSF} - \mathrm{CF}, \quad (8)$$

where SSF is sea salt fraction, CF is crust fraction, and AF is anthropogenic fraction. Note that if SSF is greater than 1, SSF is recalculated as the difference between 1 and CF, and if CF is greater than 1, CF is recalculated as the difference between 1 and SSF.

### 2.4.2 Principal component analysis

Principal component analysis has been widely used in precipitation chemical studies to determine the effect of natural and anthropogenic sources on chemical composition of precipitation (Balasubramanian et al., 2001; Cao et al., 2009; Migliavacca et al., 2005; Zhang et al., 2007). In this study, principal component analysis was also used to examine the various source of major ions in precipitation at the five remote sites on the TP. Varimax-rotated principal component analysis was performed using "principal" function in package "psych" of R 3.2.0 (R Core Team, 2015; http://www.R-project.org).

### 2.4.3 Backward trajectory analysis

To identify the long range transport of water-soluble ions in precipitation, seven-day backward trajectories arriving at the sampling sites for each individual precipitation event were calculated. Backward trajectories were calculated using TrajStat (version 1.4.4R4, http://www.meteothinker.com/TrajStatProduct.html), which is a Geographic Information System based software, including a trajectory calculation module of HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model; http://www.arl.noaa.gov/ready/hysplit4.html) (Wang et al., 2009). The input meteorological data was the Global Data Assimilation System (GDAS) meteorological data archives of the Air Resource Laboratory, National Oceanic and Atmospheric Administration (NOAA) (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1). All backward trajectories were calculated at 6-h interval (00:00, 06:00, 12:00, 18:00 UTC) at each sampling day, with an arrival height of 500 m above the ground. Then, cluster analysis was performed using of the trajectories during the one year-round sampling period at each site using TrajStat (version 1.4.4R4).

## 3 Results

### 3.1 Chemical composition of atmospheric precipitation

Figure 2 shows the seasonal dynamics of ion concentrations in precipitation at the five remote ~~stations~~ sites on the ~~Tibetan Plateau~~TP. Wet deposition of all ions mainly occurs in summer at all sites~~stations~~. Compared to the sites ~~stations~~ with relatively higher precipitation amounts, e.g. Ngari Station and Muztagh Ata Station, the sites ~~stations~~ with relatively lower precipitation amounts had relatively higher ion concentrations, e.g. Southeast Tibet Station and Nam Co Station (Fig. 2, Table 2). $Ca^{2+}$ had the highest annual VWM concentration in precipitation at most sites ~~stations~~ (except for Nam Co Station),

with the highest proportion accounting for measured ions of 54.6% at Southeast Tibet Station (Fig. 2 and 3). At Nam Co Station, $NH_4^+$ in precipitation had the highest proportion accounting for measured ions of 3~~8.2~~9.5%, higher than those at the other sites ~~stations~~ (ranging 12.9% at Southeast Tibet Station to 18.9% at Muztagh Ata Station) (Fig. 3). Compared to $NH_4^+$, $NO_3^-$ had much lower proportion accounting for measured ions in precipitation, ranging 0.~~4~~6% at Qomolangma Station to 14~~.8~~% at Nam Co Station (Fig. 3). The order of the average VWM of ion deposition at the five stations was: $Ca^{2+}$ > $NH_4^+$ > $SO_4^{2-}$ > $Cl^-$ > $Na^+$ > $Mg^{2+}$ > $NO_3^-$ > $K^+$ (Table ~~1~~2). All major ion concentrations in precipitation on the ~~Tibetan Plateau~~TP were much lower than those in northern and southern China (Table 2).

### 3.2 Wet deposition of atmospheric inorganic N

~~The seasonal dynamics inorganic of N wet deposition at most stations appeared the shape of single peak type (Fig. 4), which were similar to the seasonal pattern of precipitation (Fig. 2). The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all stations (Fig. 4).~~ At Southeast Tibet Station, Nam Co Station, Qomolangma Station, Ngari Station, and Muztagh Ata Station, the $NH_4^+$-N wet deposition was 0.63, 0.~~91~~68, ~~1.61~~0.92, 0.36 and 1.25 kg N $ha^{-1}$ $yr^{-1}$, respectively; the $NO_3^-$-N wet deposition was 0.28, 0.~~35~~24, 0.~~04~~03, 0.08 and 0.3 kg N $ha^{-1}$ $yr^{-1}$, respectively; and the inorganic N deposition was 0.91, ~~1.26~~0.92, ~~1.64~~0.94, 0.44 and 1.55 kg N $ha^{-1}$ $yr^{-1}$, respectively (Table 3). Besides the above five sites of the TORP network, previous site-scale *in situ* measurements of inorganic N wet deposition at other sites ~~observations~~ on the ~~Tibetan Plateau~~TP ~~from previous studies~~ were also collected, e.g. at Waliguan (Tang et al., 2000), Wudaoliang (Yang et al., 1991), Lhasa (Zhang et al., 2003), Naidong (Jia, 2008), Biru (Jia, 2008), Jiangda (Jia, 2008), and Lijiang (N. N. Zhang et al., 2012). Combining site-scale *in situ* measurements in our study ~~our results~~ with ~~these~~ those in previous studies~~observations~~, the average wet deposition of atmospheric $NH_4^+$-N,

$NO_3^-$-N and inorganic N on the ~~Tibetan Plateau~~TP were estimated to be 1.~~17~~09, 0.~~58~~57 and 1.~~75~~66 kg N $ha^{-1}$ $yr^{-1}$, respectively, and the estimated $NH_4^+$-N:$NO_3^-$-N ratio in precipitation on the ~~Tibetan Plateau~~TP was 2:1. Both $NH_4^+$-N and $NO_3^-$-N wet deposition on the ~~Tibetan Plateau~~TP were much lower than those in northern and southern China (Table 3).

### 3.3 Seasonal dynamics of inorganic N wet deposition

The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all sites (Fig. 4). Both concentrations of $NH_4^+$ and $NO_3^-$ did not exhibited any clear seasonal pattern (Fig. 2).~~T~~ The seasonal dynamics ~~inorganic of~~ inorganic N wet deposition at most stations appeared the shape of single peak type (Fig. 4)~~, which~~. The seasonal pattern of inorganic N wet deposition were similar to the seasonal pattern of precipitation, rather than that of $NH_4^+$ or $NO_3^-$ concentration (Fig. 2). ~~The inorganic N wet deposition mainly occurred as the form of $NH_4^+$-N during the summer at all stations (Fig. 4).~~

~~At the five stations, inorganic N wet deposition mainly occurred during summer (Fig. 4). The seasonal patterns of inorganic N wet deposition at most stations appeared the shape of single peak type (Fig. 4), which were more similar to that of precipitation, rather than that of $NH_4^+$ or $NO_3^-$ concentration (Fig. 2). Nearly all $Ca^{2+}$ in precipitation came from crust at the five sites, with the CF value being above 90%.~~

### 3.4 Source assessment of wet deposition of inorganic N and other ions

#### 3.4.1 Enrichment factors

Table 4 shows the EFs of precipitation constituents at the five sites relative to seawater and soil. If the EF value of an ion in precipitation is much higher (lower) than 1, the ion is considered to be enriched

(diluted) relative to the reference source. Among the five sites, $Cl^-$ had a relatively lower $EF_{sea}$ value, ranging from 0.50 (Nam Co Station) to 0.90 (Qomolangma Station), but a relatively higher $EF_{soil}$ value, ranging from 42.4 (Muztagh Ata Station) to 286 (Qomolangma Station). Different from $Cl^-$, $NH_4^+$ in precipitation was enriched relative to both marine origin and soil reference source at all sites, because its $EF_{sea}$ values ranged from 11629 to 80684, and its $EF_{soil}$ from 350 to 2378. Similar to $NH_4^+$, $NO_3^-$ also had a relatively high value of both $EF_{sea}$ and $EF_{soil}$ at all five sites.

Table 5 shows the source contributions for major ions in precipitation of the five remote sites in this study. Almost all $Cl^-$ and $Na^+$ in precipitation on the TP appeared to be of marine origin, with SSF value above 95% at the five sites. Nearly all $Ca^{2+}$ in precipitation came from crust at the five sites, with the CF value being above 90%. Among the five sites, anthropogenic sources contributed at least 99% of $NH_4^+$ in precipitation, and $NO_3^-$ in precipitation was also mainly influenced by anthropogenic activities, with AF values ranging from 95.3% to 99.9%.

### 3.4.2 Principal component analysis

Table 6 shows the 1st, 2nd and 3rd component of principal component analysis, which account for at least 85% of the total variance across the five sites. $Na^+$ and $Cl^-$ were mainly explained by the same component at all sites. Principal component analysis shows that the variances of $Ca^{2+}$ and $Na^+$ were represented by different components at four of five sites (except Southeast Tibet Station) (Table 6). The common variance of $Ca^{2+}$, $Mg^{2+}$ and $SO_4^{2-}$ as 1st component represents the largest proportion of the total specie variation at the 3 sites (Nam Co Station, Ngari Station and Muztagh Ata Station) in the central and western TP (Table 6). At Qomolangma Station, $Na^+$, $Cl^-$, $K^+$ and $NH_4^+$ as 1st component represents the largest proportion of the total specie variation (Table 6). Except for Qomolangma Station,

at the other four sites, the variances of $NH_4^+$ were mainly represented by 3rd component (Table 6). At Southeast Tibet Station, both $Ca^{2+}$ and $Na^+$ variances were mostly represented by the 1st component, but $NO_3^-$ variances were mainly represented by the 2nd component (Table 6). At Nam Co Station, Qomolangma Station and Ngari Station, $NO_3^-$ variances were mainly represented by the 3nd component, which were different with that of both $Ca^{2+}$ and $Na^+$ (Table 6). However, $NO_3^-$ variances were mainly represented by the 1st component at Muztagh Ata Station (Table 6).

### 3.4.3 Backward trajectory analysis

Fig. 5 shows the seven-day backward trajectories of air mass arriving at the five remote sites at the sampling days. The transport pathways of air masses were various with the different sites (Fig. 5). The cluster trajectory results showed that at Muztagh Ata Station, nearly all air masses at sampling days were transported from Central Asia and Middle East (Fig. 5a). Different to Muztagh Ata Station, almost all air masses at Nam Co Station were transported from South Asia (Fig. 5d). For Ngari Station, Qomolangma Station, and Southeast Tibet Station, the air masses at sampling days were mainly transported from South Asia, with the proportion of 90%, 79.8% and 90.6%, respectively (Figs. 5b-5e). Besides South Asia, Central Asia, Qaidam Basin and Middle East was the second source of air masses at sampling days for Ngari Station, Qomolangma Station, and Southeast Tibet Station, respectively (Figs. 5b-5e).

## 4 Discussion

### 4.1 Wet deposition of atmospheric inorganic N on the ~~Tibetan Plateau~~TP

According to our field observations, wet deposition of atmospheric inorganic N on the western ~~Tibetan Plateau~~TP was lower than that on the eastern ~~Tibetan Plateau~~TP. For example, the rates of inorganic N wet deposition at Ngari Station and Muztagh Ata Station, located on the western ~~Tibetan Plateau~~TP, were 0.44 and 1.55 kg N $ha^{-1}$ $yr^{-1}$, respectively, which were lower than those at the stations on the eastern ~~Tibetan Plateau~~TP, e.g. Jiangda (1.91 kg N $ha^{-1}$ $yr^{-1}$), Lijiang (1.89 kg N $ha^{-1}$ $yr^{-1}$) and Waliguan (2.92 kg N $ha^{-1}$ $yr^{-1}$) (Table 3). However, the concentrations of inorganic N in precipitation at the sites ~~stations~~ on the western ~~Tibetan Plateau~~TP were comparable to those at the sites ~~stations~~ on the eastern ~~Tibetan Plateau~~TP. For instance, the annual average concentrations of $NH_4^+$ in precipitation at Ngari Station and Muztagh Ata Station were 20.5 and 42.0 μeq $L^{-1}$, respectively, which were higher than those at stations on the eastern ~~Tibetan Plateau~~TP, e.g. Lijiang (11.4 μeq $L^{-1}$) and Lhasa (14.3 μeq $L^{-1}$) (Table 2). Meanwhile, compared to Lijiang and Lhasa, Ngari Station and Muztagh Ata had lower annual precipitation rates of 124.6 and 213.6 mm $yr^{-1}$ (Table 2). Therefore, compared to the eastern ~~Tibetan Plateau~~TP, the western ~~Tibetan Plateau~~TP had relatively lower inorganic N deposition, probably due to its lower precipitation rather than its comparable inorganic N concentration in precipitation.

Wet deposition of inorganic N for the entire ~~Tibetan Plateau~~TP was much lower than that in northern and southern China (Table 3). The average wet deposition of atmospheric inorganic N (sum of $NH_4^+$-N and $NO_3^+$-N) for the ~~Tibetan Plateau~~TP was estimated to be 1.~~75~~ 66 kg N $ha^{-1}$ $yr^{-1}$. This was much lower than the inorganic N wet deposition in the cities of both northern and southern China, e.g. Beijing, Tianjin, Tangshan, Dalian, Nanjing, Hangzhou, Ningbo, Shanghai, Shenzhen and Guiyang (Table 3). Moreover, the inorganic N wet deposition on the ~~Tibetan Plateau~~TP was also lower than that in the forest ecosystems of eastern China, e.g. TieShanPing, LiuChongGuan, LeiGongShan, CaiJiaTang and

XiLiuHe (Table 3). Overall, compared to eastern China, the ~~Tibetan Plateau~~TP had relatively lower inorganic N wet deposition, probably due to following two reasons. First, except for Southeast Tibet Station and Lijiang, most N observation sites on the ~~Tibetan Plateau~~TP are located in typical arid and semi-arid regions, with annual precipitation ranging from 124.6 mm $yr^{-1}$ at Ngari Station to 582 mm $yr^{-1}$ at Biru. Compared to this, annual precipitation rates at sites in eastern China are much higher, particularly in southern China, where annual precipitation ranges from 825.5 mm $yr^{-1}$ at Shanghai to 1769 mm $yr^{-1}$ at Shenzhen (Table 3). Second, the average annual concentration of inorganic N ($NH_4^+$-N, $NO_3^-$-N) in precipitation on the ~~Tibetan Plateau~~TP was much lower than that in eastern China, especially at cities in northern China (Table 2). This is probably because the effects of anthropogenic activities in eastern China are much more intense than on the ~~Tibetan Plateau~~TP, which is referred to as "The Third Pole" and has an average altitude exceeding 4,000 meters above sea level (Qiu, 2008; Yao et al., 2012).

### 4.2 Source assessment ~~contributions~~ of atmospheric inorganic N wet deposition ~~ions in precipitation~~ on the ~~Tibetan Plateau~~TP

To analyze the source contributions of major ions wet deposition, EF was applied using Na and Ca as reference element for seawater and continental crust, respectively. Here, Na and Ca in precipitation on the TP was hypothesized mainly coming seawater and continental crust, respectively. This assumption was partly confirmed by the results of principal component analysis in this study (Table 6). Principal component analysis shows that the variances of $Ca^{2+}$ and $Na^+$ were represented by different components at four of five sites (except Southeast Tibet Station), indicating different source of $Ca^{2+}$ and $Na^+$ in precipitation on the TP (Table 6). Moreover, $Na^+$ and $Cl^-$ were mainly explained by the same component at all sites. This indicates that $Na^+$ and $Cl^-$ were likely contributed by the same source:

sea-salt (Table 6). This assumption was also confirmed by the relatively high Pearson correlation between $Na^+$ and $Cl^-$ at all five sites (Table S1 in the supplementary material). At Southeast Tibet Station, both $Ca^{2+}$ and $Na^+$ variances were mostly represented by the 1st component (Table 6). This probably because South Asia is also an important source of dust aerosols in the southeastern TP during the during the monsoon period (Zhao et al., 2013).

EF analysis results showed that at all the five sites, both $NH_4^+$ and $NO_3^-$ in precipitation were mainly contributed by anthropogenic sources (Table 5). This was also confirmed by principal component analysis. Different with $Ca^{2+}$ and $Na^+$, $NH_4^+$ variances were mainly represented by 3rd component at four in five sites (except for Qomolangma Station) (Table 6). Except for Muztagh Ata Station, at the other four stations, $NO_3^-$ variances were also represented by different component with that of $Ca^{2+}$ and $Na^+$ variances (Table 6). This indicates that the source of inorganic N wet deposition was probably different with the sources of $Ca^{2+}$ or $Na^+$. Meanwhile, at all five sites, Na and Ca mainly came from seawater and continental crust, respectively. Therefore, inorganic N wet deposition at the five sites on the TP was mainly influenced by anthropogenic activities.

TP is mainly controlled by westerlies and Indian monsoon (Yao et al., 2013). The northern TP is mainly influenced by westerlies, and the ~~Table 4 shows the EFs of precipitation constituents at the five sites relative to seawater and soil. If the EF value of an ion in precipitation is much higher (lower) than 1, the ion is considered to be enriched (diluted) relative to the reference source. Among the five sites, $Cl^-$ had a relatively lower $EF_{sea}$ value, ranging from 0.50 (Nam Co Station) to 0.85 (Qomolangma Station), but a relatively higher $EF_{soil}$ value, ranging from 42.4 (Muztagh Ata Station) to 365 (Qomolangma Station). This indicates that $Cl^-$ in precipitation on the Tibetan Plateau was mainly influenced by marine origin, rather than local source. Different from $Cl^-$, $NH_4^+$ in precipitation was enriched relative~~

~~to both marine origin and soil reference source at all stations, because its $EF_{sea}$ values ranged from 10595 to 84974, and its $EF_{soil}$ from 350 to 2164. Similar to $NH_4^+$, $NO_3^-$ also had a relatively high value of both $EF_{sea}$ and $EF_{soil}$ at all five stations. This indicates that, for $NH_4^+$ and $NO_3^-$ in precipitation on the Tibetan Plateau, contributions from marine and soil sources could be negligible. Thus, anthropogenic activities can be considered as the major sources of inorganic N deposition on the Tibetan Plateau.~~

~~Table 5 shows the source contributions for major ions in precipitation of the five remote sites in our study. Almost all $Cl^-$ and $Na^+$ in precipitation on the Tibetan Plateau appeared to be of marine origin, with SSF value above 95% at all five stations. The main source of $Mg^{2+}$ varied between sites. The primary contribution of $Mg^{2+}$ was marine at Southeast Tibet Station and Qomolangma Station, but was crust at Muztagh Ata Station. Meanwhile, the $Mg^{2+}$ in precipitation at Nam Co Station and Ngari Station was mainly contributed by both marine and crustal sources.~~ ~~Nearly all $Ca^{2+}$ in precipitation came from crust at the five sites, with the CF value being above 90%.~~ ~~$K^+$ in precipitation mainly originated from crust (84.5%–92.6%), with a little of marine origin (7.4%–15.5%). Precipitation $SO_4^{2-}$ came from anthropogenic sources at most sites, except at Qomolangma Station, where $SO_4^{2-}$ in precipitation was mainly of marine origin, with an SSF value of 88.0%. Among the five sites, anthropogenic sources contributed at least 99% of $NH_4^+$ in precipitation, and $NO_3^-$ in precipitation was also mainly influenced by anthropogenic activities, with AF values ranging from 94.8% to 99.9%.~~

~~At the five stations, inorganic N wet deposition mainly occurred during summer (Fig. 4). The seasonal patterns of inorganic N wet deposition at most stations appeared the shape of single peak type (Fig. 4), which were more similar to that of precipitation, rather than that of $NH_4^+$ or $NO_3^-$ concentration (Fig. 2).~~ ~~S~~southern ~~Tibetan Plateau~~TP is mainly controlled by the Indian monsoon in summer seasons, but

by the westerlies during non-monsoon seasons (Yao et al., 2013). N emissions from anthropogenic activities in Central Asia might be transported to the northern TP by westerlies. N emissions from anthropogenic activities in Southhern Asia (e.g. India) might be transported to the southern Tibetan PlateauTP by Indian monsoon. After China and USA, India has been the third largest producer and consumer of fertilizers due to intensification of agriculture, resulting in high anthropogenic N emissions (Aneja et al., 2012). At present, reactive N emissions from crop and livestock farming in India were second only to that in China (Aneja et al., 2012). For instance, ammonia ($NH_3$) emissions from livestock and fertilizer applications in India in 2003 was estimated as 1705 Gg $yr^{-1}$ and 1697 Gg $yr^{-1}$, respectively (Aneja et al., 2012). Moreover, in India, field burning of crop residue (FBCR) is another critical anthropogenic activity leading to N emissions. N emissions in India due to FBCR showed increase trend in the past 3 decades (Sahai et al., 2011). In 2010, 6300 Gg of dry biomass are estimated to be subjected to FBCR in India, resulting in 350 Gg N emissions (Sahai et al., 2011). Besides Indian monsoon, biomass-burning emissions in South Asia could be across the Himalayas and transported to the Tibetan PlateauTP by the mountain/valley wind (Cong et al., 2015).

We applied backward trajectory analysis to identify the long range transport of atmospheric inorganic N wet deposition at the five sites on the TP (Fig. 5). There is large spatial heterogeneity of air mass transport pathways across the five sites. At Muztagh Ata Station, wet deposition was mainly transported from Central Asia and Middle East (Fig. 5a). It seems that air mass from South Asia has no influence on the wet deposition at Muztagh Ata Station. This is probably because Muztagh Ata Station is located on the northwestern TP, where is almost completely controlled by westerlies, rather than Indian Monsoon (Yao et al., 2013). Thus, anthropogenic activities in Central Asia and Middle East are the principal source of the inorganic N wet deposition at Muztagh Ata Station. Except for

Muztagh Ata Station, inorganic N wet deposition at the other four sites was mainly influenced by anthropogenic activities in South Asia and was probably transported by India Monsoon (Figs. 5b-5e). At Ngari Station, 90.0% of wet deposition was transported from Nepal and North India through Indian Monsoon, and 10.0% wet deposition was transported from Central Asia and Qaidam Basin through westerlies (Fig. 5b). At Qomolangma Station and Nam Co Station, inorganic N wet deposition was mainly influenced by the anthropogenic activities in the northeastern India and Bangladesh (Figs. 5c and 5d). At Southeast Tibet Station, 90.6% of wet deposition was transported from India, Bangladesh and Myanmar by India Monsoon, and the other 9.4% came from the western TP and Middle East (Fig. 5e).~~Among the five observation stations in our study, Qomolangma Station is the most southerly station (Fig. 1), and is located on the northern slope of the Himalayas. Therefore, inorganic N wet deposition at Qomolangma Station probably could be more severely influenced by anthropogenic activities in Southern Asia than that at the other four stations. As expected, among the five stations, Qomolangma Station had highest value of inorganic N wet deposition, $NH_4^+$ concentration and $NH_4^+$-N:$NO_3^-$-N ratio (Fig. 2 and 4, Table 3).~~

### 4.3 Comparison of inorganic N wet deposition on the ~~Tibetan Plateau~~TP with previous estimations

Long-term dataset series of N deposition have been established based on observations (Lu and Tian, 2007, 2014, 2015) or model simulations (Dentener et al., 2006). These datasets have been used to estimate global or regional N deposition (Dentener et al., 2006; Lu and Tian, 2007) and drive ecosystem models to examine the ecological effects of elevated N deposition (Lu and Tian, 2013). Thus, reliable N deposition datasets are a prerequisite for N deposition estimation or driving ecosystem

models. Here, the estimation of N wet deposition on the ~~Tibetan Plateau~~TP based on our field observations is compared with previous estimations via limited observations or simulations.

Lu and Tian (2007) estimated the inorganic N wet deposition as ranging from 4.16 kg N $ha^{-1}$ $yr^{-1}$ in Tibet Autonomous Region (on the western ~~Tibetan Plateau~~TP) to 4.76 kg N $ha^{-1}$ $yr^{-1}$ in Qinghai Province (on the eastern ~~Tibetan Plateau~~TP). Recently, Jia et al. (2014) estimated the inorganic N wet deposition during the 2000s as ranging from 6.11 kg N $ha^{-1}$ $yr^{-1}$ in Tibet Autonomous Region to 7.87 kg N $ha^{-1}$ $yr^{-1}$ in Qinghai Province. Those estimations were much higher even than the highest record of inorganic N wet deposition observations on the ~~Tibetan Plateau~~TP (3.08 kg N $ha^{-1}$ $yr^{-1}$ at Biru during 2006-2007) (Table 3). In this study, combing *in situ* measurements at 5 sites in this study and 7 sites in previous ~~observations~~ studies (Table 2), the average wet deposition of atmospheric $NH_4^+$-N, $NO_3^+$-N and inorganic N on the ~~Tibetan Plateau~~TP were estimated to be 1.~~17~~09, 0.~~58~~57 and 1.~~75~~66 kg N $ha^{-1}$ $yr^{-1}$, respectively. According to our study, both Lu and Tian (2007) and Jia et al. (2014) highly overestimated inorganic N wet deposition on the ~~Tibetan Plateau~~TP, probably due to following two reasons. First, the observations used in previous ~~studies~~ regional-scale estimations were limited, with fewer observation sites than used in the present study. For example, there were only four sites in Tibet Autonomous Region and one site in Qinghai Province used in the estimation of Jia et al. (2014). Such limited observations lead to uncertainty in the conclusions drawn regarding inorganic N wet deposition on the entire ~~Tibetan Plateau~~TP. Second, the Kriging interpolation technique was used in both Lu and Tian (2007) and Jia et al. (2014) to estimate the spatial pattern of inorganic N wet deposition in China. However, observation sites are sparsely distributed on the ~~Tibetan Plateau~~TP, and the estimation of inorganic N wet deposition is largely influenced by N deposition observations in the surrounding regions of much lower altitude. The average altitude of the ~~Tibetan Plateau~~TP is above 4000 m, where

both the climate and anthropogenic activities are substantially different with those in lower altitude areas. For example, the average inorganic N wet deposition was 1.~~75~~ 66 kg N $ha^{-1}$ $yr^{-1}$, which was much lower than that in northern and southern China (Table 3). The interpolations at the national scale in Lu and Tian (2007) and Jia et al. (2014)~~previous studies~~ probably overestimated inorganic N wet deposition on the ~~Tibetan Plateau~~TP. In addition, we also estimated the inorganic N wet deposition for the entire ~~Tibetan Plateau~~TP using Kriging interpolation, but only based on the site-scale *in situ* measurements ~~observations only~~ on the ~~Tibetan Plateau~~TP (12 sites, including: 5 sites in this study and 7 sites in previous ~~observations~~field observations), rather than the observations in the surrounding regions of much lower altitude. The inorganic N wet deposition for the entire ~~Tibetan Plateau~~TP estimation based on the Kriging interpolation in our study is 1.~~86~~ 76 kg N $ha^{-1}$ $yr^{-1}$ (Fig. S1 and spatial data as NetCDF file in the supplementary material~~, Fig. S1~~), which is much lower than that in previous interpolation studies (Lu and Tian, 2007; Jia et al., 2014), but is comparable with the averaged inorganic N wet deposition among the 12 sites (1.~~75~~ 66 kg N $ha^{-1}$ $yr^{-1}$) (Table 2).

Atmospheric chemistry transport models are commonly used to calculate current and future N deposition. Dentener et al. (2006) used 23 atmospheric chemistry transport models to assess both global and regional N deposition. Compared to observation records, Dentener et al. (2006) underestimated inorganic N wet deposition over the whole of China (Lu and Tian, 2007), but overestimated it for the ~~Tibetan Plateau~~TP. According to Dentener et al. (2006), the $NH_4^+$, $NO_3^-$ and inorganic N wet deposition on the ~~Tibetan Plateau~~TP are 1.97, 0.99 and 2.96 kg N $ha^{-1}$ $yr^{-1}$, respectively—nearly double that of N deposition estimated in our study. Based on site-scale *in situ* measurements~~field observations~~, we provide a more accurate regional-scale estimation of inorganic N wet deposition on the TP, which can be used as background information in research focusing on the

responses of alpine ecosystems to elevated N deposition ~~on the Tibetan Plateau~~. Besides assessment of N deposition, N deposition simulated by atmospheric chemistry transport models is usually used to drive large-scale ecosystem models for integrated ecosystem assessment (Xu-Ri et al., 2012; Zaehle, 2013). The ecological effects of N addition are probably not only influenced by the quantity of N deposition, but also by the proportions of each component, e.g. the $NH_4^+$-N:$NO_3^-$-N ratio. For example, in African savannas, plants demonstrate N uptake preference, which is likely influenced by the $NH_4^+$-N:$NO_3^-$-N ratio in their native habitats (Wang and Macko, 2011). However, in most current N fertilization experiments, the N forms of fertilizer are $NH_4NO_3$, $NH_4^+$-N or $NO_3^-$-N (Liu and Greaver, 2009), with the $NH_4^+$-N:$NO_3^-$-N ratio of N wet deposition at experimental sites not considered. Our work shows that the estimated $NH_4^+$-N:$NO_3^-$-N ratio of inorganic N wet deposition on the ~~Tibetan Plateau~~TP is 2:1, which is consistent with the modelled estimation of Dentener et al. (2006), but lower than the $NH_4^+$-N:$NO_3^-$-N ratio of 2.5 in forest ecosystems in eastern China (Du et al., 2014). This $NH_4^+$-N:$NO_3^-$-N ratio (2:1) is recommended to be considered when N fertilization experiments are conducted in alpine ecosystems on the ~~Tibetan Plateau~~TP.

### 4.4 Uncertainty and recommendations

Combining our *in situ* measurements at five remote sites~~field observations~~ and previous site-scale field observations~~studies~~, the inorganic N wet deposition on the ~~Tibetan Plateau~~TP was quantitatively assessed in this study. The assessment is conducive to accurately estimating N wet deposition for the entire nation of China, and provides background information ~~on~~ of N wet deposition for the studies ~~research~~ focusing on the alpine ecological effects of elevated N deposition. Despite this, there are uncertainties in the estimation of N deposition on the ~~Tibetan Plateau~~TP due to ~~the~~ following reasons. First, total N deposition comprises wet deposition (in the form of precipitation) and dry deposition (in

the form of gases and particles). Considering the whole of China, dry deposition contributes 30% to total inorganic N deposition (Lu and Tian, 2007, 2014, 2015). In northern China, this ratio is much higher, at 60% (Pan et al., 2012). However, in this study, we only estimated the inorganic N wet deposition on the ~~Tibetan Plateau~~TP, with the situation regarding dry deposition remaining unclear. Thus, investigation of N dry deposition is critical for assessing total N deposition on the ~~Tibetan Plateau~~TP. Second, the ~~Tibetan Plateau~~TP covers an area of about 2.57 million $km^2$, occupying approximately 1/4 of the land area of China (Zhang et al., 2002). Precipitation on the ~~Tibetan Plateau~~TP is influenced by both the Indian monsoon and westerlies, leading to spatial variation in the origins of N wet deposition. Therefore, it is necessary to establish N wet deposition observation sites in different climatic zones. Third, besides spatial heterogeneity, N deposition on the ~~Tibetan Plateau~~TP also possesses temporal heterogeneity. Inorganic N wet deposition on the ~~Tibetan Plateau~~TP has increased during recent decades, as recorded in ice cores (Hou et al., 2003;Kang et al., 2002a, b;Thompson et al., 2000;Zhao et al., 2011;Zheng et al., 2010) and sediment cores of alpine lakes (Choudhary et al., 2013;Hu et al., 2014). The long-term trend and inter-annual variability of inorganic N wet deposition on the TP can't be quantitatively characterized by the short-term *in situ* measurements in this study. Overall, critical questions remain open regarding the quantitative understanding of N deposition on the TP. Therefore, to deepen our understanding N deposition on the TP, it is essential to perform long-term *in situ* measurements of N wet and dry deposition in various climate zones in the future.

~~To clarify the temporal trend of N deposition, long-term monitoring is essential in the future.~~

## 5 Conclusion

Alpine ecosystems on the ~~Tibetan Plateau~~TP are sensitive to elevated N deposition, and the inorganic N ~~wet~~ deposition has been increasing since the mid-20th century. However, the amount of inorganic N wet deposition on the Tibetan remains unclear, due to a paucity of *in situ* measurement~~observations~~. In this study, using stations in the TORP network, we ~~investigated~~ conducted~~the~~ *in situ* measurements of major ions wet deposition ~~of major ions~~ at five remote ~~stations~~sites, situated mainly on the central and western ~~Tibetan Plateau~~TP. Among the five ~~stations~~sites, both $NH_4^+$-N and $NO_3^-$-N were mainly contributed by anthropogenic sources. Combining site-scale *in situ* measurements in our ~~field observations~~ and ~~results from~~ previous studies, the average wet deposition of atmospheric $NH_4^+$-N, $NO_3^-$-N and inorganic N on the ~~Tibetan Plateau~~TP are estimated to be 1.~~17~~09, 0.~~58~~57 and 1.~~75~~66 kg N $ha^{-1}$ $yr^{-1}$, respectively. Considering the entire ~~Tibetan Plateau~~TP, according to our results, previous ~~studies~~ regional-scale assessment have highly overestimated inorganic N wet deposition, either through simulations with atmospheric chemistry transport models (Dentener et al., 2006) or interpolations based on limited field observations for the whole of China (Jia et al., 2014; Lu and Tian, 2007). The $NH_4^+$-N:$NO_3^-$-N ratio in precipitation on the ~~Tibetan Plateau~~TP was found to be 2:1, which is consistent with model simulations (Dentener et al., 2006). To clarify the total N deposition on the ~~Tibetan Plateau~~TP more clearly, we recommend conducting long-term monitoring of both wet and dry deposition of N in various climate zones in the future work.


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**Table 1. Descriptions of the five precipitation sampling ~~stations~~ sites on the ~~Tibetan Plateau~~TP.**

| Station name | Station name expanded | Latitude | Longitude | Altitude<br>m a.s.l. | Annual mean temperature<br>℃ | Annual precipitation<br>mm $yr^{-1}$ | Vegetation type | References |
|---|---|---|---|---|---|---|---|---|
| Southeast Tibet Station | Southeast Tibet Observation and Research Station for the Alpine Environment, Chinese Academy of Sciences | 29°46′N | 94°44′E | 3326 | 5.6 | 800–1000 | Subalpine coniferous forest and temperate deciduous conifer mixed forest | Wang et al., (2010) |
| Nam Co Station | Nam Co Monitoring and Research Station for Multisphere Interactions, Chinese Academy of Sciences | 30°47′N | 90°58′E | 4730 | –0.6 | 414.6 | Alpine meadow and alpine steppe | Zhang et al., (2011) |
| Qomolangma Station | Qomolangma Atmospheric and Environmental Observation and Research Station, Chinese Academy of Sciences | 28°13′N | 86°34′E | 4300 | 3.9 | 402.8 | Alpine meadow and alpine steppe | Gao et al., (2014) and M. Li et al., (2007) |
| Ngari Station | Ngari Desert Observation and Research Station | 33°24′N | 79°43′E | 4264 | - | 124.6 | Desert steppe | This study |
| Muztagh Ata Station | Muztagh Ata Westerly Observation and Research Station | 38°17′N | 75°1′E | 3650 | - | 213.6 | Alpine steppe | This study |

**Table 2. Annual mean concentrations of major ions (μeq $L^{-1}$) in precipitation at five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP and other sites in China.** Units of precipitation are millimeters (mm $yr^{-1}$). VWM indicates volume-weighted mean.

| Area | Sites | Represents | Periods | Precipitation | $NH_4^+$ | $Na^+$ | $K^+$ | $Mg^{2+}$ | $Ca^{2+}$ | $NO_3^-$ | $Cl^-$ | $SO_4^{2-}$ | Data type | Reference |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Tibetan Plateau | Southeast Tibet | Remote site | 2011–2012 | 914.6 | 4.9 | 3.8 | 1.0 | 0.9 | 20.8 | 2.2 | 2.5 | 2.1 | VWM | This study |
| | Nam Co | Remote site | 2011–2012 | ~~517~~382.5 | 12.~~6~~7 | 1.~~8~~9 | 0.4 | 0.9 | ~~8.6~~7.<br>9 | 4.~~9~~5 | 1.~~0~~1 | 2.~~8~~9 | VWM | This study |
| | Qomolangma | Remote site | 2011–2012 | ~~402.8~~258 | 2~~8.5~~5<br>.4 | ~~32.1~~2<br>6.0 | ~~7.1~~4.<br>5 | ~~1.6~~1.<br>7 | ~~48.9~~5<br>3.4 | 0.~~6~~8 | ~~31.7~~2<br>7.1 | 3.2 | VWM | This study |
| | Ngari | Remote site | 2013 | 124.6 | 20.5 | 12.4 | 1.8 | 4.8 | 50.9 | 4.8 | 11.9 | 11.6 | VWM | This study |
| | Muztagh Ata | Remote site | 2011 | 213.6 | 42.0 | 10.1 | 3.0 | 11.3 | 119.4 | 9.9 | 8.9 | 17.4 | VWM | This study |
| | Waliguan | Remote site | 1997 | 388 | 45.5 | 8.7 | 3.8 | 12.1 | 34.0 | 8.3 | 6.1 | 24.0 | Mean | Tang et al. (2000) |
| | Wudaoliang | Remote site | Aug. 1989 | 266.5[a] | 27.1 | 21.7 | 6.2 | | | 13.2 | 25.6 | 29.2 | Mean | Yang et al. (1991) |
| | Lhasa | Remote city | 1998–2000 | 250-500 | 14.3 | 11.2 | 5.1 | 10.9 | 197.4 | 6.9 | 9.7 | 5.2 | Mean | Zhang et al. (2003) |
| | Lijiang | City | 1989–2006 | 900 | 11.4 | 2.5 | | 7.7 | 50.2 | 3.6 | 11.6 | 32.6 | Mean | Zhang et al. (2012a) |
| | | | | **Average** | ~~23.0~~2<br>**2.6** | ~~11.6~~1<br>**0.9** | **3.~~5~~2** | **6.3** | **66.~~38~~** | **6.0** | **1~~2.1~~1<br>.6** | **14.~~2~~3** | | |
| Northern China | Beijing | City | 2001–2005 | 441 | 236.0 | 22.5 | 13.8 | 48.4 | 209.0 | 106.0 | 34.9 | 314.0 | VWM | F. Yang et al. (2012) |
| | Dalian | City | 2007 | 602 | 107.8 | 36.2 | 6.87 | 25.29 | 78.92 | 51.38 | 59.83 | 168.0 | VWM | Zhang et al. (2012e) |
| | Nanjing | City | 1992–2003 | 648-1242 | 193.2 | 23.0 | 12.1 | 31.7 | 295.4 | 39.6 | 142.6 | 241.8 | VWM | Tu et al. (2005) |
| | Tianshan Mountain | Remote site | 1995–1996 | | | 55.7 | 14.9 | 15.8 | 78.0 | 22.3 | 40.9 | 88.1 | Mean | Hou (2001) |
| Southern China | Hangzhou | City | 2006–2008 | 1435 | 79.9 | 12.2 | 4.2 | 7.1 | 51.9 | 38.4 | 13.9 | 110.0 | VWM | Xu et al. (2011) |
| | NingBo | City | 2010–2011 | 1374.7 | 46.2 | 22.4 | 7.0 | 9.3 | 31.5 | 38.7 | 31.0 | 72.6 | VWM | Ding et al. (2012) |
| | Shanghai | City | 2005 | 825.5 | 80.7 | 50.1 | 14.9 | 29.6 | 204.0 | 49.8 | 58.3 | 199.6 | VWM | K. Huang et al. (2008) |

| | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Shenzhen | City | 1986–2006 | 1769 | 35.2 | 40.3 | 7.2 | 9.7 | 77.7 | 22.1 | 37.9 | 74.3 | Mean | Y. L. Huang et al. (2008) |
| Guiyang | City | 2008–2009 | 1171 | 112.8 | 13.9 | 9.6 | 10.5 | 182.9 | 7.3 | 20.7 | 265.6 | VMW | Xiao et al. (2013) |

[a] Precipitation amount data was obtained from Yang et al. (2010).

**Table 3. Annual inorganic nitrogen wet deposition (kg N ha$^{-1}$ yr$^{-1}$) at five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP, as well as other sites in China.** Units of precipitation are millimeters (mm yr$^{-1}$). DIN means inorganic nitrogen (sum of $NH_4^+$-N and $NO_3^-$-N). VWM indicates volume-weighted mean.

| Area | Sites | Represents | Periods | Precipitation | $NH_4^+$-N | $NO_3^-$-N | DIN | Data type | Reference |
|---|---|---|---|---|---|---|---|---|---|
| Tibetan Plateau | Southeast | Remote site | 2011–2012 | 914.6 | 0.63 | 0.28 | 0.91 | VWM | This study |
| | Nam Co | Remote site | 2011–2012 | ~~517~~382.5 | 0.~~91~~68 | 0.~~35~~24 | ~~1.26~~0.92 | VWM | This study |
| | Qomolangma | Remote site | 2011–2012 | ~~402.8~~258 | ~~1.61~~0.92 | 0.~~04~~03 | ~~1.6~~0.94 | VWM | This study |
| | Ngari | Remote site | 2013 | 124.6 | 0.36 | 0.08 | 0.44 | VWM | This study |
| | Muztagh Ata | Remote site | 2011 | 213.6 | 1.25 | 0.30 | 1.55 | VWM | This study |
| | Waliguan | Remote site | 1997 | 388 | 2.47 | 0.45 | 2.92 | Mean | Tang et al. (2000) |
| | Wudaoliang | Remote site | Aug. 1989 | 266.5[a] | 1.55 | 1.11 | 2.66 | Mean | Yang et al. (1991) |
| | Lhasa | Remote city | 1998–2000 | 250-500 | 0.54 | 0.45 | 0.99 | Mean | Zhang et al. (2003) |
| | Naidong | Remote city | 2006–2007 | 451 | 0.91 | 0.82 | 1.72 | VWM | Jia (2008) |
| | Biru | Remote city | 2006–2007 | 582 | 1.22 | 1.86 | 3.08 | VWM | Jia (2008) |
| | Jiangda | Remote city | 2006–2007 | 547 | 1.11 | 0.80 | 1.91 | VWM | Jia (2008) |
| | Lijiang | Remote city | 1989–2006 | 900 | 1.43 | 0.46 | 1.89 | Mean | N. N. Zhang et al. (2012) |
| | | | | **Average** | **1.~~17~~09** | **0.~~58~~57** | **1.~~75~~66** | | |
| Northern China | Beijing | City | 2001–2005 | 441 | 14.57 | 6.54 | 21.12 | VWM | F. Yang et al. (2012) |
| | Dalian | City | 2007 | 602 | 9.08 | 4.33 | 13.41 | VWM | X. Y. Zhang et al. (2012) |
| | Nanjing | City | 1992–2003 | 648-1242 | 25.56 | 5.23 | 30.79 | VWM | Tu et al. (2005) |
| | Beijing | City | 2008–2010 | 572 | | | 27.9 | VWM | Pan et al. (2012) |
| | Tianjin | City | 2008–2010 | 544 | | | 18.1 | VWM | Pan et al. (2012) |
| | Baoding | Industrial | 2008–2010 | 513 | | | 23.1 | VWM | Pan et al. (2012) |
| | Tanggu | Industrial | 2008–2010 | 566 | | | 28.2 | VWM | Pan et al. (2012) |
| | Tangshan | Industrial | 2008–2010 | 610 | | | 21.6 | VWM | Pan et al. (2012) |
| | Yangfang | Suburban | 2008–2010 | 404 | | | 20.7 | VWM | Pan et al. (2012) |
| | Cangzhou | Suburban | 2008–2010 | 605 | | | 22.6 | VWM | Pan et al. (2012) |
| | Luancheng | Agricultural | 2008–2010 | 517 | | | 22.2 | VWM | Pan et al. (2012) |
| | Yucheng | Agricultural | 2008–2010 | 566 | | | 24.8 | VWM | Pan et al. (2012) |
| | Xinglong | Rural | 2008–2010 | 512 | | | 16.3 | VWM | Pan et al. (2012) |
| Southern China | TieShanPing | Remote site | 1999–2004 | 1228 | 25.50 | 9.80 | 35.30 | VWM | Chen and Mulder (2007) |
| | LiuChongGuan | Remote site | 1999–2004 | 854 | 2.40 | 1.30 | 3.70 | VWM | Chen and Mulder (2007) |
| | LeiGongShan | Remote site | 1999–2004 | 1714 | 3.70 | 2.60 | 6.30 | VWM | Chen and Mulder (2007) |
| | CaiJiaTang | Remote site | 1999–2004 | 1232 | 21.10 | 12.70 | 33.80 | VWM | Chen and Mulder (2007) |
| | LiuXiHe | Remote site | 1999–2004 | 1620 | 4.30 | 7.50 | 11.80 | VWM | Chen and Mulder (2007) |
| | Hangzhou | City | 2006–2008 | 1435 | 16.1 | 7.7 | 23.77 | VWM | Xu et al. (2011) |
| | Ningbo | City | 2010–2011 | 1374.7 | 8.9 | 7.4 | 16.34 | VWM | Ding et al. (2012) |
| | Shanghai | City | 2005 | 825.5 | 9.3 | 5.8 | 15.08 | VWM | K. Huang et al. (2008) |
| | Shenzhen | City | 1986–2006 | 1769 | 8.7 | 5.5 | 14.19 | Mean | Y. L. Huang et al. (2008) |
| | Guiyang | City | 2008–2009 | 1171 | 18.5 | 1.2 | 19.69 | VMW | Xiao et al. (2013) |

Notes: Tang et al. (2000), Yang et al. (1991), Zhang et al. (2003), N. N. Zhang et al. (2012), F. Yang et al. (2012), X. Y. Zhang et al. (2012), Xu et al. (2011), Ding et al. (2012), K. Huang et al. (2008), Y. L. Huang et al. (2008) and Xiao et al. (2013) reported the concentrations of $NH_4^+$-N and $NO_3^-$-N in precipitation, but did not calculate nitrogen wet deposition.

903 For these previous studies, we recalculated the annual inorganic nitrogen wet deposition according to the reported
904 concentrations of $NH_4^+$-N and $NO_3^-$-N in precipitation and annual precipitation. [a] Precipitation amount data was obtained
905 from Yang et al. (2010).

906

**Table 4. Enrichment factors relative to seawater and soil for precipitation constituents of five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP.**

| | Southeast Tibet Station | | Nam Co Station | | Qomolangma Station | | Ngari Station | | Muztagh Ata Station | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $EF_{sea}$ | $EF_{soil}$ | $EF_{sea}$ | $EF_{soil}$ | $EF_{sea}$ | $EF_{soil}$ | $EF_{sea}$ | $EF_{soil}$ | $EF_{sea}$ | $EF_{soil}$ | $[X/Na^+]_{sea}$ | $[X/Ca^{2+}]_s$ |
| $a^+$ | 1.0 | 0.36 | 1.0 | 0.~~42~~48 | 1.0 | ~~1.3~~0.99 | 1.0 | 0.5 | 1.0 | 0.17 | 1.0000 | 0.5690 |
| $H_4^+$ | 15707 | 350 | ~~8497~~480684 | ~~2164~~2378 | ~~1059~~511629 | ~~859~~701 | 19706 | 594 | 49751 | 519 | 0.0001[a] | 0.0006[c] |
| $^+$ | 11.5 | 0.18 | 8.5~~10.0~~ | 0.~~18~~17 | ~~10.0~~7.83 | 0.~~56~~33 | 6.4 | 0.13 | 13.5 | 0.10 | 0.0220 | 0.5040 |
| $g^{2+}$ | 1.1 | 0.05 | 2.~~2~~0 | 0.~~11~~12 | 0.~~22~~29 | 0.03 | 1.7 | 0.10 | 4.9 | 0.10 | 0.2270 | 0.5610 |
| $a^{2+}$ | 126.2 | 1.0 | ~~110~~95.3 | 1.0 | ~~34.7~~46.6 | 1.0 | 93.1 | 1.0 | 269.5 | 1.0 | 0.0440 | 1.0000 |
| $l^-$ | 0.57 | 67.3 | 0.50 | ~~67.1~~77.8 | 0.~~85~~90 | ~~365~~286 | 0.8 | 132 | 0.76 | 42.2 | 1.1600 | 0.0031 |
| $O_3^-$ | 23842 | 154 | ~~113217~~98242 | ~~837~~840 | ~~832~~1237 | ~~19.6~~21.7 | 15869 | 139 | 40619 | 123 | 0.0000[b] | 0.0021[d] |
| $O_4^{2-}$ | 4.7 | 13.0 | ~~13~~12.~~0~~7 | 4~~1.5~~6.7 | ~~0.83~~1.03 | ~~8.4~~7.76 | 7.7 | 29.2 | 14.3 | 18.6 | 0.1210 | 0.0188[e] |

[a] Marine nitrogen ions regarded as entire $NH_4^+$.

[b] Marine nitrogen ions regarded as entire $NO_3^-$.

[c] Soil nitrogen regarded as entire range of $NH_3$ compounds.

[d] Soil nitrogen regarded as entire range of $NO_3$ compounds.

[e] Soil sulfur regarded as entire range of $SO_4$ compounds.

**Table 5. Source contributions (%) for major ions in precipitation of five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP.** SSF indicates sea salt fraction; CF indicates crust fraction; AF indicates anthropogenic fraction.

| | Southeast Tibet Station | | | Nam Co Station | | | Qomolangma Station | | | Ngari Station | | | Muztagh Ata Station | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | SSF | CF | AF | SSF | CF | AF | SSF | CF | AF | SSF | CF | AF | SSF | CF | AF |
| $NH_4^+$ | 0.0 | 0.3 | **99.7** | 0.0 | 0.0 | **100~~.0~~** | 0.0 | 0.1 | **99.~~9~~8** | 0.0 | 0.2 | **99.8** | 0.0 | 0.2 | **99.8** |
| $NO_3^-$ | 0.0 | 0.6 | **99.3** | 0.0 | 0.1 | **99.9** | 0.1 | ~~5.1~~4.6 | **~~94.8~~5.3** | 0.0 | 0.7 | **99.3** | 0.0 | 0.8 | **99.2** |
| $SO_4^{2-}$ | 21.3 | 7.7 | **71.0** | 7.~~7~~9 | 2.41 | **~~89.9~~90.0** | **~~88.0~~7.1** | 12.~~0~~9 | | 12.9 | 3.4 | **83.7** | 7.0 | 5.4 | **87.6** |
| $Ca^{2+}$ | 0.8 | **99.2** | | ~~0.9~~1.0 | **99.~~1~~0** | | 2.~~9~~1 | **97.~~1~~9** | | 1.1 | **98.9** | | 0.4 | **99.6** | |
| $K^+$ | 8.7 | **91.3** | | 1~~0.0~~1.8 | **~~90.0~~88.2** | | 1~~0.0~~2.8 | **~~90.0~~87.2** | | 15.5 | **84.5** | | 7.4 | **92.6** | |
| $Mg^{2+}$ | **92.9** | 7.1 | | 4~~4.8~~9.0 | **5~~5.2~~1.0** | | 0**~~100~~** | **100** | | **59.2** | 40.8 | | 20.2 | **79.8** | |
| $Cl^-$ | **98.5** | 1.5 | | **98.~~5~~7** | 1.~~5~~3 | | **99.7** | 0.3 | | **99.2** | 0.8 | | **97.6** | 2.4 | |
| $Na^+$ | **100** | | | **100** | | | **100** | | | **100** | | | **100** | | |

Table 6: Varimax-rotated principal component analysis of major ions in precipitation at five remote sites on the TP. PC1, PC2 and PC3 indicates the 1st, 2nd and 3rd component, respectively. CT means communality. *N* indicates the number of precipitation samples at each site. Boldfaced values are the largest value among the 3 component for each ion.

| | Southeast Tibet (*N* = 53) | | | | Nam Co (*N* = 27) | | | | Qomolangma (*N* = 30) | | | | Ngari (*N* = 39) | | | | Muztagh Ata (*N* = 19) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | RC1 | RC2 | RC3 | CT | RC1 | RC2 | RC3 | CT | RC1 | RC2 | RC3 | CT | RC1 | RC2 | RC3 | CT | RC1 | RC2 | RC3 | CT |
| $Na^+$ | **0.94** | 0.02 | 0.24 | 0.94 | 0.62 | **0.73** | 0.00 | 0.93 | **0.91** | 0.28 | 0.10 | 0.92 | 0.62 | **0.73** | 0.06 | 0.92 | 0.22 | **0.96** | 0.08 | 0.98 |
| $NH_4^+$ | 0.25 | 0.22 | **0.93** | 0.98 | 0.36 | 0.13 | **0.89** | 0.94 | **0.69** | -0.15 | 0.24 | 0.56 | -0.11 | 0.57 | **0.75** | 0.90 | -0.19 | 0.14 | **0.96** | 0.98 |
| $K^+$ | **0.88** | 0.10 | 0.35 | 0.91 | 0.11 | **0.93** | 0.07 | 0.89 | **0.84** | 0.35 | 0.27 | 0.89 | 0.15 | **0.84** | 0.40 | 0.89 | 0.47 | **0.79** | -0.05 | 0.85 |
| $Mg^{2+}$ | **0.77** | 0.45 | 0.29 | 0.89 | **0.89** | 0.19 | 0.38 | 0.97 | 0.39 | **0.88** | 0.18 | 0.97 | **0.67** | 0.56 | 0.09 | 0.78 | **0.90** | 0.38 | -0.16 | 0.99 |
| $Ca^{2+}$ | **0.66** | 0.46 | 0.21 | 0.70 | **0.76** | 0.11 | 0.57 | 0.92 | -0.02 | **0.96** | 0.03 | 0.92 | **0.85** | 0.17 | 0.15 | 0.77 | **0.85** | 0.35 | -0.25 | 0.90 |
| $Cl^-$ | **0.91** | 0.10 | -0.06 | 0.85 | 0.05 | **0.95** | 0.20 | 0.95 | **0.92** | 0.25 | 0.11 | 0.92 | 0.37 | **0.83** | 0.00 | 0.82 | 0.25 | **0.92** | 0.17 | 0.94 |
| $NO_3^-$ | 0.03 | **0.95** | 0.09 | 0.91 | 0.32 | 0.13 | **0.92** | 0.96 | 0.26 | 0.16 | **0.94** | 0.98 | 0.36 | -0.01 | **0.84** | 0.84 | **0.88** | 0.12 | -0.18 | 0.83 |
| $SO_4^{2-}$ | 0.24 | **0.89** | 0.18 | 0.89 | **0.80** | 0.10 | 0.52 | 0.92 | **0.64** | **0.64** | 0.31 | 0.90 | **0.91** | 0.22 | 0.15 | 0.90 | **0.87** | 0.31 | 0.11 | 0.87 |
| Variance (%) | 46 | 27 | 15 | | 33 | 30 | 30 | | 43 | 30 | 15 | | 34 | 33 | 19 | | 43 | 35 | 14 | |
| Cumulative (%) | 46 | 73 | 88 | | 33 | 63 | 93 | | 43 | 74 | 88 | | 34 | 66 | 85 | | 43 | 78 | 92 | |

## Figure captions

**Figure 1. Map of the precipitation nitrogen wet deposition sampling sites on the ~~Tibetan Plateau~~TP.** The red points indicate the five remote sampling stations of this study. The black points indicate the sampling sites from previous records. Southeast Tibet Station is short for Southeast Tibet Observation and Research Station for the Alpine Environment, Chinese Academy of Sciences; Nam Co Station is short for Nam Co Monitoring and Research Station for Multisphere Interactions, Chinese Academy of Sciences; Qomolangma Station is short for Qomolangma Atmospheric and Environmental Observation and Research Station, Chinese Academy of Sciences; Ngari Station is short for Ngari Desert Observation and Research Station; and Muztagh Ata Station is short for Muztagh Ata Westerly Observation and Research Station.

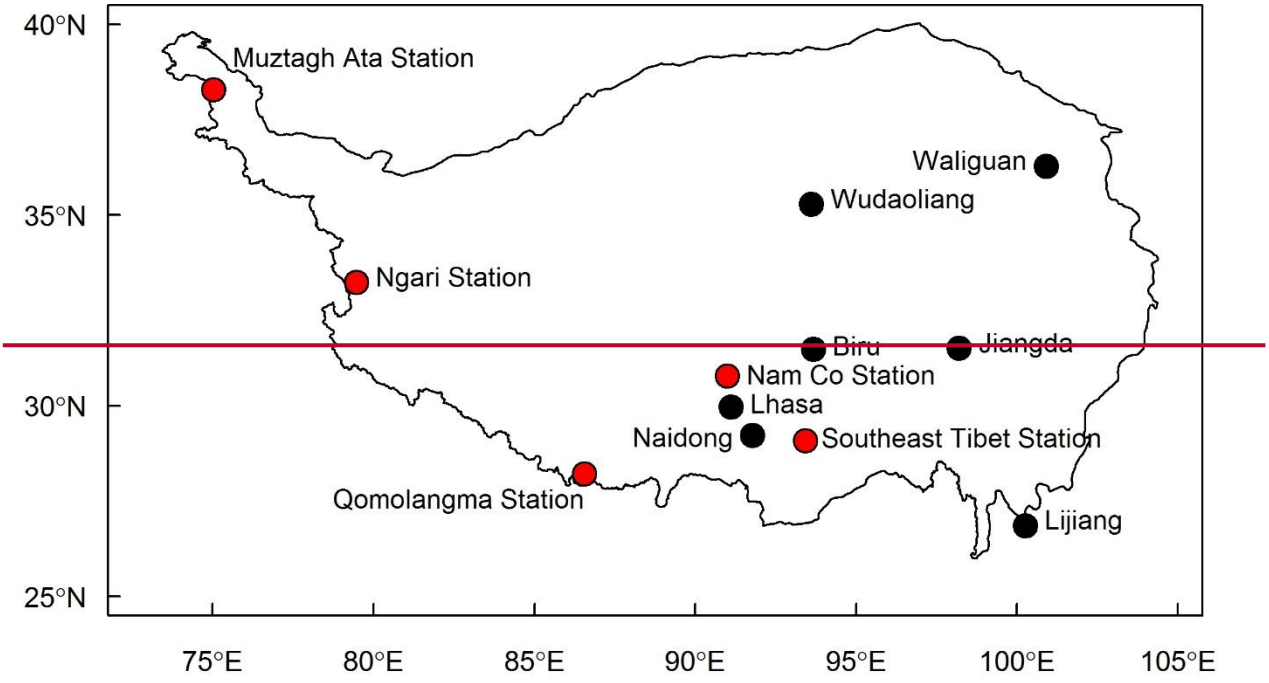
**Figure 2. Seasonal dynamics of ion concentrations (unit: μeq $L^{-1}$) and precipitation (unit: mm) at five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP.** The sampling times of the five stations were as follows: Southeast Tibet Station, November 2011 to October 2012; Nam Co Station, August 2011 to July 2012; Qomolangma Station, April 2011 to March 2012; Ngari Station, January 2013 to December 2013; Muztagh Ata Station, January 2011 to December 2011.

**Figure 3. Annual average volume-weighted concentration percentages of measured ions in precipitation (unit: μeq $L^{-1}$/μeq $L^{-1}$) at five remote sites ~~stations~~ on the ~~Tibetan Plateau~~TP.**

**Figure 4. Seasonal dynamics of inorganic N wet deposition at five remote ~~stations~~ sites on the ~~Tibetan Plateau~~TP.** The sampling time windows of those stations are same to Fig. 2.

**Figure 5. Seven-day backward trajectories at five remote sites on the TP.** Black lines show the backward trajectories calculated at 6-h interval (00:00, 06:00, 12:00, 18:00 UTC) at sampling days, with an arrival height of 500 m above the ground. Red lines show the clustering trajectories.

**Figure 1.**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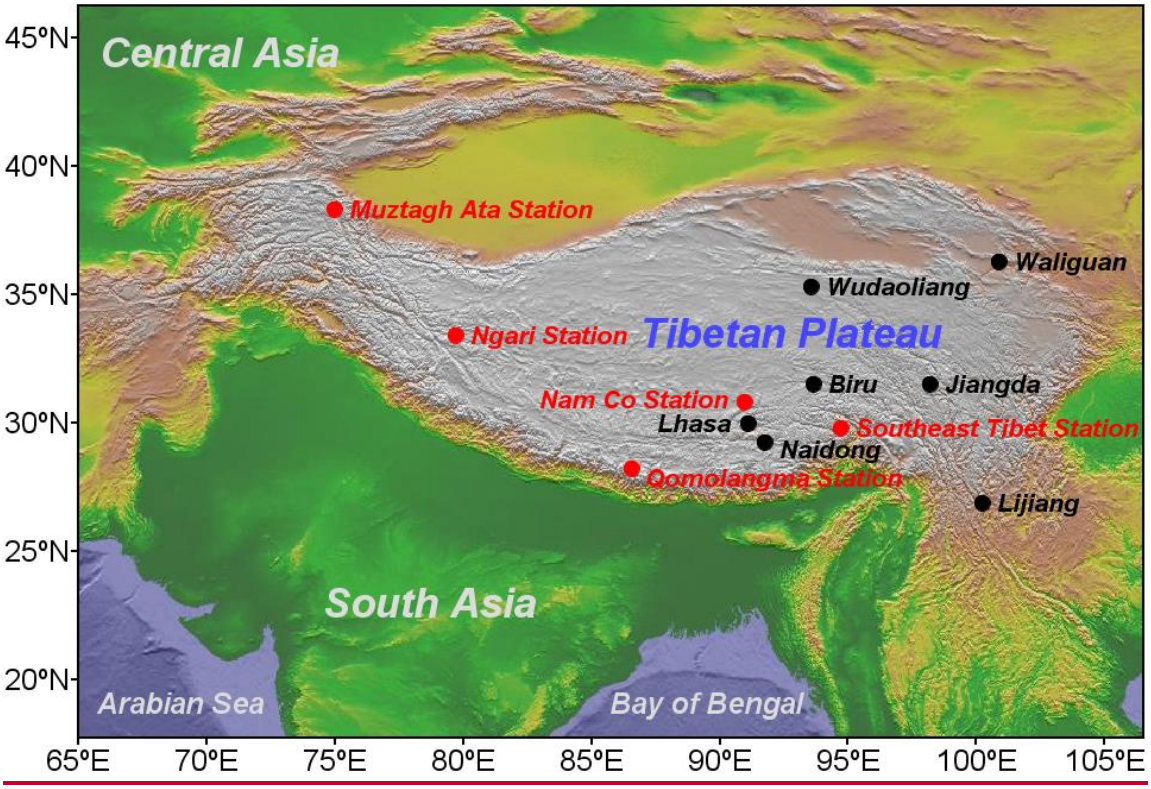
35°N
30°N
25°N
75°E
80°E
85°E
90°E
95°E
100°E
105°E
Muztagh Ata Station
Waliguan
Wudaoliang
Ngari Station
Biru
Jiangda
Nam Co Station
Lhasa
Naidong
Southeast Tibet Station
Qomolangma Station
Lijiang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40°N
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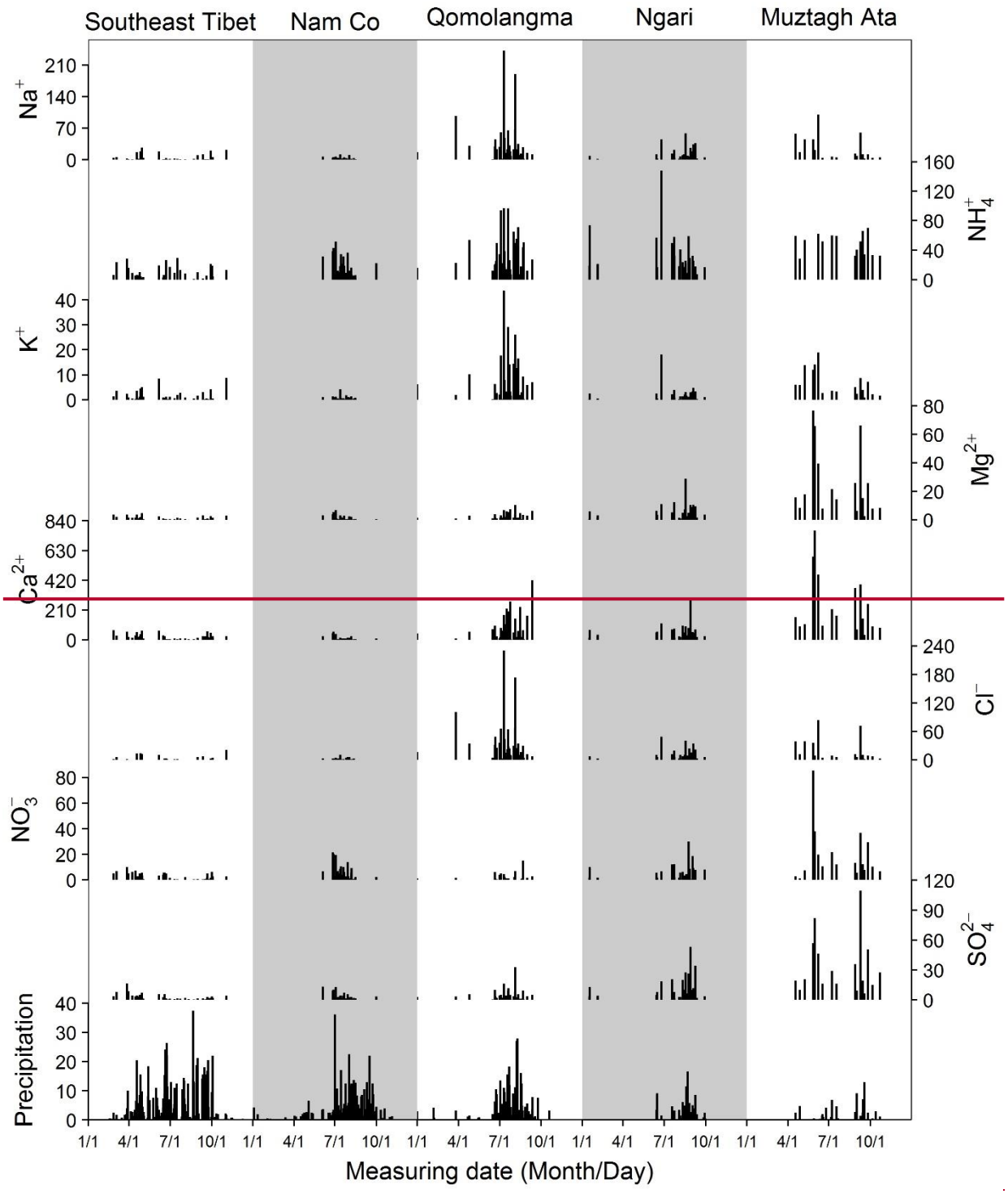
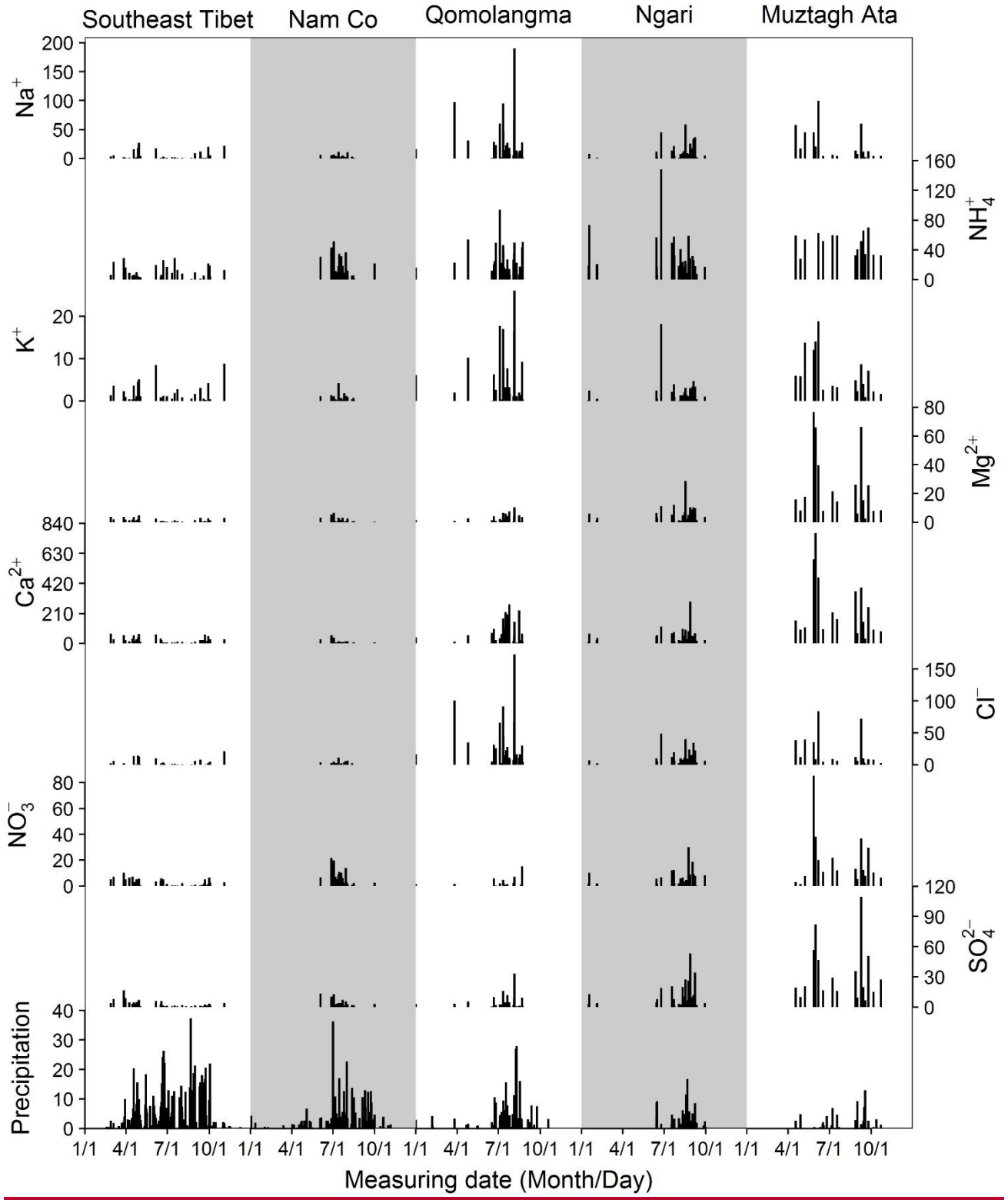
65°E
70°E
75°E
80°E
85°E
90°E
95°E
100°E
105°E
Central Asia
Muztagh Ata Station
Waliguan
Wudaoliang
Ngari Station
Tibetan Plateau
Biru
Jiangda
Nam Co Station
Lhasa
Southeast Tibet Station
Naidong
Qomolangma Station
Lijiang
South Asia
Arabian Sea
Bay of Bengal

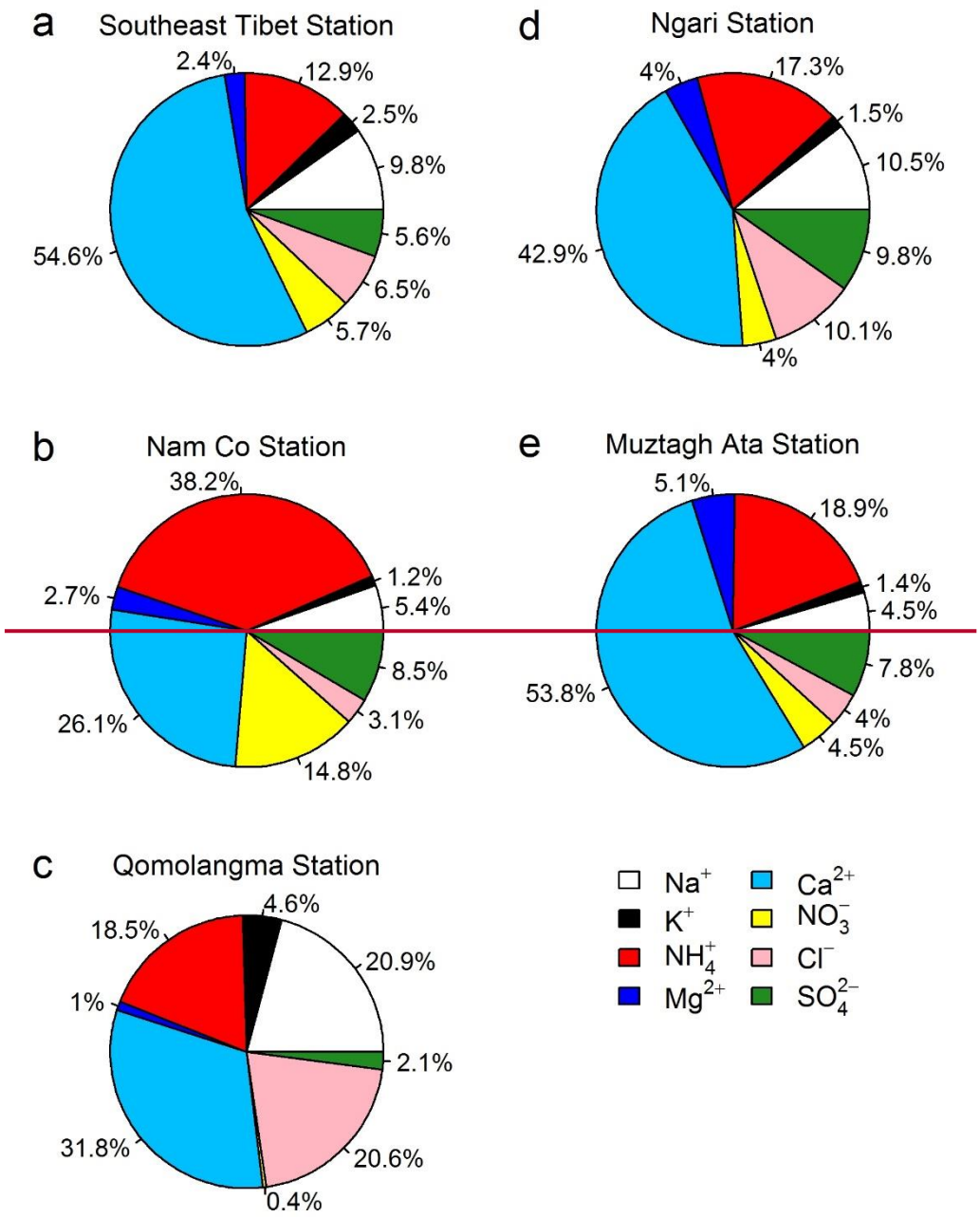
**Figure 2.**

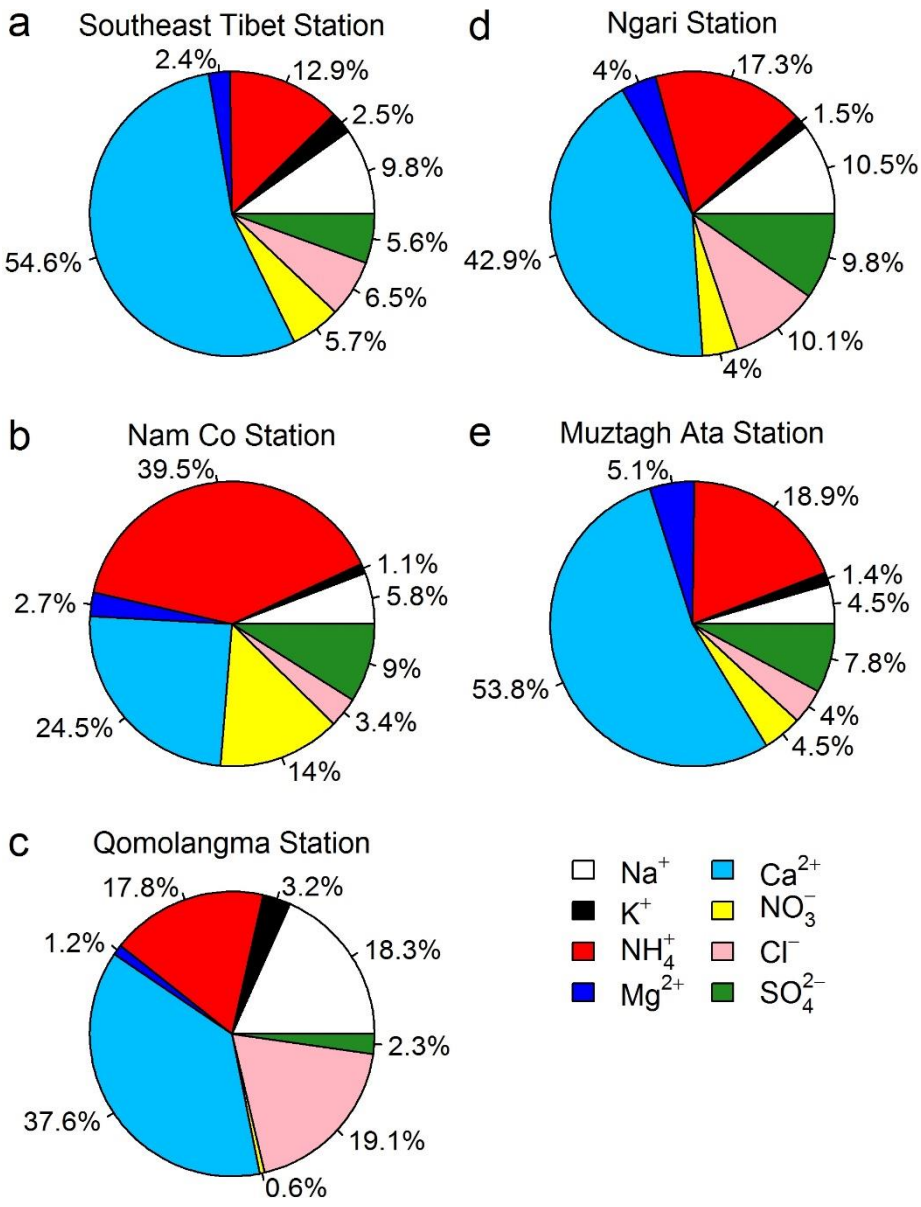

Southeast Tibet
Nam Co
Qomolangma
Ngari
Muztagh Ata
Na+
K+
Ca2+
NO3−
Precipitation
NH4+
Mg2+
Cl−
SO4 2−
1/1 4/1 7/1 10/1
Measuring date (Month/Day)


Southeast Tibet
Nam Co
Qomolangma
Ngari
Muztagh Ata
Na+
K+
Ca2+
NO3−
Precipitation
NH4+
Mg2+
Cl−
SO42−
1/1 4/1 7/1 10/1
Measuring date (Month/Day)

952

**Figure 3.**


a
Southeast Tibet Station
2.4%
12.9%
2.5%
9.8%
5.6%
6.5%
5.7%
54.6%
d
Ngari Station
4%
17.3%
1.5%
10.5%
9.8%
10.1%
4%
42.9%
b
Nam Co Station
38.2%
1.2%
5.4%
8.5%
3.1%
14.8%
26.1%
2.7%
e
Muztagh Ata Station
5.1%
18.9%
1.4%
4.5%
7.8%
4%
4.5%
53.8%
c
Qomolangma Station
4.6%
20.9%
2.1%
20.6%
0.4%
31.8%
1%
18.5%
Na+
K+
NH4+
Mg2+
Ca2+
NO3−
Cl−
SO42−


a
Southeast Tibet Station
2.4%
12.9%
2.5%
9.8%
5.6%
6.5%
5.7%
54.6%
d
Ngari Station
4%
17.3%
1.5%
10.5%
9.8%
10.1%
4%
42.9%
b
Nam Co Station
39.5%
1.1%
5.8%
9%
3.4%
14%
24.5%
2.7%
e
Muztagh Ata Station
5.1%
18.9%
1.4%
4.5%
7.8%
4%
4.5%
53.8%
c
Qomolangma Station
17.8%
3.2%
18.3%
2.3%
19.1%
0.6%
37.6%
1.2%
Na+
K+
NH4+
Mg2+
Ca2+
NO3−
Cl−
SO42−

955

956

**Figure 4.**

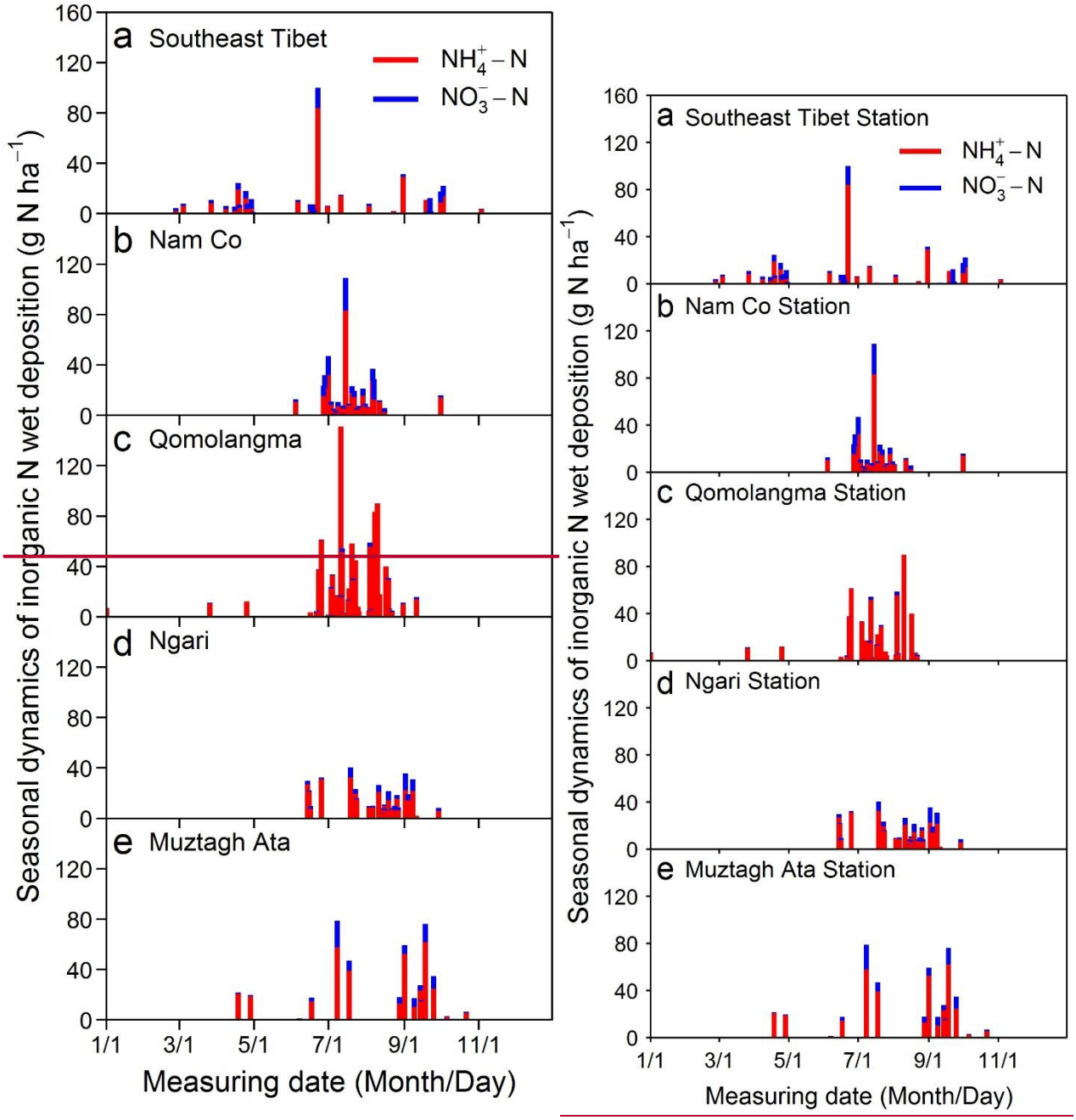
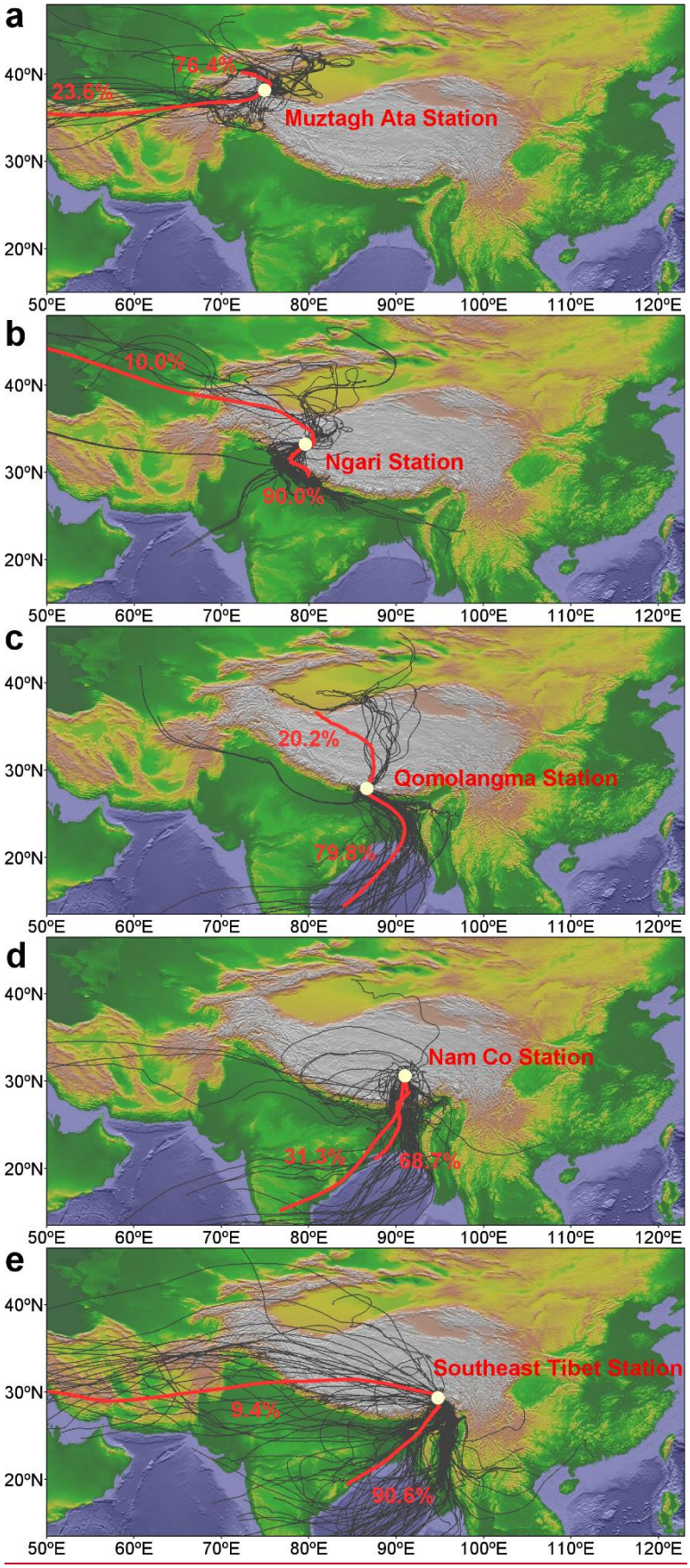

a Southeast Tibet
b Nam Co
c Qomolangma
d Ngari
e Muztagh Ata
$NH_4^+ - N$
$NO_3^- - N$
Seasonal dynamics of inorganic N wet deposition (g N ha$^{-1}$)
Measuring date (Month/Day)
1/1
3/1
5/1
7/1
9/1
11/1
0
40
80
120
160
a Southeast Tibet Station
b Nam Co Station
c Qomolangma Station
d Ngari Station
e Muztagh Ata Station

**Figure 5.**


a
76.4%
23.6%
Muztagh Ata Station
b
10.0%
Ngari Station
90.0%
c
20.2%
Qomolangma Station
79.8%
d
Nam Co Station
31.3%
68.7%
e
Southeast Tibet Station
9.4%
90.6%
40°N
30°N
20°N
50°E
60°E
70°E
80°E
90°E
100°E
110°E
120°E