Interactive comment on "Wet deposition of atmospheric inorganic nitrogen at five remote stations on the Tibetan Plateau" *by* Y. W. Liu et al.

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.

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.

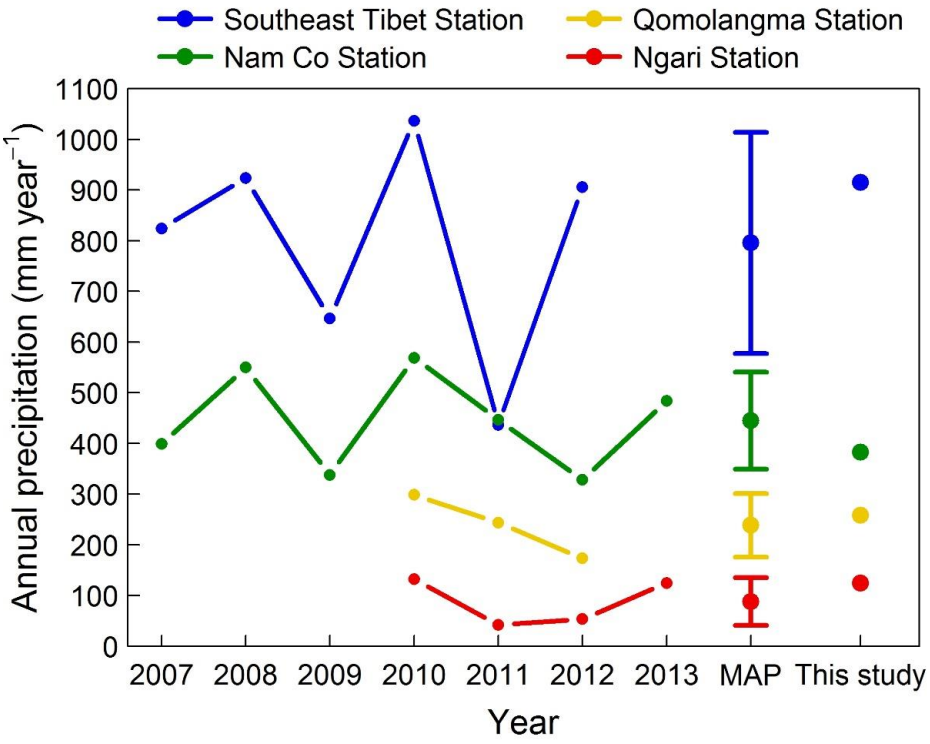

Figure R1. Annual precipitation at four sites on the TP. MAP indicates mean annual precipitation (mean±1 standard deviation).

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***". 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.***"

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 R1) and (2) backward trajectories (Fig. R2) 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:

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.

Table R1: 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	

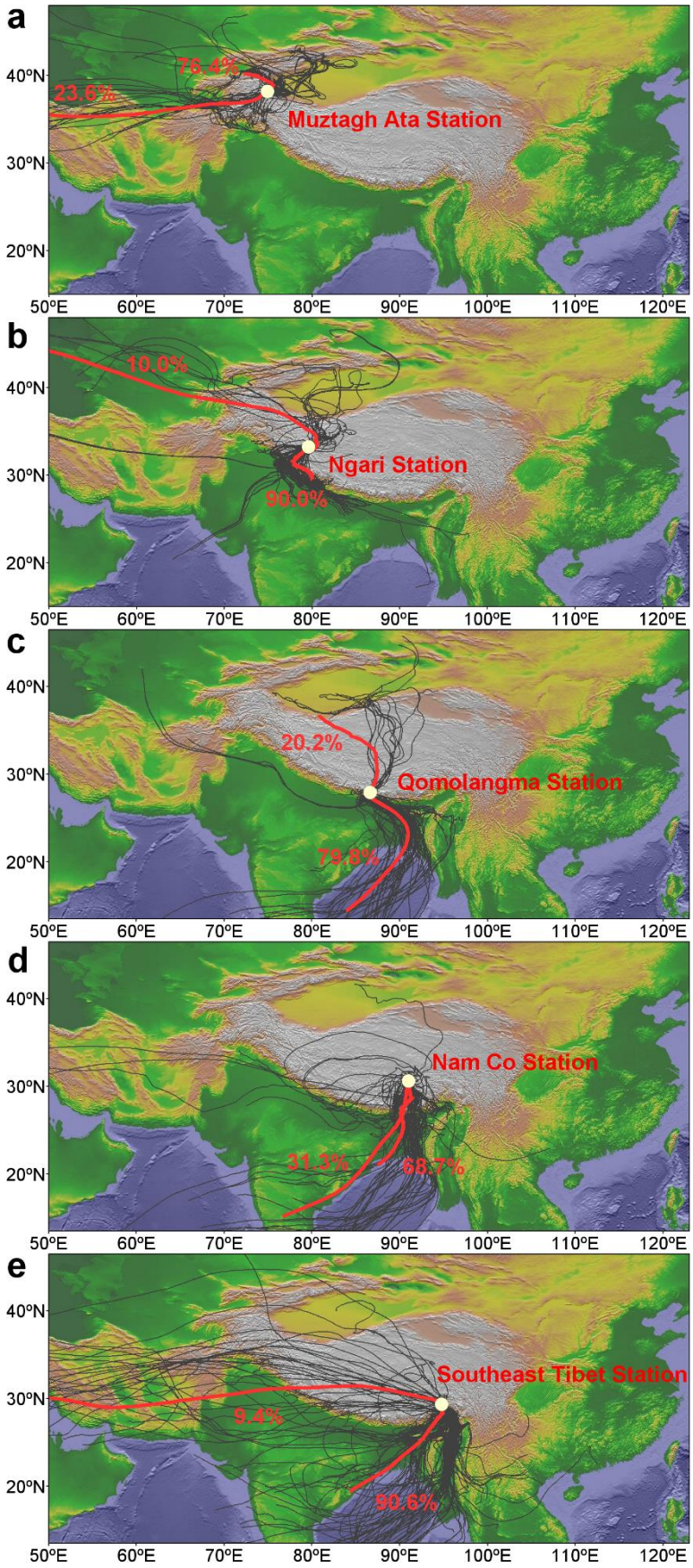

Figure R2 Seven-day backward trajectories at five remote sites on the TP. Black lines show the backward trajectories calculated at 6-h interval (00:00, 06:00, 12:00, 18:00 UTC) at sampling days, with an arrival height of 500 m above the ground. Red lines show the clustering trajectories.

Reference

Liu, Y. W., Xu-Ri, Xu, X. L., Wei, D., Wang, Y. H., and Wang, Y. S.: Plant and soil responses of an alpine steppe on the Tibetan Plateau to multi-level nitrogen addition, Plant Soil, 373, 515-529, doi: 10.1007/s11104-013-1814-x, 2013.