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 #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 R1 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 R2). 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 3.1×10^5 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. R1 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. R1). At the other four sites, the air masses at sampling days were mainly transported from South Asia (Fig. R1). 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 R1). 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 R1). 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 R1). 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 R1). 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. 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 and backward trajectory analysis. Section 4.2 was almost rewrote 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	

Table R2: Pearson correlations matrix of ionic concentrations in precipitation at five remote sites on the TP. *N* indicates the number of precipitation samples at each site. Only statistically significant correlation coefficient are shown ($P < 0.05$).

Sites		Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Southeast Tibet	Na^+	1							
(*N* = 53)	NH_4^+	0.45	1						
	K^+	0.89	0.57	1					
	Mg^{2+}	0.81	0.55	0.77	1				
	Ca^{2+}	0.69	0.41	0.64	0.82	1			
	Cl^-	0.8	—	0.83	0.68	0.48	1		
	NO_3^-	—	0.31	—	0.44	0.42	—	1	
	SO_4^{2-}	—	0.43	0.4	0.61	0.53	0.35	0.81	1
Nam Co	Na^+	1							
(*N* = 27)	NH_4^+	—	1						
	K^+	0.69	—	1					
	Mg^{2+}	0.69	0.69	—	1				
	Ca^{2+}	0.52	0.75	—	0.91	1			
	Cl^-	0.74	—	0.84	—	—	1		
	NO_3^-	—	0.91	—	0.67	0.8	—	1	
	SO_4^{2-}	0.55	0.77	—	0.9	0.88	—	0.71	1
Qomolangma	Na^+	1							
(*N* = 30)	NH_4^+	0.45	1						
	K^+	0.83	0.63	1					
	Mg^{2+}	0.61	—	0.69	1				
	Ca^{2+}	—	—	—	0.85	1			
	Cl^-	1	0.47	0.83	0.58	—	1		
	NO_3^-	0.42	—	0.5	0.41	—	0.42	1	
	SO_4^{2-}	0.8	—	0.82	0.84	0.57	0.78	0.55	1
Ngari	Na^+	1							
(*N* = 39)	NH_4^+	0.35	1						
	K^+	0.7	0.77	1					
	Mg^{2+}	0.89	0.28	0.54	1				
	Ca^{2+}	0.56	—	0.44	0.59	1			
	Cl^-	0.81	0.37	0.68	0.65	0.39	1		
	NO_3^-	0.32	0.47	—	0.38	—	—	1	
	SO_4^{2-}	0.72	—	0.4	0.63	0.84	0.52	0.4	1
Muztagh Ata	Na^+	1							
(*N* = 19)	NH_4^+	—	1						
	K^+	0.82	—	1					
	Mg^{2+}	0.56	—	0.71	1				
	Ca^{2+}	0.49	—	0.74	0.92	1			
	Cl^-	0.96	—	0.74	0.56	—	1		
	NO_3^-	—	—	0.51	0.88	0.77	—	1	
	SO_4^{2-}	0.5	—	0.6	0.9	0.81	0.56	0.68	1

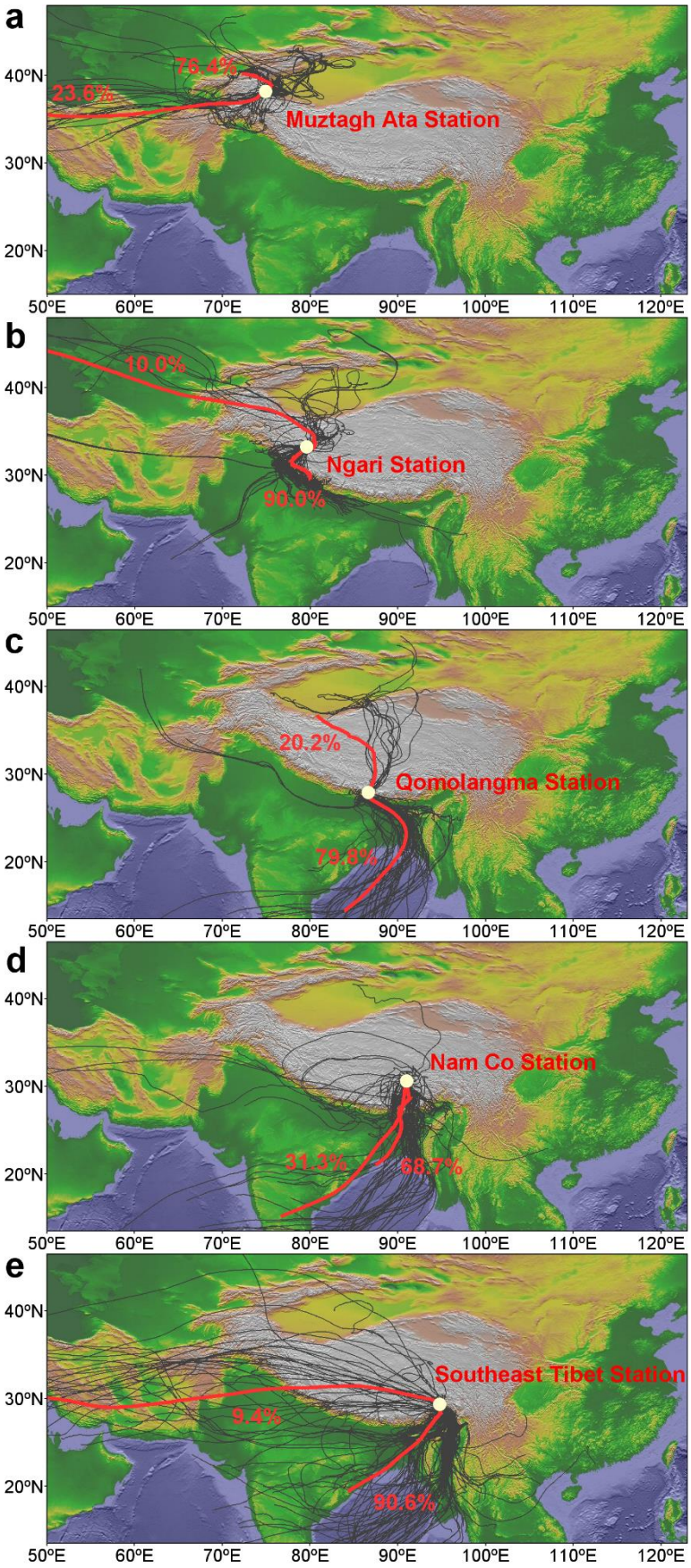

Figure R1 Seven-day backward trajectories at five remote sites on the TP. Black lines show the backward trajectories calculated at 6-h interval (00:00, 06:00, 12:00, 18:00 UTC) at sampling days, with an arrival height of 500 m above the ground. Red lines show the clustering trajectories.

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