Supplement of Atmos. Chem. Phys., 20, 11551–11567, 2020
https://doi.org/10.5194/acp-20-11551-2020-supplement

Supplement of

Characterizing the spatiotemporal nitrogen stable isotopic composition of ammonia in vehicle plumes

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Section S1. Denuder and Filter Preparation, Extraction, and Handling Protocol

All denuders and filters were cleaned and coated fresh daily. Denuder and filter coating solutions were produced using ACS certified reagents, and all water used for cleaning, coating, and extraction was of ultra-high purity grade (>18.2 MΩ). Denuders were coated using 10 mL of the appropriate solution and air-dried for at least 1 h. PTFE and Nylon filters were rinsed and sonicated with 20 mL of water for 30 minutes three times and dried in an oven at 50 ºC for at least thirty minutes. The cellulose filters were not rinsed before use due to disintegration issues and were directly impregnated with a 0.5 mL of the 5 % citric acid (w/v) solution and dried with ultra-high purity nitrogen (>99.9 %). The denuder-filter packs were loaded, and the inlet and outlet were capped and transported to the field site. Immediately after collection, the cartridges were capped and transported back to the laboratory for extraction. Periodically, field blanks were taken for all sample types (i.e., filters and denuders), representing approximately 10 % of the total number of collected samples. Replicate samples (e.g., side-by-side samples) were periodically conducted at the near-highway site during summer to determine measurement precision. The ALPHA samplers were prepared in the laboratory, capped, and then transported to the field site. After collection, the passive samplers were immediately capped and transported back to the laboratory for extraction.

Collections on the denuders and filters were extracted using water. The PTFE filters were pre-wetted with 500 µL of ethanol to wet its hydrophobic surface before extraction. All filter samples were sonicated for 1 h. Filters were then removed using cleaned forceps, and the extraction solutions were then passed through a 0.22 µm syringe filter to remove loose filter pieces and potential microbial contaminants. After extraction, all solutions were placed in a freezer at -30 ºC until subsequent concentration and isotopic analysis.

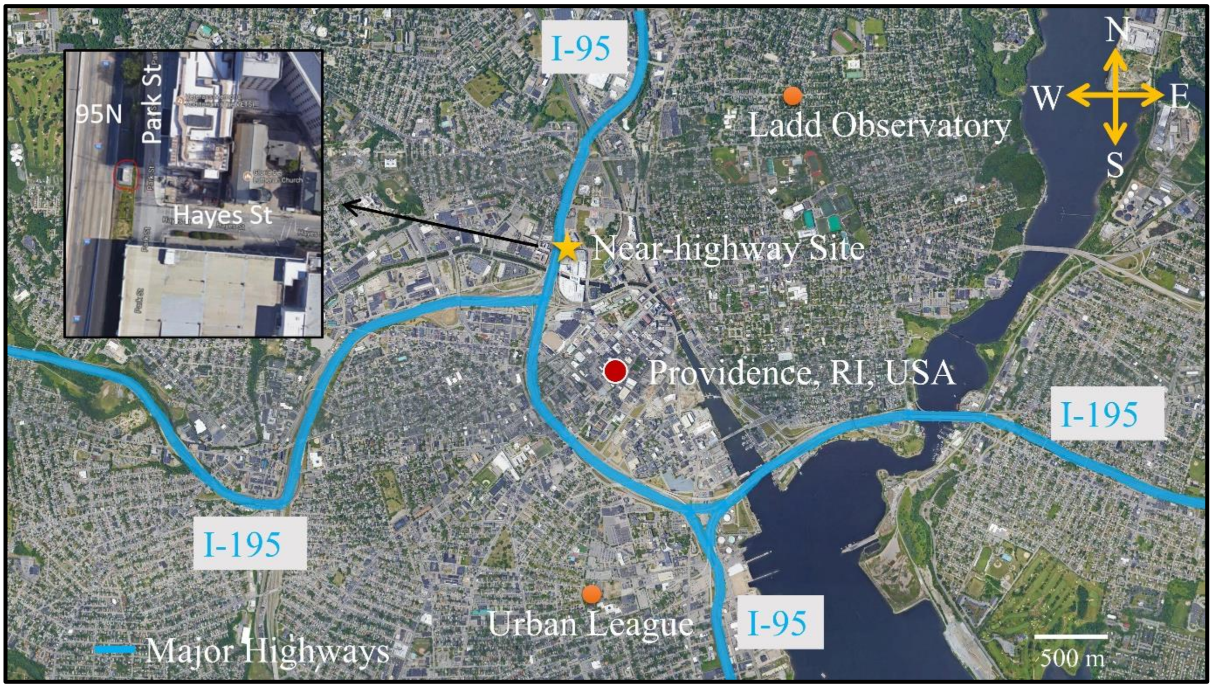

Figure S1: Google Earth image of Providence, RI, US with the locations of the near-highway stationary monitoring site (star), meteorological data monitoring locations (circles), and major interstate highway routes (turquoise). Copyright © 2020 Google.

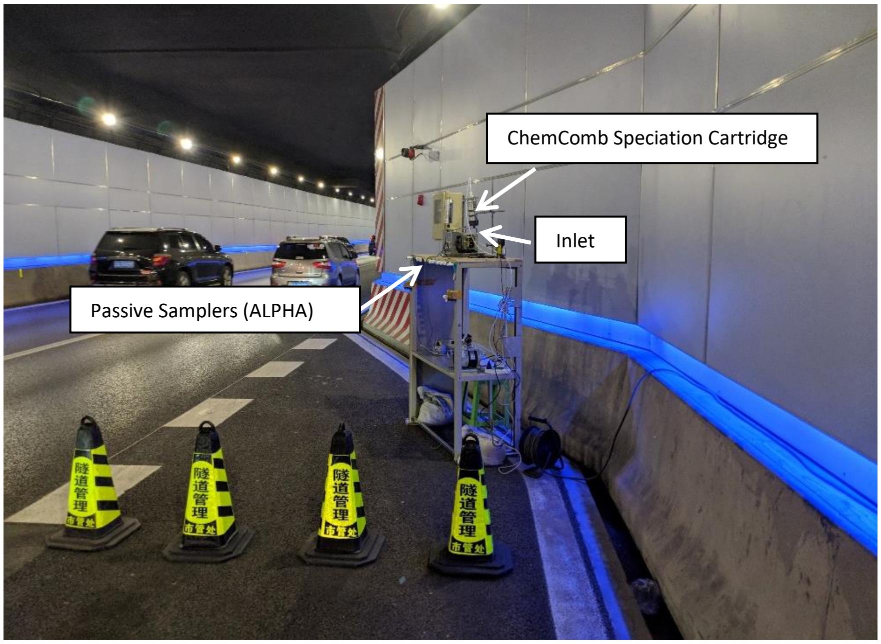

Figure S2: Image of the sampling set-up for collection of NH_3 from fresh vehicle traffic plumes in a tunnel at Shenyang, Liaoning, China.

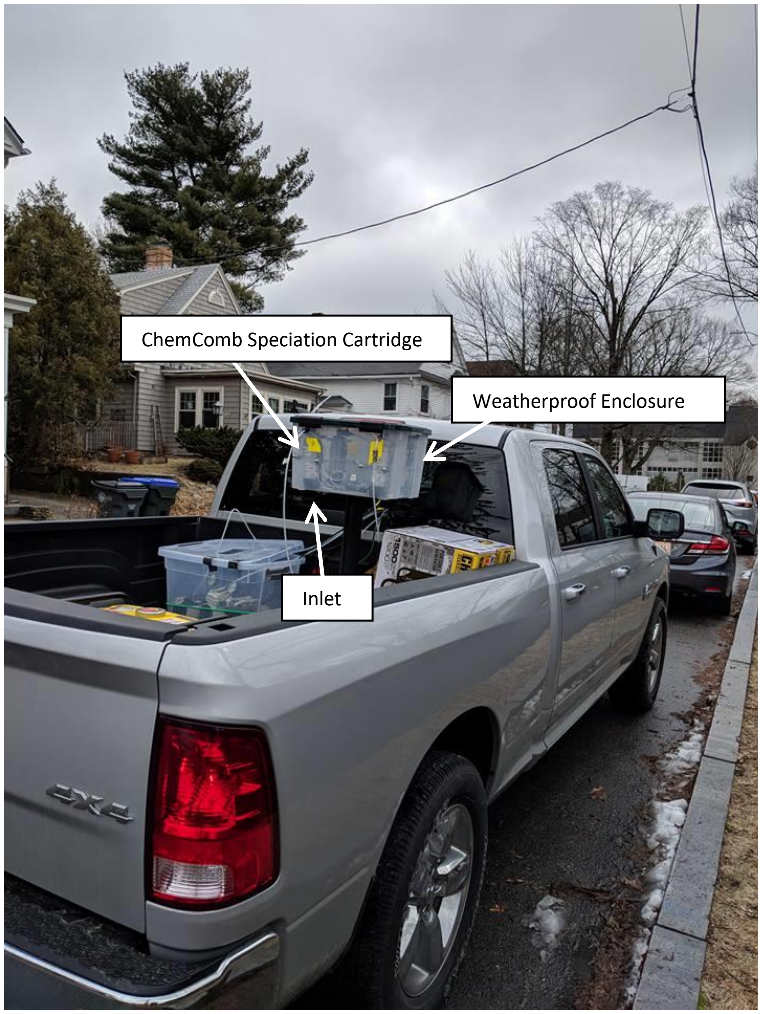

Figure S3: Image of the mobile lab for on-road collections of NH_3 from fresh vehicle traffic plumes in the northeastern US. The collection of NH_3 was conducted using a ChemComb Speciation Cartridge, which was held in a weatherproof enclosure, and the sampling inlet was directly exposed to ambient air to limit the inlet loss of NH_3.

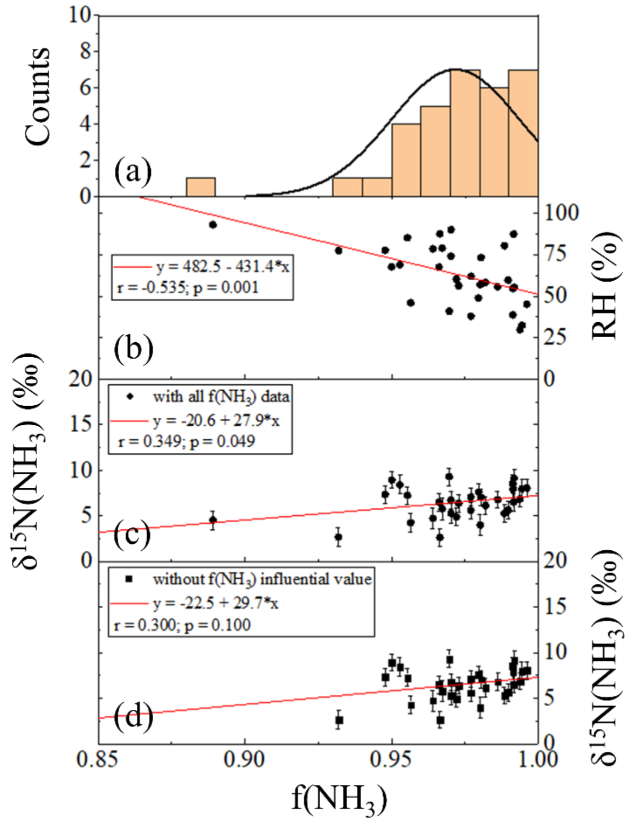

Figure S4: The summer near-highway monitoring (Providence, RI, US) $f(NH_3)$ data including (a) $f(NH_3)$ distribution and linear relations between (b) relative humidity (RH) and $f(NH_3)$, (c) $\delta^{15}N(NH_3)$ and $f(NH_3)$ (all data), and (d) $\delta^{15}N(NH_3)$ and $f(NH_3)$ (without influential $f(NH_3)$ value).

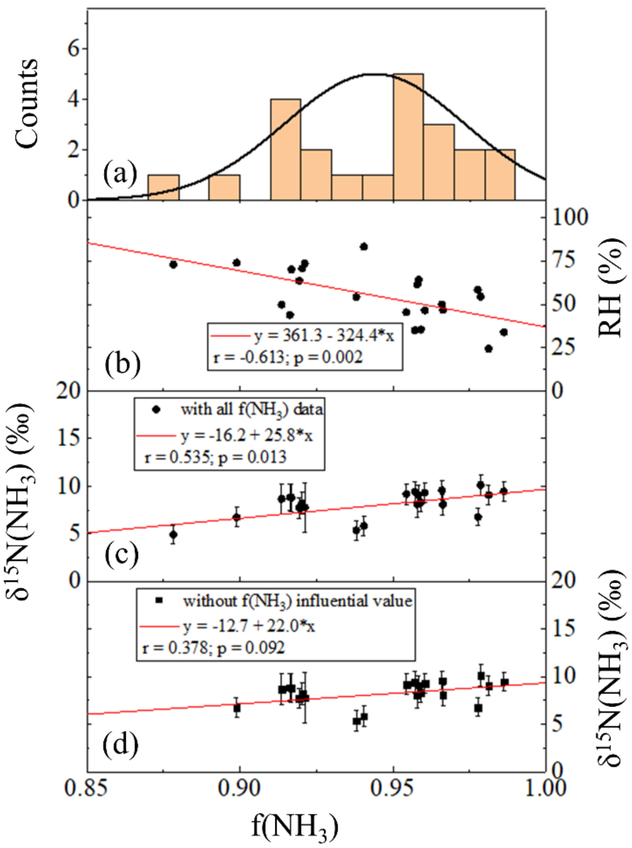

Figure S5: The winter near-highway monitoring (Providence, RI, US) $f(NH_3)$ data including (a) $f(NH_3)$ distribution and linear relations between (b) relative humidity (RH) and $f(NH_3)$, (c) $\delta^{15}N(NH_3)$ and $f(NH_3)$ (all data), and (d) $\delta^{15}N(NH_3)$ and $f(NH_3)$ (without influential $f(NH_3)$ value).

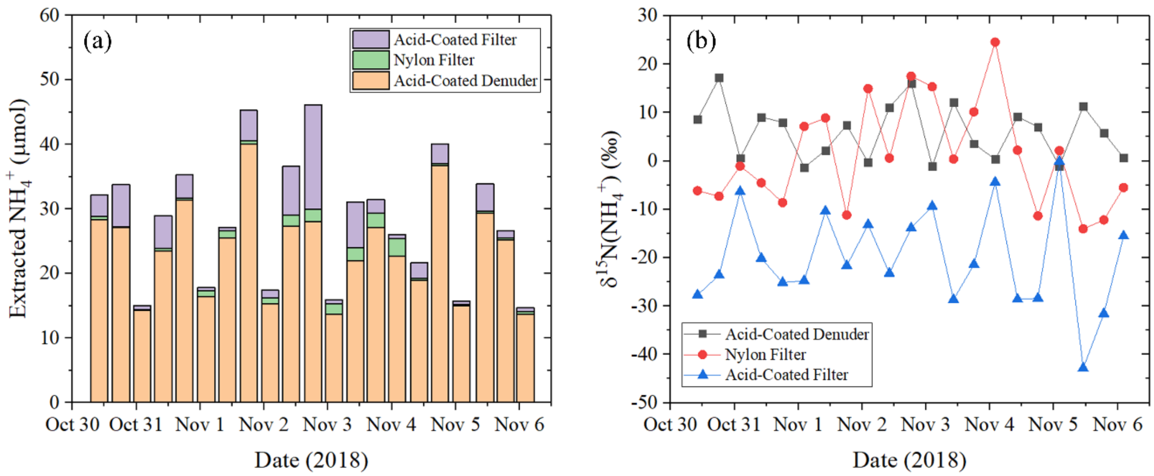

Figure S6: Comparison between the extracted NH_4^+ (a) and the measured $\delta^{15}N$ (b) of the different collection media, including the acid-coated denuder, nylon filter, and acid-coated filter for collection conducted in the Shenyang Tunnel.

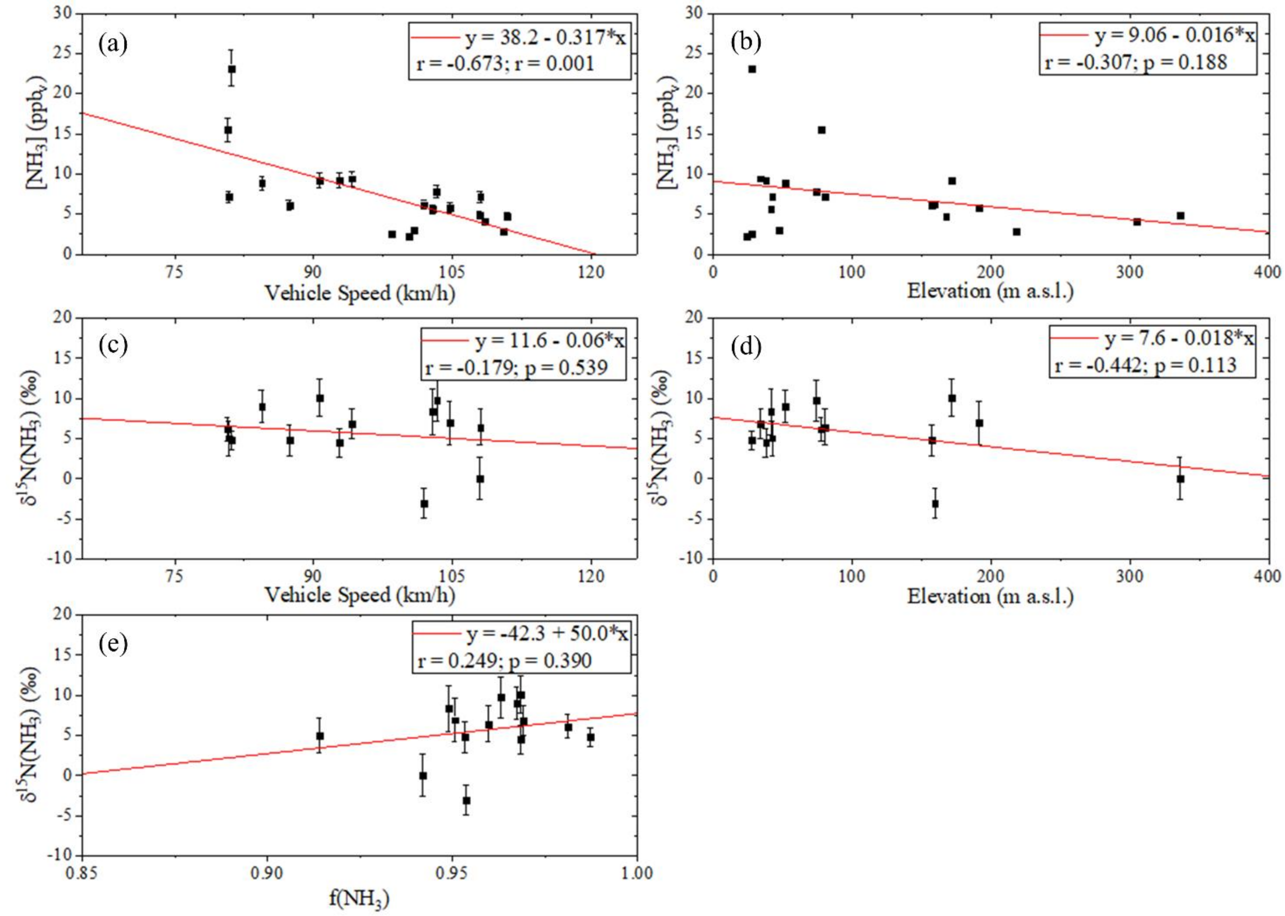

Figure S7: Linear relations from mobile on-road measurements in the northeastern US between (a) $[NH_3]$ and vehicle speed, (b) $[NH_3]$ and elevation, (c) $\delta^{15}N(NH_3)$ and vehicle speed, (d) $\delta^{15}N(NH_3)$ and elevation, and (e) $\delta^{15}N(NH_3)$ and $f(NH_3)$.

Table S1: Summary of ISORROPIA modeled of NH_x speciation conducted for samples collected in the tunnel from Shenyang, China. Model inputs included [NH_x], [NO_3^T], [SO_4^T], relative humidity (RH) and temperature (Temp) and model outputs included [NH_3] and [pNH_4^+] from which f(NH_3) was calculated. All concentrations are reported in units of μmol*m^{-3}.

	Model Input					Model Output		
Average Collection Date	[NH_x]	[NO_3^T]	[SO_4^T]	RH(%)	Temp (K)	[NH_3]	[pNH_4^+]	f(NH_3)
10/31/18 18:07	7.317	0.041	0.027	39.5	293.1	7.225	0.091	0.987
11/1/18 2:12	3.656	0.065	0.062	33.6	290.4	3.471	0.185	0.949
11/1/18 10:12	5.775	0.157	0.083	32.2	291.0	5.455	0.320	0.945
11/1/18 18:12	9.279	0.000	0.030	31.5	292.7	9.219	0.060	0.994
11/2/18 2:14	3.675	0.063	0.051	36.4	289.5	3.513	0.162	0.956
11/2/18 10:14	7.545	0.324	0.099	37.4	291.6	7.026	0.519	0.931
11/2/18 18:23	9.400	0.388	0.058	37.0	294.9	8.901	0.498	0.947
11/3/18 2:19	3.466	0.363	0.069	41.9	290.4	2.969	0.497	0.857
11/3/18 10:19	6.231	0.381	0.081	36.7	293.1	5.693	0.537	0.914
11/3/18 18:01	7.399	0.413	0.108	40.3	294.1	6.776	0.623	0.916
11/4/18 1:57	4.959	0.352	0.111	45.8	292.1	4.390	0.569	0.885
11/4/18 10:22	4.496	0.034	0.046	41.9	290.9	4.373	0.122	0.973
11/4/18 18:04	9.112	0.026	0.041	44.3	290.2	9.004	0.108	0.988
11/5/18 2:05	3.020	0.035	0.028	30.3	289.8	2.932	0.087	0.971
11/5/18 11:02	6.150	0.047	0.035	27.8	289.6	6.034	0.116	0.981
11/5/18 18:48	7.031	0.041	0.028	24.7	289.9	6.936	0.095	0.986
11/6/18 2:14	2.876	0.056	0.040	20.6	289.7	2.744	0.133	0.954