Supplementary Information for:

Wintertime Spatial Distribution of Ammonia and its Emission Sources in the Great Salt Lake Region

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Content:

Section S1 Study area

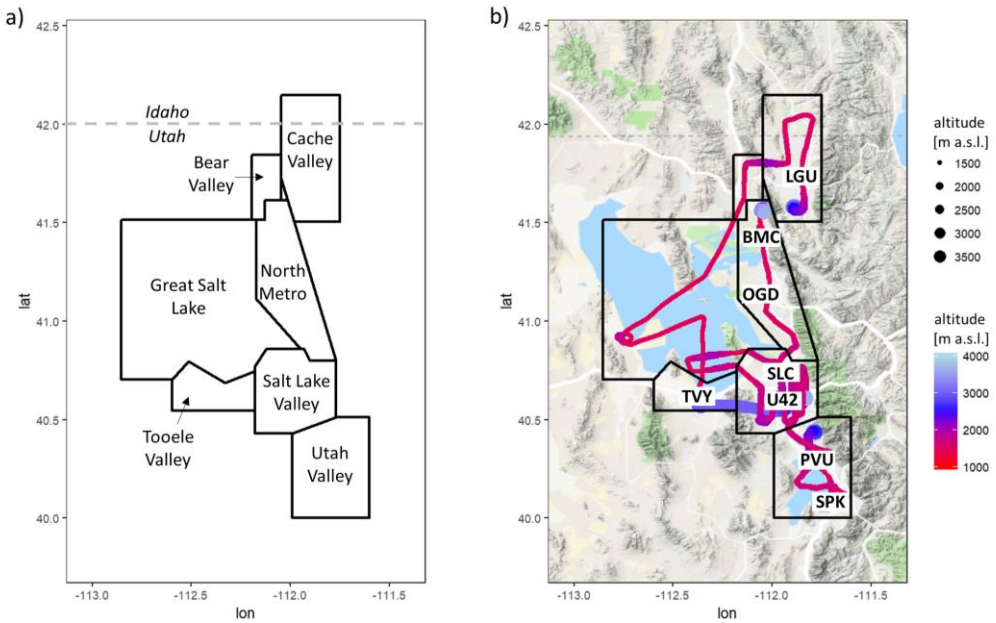

Figure S1: Overview of the study area: a) Division into sub-regions used for the analysis of Twin Otter measurements. b) Location of a typical flight path within the defined sub-regions. The flightpath is color-coded by the flight altitude, labels show the location of the Salt Lake City International Airport (SLC) and the airports for missed approaches (OGD = Ogden-Hinckley Airport, BMC = Brigham City Municipal Airport, LGU = Logan-Cache Airport, TVY = Tooele Valley Airport, U42 = South Valley Regional Airport, PVU = Provo Municipal Airport, SPK = Spanish Fork-Springville Airport).

Section S2 Observations of NH_3 from Twin Otter aircraft

Section S2.1 Surface level NH_3 from Twin Otter (missed approaches)

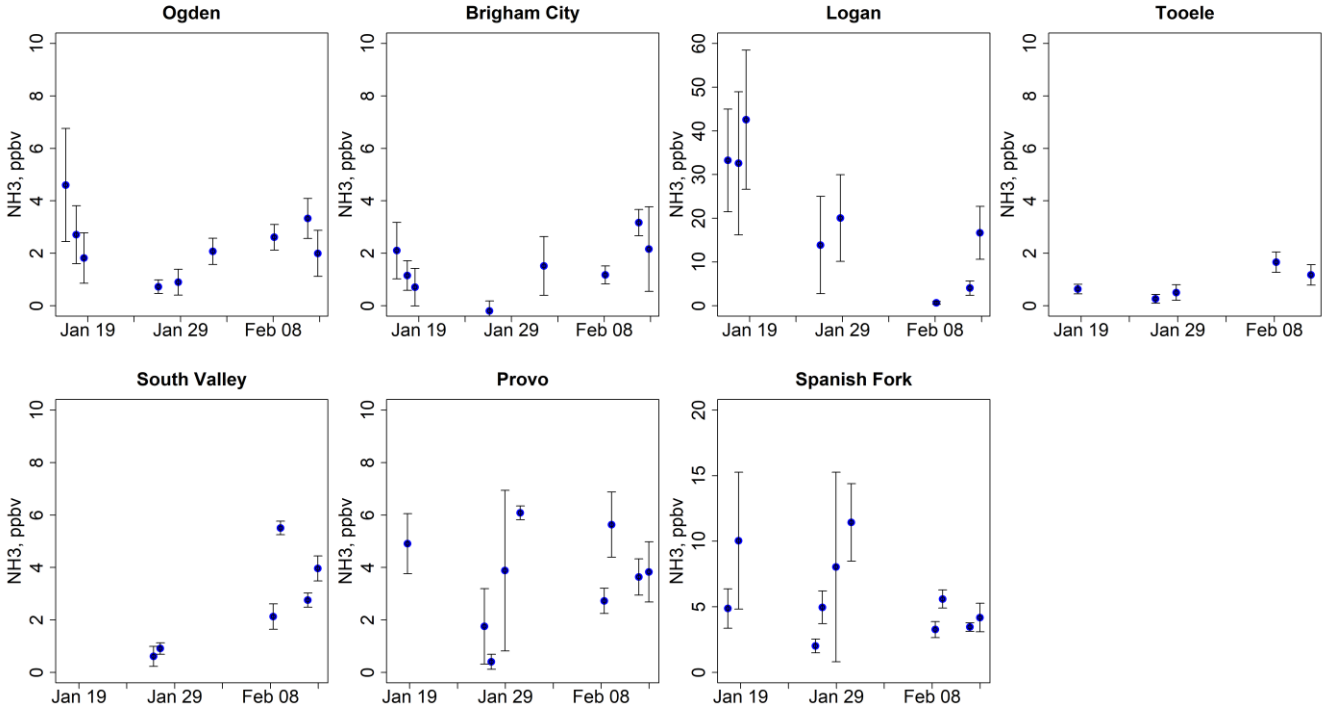

Figure S2: Mean NH_3 mixing ratios (+ standard deviation) during missed approaches. NH_3 mixing measured over the airport runway, extended by 0.01° latitude to North and South and by 0.01° longitude to West and East, were taken into account. Airports: OGD = Ogden-Hinckley Airport, BMC = Brigham City Municipal Airport, LGU = Logan-Cache Airport, TVY = Tooele Valley Airport, U42 = South Valley Regional Airport, PVU = Provo Municipal Airport, SPK = Spanish Fork-Springville Airport.

Section S2.2 Ground site and Twin Otter comparison

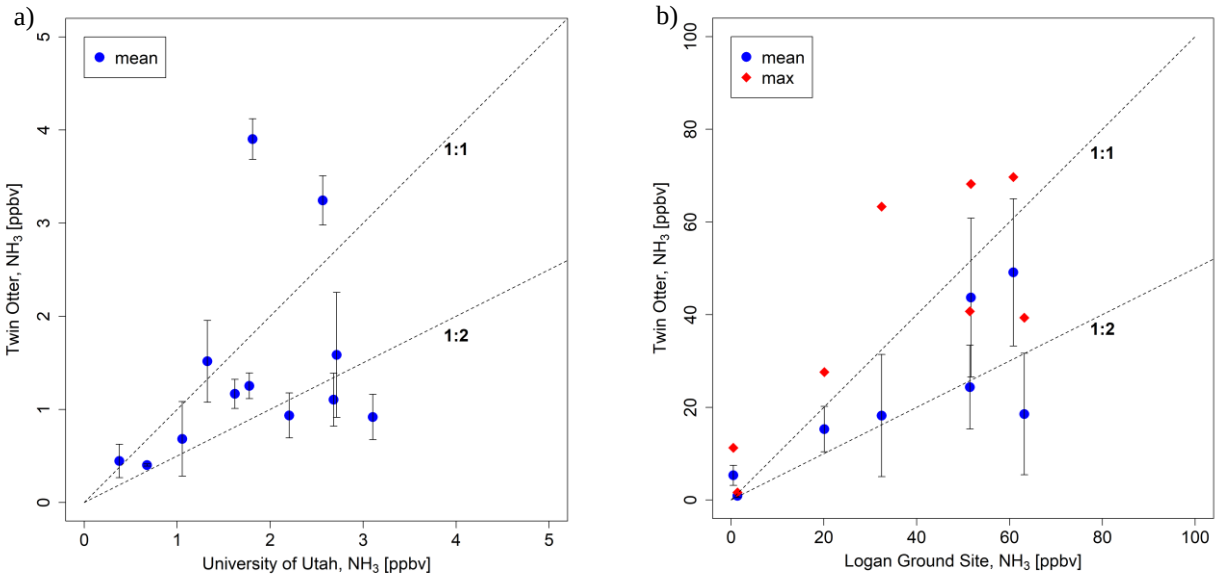

Figure S3: Comparison of NH_3 mixing ratios between Twin Otter and ground site observations. a) Comparison with the University of Utah ground site: NH_3 mixing ratios were averaged when the Twin Otter was overflying the ground site within a distance of 1 km before and after passing the ground site. b) Comparison with the Logan ground site: Twin Otter data shown represents the missed approaches at Logan airport, located approximately 3 km NW of the Logan ground site. Mean values (blue dots, with standard deviations) are the average NH_3 mixing ratios between 1 km before and 1 km after the start and end of the runway. Maximum values (red diamonds) were observed directly above the runway and are most representative for the NH_3 mixing ratios at ground level.

Section S3 UDAQ emission inventory

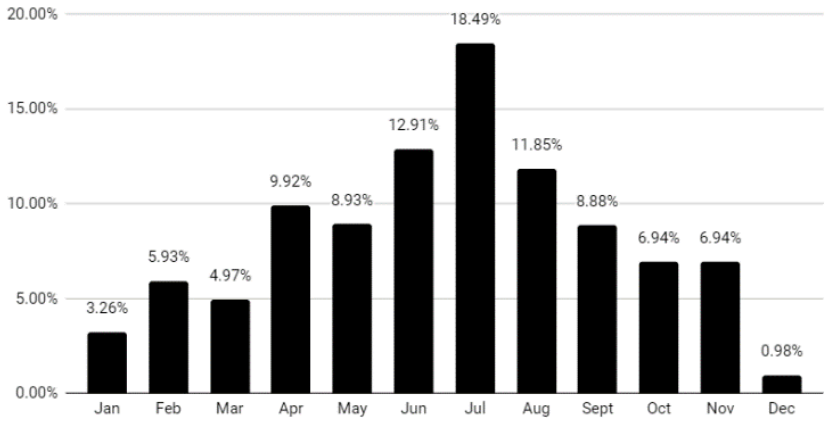

Figure S4: Monthly temporal profile of NH_3 livestock emissions used in the UDAQ emission inventory. The monthly profile redistributes the annual total NEI emissions over the year and is determined through inverse modelling as described in Gilliland et al. (2006).

Section S4 USU inventory

Table S1: Utah State University (USU) NH_3 emission inventory for Cache Valley and separated by county (Moore, 2007). Percentages may not sum to 100.0 due to rounding.

	CACHE COUNTY				FRANKLIN COUNTY				CACHE VALLEY (both counties)			
	Winter		Summer		Winter		Summer		Winter		Summer	
SOURCE	kg/d	%	kg/d	%	kg/d	%	kg/d	%	kg/d	%	kg/d	%
Dairy cattle	4,495.9	82.4	4,495.9	83.1	6,252.5	89.6	6,252.5	90	10,748.4	86.4	10,748.4	87
Beef cattle	143	2.6	98.2	1.8	82.9	1.2	49.7	0.7	225.9	1.8	147.9	1.2
Swine	54.7	1	54.7	1	13.8	0.2	13.8	0.2	68.5	0.6	68.5	0.6
Poultry	546.7	10	546.7	10.1	609	8.7	609	8.8	1,155.7	9.3	1,155.7	9.4
Subtotal	*5,240.3*	*96*	*5,195.5*	*96*	*6,958.2*	*99.7*	*6,925.0*	*99.7*	*12,198.5*	*98.1*	*12,120.5*	*98.2*
Automobiles	196.7	3.6	196.7	3.6	21.6	0.3	21.6	0.3	218.3	1.8	218.3	1.8
Industry/WWTP	18.1	0.3	18.1	0.3	0.05	<0.1	0.05	< 0.1	18.2	0.2	18.2	0.2
Total	*5,455.0*	*99.9*	*5,410.2*	*99.9*	*6,979.9*	*100*	*6,946.7*	*100*	*12,434.9*	*100.1*	*12,357.0*	*100.2*

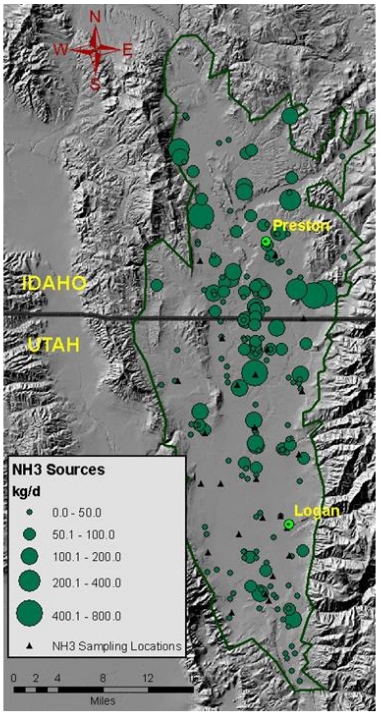

Figure S5: Map overlay of NH_3 sources in Cache Valley used in the Utah State University (USU) NH_3 emission inventory (Moore, 2007).

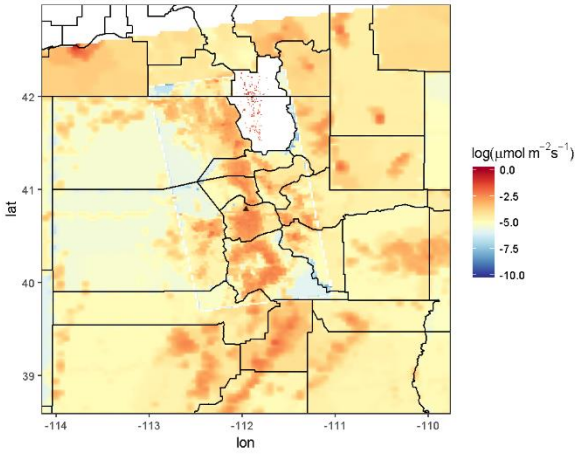

Figure S6: Emission inventory map used in the footprint model approach, where emissions in Cache Valley are substituted by the USU NH_3 emissions (white are with point sources).

Section S5 Footprint model

Section S5.1 Modelled and measured NH_x enhancements

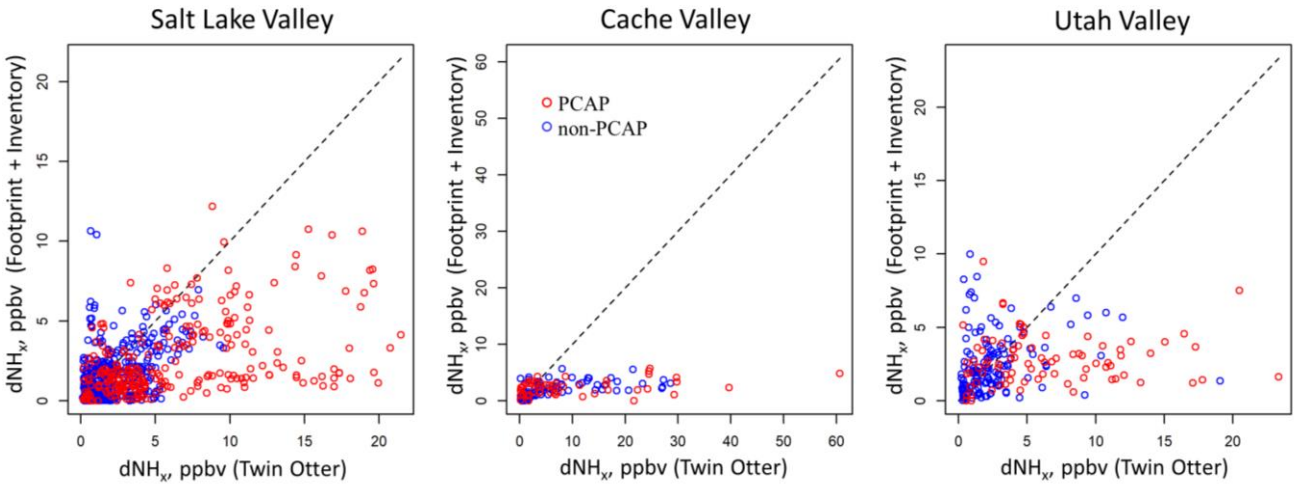

Figure S7. Modelled (Footprint + Inventory) vs. measured (Twin Otter) NH_x enhancements (dNH_x) for Salt Lake Valley, Cache Valley and Utah Valley during PCAP (red) and non-PCAP period (blue). NH_3 emission from gas furnace combustion and human perspiration were removed from the area sources sector of the original UDAQ NH_3 emission inventory.

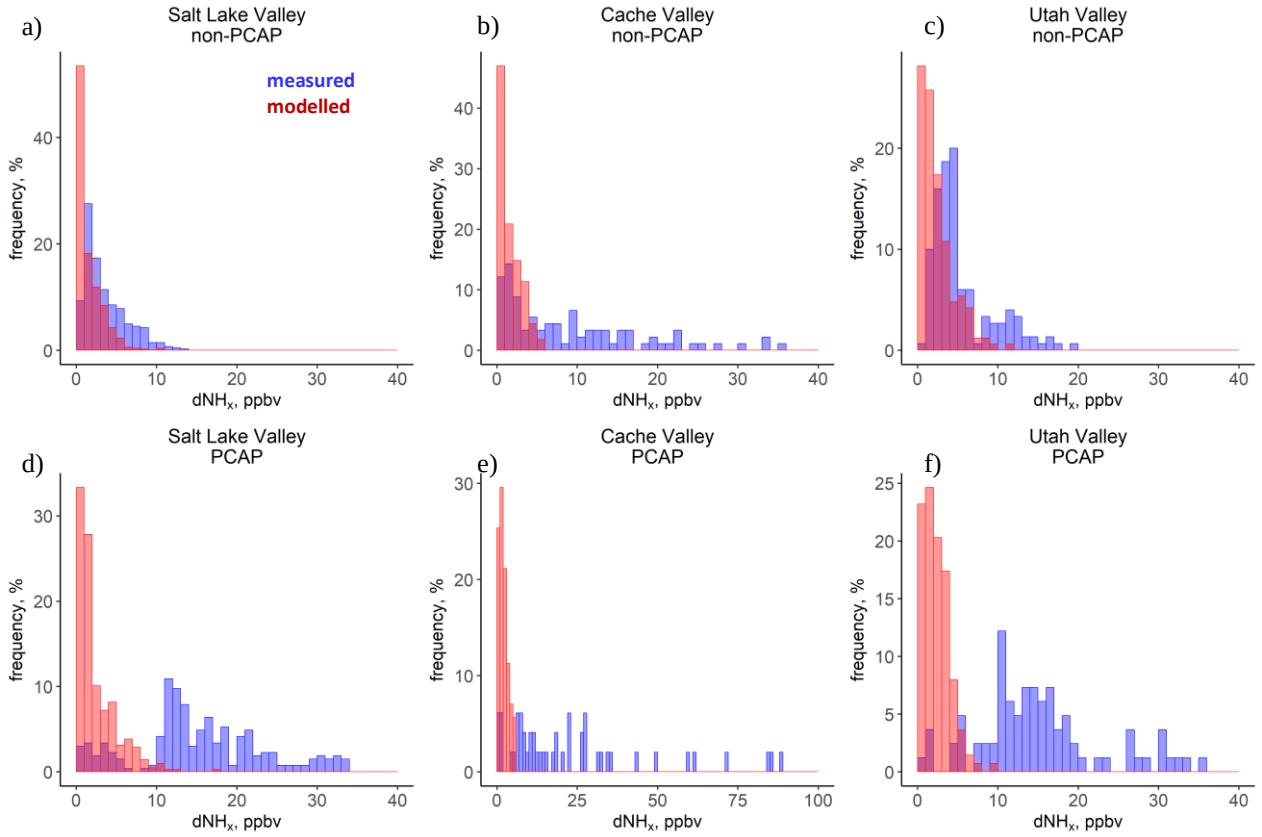

Figure S8: Frequency distribution of measured (blue) and modelled (red) NH_x enhancements (dNH_x) for Salt Lake Valley, Cache Valley and Utah Valley (x-axis is linear scale) for (a-c) non-PCAP and (d-f) PCAP conditions.

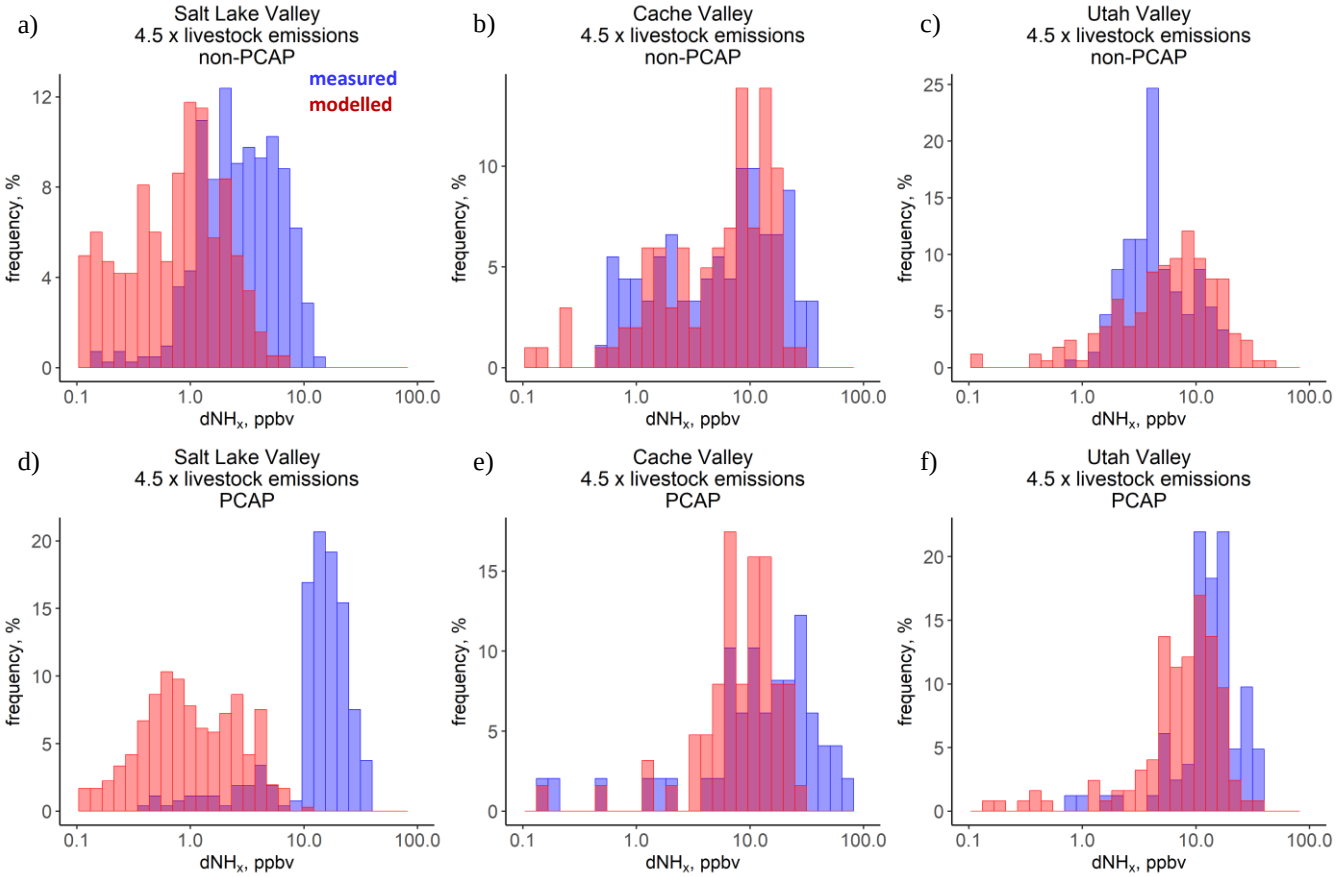

Figure S9: Frequency distribution of measured (blue) and modelled (red) NH_x enhancements (dNH_x) for Salt Lake Valley, Cache Valley and Utah Valley using enhanced livestock emissions by a factor of 4.5 for (a-c) non-PCAP and (d-f) PCAP conditions.

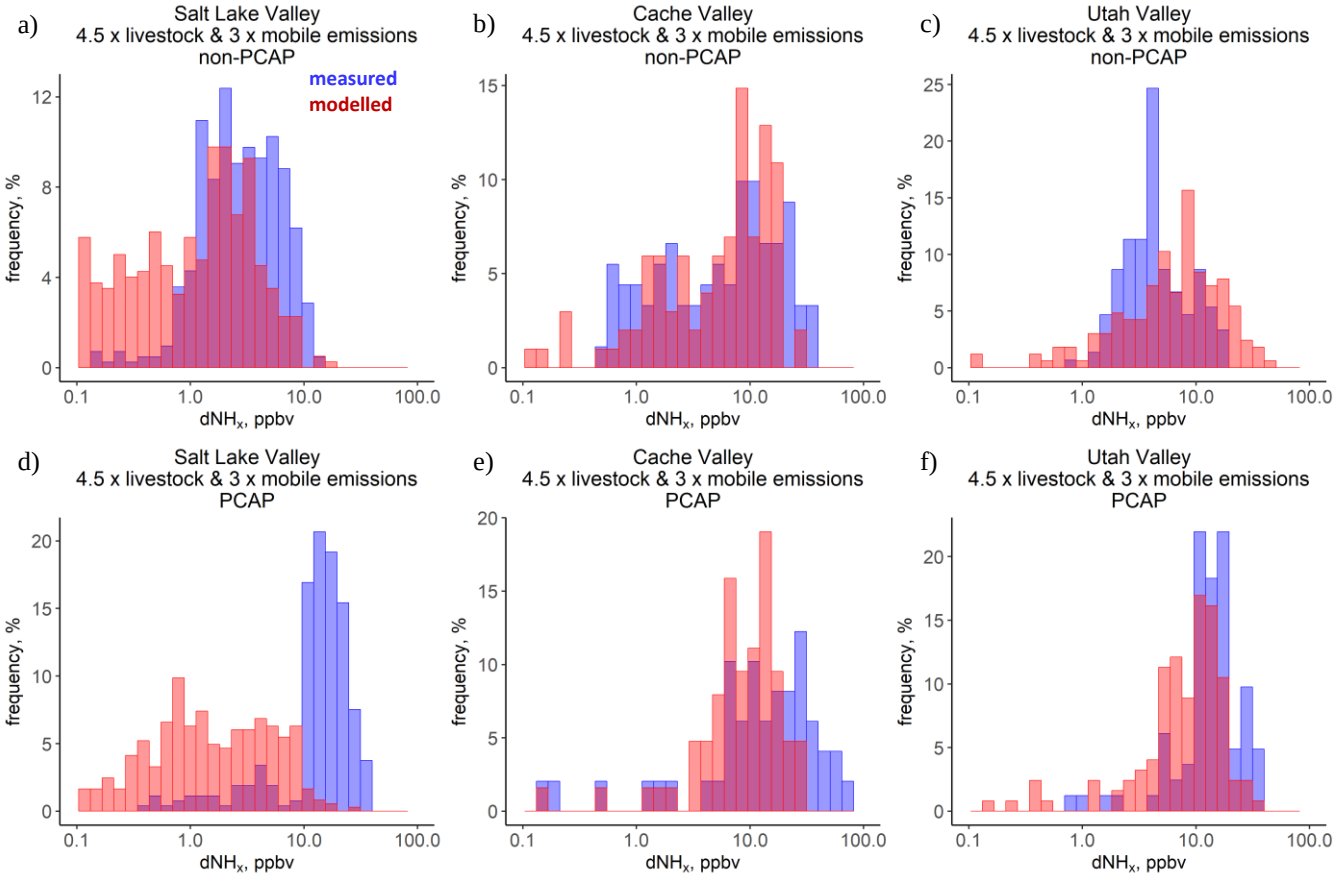

Figure S10: Frequency distribution of measured (blue) and modelled (red) NH_x enhancements (dNH_x) for Salt Lake Valley, Cache Valley and Utah Valley using enhanced livestock (factor of 4.5) and enhanced mobile (factor of 3) emissions for (a-c) non-PCAP and (d-f) PCAP conditions.

	All			non-PCAP			PCAP		
	original	f_{ls}	$f_{ls} + f_{mob}$	original	f_{ls}	$f_{ls} + f_{mob}$	original	f_{ls}	$f_{ls} + f_{mob}$
BV	4.2E-12	-	-	1.5E-09	-	-	4.5E-07	-	-
CV	9.7E-11	**1.3E-01**	**1.1E-01**	3.2E-07	**2.3E-01**	**2.1E-01**	3.7E-06	**4.4E-01**	**4.0E-01**
GSL	2.2E-57	-	-	6.5E-40	-	-	1.6E-27	-	-
NM	1.4E-22	-	-	7.2E-08	-	-	1.0E-17	-	-
SLV	4.8E-25	1.0E-75	1.5E-33	2.9E-10	1.9E-37	3.8E-15	2.4E-29	3.4E-54	2.6E-30
TC	4.3E-11	-	-	5.8E-08	-	-	1.2E-04	-	-
UV	3.4E-03	8.9E-20	8.9E-20	**4.9E-01**	9.5E-23	4.2E-24	3.3E-11	1.4E-02	3.6E-03

Table S2: p-values obtained from the Mann-Whitney-Wilcoxon test comparing modelled and measured dNH_x values Shown are the results for the regions in the study area using all data and only data during non-PCAP and PCAP conditions. In addition, the effect of increasing livestock emissions by a constant factor (f_{ls} = 4.5) and further increase of mobile emissions (f_{mob} = 3) is illustrated for Cache Valley, Salt Lake Valley and Utah Valley. If the p-value is <0.05, the null hypothesis, that both modelled and measured data are drawn from same distribution, is rejected. Values in bold indicate cases where the null hypothesis is not rejected and therefore a similarity between modelled and measured distributions is likely.

Section S5.2 Comparison with modelled and measured NO_y enhancements

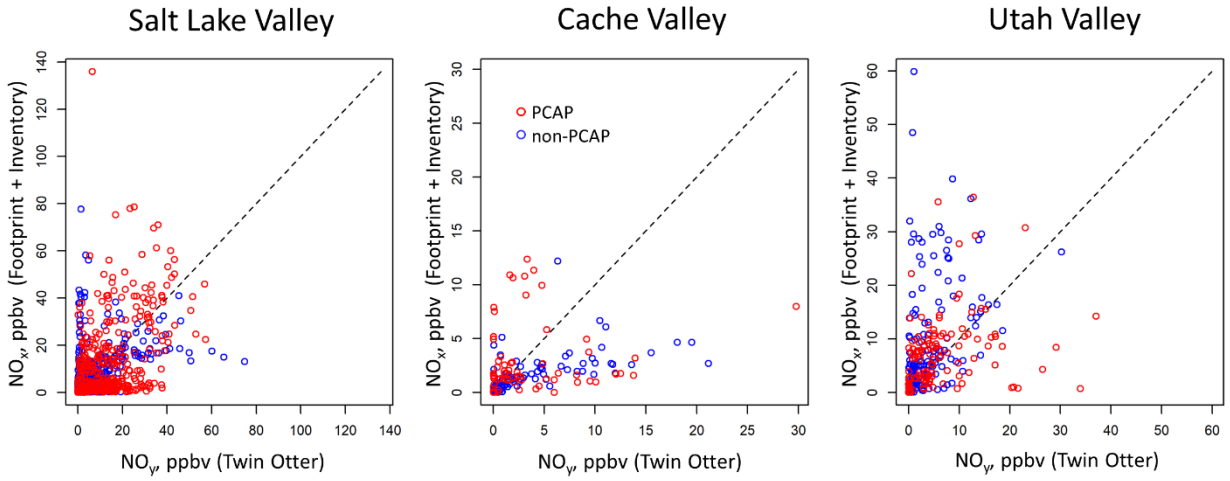

Figure S11. Modelled (Footprint + Inventory) vs. measured (Twin Otter) NO_y enhancements (dNO_y) for Salt Lake Valley, Cache Valley and Utah Valley during PCAP (red) and non-PCAP period (blue).

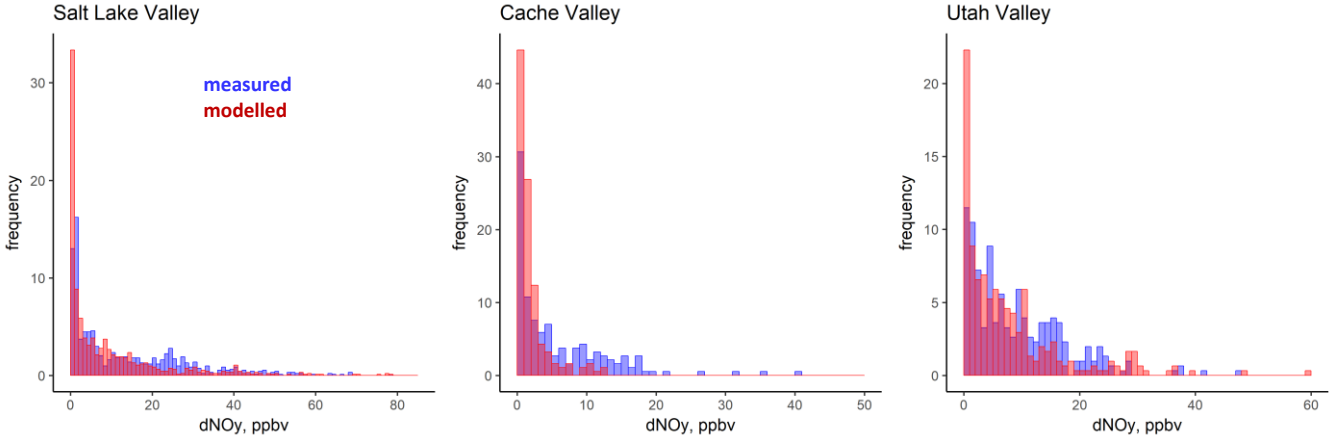

Figure S12: Frequency distribution of measured (blue) and modelled (red) NO_y enhancements (dNO_y) for Salt Lake Valley, Cache Valley and Utah Valley (x-axis is linear scale).

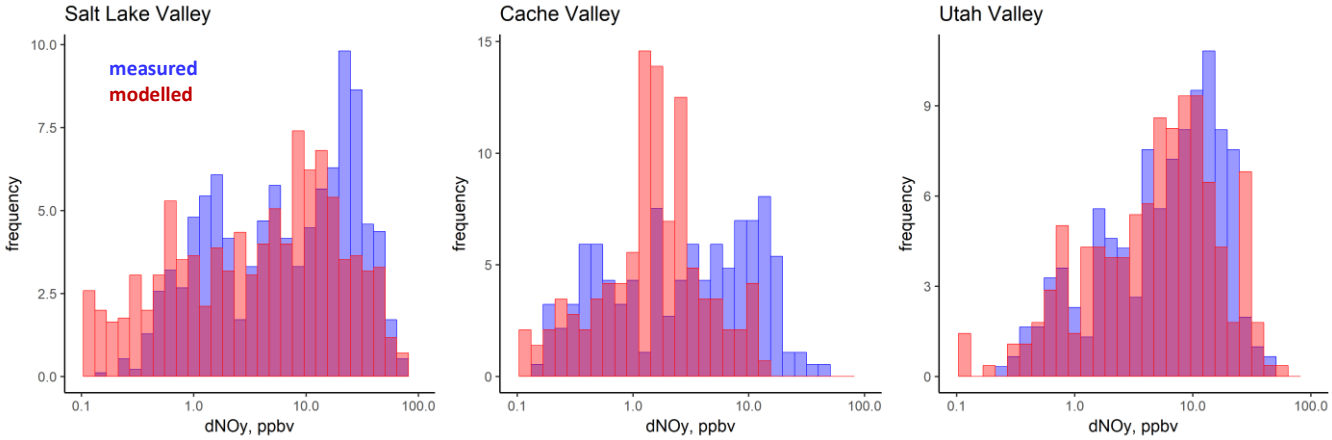

Figure S13: Frequency distribution of measured (blue) and modelled (red) NO_y enhancements (dNO_y) for Salt Lake Valley, Cache Valley and Utah Valley (x-axis is logarithmic scale).

Section S5.3 Background mixing ratio determination

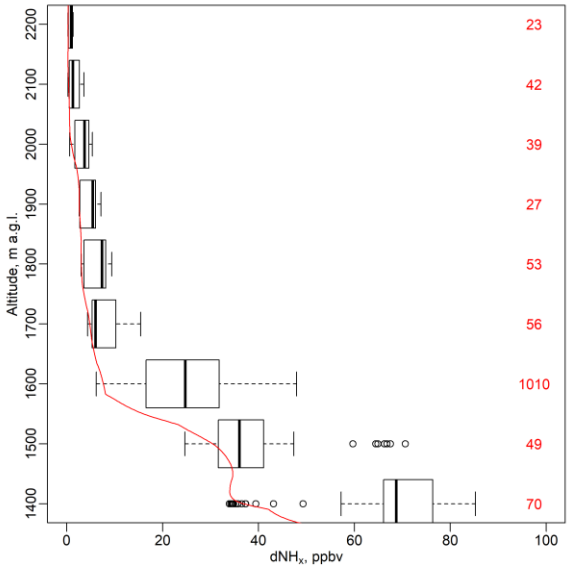

Figure S14: Example altitude profile for a research flight in Cache Valley used for the calculation of background mixing ratios. The red background profile was derived by calculating the moving 1st percentile (in 1-m altitude increments) with a layer depth of 50 m. In a second step, the 1st percentile altitude profile was further smoothed by a moving average (again in 1-m altitude increments and with a layer depth of 50 m). The shown boxplots visualize the variation of observed NH_x with altitude. Since they are defined for layers of 100 m depth, they do not necessarily match with the calculated background profile. Red numbers are the number of measurement points available in each boxplot.

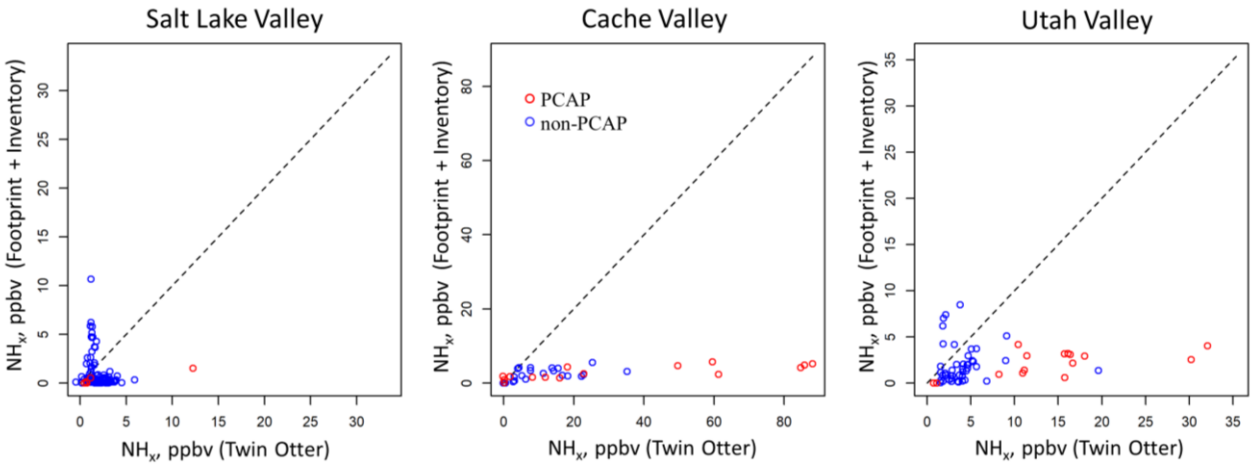

Figure S15 Modelled (Footprint + Inventory) vs. measured (Twin Otter) NH_x mixing ratios for Salt Lake Valley, Cache Valley and Utah Valley during PCAP (red) and non-PCAP period (blue). NH_3 emission from gas furnace combustion and human perspiration were removed from the area sources sector of the original UDAQ NH_3 emission inventory. In contrast to Figure S7, only those data point are shown where the back trajectories at the exit of the inventory domain met the following conditions: at least 180 out of the 200 trajectories originate in greater than 1000 m a.g.l. and/or UDAQ NH_3 emission are below 10^{-4} µmol m^{-2} s^{-1}. As a result it is assumed that the NH_3 mixing ratio at the domain entry is zero ppb and the modelled NH_3 values can be regarded as absolute mixing ratios instead of enhancements. For that reason, the background was mixing ratios was not subtracted from the measured NH_x values, representing absolute NH_3 mixing ratios as well.

Section S6 Inter-valley exchange

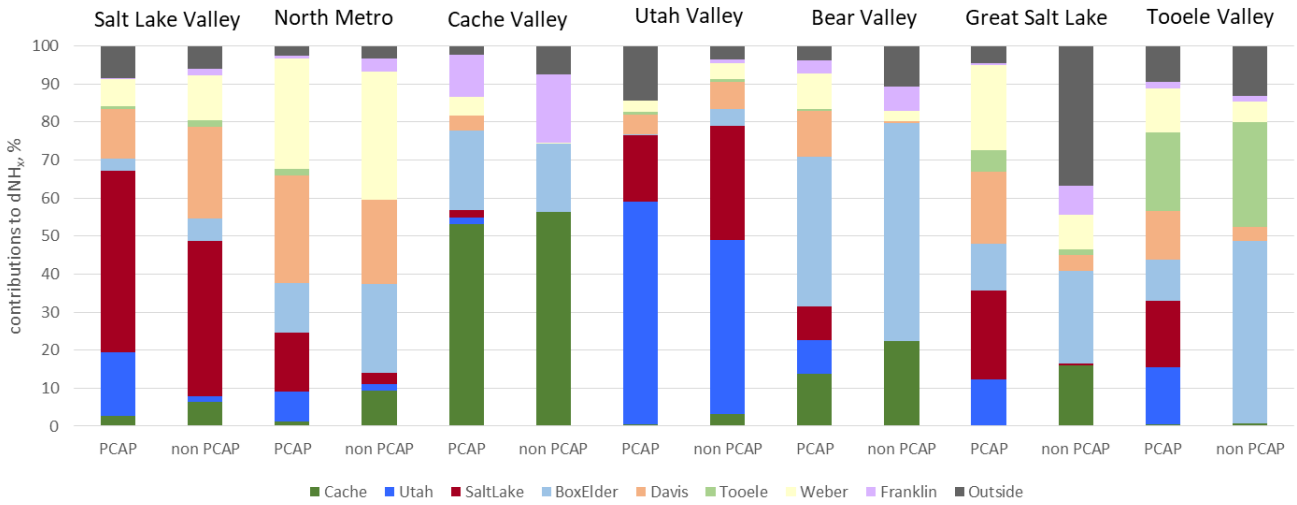

Figure S16: Inter-valley exchange of NH_x: Contributions from different counties to dNH_x at the Twin Otter locations in all sub-regions of the study period. The inter-valley exchange was evaluated by segregating contributions from the footprint model (see contributions map in Figure 5 in main text) into counties of origin for each run of the footprint model (i.e. every 2 min of Twin Otter flight path).

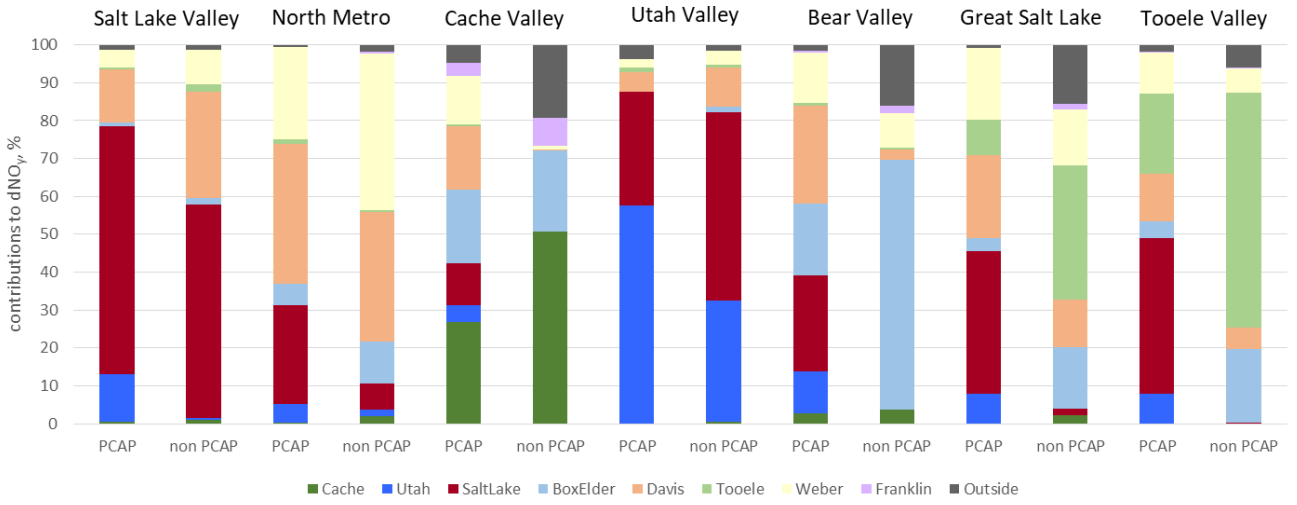

Figure S17: Inter-valley exchange of NO_y: Contributions from different counties to dNO_y at the Twin Otter locations in all sub-regions of the study period. The inter-valley exchange was evaluated by segregating contributions from the footprint model (see contributions map in Figure 5 in main text) into counties of origin for each run of the footprint model (i.e. every 2 min of Twin Otter flight path).

Section S7 Source apportionment

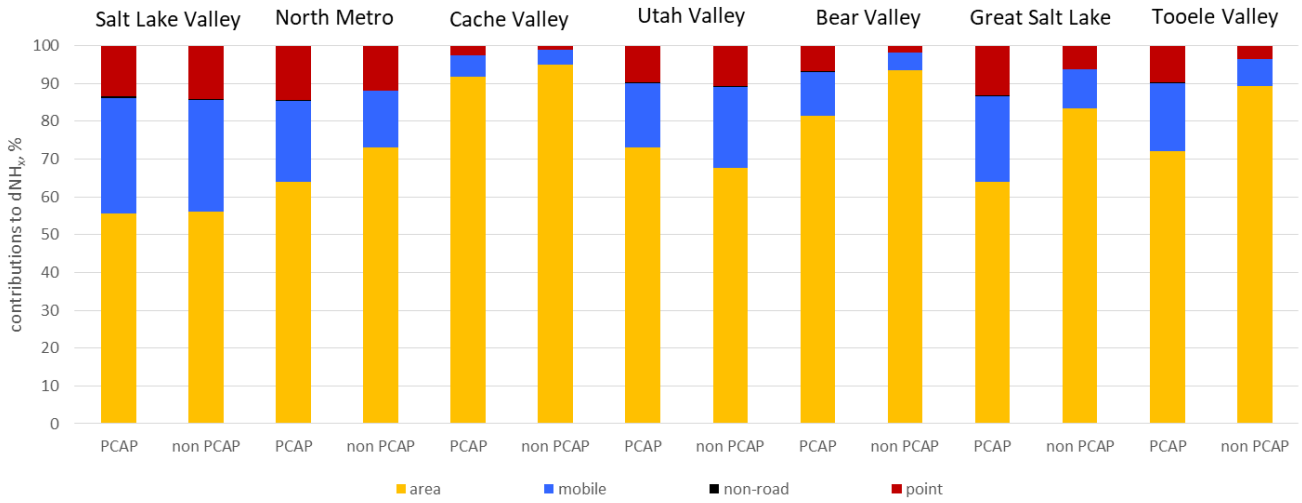

Figure S18: Sector contribution to dNH_x: Contributions from different emission sectors to dNH_x at the Twin Otter locations in all sub-regions of the study period. For Salt Lake Valley, dNH_z contributions from the area sector were by 10 % lower during PCAP and by 15 % lower during non-PCAP periods, when using the STIL footprints from the UU ground site. While point source contributions were approximately the same, contributions from area sources were higher by that value.

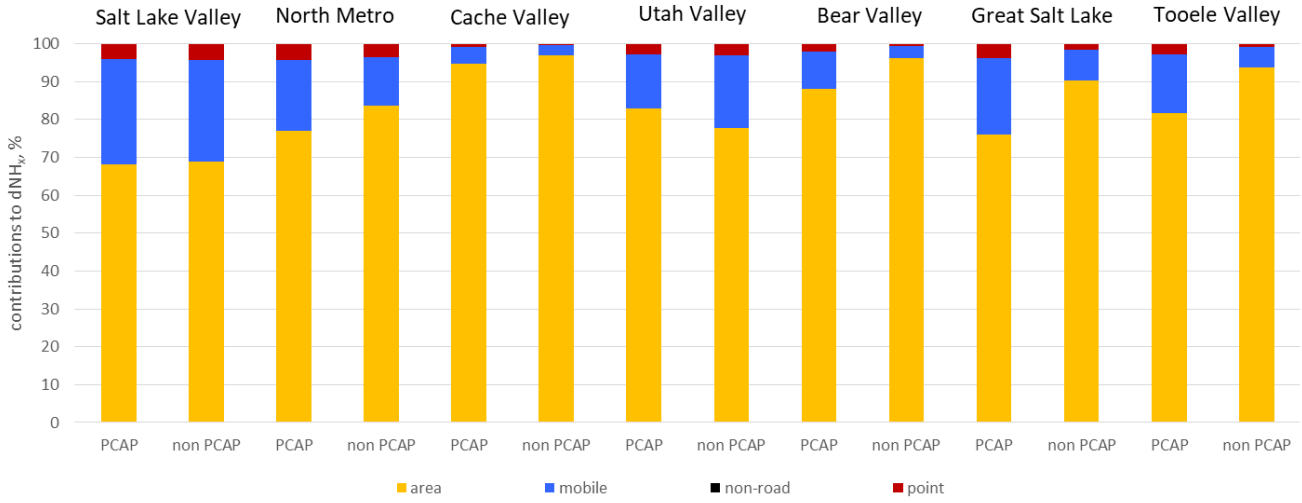

Figure S19: Sector contribution to dNH_x (scaled emissions): Contributions from different emission sectors to dNH_x at the Twin Otter locations in all sub-regions of the study period. To account for the observed underestimation of emission sources in the UDAQ inventory, area source and mobile emissions were increased by a factor 4.5 and 3, respectively.

References

Gilliland, A. B., Wyat Appel, K., Pinder, R. W., & Dennis, R. L. (2006). Seasonal NH3emissions for the continental united states: Inverse model estimation and evaluation. *Atmospheric Environment*, *40*(26), 4986–4998. https://doi.org/10.1016/j.atmosenv.2005.12.066

Moore, K. D. (2007). *Derivation of agricultural gas-phase ammonia emissions and application to the Cache Valley*. Utah State University, Logan, UT.