Validation of IASI NH_3 columns at the single-pixel scale from airborne- and ground-based measurements

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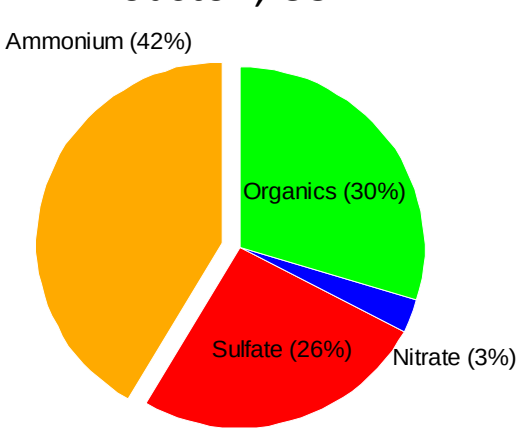
Field data:
Armin Wisthaler (P3-B NH_3, Innsbruck)
John Nowak (C-130 NH_3, Aerodyne)
Da Pan, Kang Sun, Lei Tao, Levi Golston, Lars Wendt (mobile NH_3, Princeton)
Cody Fleishcer (mobile NH_3, Aerodyne)
Rainer Volkamer (mobile FTIR NH_3, Univ. Colorado)
Anne Thompson, Bill Brune (sondes, Penn State)
Steve Brown (BAO tall tower, NOAA CSD)
Jennifer Murphy (NH_3 BAO tower, Univ. Toronto)
Erik Olson (sondes, Univ. Wisc.)
NASA DISCOVER-AQ, NSF FRAPPE science teams

Funding
NASA NNX-NNX14AT36G
NSF EEC-0540832
O3MSAF Visiting Scientist

Why NH_3?

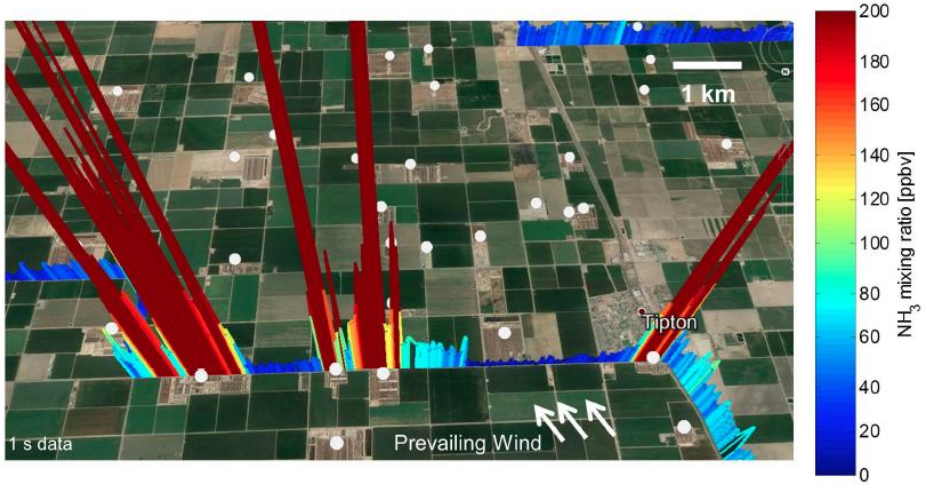
- human health: key precursor for PM2.5
- ecological: nitrogen deposition, eutrophication
- chemistry: aerosol nucleation, growth, composition
- climate: ammoniated aerosols cool locally



Molecular composition
(assuming M_{OM} = 100 g/mol)

Challenges

- NH_3 incredibly difficult to measure
  - low mole fractions (~ pptv-ppbv)
  - large dynamic range
  - adsorption to inlets/instruments
  - gas/aerosol phase partitioning



Limited datasets in which to validate recent advances in satellite NH_3

satellite NH_3 validation advances

IASI:

in-situ: m=0.18, r=0.82, ±1 hour in [NH_3] *(Van Damme et al., AMT, 2015)*

FTIR NDAAC: m=0.74, r=0.7 at 90 min., 25 km *(Dammers et al., ACPD, 2016)*

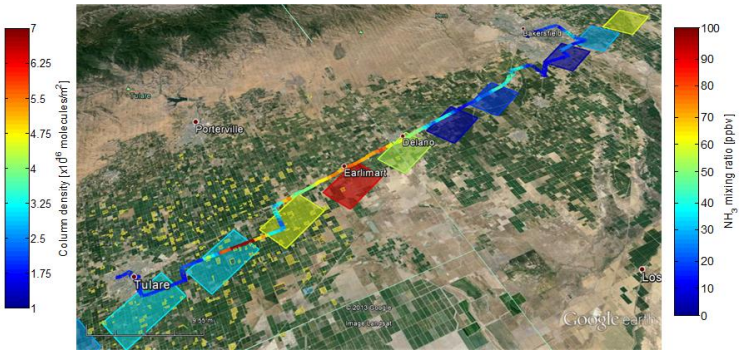
NASA TES:

15% with in-situ @ select pixels but at high NH_3 (mid 10^{16} molec NH_3 cm^{-2})

(Sun et al., JGR-A, 2015)

NOAA CrIS:

simulated profiles, qualitative agreement at pixel *(Shephard & Cady-Pereira, AMT, 2015)*



NASA AIRS:

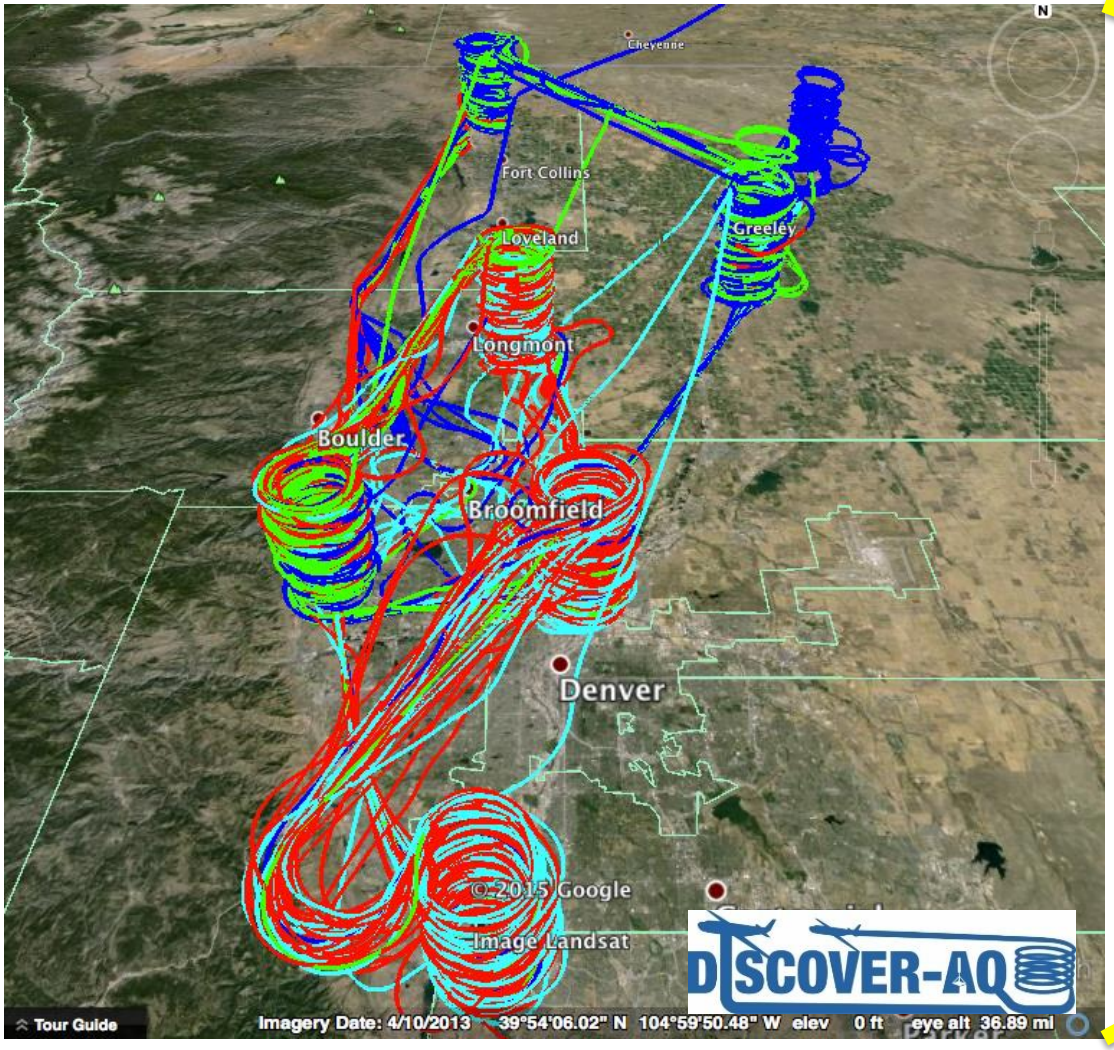
qualitative comparisons with aircraft *(Warner, ACPD, 2015)*

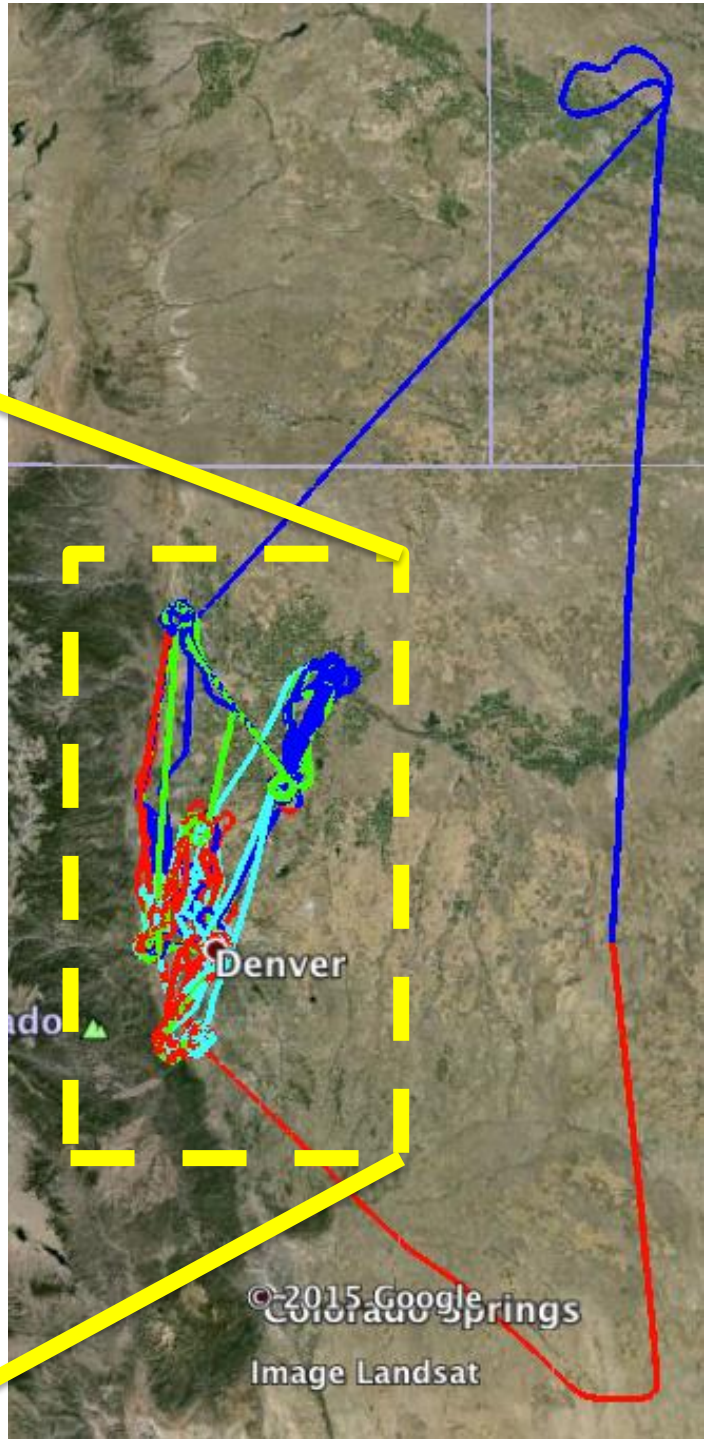
Validation limited to spatial/monthly averaging, chemical transport model simulations, or in regions of very high-concentrations

To what extent do NH_3 columns intercompare with in-situ at pixel scales? … at moderate mole fractions?

NASA DISCOVER-AQ (DAQ)

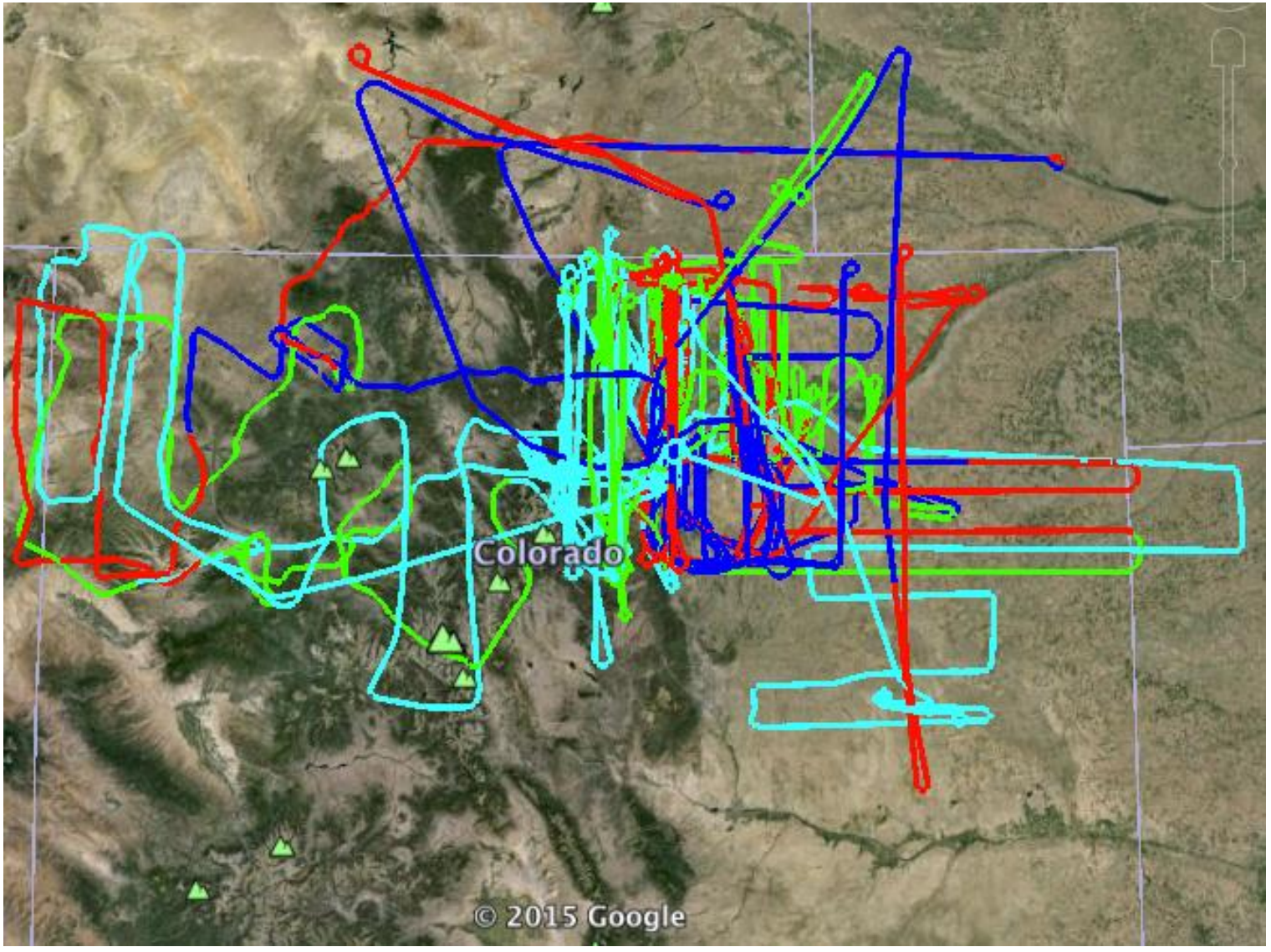
- repeated spiral profiles along same circuit
- take off at ~ 0830 local (July-Aug 2014)
- 2-3 circuits per day
- excellent flight pattern for IASI validation



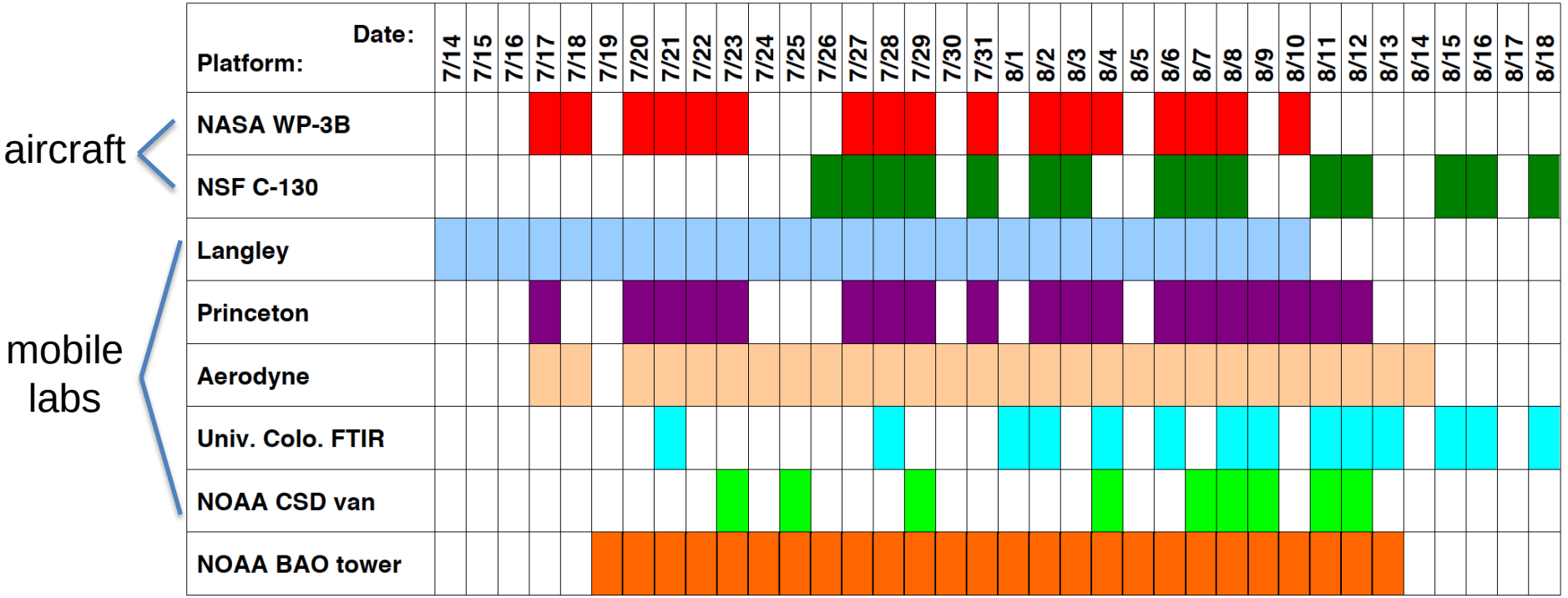


NSF FRAPPE

- focus on (largely) horizontal gradients and temporal evolution of the gradients
- takeoff ~ 0900 local
- constant altitude legs (some vertical profiles)



Additional in-situ NH_3 measurements



NH_3 measurements during DISCOVER-AQ/FRAPPE in Colorado:

platform	avg. uncertainty	method	PI
P3-B	30%	PTRMS	A. Wisthaler (Innsbruck)
C-130	34% + 0.39 ppbv	TDL	J. Nowak (Aerodyne)
Aerodyne mob.	34% + 0.39 ppbv	TDL	C. Floerchinger (Aerodyne)
Princeton mob.	20% + 0.4 ppbv	Open-path	M. Zondlo (Princeton)
CU column	10%	FTIR	R. Volkamer (CU)
NOAA/CSD	20% + 0.3 ppbv	Picarro	A. Neuman (NOAA CSD)
NOAA/BAO	20% + 0.3 ppbv	Picarro	J. Murphy (Univ. Toronto) / S. Brown (NOAA CSD)

IASI MetOp/A NH_3 during DAQ/FRAPPE

Time: 7/14/14-8/18/14

Spatial: 38-42ºN to 101-109.5ºW

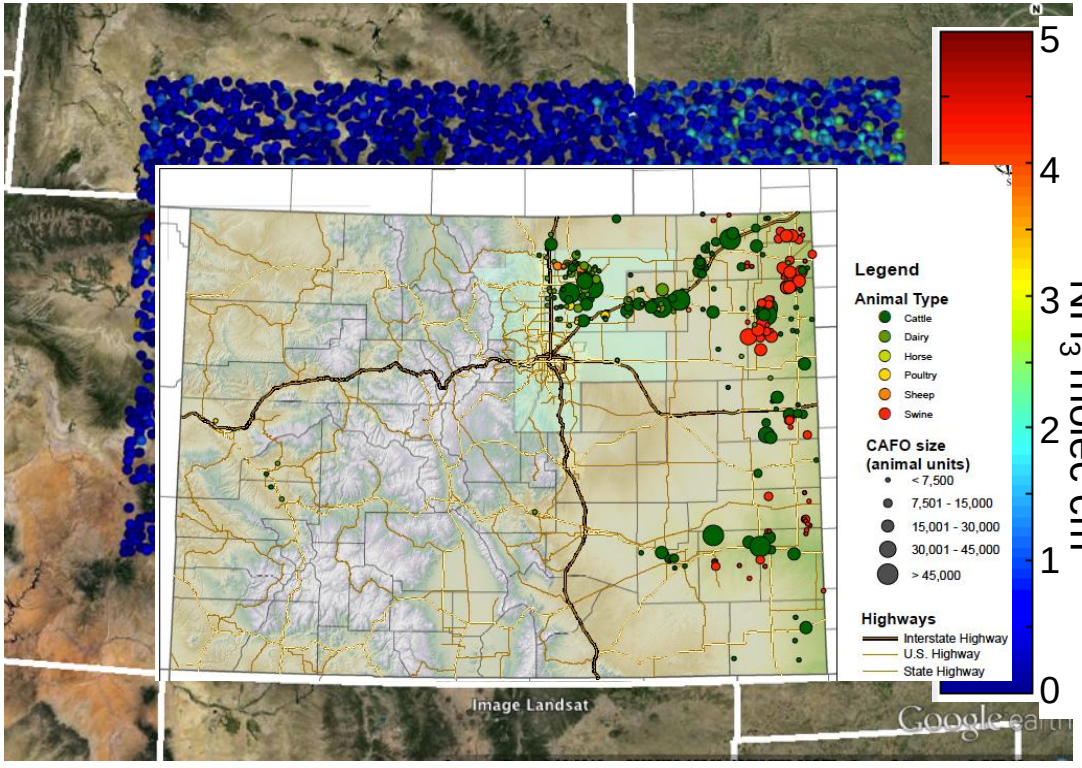
IASI Neural Network (*Whitburn et al., JGR-A, 2016*)

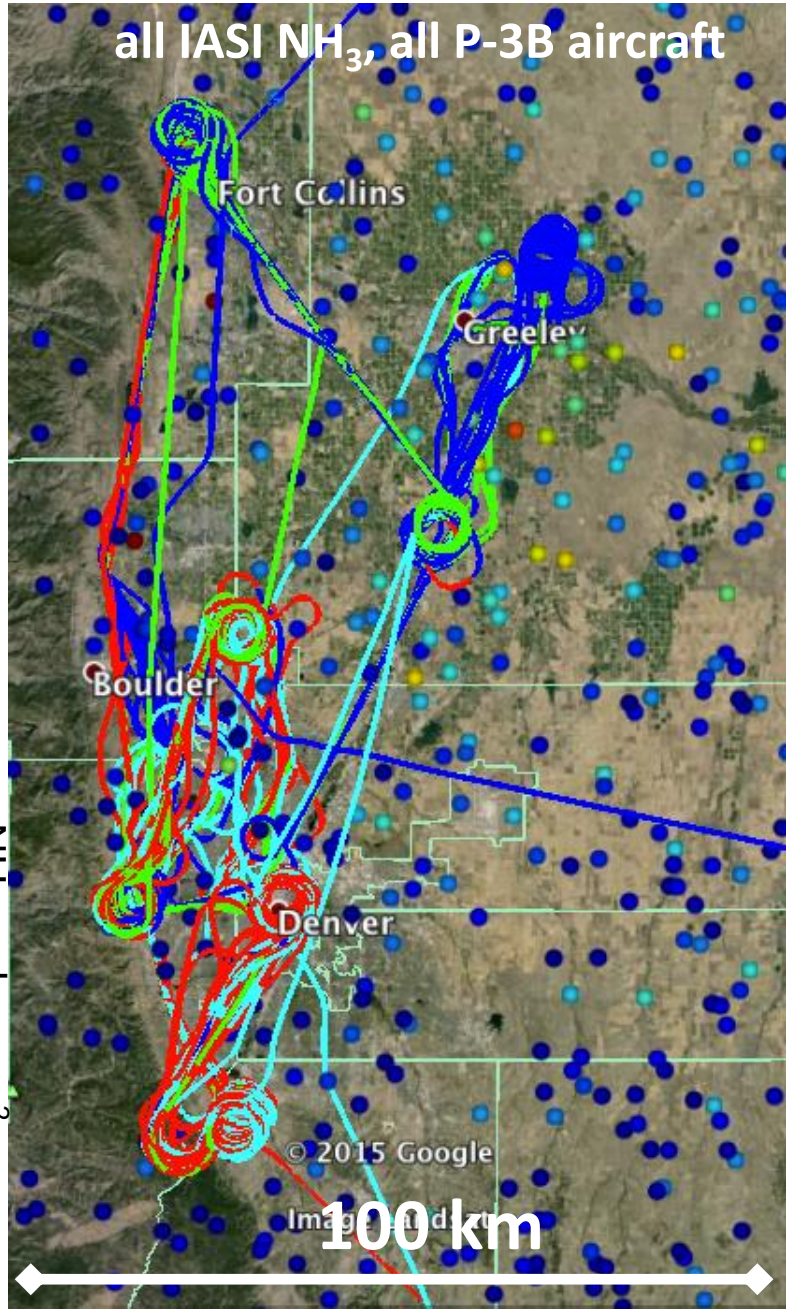
N=11,760 IASI NH_3 measurements (AM only)

N=6,371 filtered IASI NH_3

(cloud < 25%, sand < 50%, T_{skin} > 265.15 K; error<200%)

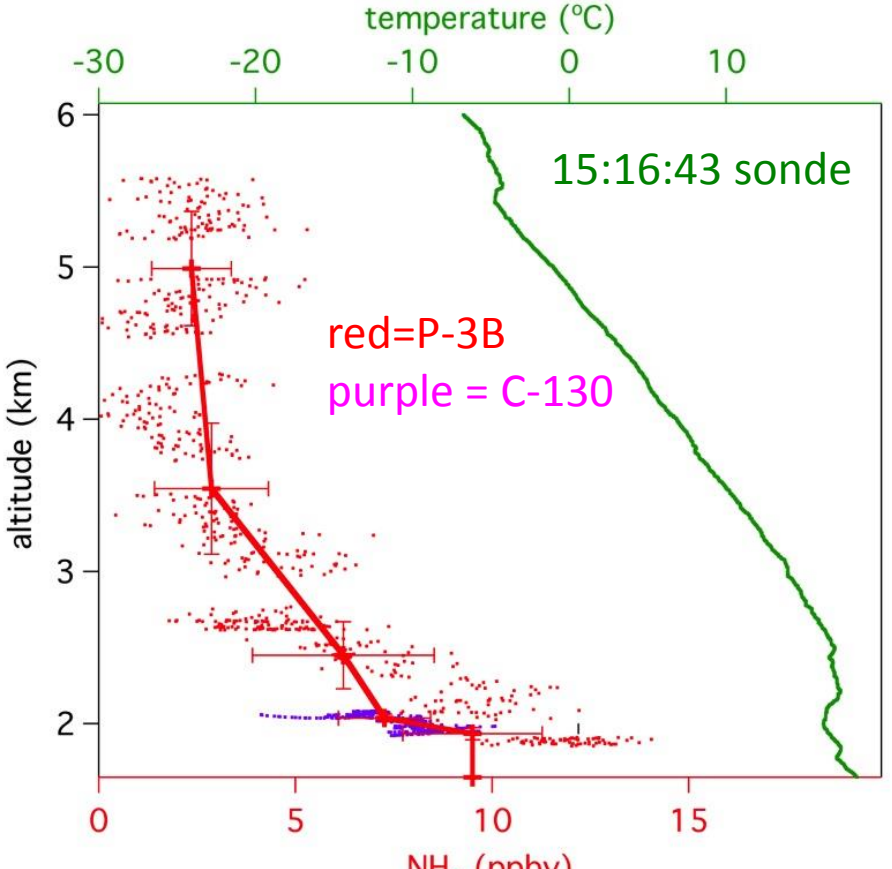
N=64 (±60 min, ±15 km with in-situ profiles)



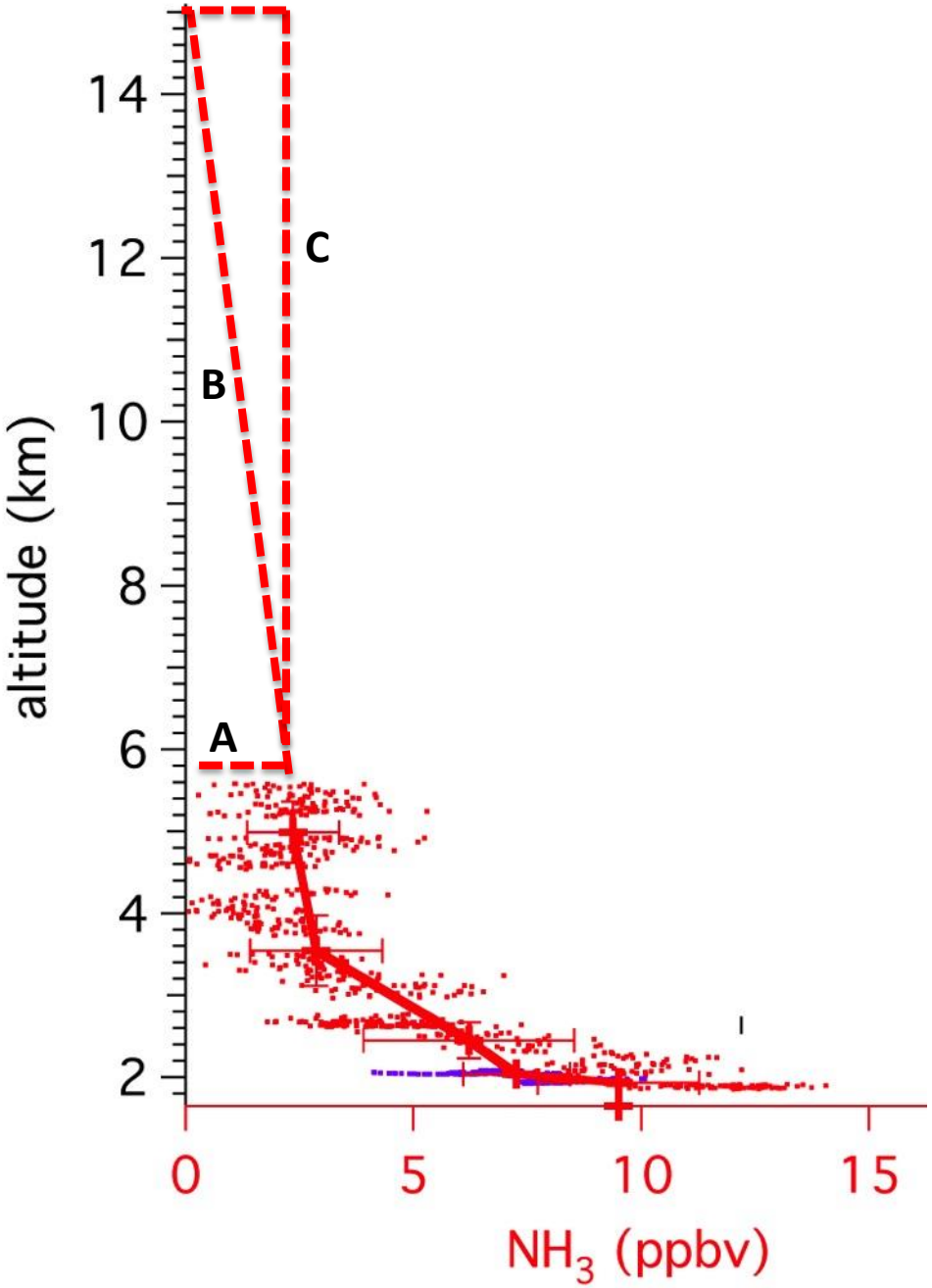


in-situ-derived column comparison method

- identify all in-situ data within space / time window of ±15 km, 60 min.
  (consistent with 4 m s^{-1} mean wind speed in boundary layer)
- data binned within ±100 m of ground and at 500 m bins above
- NH_3 molecular density derived from nearly sonde
- trapezoidal integration of column
- “profile”: in-situ data < 500 m of ground and at least >1.5 km altitude
- profile uncertainties derived from in-situ measurement uncertainties



Aircraft-derived column uncertainties



Assumptions:

1) no NH_3 at z>15 km
(i.e. in stratosphere)

2) extrapolate lowest NH_3 bins to ground

3) no NH_3 spatiotemporal gradients in window

4) mid/upper troposphere for NH_3(z) for z>5 km:

Case A:
No NH_3 above highest plane altitude
Total column = 2.6×10^{16} molec cm^{-2}

Case B:
Linear NH_3 decrease to tropopause
Total column = 3.7×10^{16} molec cm^{-2}

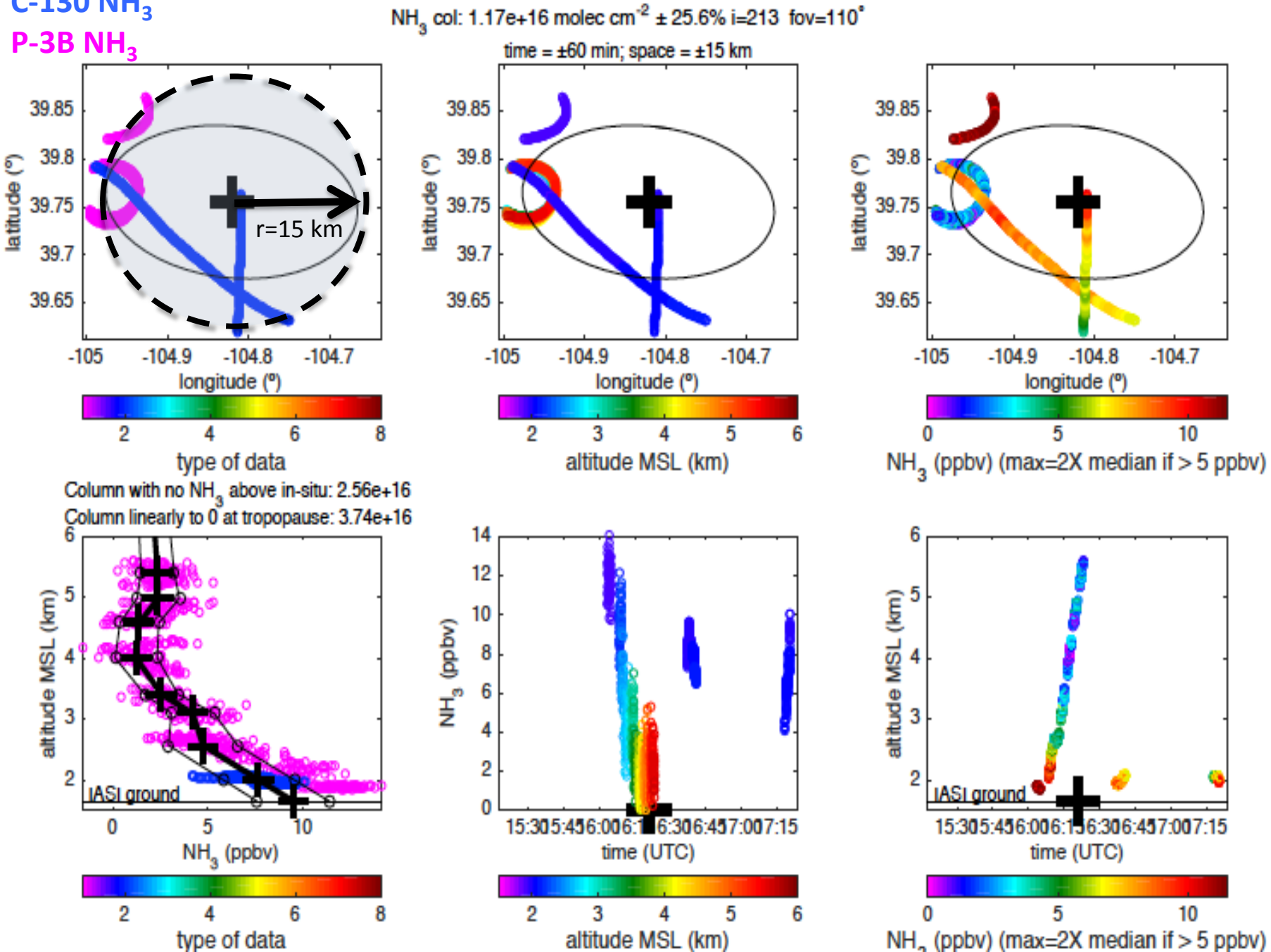
Case C:
Constant NH_3 from 5 km to tropopause
Total column = 5.5×10^{16} molec cm^{-2}

IASI column = $1.2 \times 10^{16} \pm 26\%$

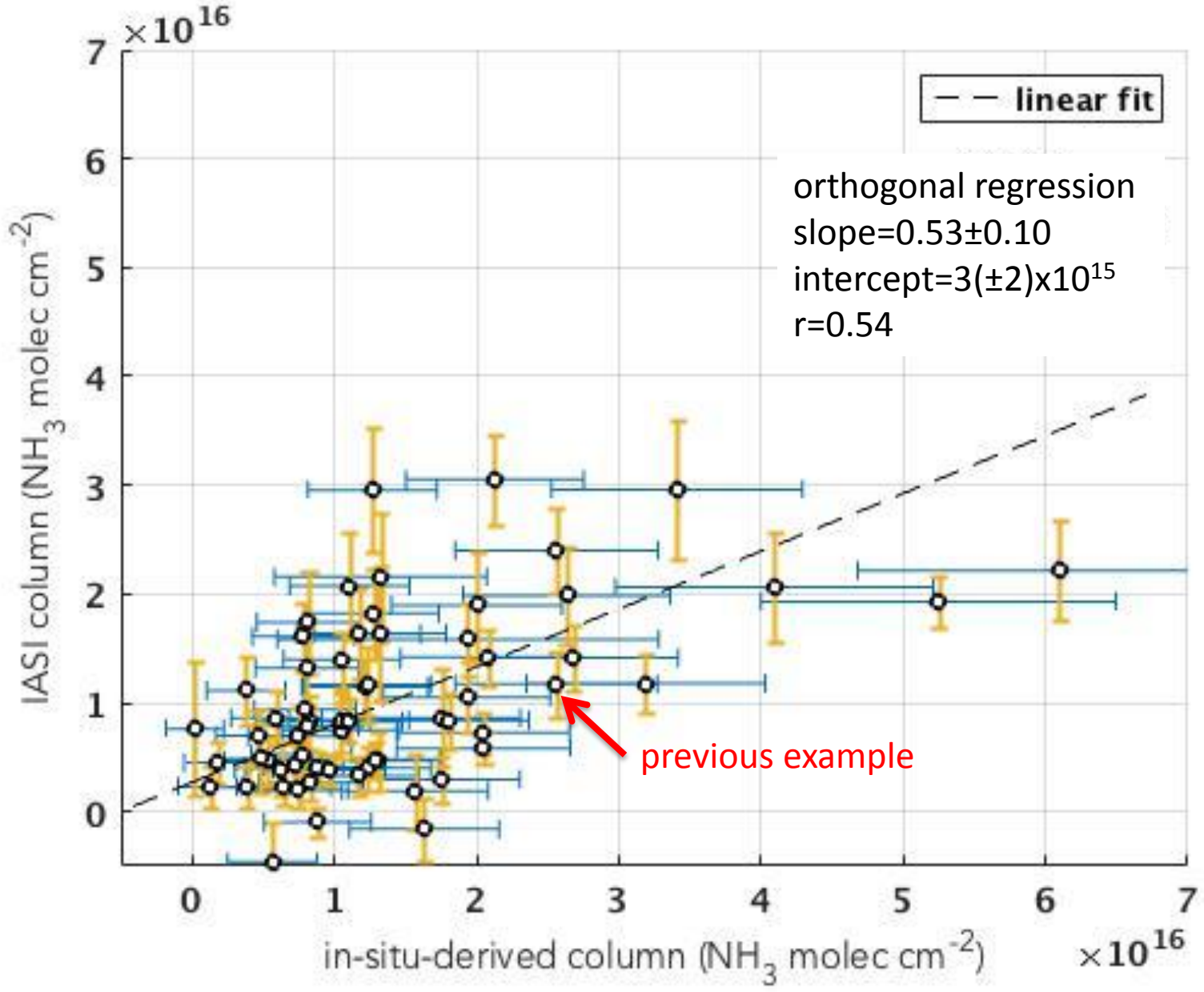
Moderate column case study

C-130 NH_3

P-3B NH_3

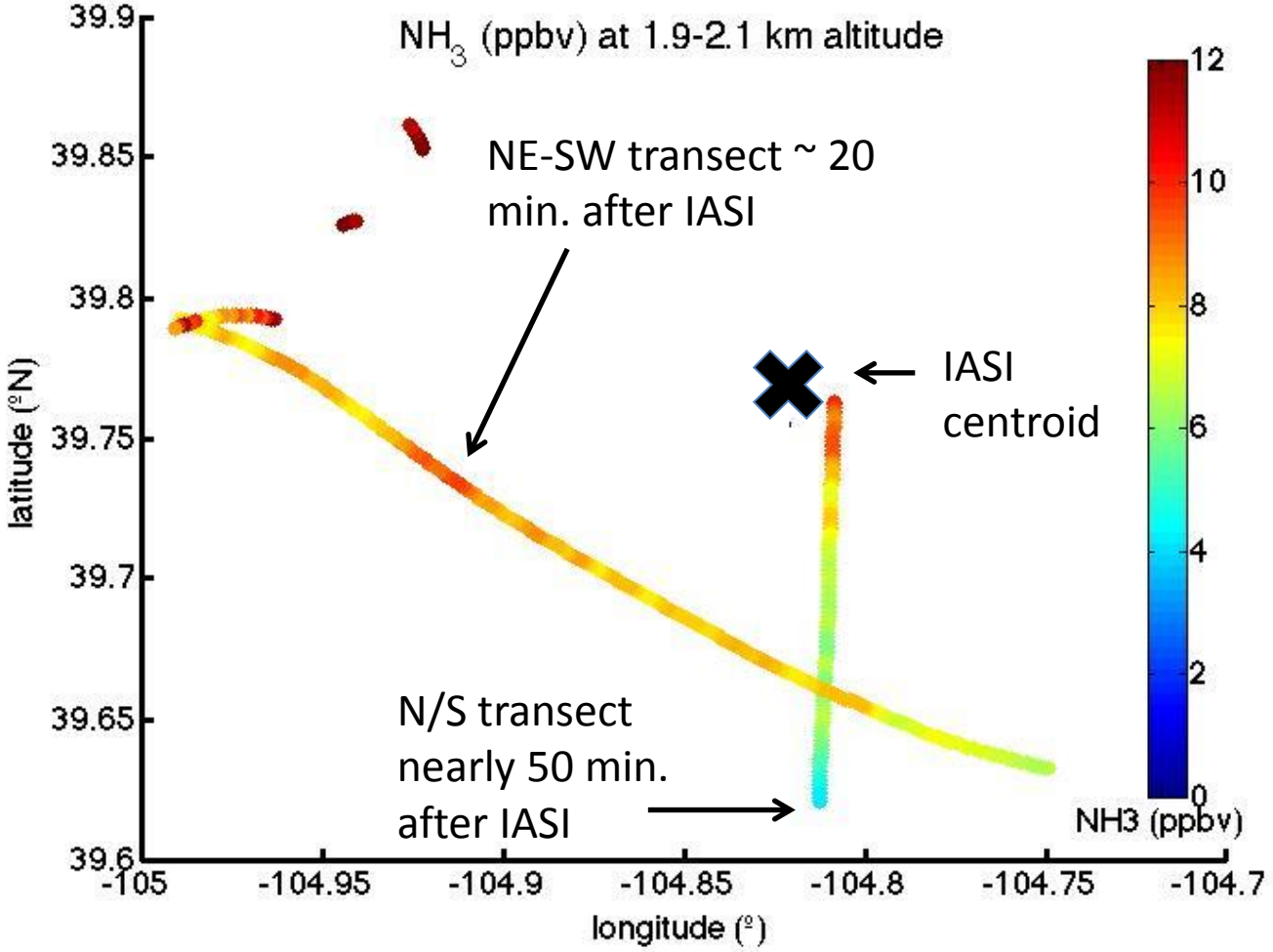


All DAQ-CO data ±1 hr, ±15 km of IASI centroid (N=64)



Revisit spatial gradient assumption

Restrict to 2.0±0.1 km altitude range where large # data available



NH_3 concentration decreases significanyly from NW➔ SE
Most of the profile is taken much further west and slightly north of IASI centroid
Lower NH_3 air to the E/S may help to explain lower IASI column than aircraft

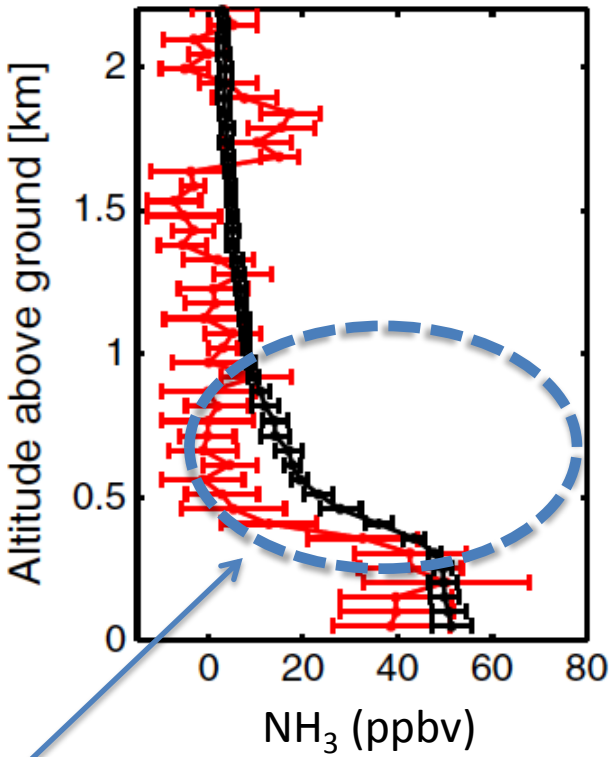
Revisit NH_3 *in-situ* sampling issues

- NH_3 is very "sticky" to instrument surfaces, degrades accuracy
- inlet problems create poorly-constrained (highly variable) uncertainties

Aircraft comparison

closed-path PTRMS (Innsbruk)

closed-path (Aerodyne TDL)

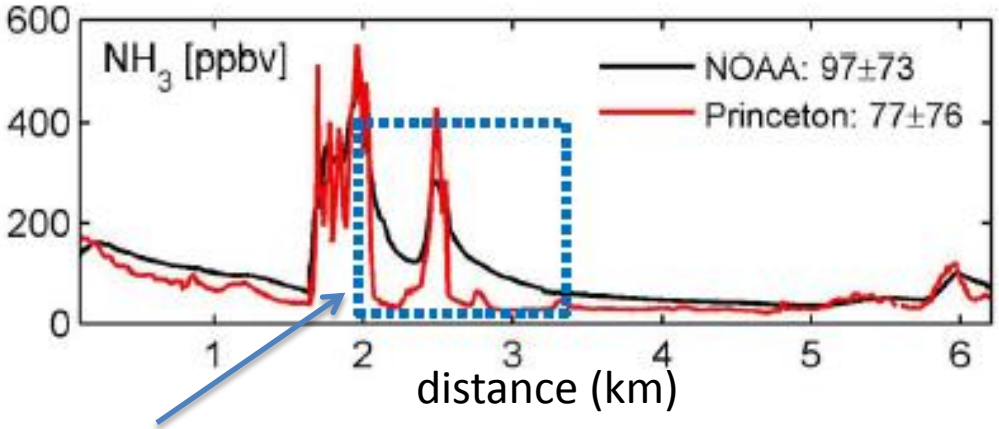


PTRMS responds faster upon spiraling from the polluted boundary layer to the clean, free troposphere but is more imprecise overall than TDL

Mobile lab intercomparison

open-path (Princeton)

closed-path (Picarro, NOAA CSD)



Open-path shows rapid changes while Picarro lags behind due to outgassing

In the presence of strong spatiotemporal gradients, need to be careful with the *in-situ* data

Sensitivity to space (Δs) and time (Δt) windows

correlation (r)

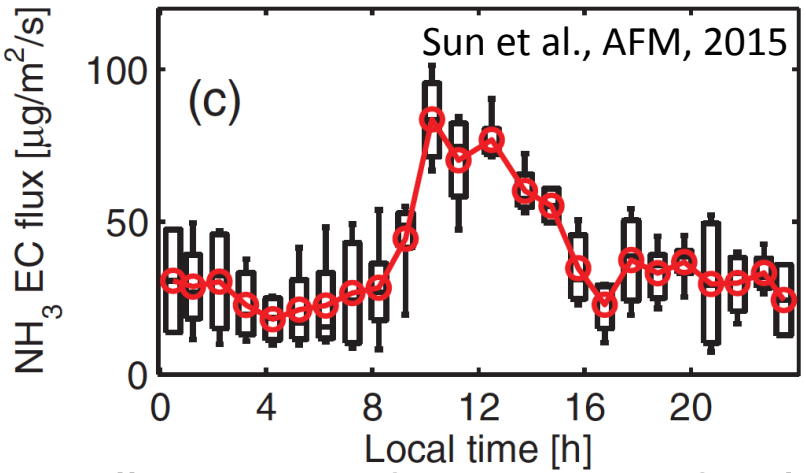
± Δ t (min.) \ ± Δ s (km)	5	15	45
20	0.75	0.63	0.19
60	0.59	0.54	0.14
180	0.60	0.53	0.27

N

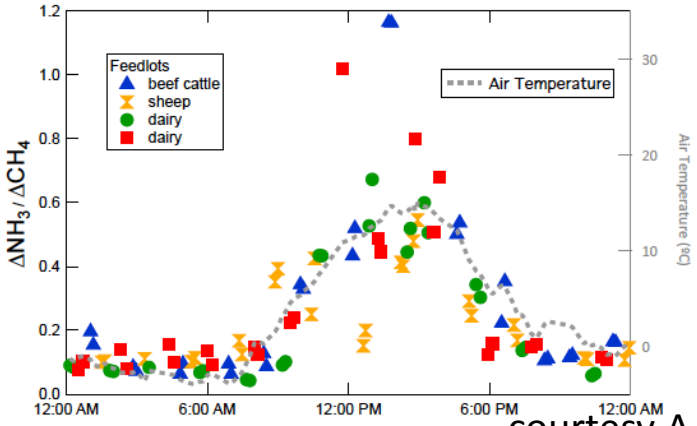
± Δ t (min.) \ ± Δ s (km)	5	15	45
20	8	28	31
60	18	64	101
180	31	229	344

- At 45 km spatial window, correlation degrades significantly at all time windows

Time window is less sensitive but emissions are strongly temp.-dependent:



diurnal NH_3 fluxes at CO feedlot



courtesy A. Neuman

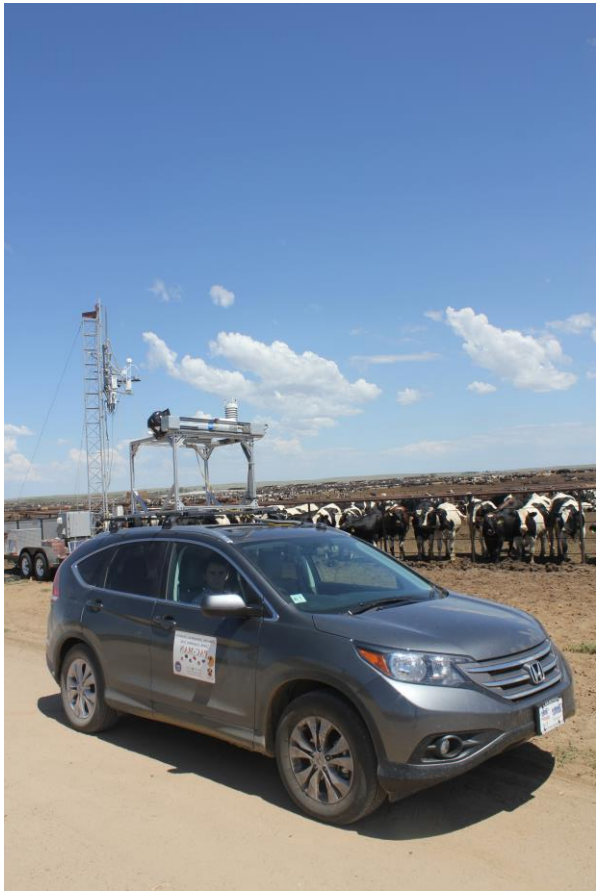
feedlot $\Delta NH_3/\Delta CH_4$ diurnal cycle

Summary / implications

- individual IASI NH_3 columns agree well with in-situ-derived columns (m=0.60 ± 0.10, r ~0.60, b=1x10^{15}), especially given the strong gradients

- first validations at low/moderate NH_3 abundances (10^{15} - 10^{16} molec cm^{-2}) (one order of magnitude lower than previously shown)

- IASI NH_3 can be used with confidence to capture spatial gradients on daily timescales and at moderate column abundances for DAQ-CO

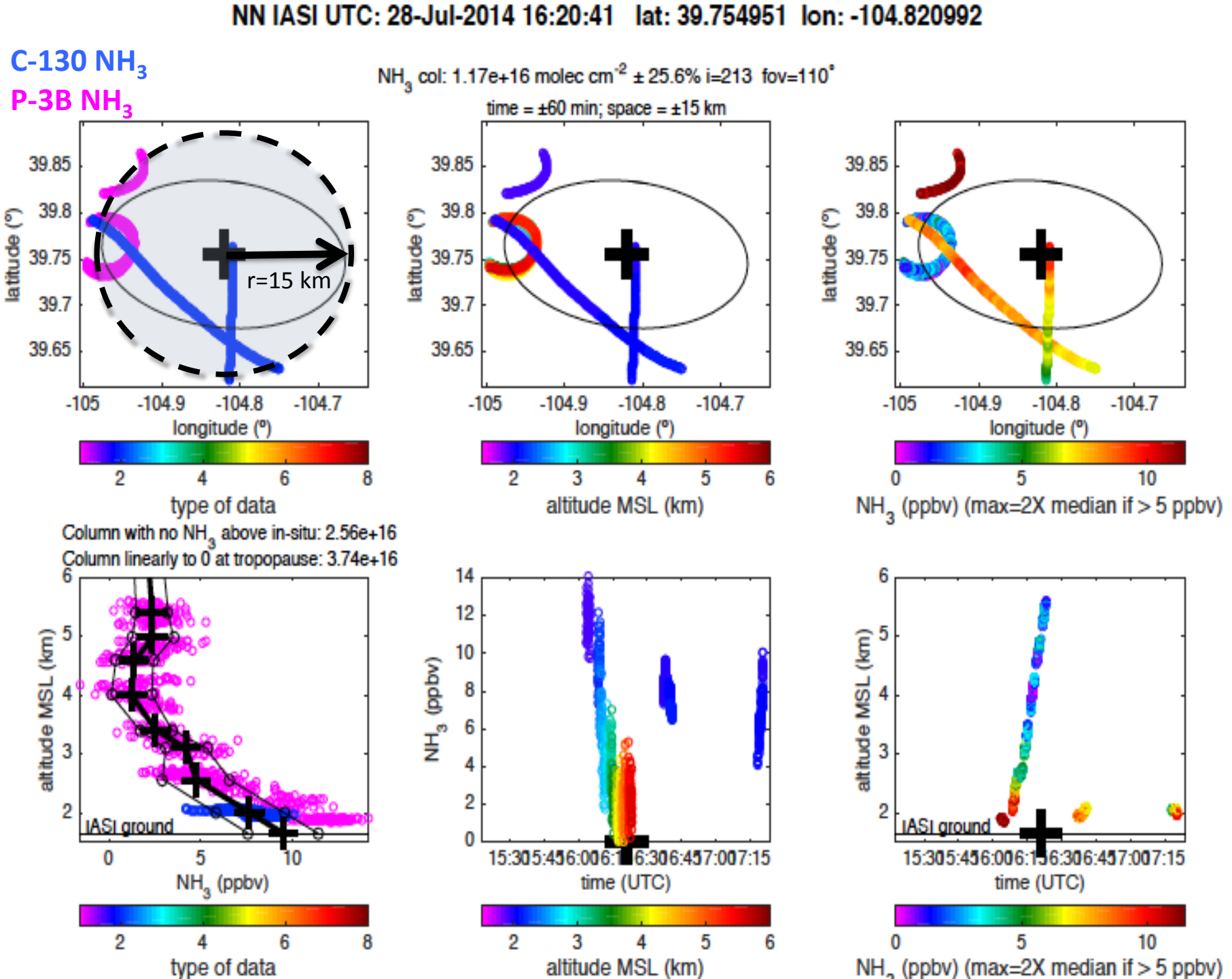


<u>Future validation needs</u>

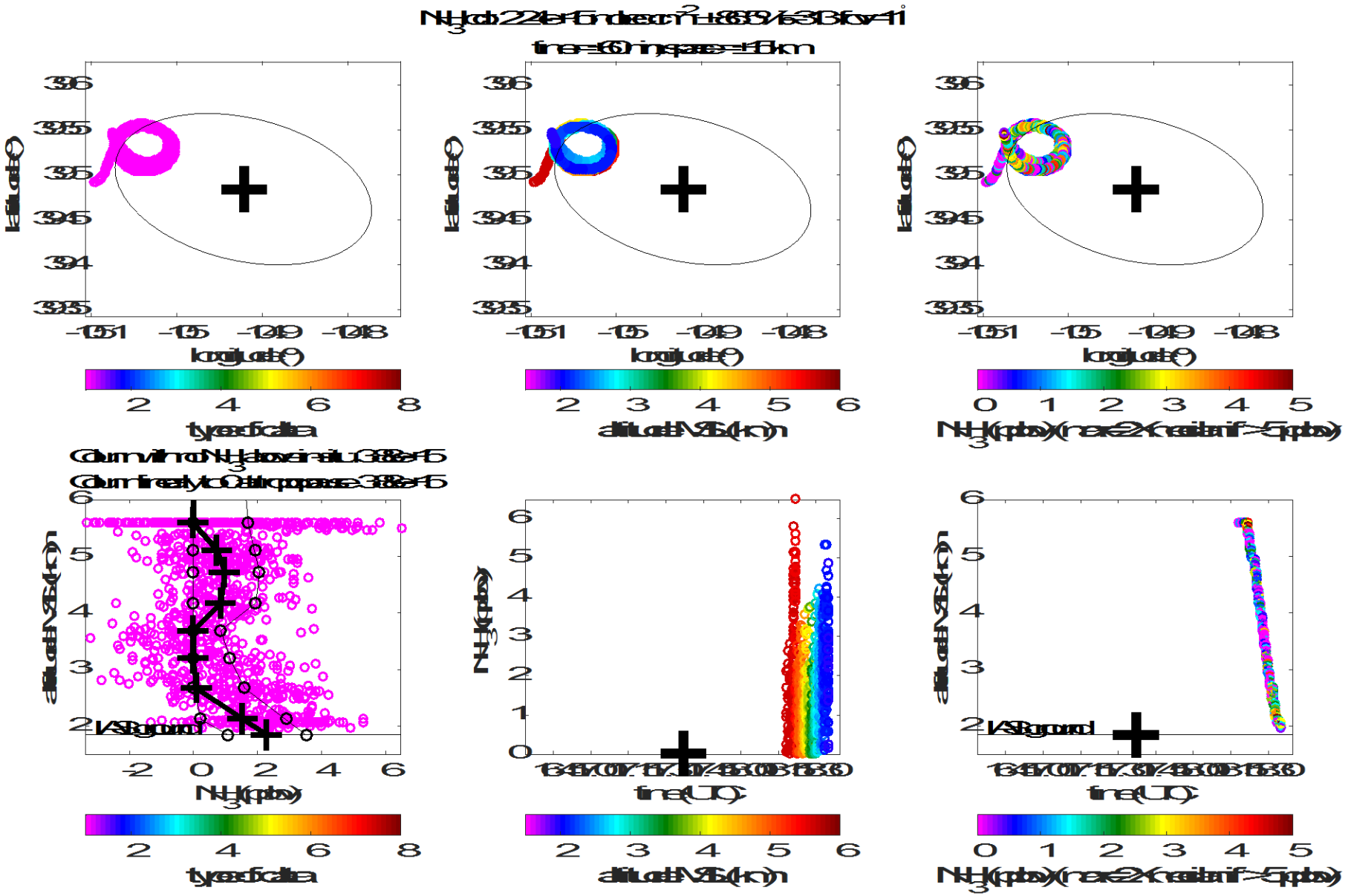
- uncertainties of free tropospheric NH_3 are significant at low column abundances

- in-situ measurement hystereses and biases problematic for columns

- sensitivity to variations in thermal contrast (ocean, night)

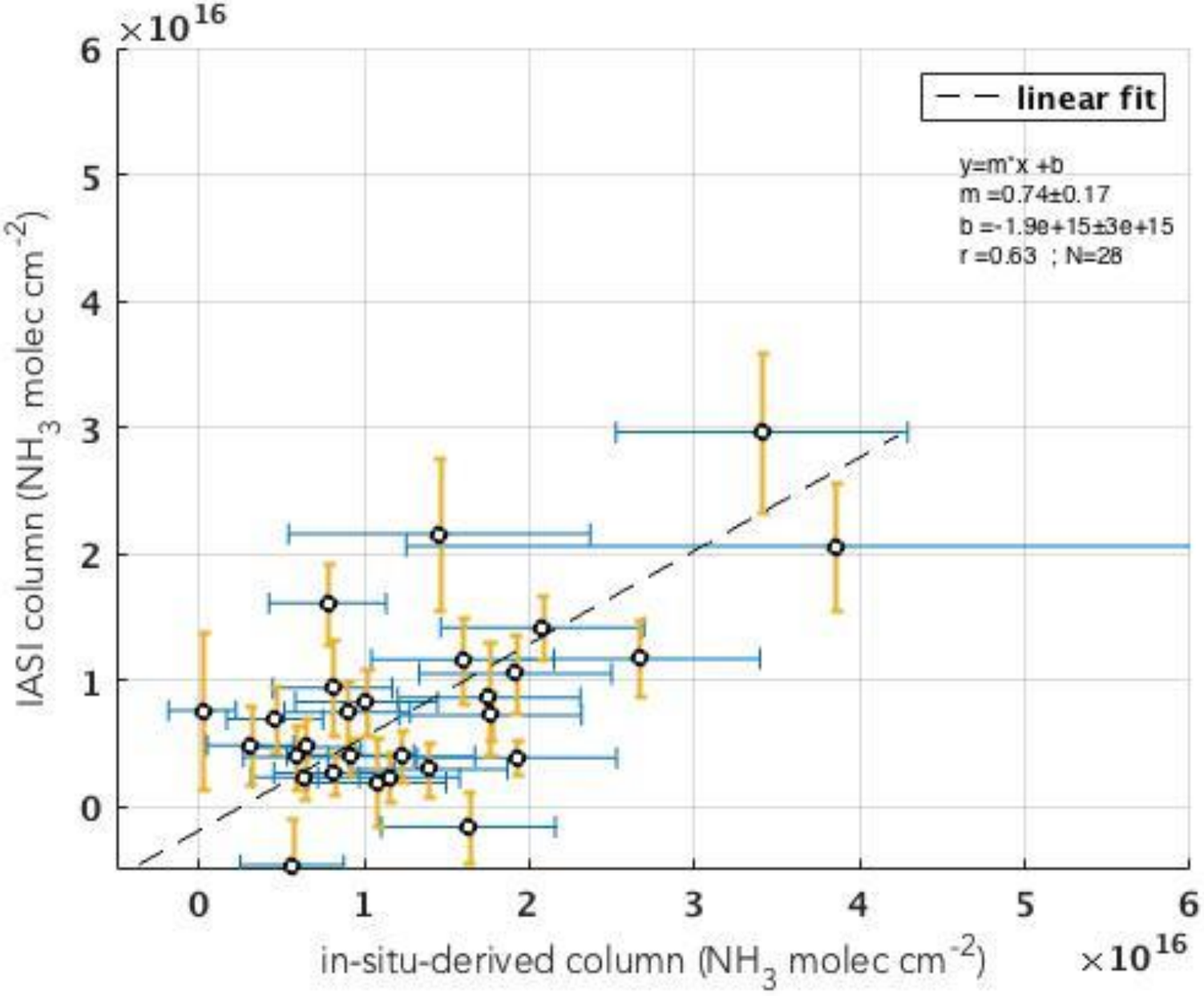

NN IASI UTC: 28-Jul-2014 16:20:41 lat: 39.754951 lon: -104.820992
C-130 NH3
P-3B NH3
NH3 col: 1.17e+16 molec cm-2 ± 25.6% i=213 fov=110°
time = ±60 min; space = ±15 km
r=15 km
latitude (°)
longitude (°)
type of data
altitude MSL (km)
NH3 (ppbv) (max=2X median if > 5 ppbv)
Column with no NH3 above in-situ: 2.56e+16
Column linearly to 0 at tropopause: 3.74e+16
IASI ground
NH3 (ppbv)
time (UTC)

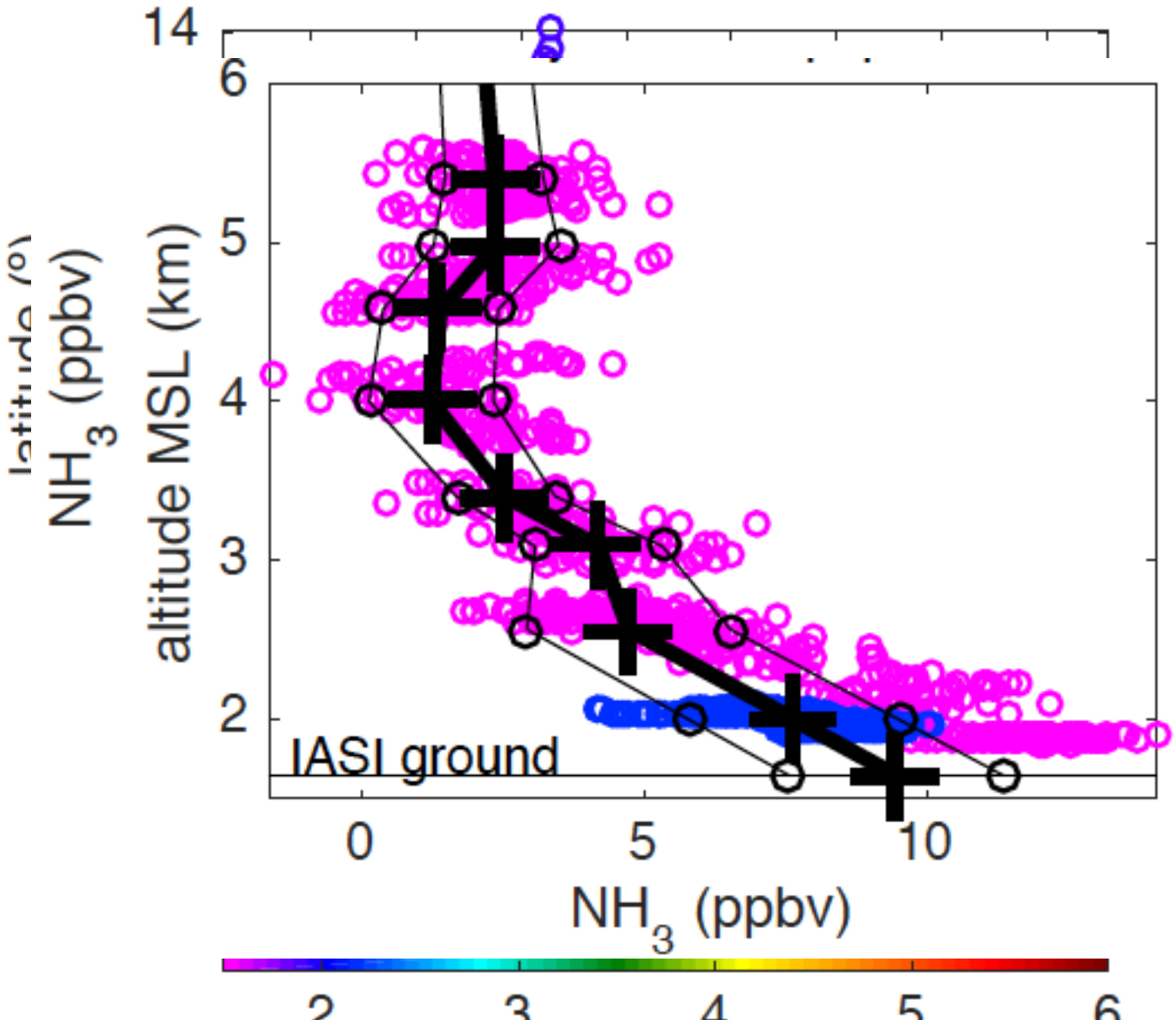
Low column (low 10^{15}) example



Validation at low columns; in-situ likely suffering from sampling biases

±15km, ±20 min


×10^16
6
5
4
3
2
1
0
IASI column (NH3 molec cm-2)
linear fit
y=m*x +b
m =0.74±0.17
b =-1.9e+15±3e+15
r =0.63 ; N=28
0
1
2
3
4
5
6
in-situ-derived column (NH3 molec cm-2)
×10^16

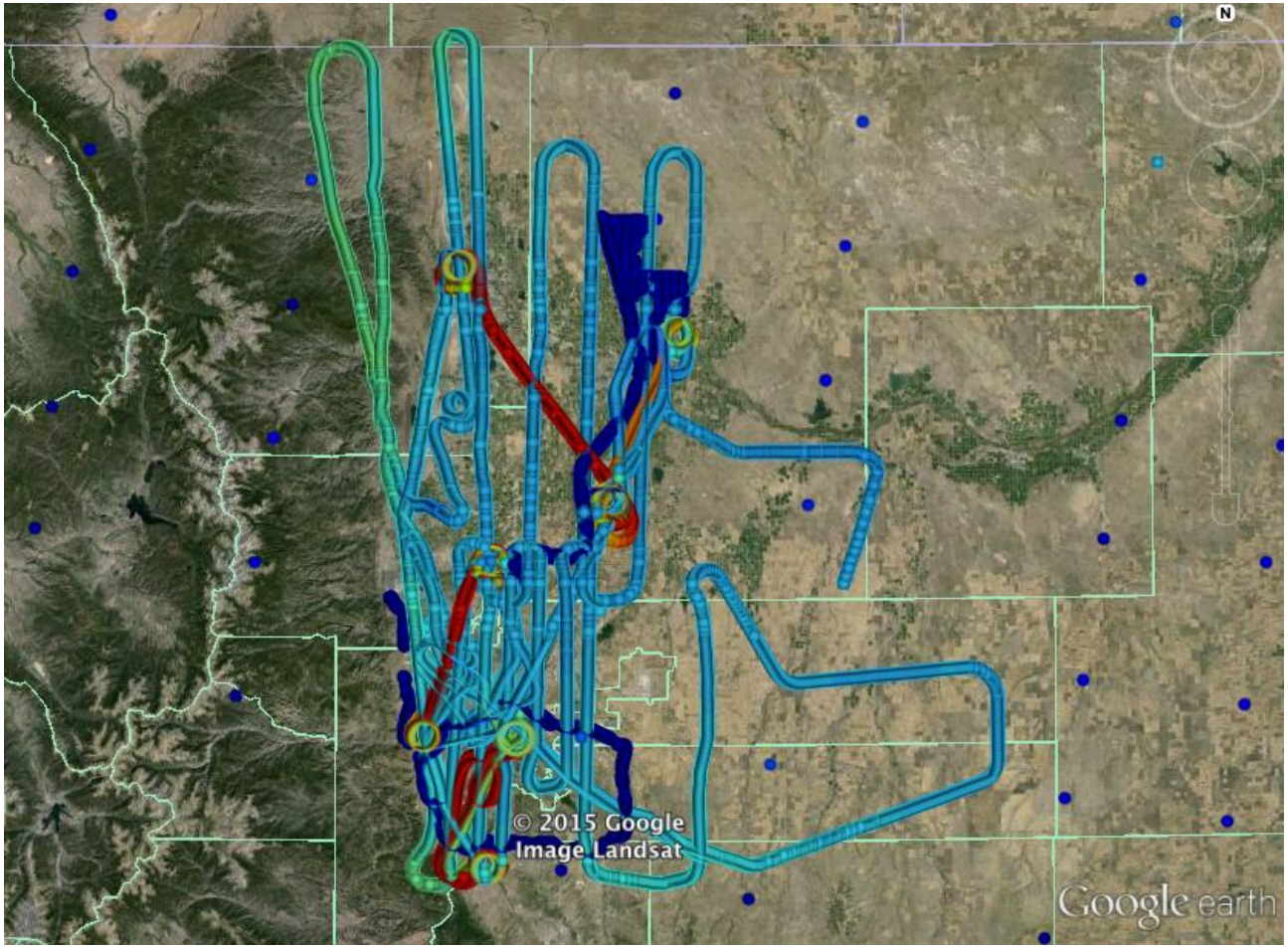

14
6
5
4
3
2
altitude MSL (km)
IASI ground
0
5
10
NH3 (ppbv)
NH3 (ppbv)
2
3
4
5
6

Case study of July 28, 2014

Extensive set of aircraft/ground measurements:

N=261 IASI; N=149 (filtered); N=8 for ±1 hour, ±15 km of aircraft data

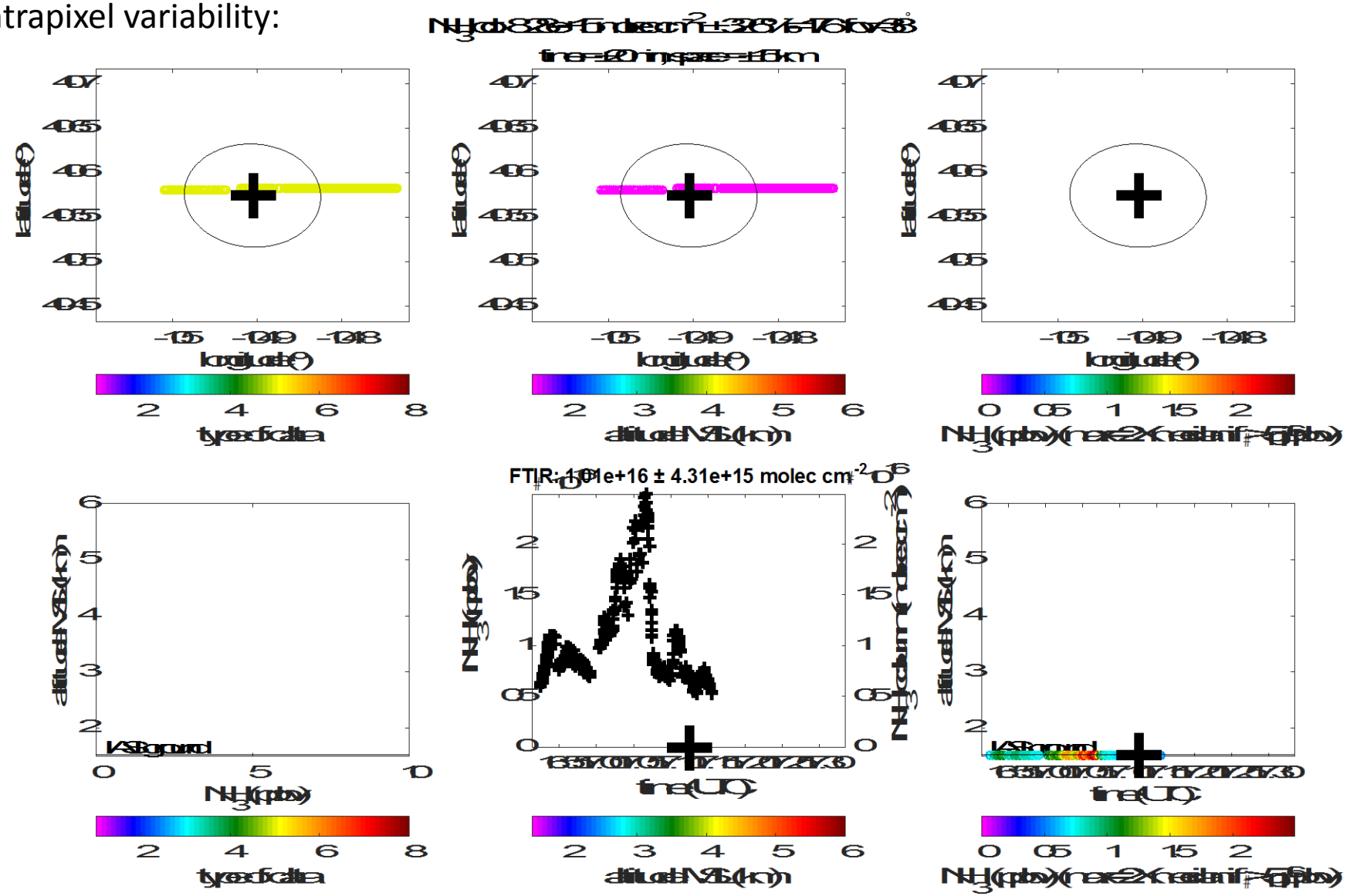
(comparable scales of Δs, Δt with wind of 4 m s^{-1})



(colors here represent ~ altitude (dk. blue = ground; red=5 km)

NN IASI UTC: 09-Aug-2014 17:12:26 lat: 40.574162 lon: -104.905441

Intrapixel variability:



CU FTIR = 1.01 x 10^{16} mean but shows large variability on 30 km transect

Field experiments

NASA DISCOVER-AQ (Colorado): Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality

- focus on vertical profiles, satellite validation, air quality modeling
- July 17 - Aug. 12, 2014
  - P3-B and B200 aircraft, mobile labs, tower, ground sites, soundings (16 P3-B flight days)

NSF FRAPPE: Front Range Air Pollution and Photochemistry Experiment

- focus on photochemistry, spatiotemporal gradients
- July 26 – Aug. 18, 2014
- separate but coordinated study on NSF C-130, mobile labs, tower (15 C-130 flight days)

Sensitivity to space (Δs) and time (Δt) windows

slope (m)

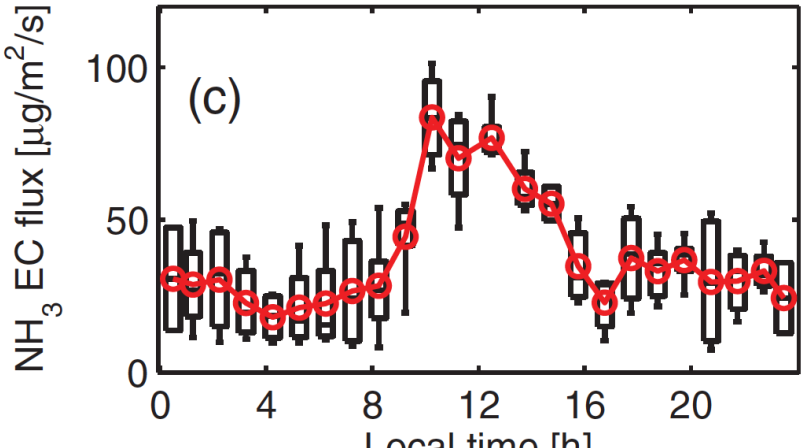
± Δ t (min.)	± Δ s (km) 5	15	45
20	0.62	0.74	0.04
60	0.16	0.53	0.14
180	0.66	0.99	0.14

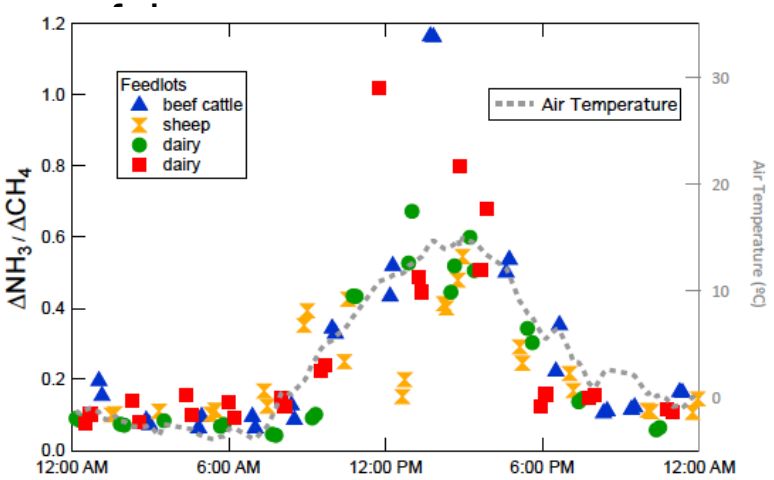
correlation (r)

± Δ t (min.)	± Δ s (km) 5	15	45
20	0.75	0.63	0.19
60	0.59	0.54	0.14
180	0.60	0.53	0.27

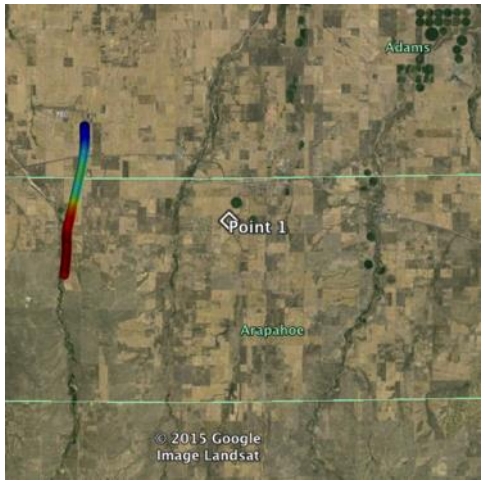
- At 45 km spatial window, correlation degrades significantly at all time windows

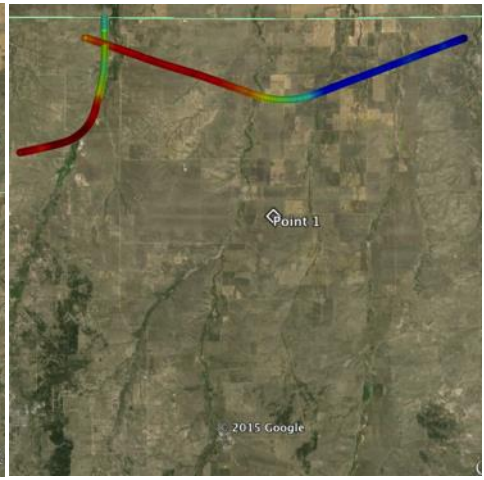
Time window is less sensitive but one must be





N=8 cases with in-situ data ±1 hr, ±15 km of an IASI centroid for July 28
(respective IASI labeled 'Point 1' in each graph below)





072814: Δs=±15 km, Δt=±1 hr (mean wind 4 m s^{-1} @ one hour = 14.4 km)
(autoscaled color here, so disregard)

Sensitivity spatiotemporal window