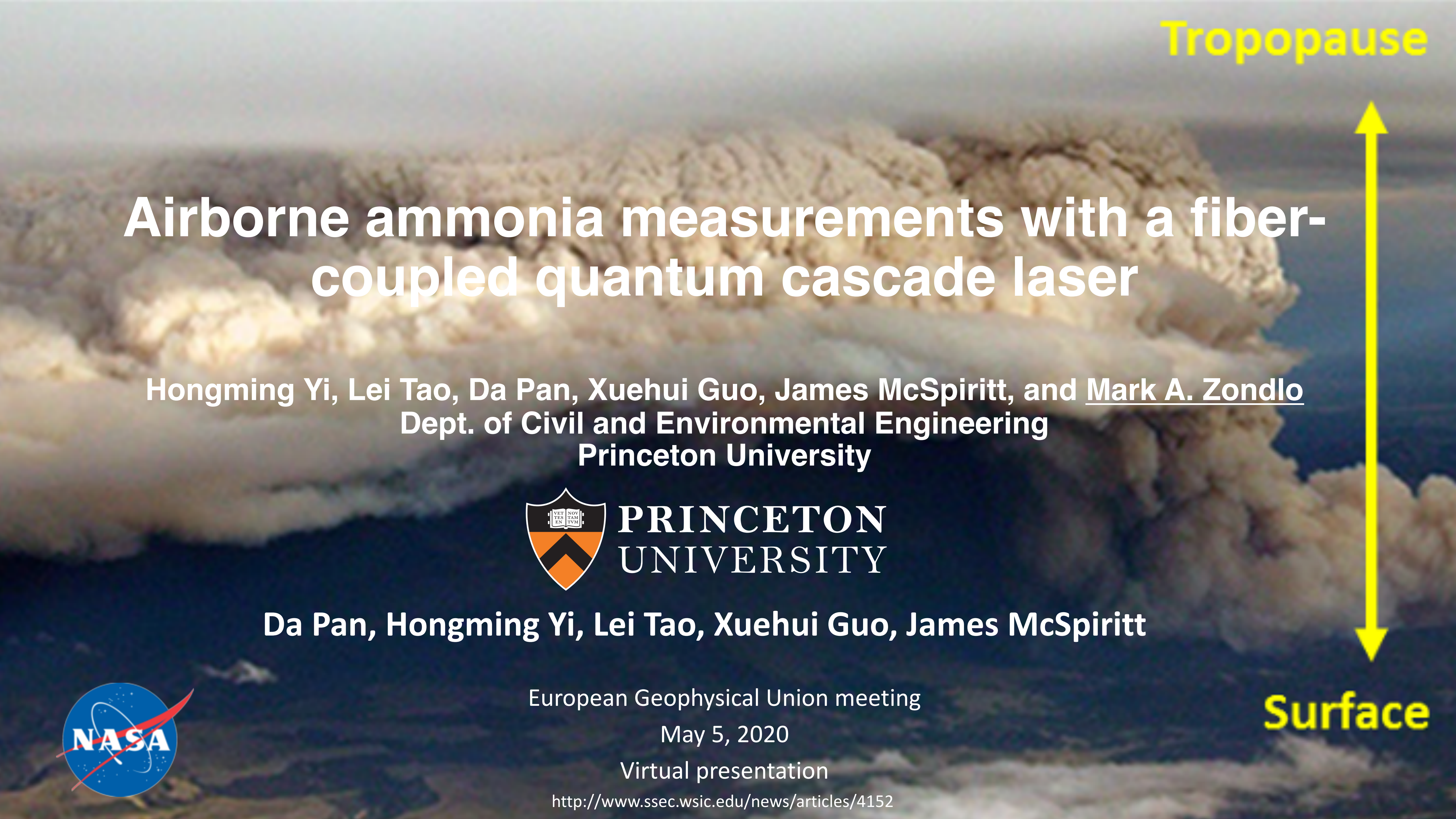
Airborne ammonia measurements with a fiber-coupled quantum cascade laser

Hongming Yi, Lei Tao, Da Pan, Xuehui Guo, James McSpiritt, and Mark A. Zondlo
Dept. of Civil and Environmental Engineering
Princeton University

PRINCETON UNIVERSITY

Da Pan, Hongming Yi, Lei Tao, Xuehui Guo, James McSpiritt

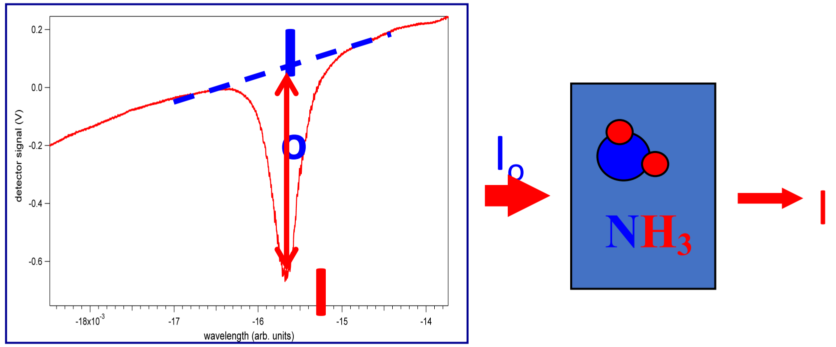


European Geophysical Union meeting
May 5, 2020
Virtual presentation

http://www.ssec.wsic.edu/news/articles/4152

Airborne-based ammonia instrument attributes

Measurement requirements	Design attributes
Fast-response	Open-path configuration (minimizes adsorption/desorption)
Accurate measurements	Open-path configuration (minimizes adsorption/desorption biases)
High sensitivity	50 m optical path; mid-IR laser at 9 µm
Laser temperature stability	Fiberized laser (laser inside cabin, light outside via fiber)
Optical cell stability over 220-310 K	Invar struts between optical cell mirrors
Mirror degradation / alignment	Replaceable optical cell from outside, can be pre-aligned with fiber optic
Mirror icing / condensation	Mirrors heated slightly above ambient
Cloud particles	Shadow zone of larger particles
Plane boundary layer artifacts	Mirrors 15” above fuselage
Optical fringing/feedback from fiber	Hollow core fiber (200 µm dia.) vs. solid core
Interstitial NH_3 inside fiber/cabin	Purge/closed N_2 flow through fiber
Ease-of-installation	Sensor attached to modified viewport plate



Beer-Lamber Law:

$$\frac{I(\lambda)}{I_o(\lambda)} = \exp(-\alpha(\lambda))$$

$$\alpha(\lambda) = S(T)\, g(\lambda,T,P)\, N\, l$$

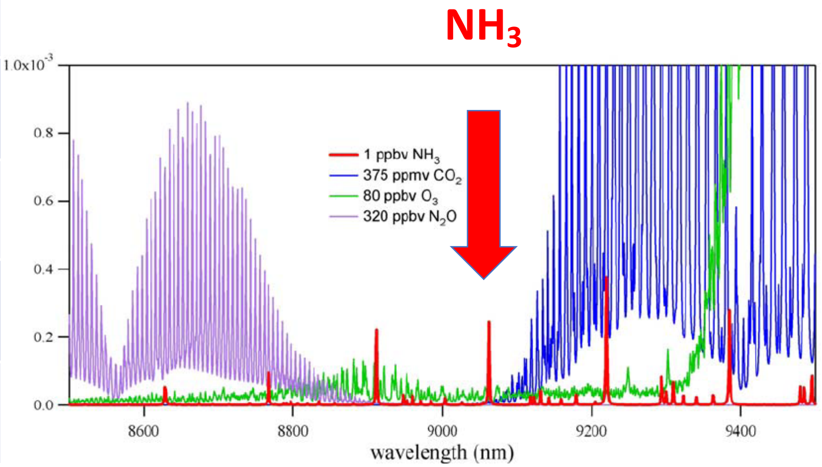
where:

S(T) is the linestrength

$g(\lambda,T,P)$ is the normalized Voigt lineshape function

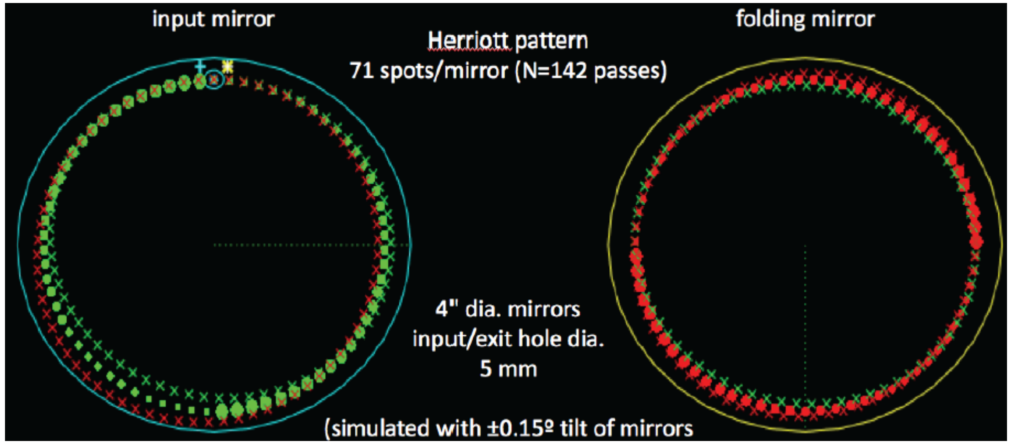
N is the absolute concentration

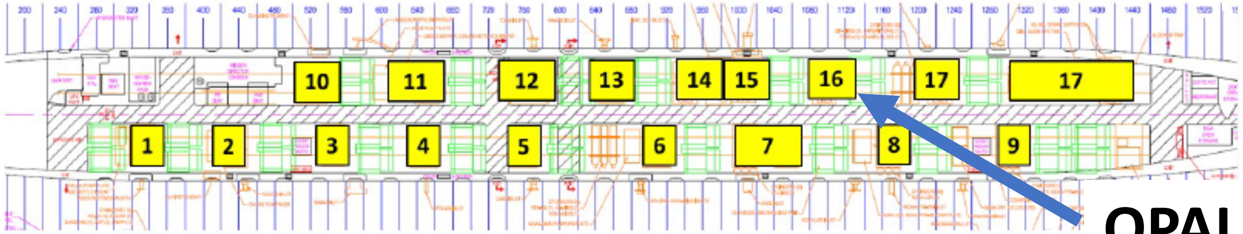
l is the pathlength



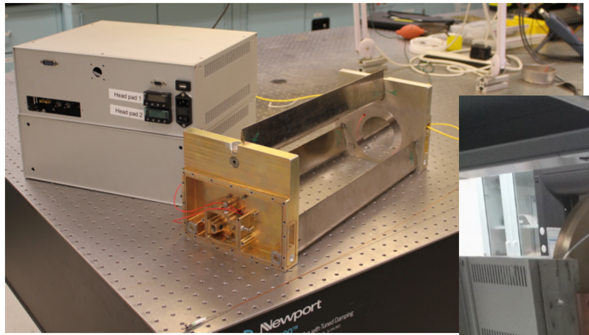
Instrumentation: Open-Path Ammonia Laser Spectrometer (OPALS)

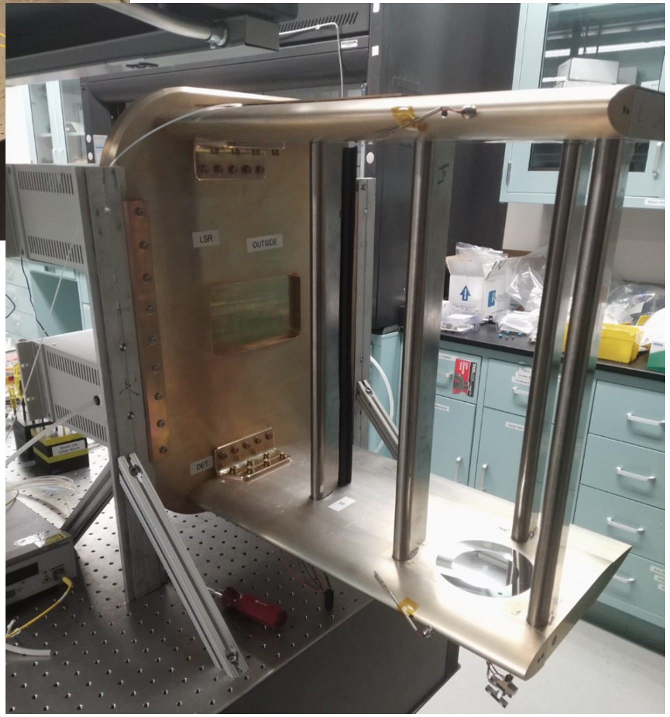
- open-path avoids adsorption hystereses, phase partitioning
- fast (1 Hz), sensitive (60 pptv $Hz^{-0.5}$), ±(20% + 0.1 ppbv) accuracy
- wavelength modulation spectroscopy
- Herriott cell w/ 4.5” Al mirrors
- builds upon QCL-based tower/mobile-lab NH_3 sensors and open-path VCSEL hygrometer for NSF G-V
- reference cell for linelocking during icing/hydrometeors

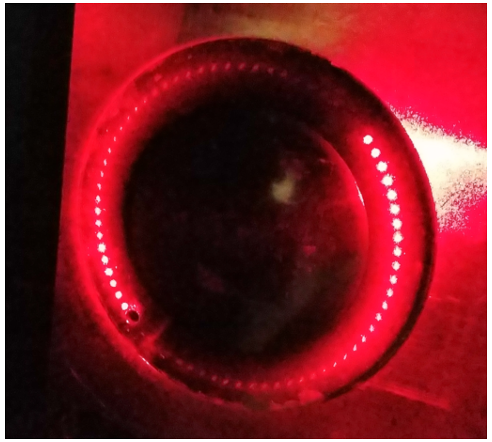


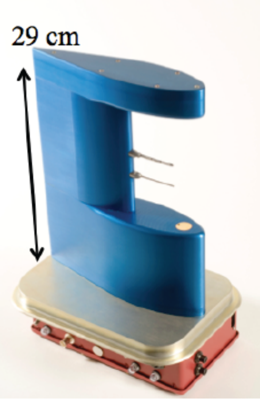


OPALS







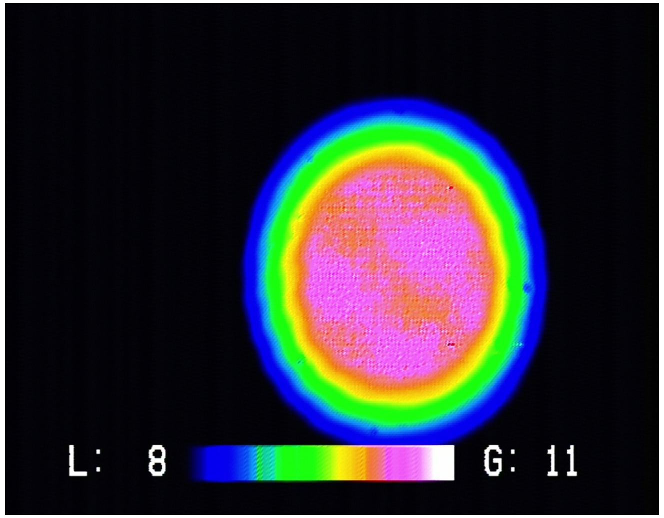


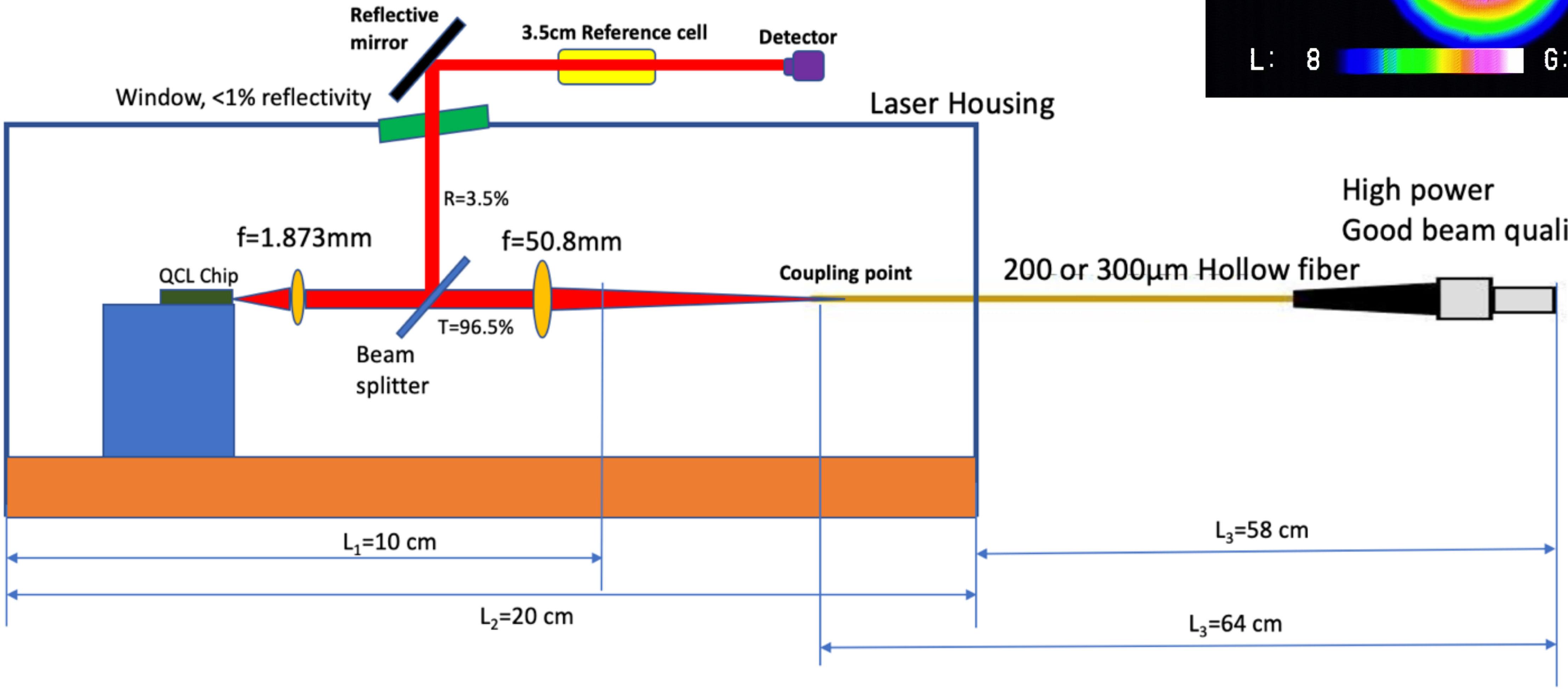
NSF G-V VCSEL hygrometer

Zondlo et al., JGR-A, 2011
Miller et al., AMT, 2014
Sun et al., Appl. Phys. B, 2013
Tao et al., Appl. Phys. B, 2015
Sun et al., Ag. For. Met., 2015
Sun et al., ES&T, 2017

Optical configuration

- 9.06 μm continuous wave, DFB quantum cascade laser (Adtech)
- hollow core 200 μm dia. fiber
- separate linelocking reference cell (ethylene, ammonia at 100 Torr)

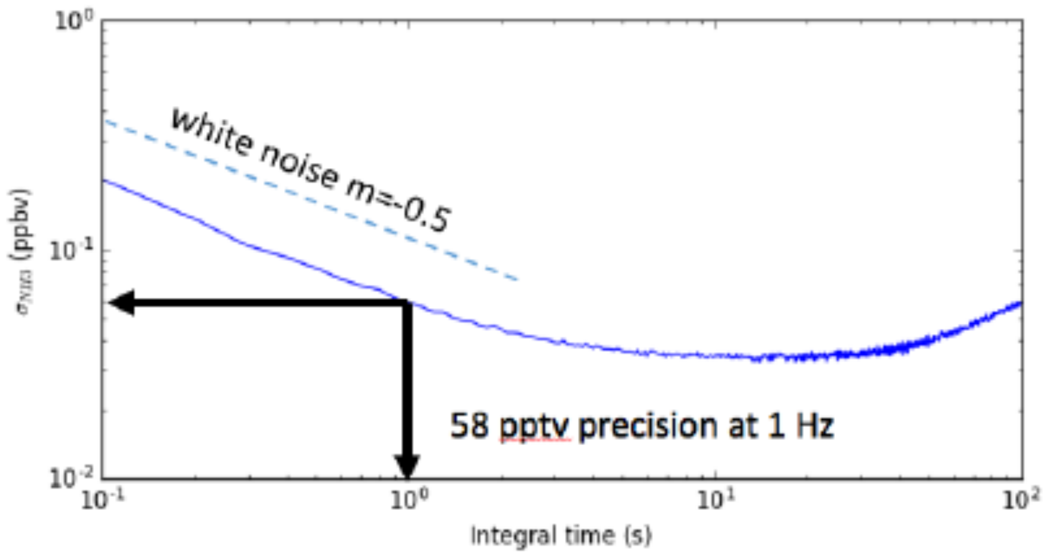


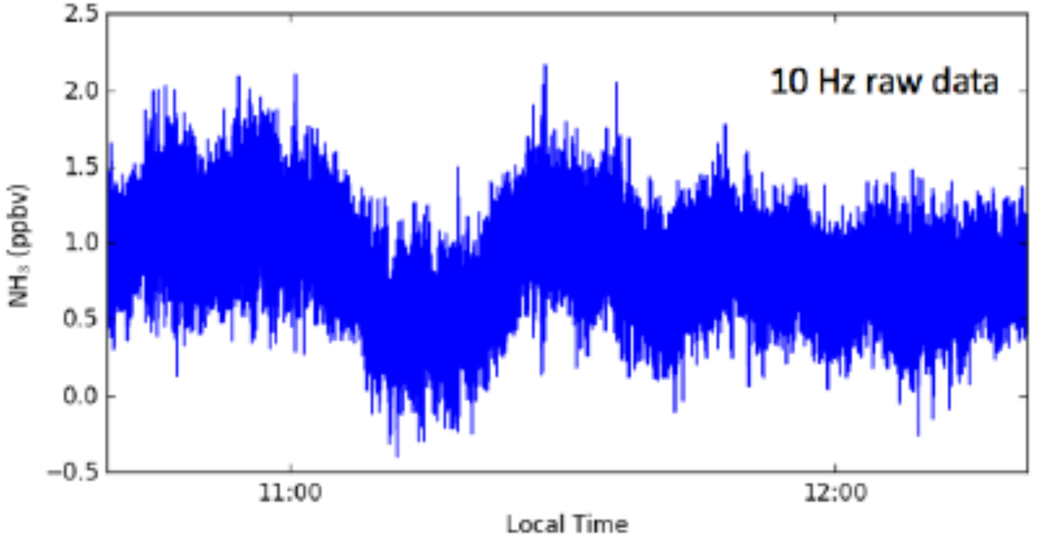


System performance: Allan plot

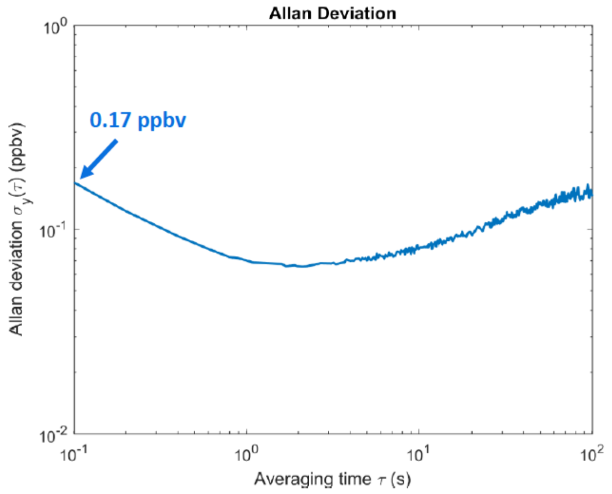
- deployment conditions on a tower at Duke Forest
- relatively low NH_3 conditions (~ 1 ppbv)
- some of the drift/noise real atmospheric signal (upper limit to instrument noise/drift)
- precision: 150 pptv at 10 Hz, 60 pptv at 1 Hz
- consistent with laboratory (though hard to get a precise and calibrated 1 ppbv NH_3 in the lab)

field data:





right, lab plot:



$$\sigma_y(\tau) = \sqrt{\frac{1}{2}\left\langle (\Delta y)^2 \right\rangle}$$

OPALS on the NASA DC-8 aircraft

OPALS = Open-Path Ammonia Laser Spectrometer

- completely self-contained on window viewport plate
- uninterrupted power supply, laptop for viewing at rack
- test flights in late 2020