High Accuracy Mobile Emissions Laboratory for Measurements of Greenhouse Gases, Isotopes, Fluxes, Pollutants

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- Overview/Motivation
- Construction of Mobile Lab
- Measurement theory
- LGR's Gas and Isotope Analyzers
- Field deployments
- Summary and Conclusions

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Need for quantification of emissions at local level

Emission targets may be established at the state or national scale, but measurement, monitoring and mitigation must be engaged at urban or individual industrial locations.

Thus reducing GHG emissions requires quantification/verification at regional and local levels.

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Novel Instruments Provide New Opportunities

- Fast Greenhouse Gas Analyzer: CH_4, CO_2, H_2O at 10 Hz
- N_2O and CO Analyzer: measurements at 20 Hz
- Isotopic CO_2 ($\delta^{13}C$, CO_2) in air at 1 Hz (continuous)
- Isotopic CO_2 ($\delta^{13}C$, CO_2): 300 ppmv - 100% (discrete)
- Water Vapor Isotopes (δ^2H, $\delta^{18}O$, H_2O) at 5 Hz
- Liquid Water Isotopes (δ^2H, $\delta^{18}O$): >133 samples/day
- Methane Carbon Isotopes ($\delta^{13}C$, CH_4) in real time
- Nitrous Oxide Isotopes ($\delta^{15}N$, $\delta^{18}N$, N_2O) in real time
- Ammonia (NH_3) in air at 1 Hz
- Carbonyl sulfide (OCS) Analyzer: 1 Hz
- Perfluorocarbon tracers

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Novel Instruments Provide New Opportunities

Christensen, et al, **Nature** 2008;456 (7222):628-30

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Novel Instruments Provide New Opportunities (NASA staff in Antarctica)

Lake Untersee, Antarctica (NASA)

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Novel Instruments Provide New Opportunities (LGR instruments for flight)

On-board aircraft: UAV and planes

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LGR's Mobile Emissions Lab provides sensitive measurements anywhere

LGR's Mobile Emissions Lab
Sponsored (in part) by:
California EPA Air Resources Board

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Trailer + LGR Gas Analyzers and assorted metrology equipment = Mobile Emissions Laboratory

- Trailer (Cargo Wagon; 7’ wide) serves as Mobile Emissions Laboratory

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Thermal insulation added inside walls of Mobile Lab

- Thermal insulation (RU13) added inside walls for operation in extreme environments.

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Thermal insulation added inside walls of Mobile Lab

- Thermal insulation (RU13) added inside walls for operation in extreme environments.

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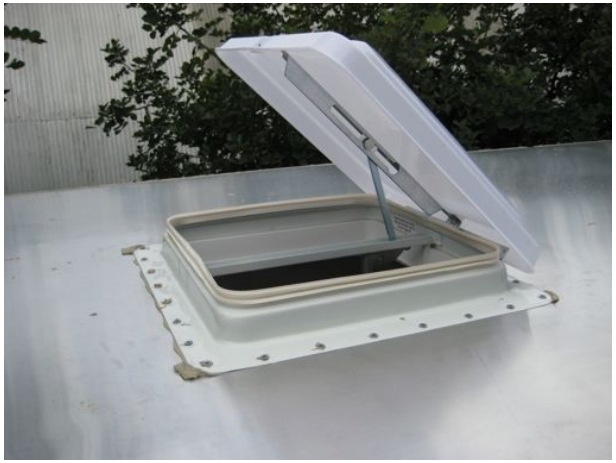
Roof mounted gas input/output ports and vents

multiple gas input/output ports

vent input/output ports



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Onboard DC power available

- Deep cycle marine batteries under floor provide power for long-term operation where AC power is limited.

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Power connections to/from Mobile Emissions Laboratory

external power input/output

breaker panels for AC input and internal power distribution

inverter/charger converts AC to DC battery power

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Accessories Inside Mobile Lab

- Multiport Inlet Unit combined with long-length inlet lines allow for measurements at multiple locations and at various distances from Lab
- Sonic anemometer provides measurements of wind direction and velocity

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Onboard PC, data logger provides centralized data acquisition

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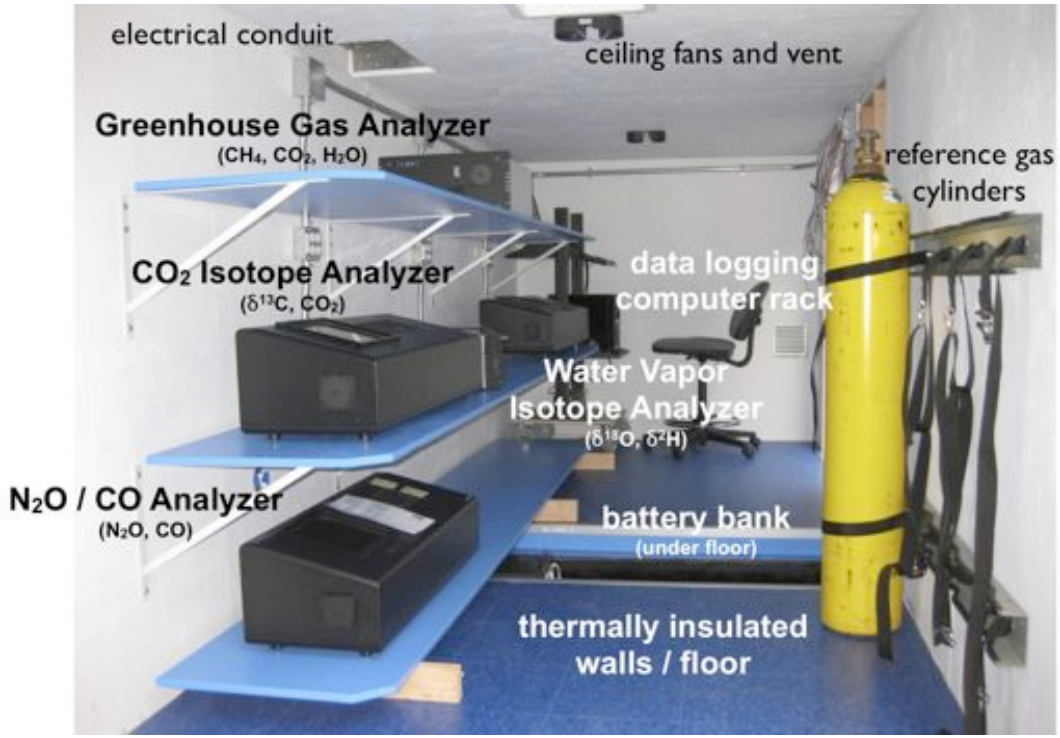
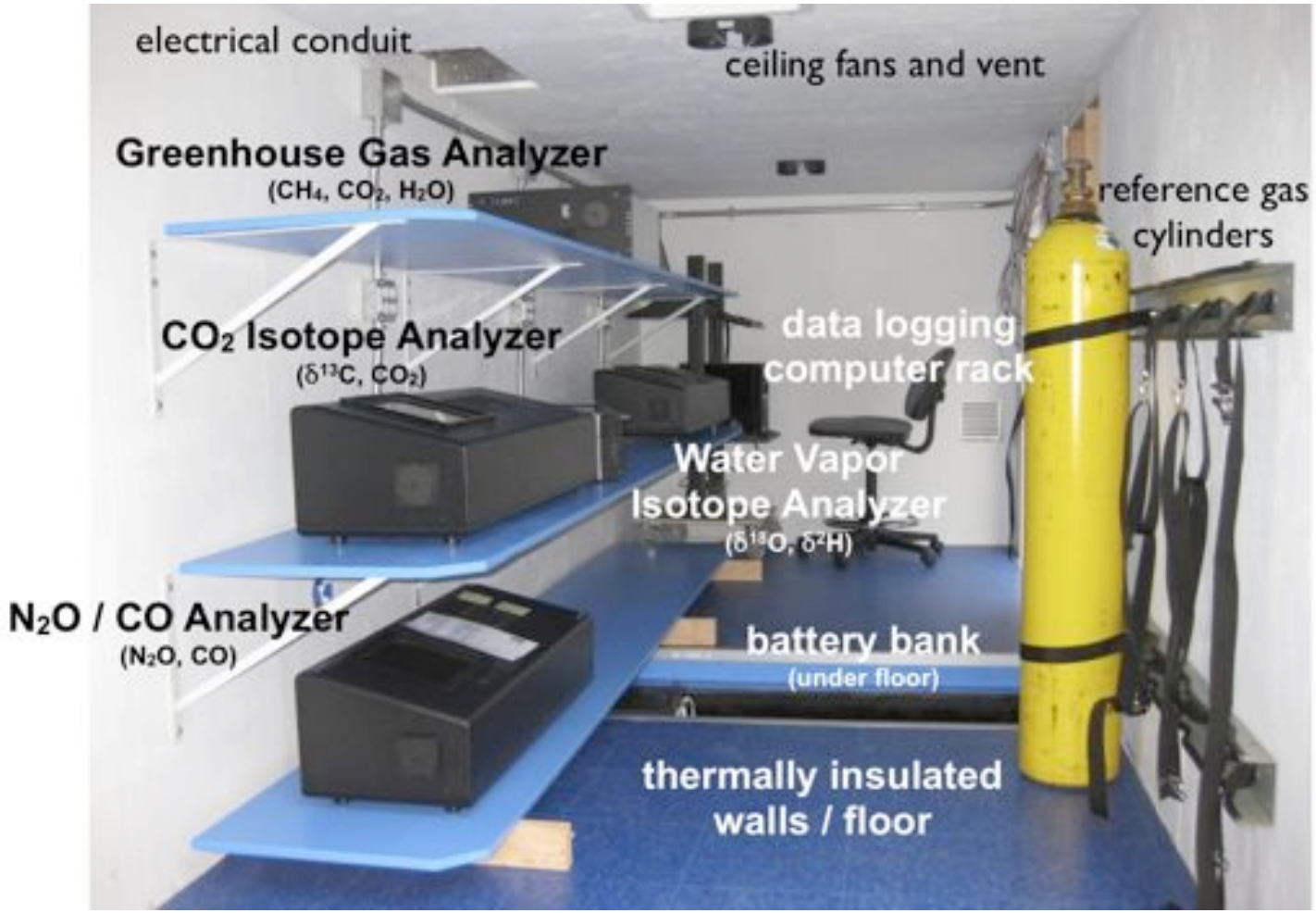


LGR's Mobile Lab includes several autonomous gas analyzers and accessories

LGR Mobile Lab provides sensitive field measurements at landfills, urban, rural locations

Mobile Emissions Lab includes several LGR Analyzers



On-board battery allows long term operation in the field

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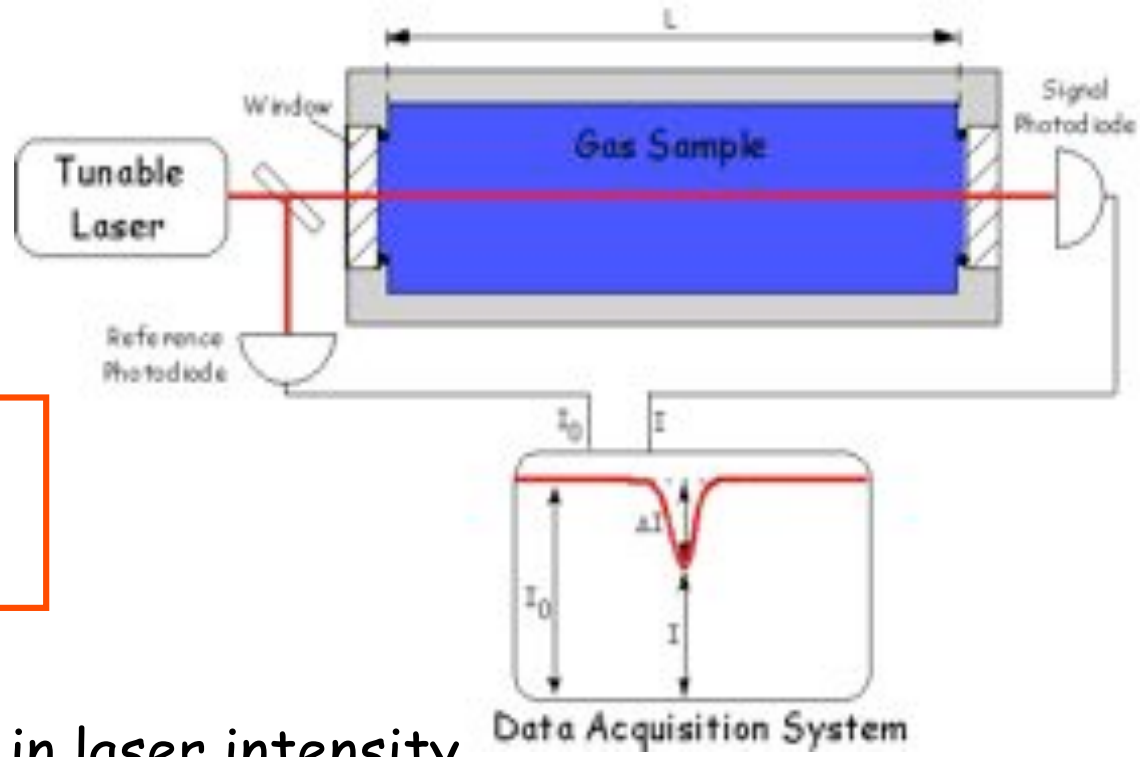
High Resolution Absorption Spectrometry: General Overview



Beer-Lambert Law:

$$\Delta I/I_o = 1-\exp(-\alpha L_{eff})$$

$\Delta I/I_o$	=	fractional change in laser intensity
S	=	absorption line strength
χ	=	mixing ratio (mole fraction)
P	=	total pressure
L_{eff}	=	effective optical path length
$\alpha(\lambda)$	=	absorption coefficient, $S\ P\ \chi\ \phi(\lambda)$

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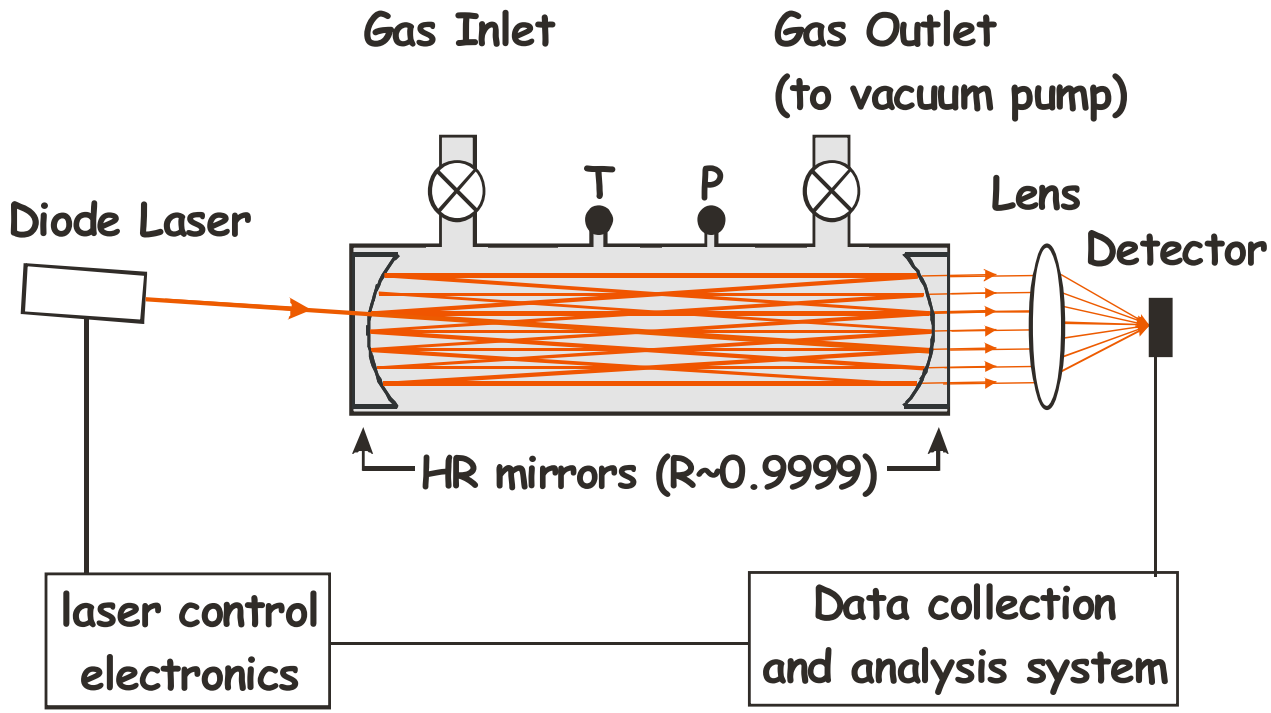
Off-Axis ICOS (2001): Cavity-Enhanced Absorption Technique



- Optical cavity provides pathlength enhancement: $L_{eff} = L / (1-R) = c\,\tau$
- Typical R = 99.995%, L_{eff} = 3-25 kilometers (or greater)
- Extraordinarily robust - alignment insensitive, telecom-grade components
- Allows for near-IR measurements of overtone and combination bands
- Measurements using mid-IR QC lasers provide extremely high sensitivity

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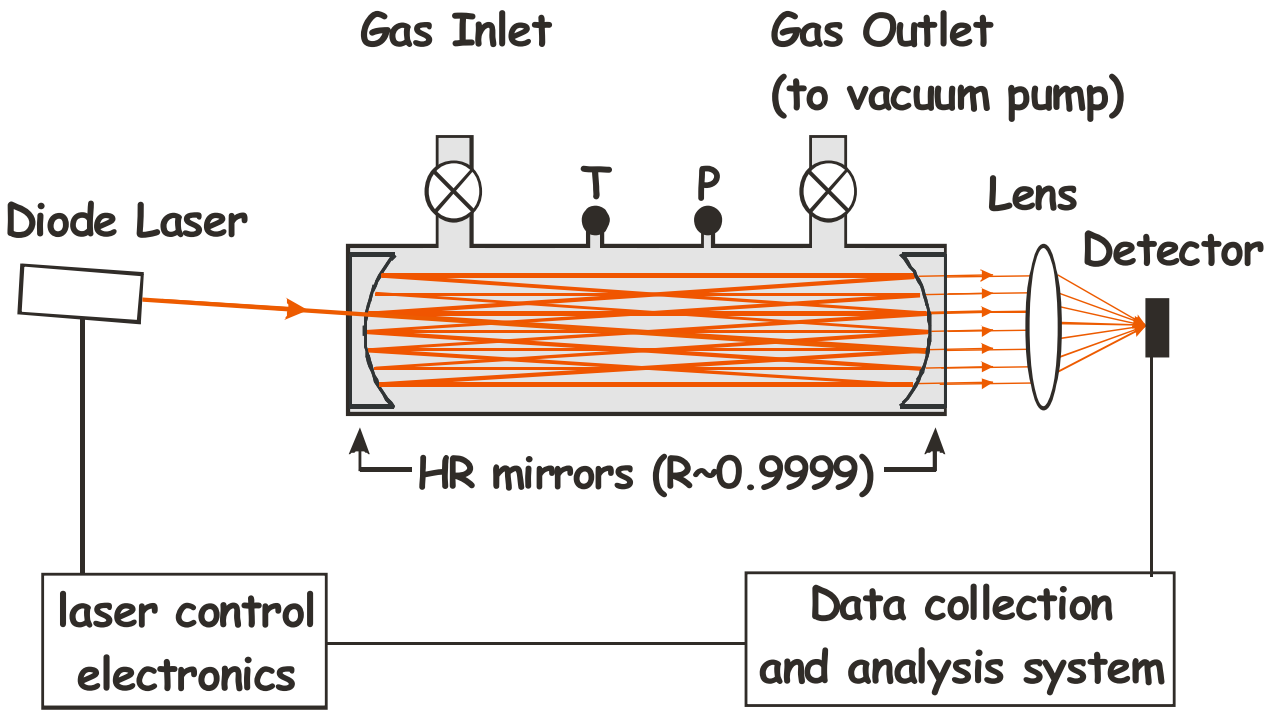
Advantages of Off-Axis ICOS (LGR patent)



- All parameters (absorption, L_{eff}, P, T) determined quickly (300-Hz, typical)
- Robust optical alignment → negligible alignment drift, mechanically stable
- Off-axis alignment spatially separates beam paths through cell
  → lengthens time/distance before beam retraces itself
  → eliminates unwanted resonance interference effects
- Off-axis alignment eliminates optical feedback from cavity to laser source

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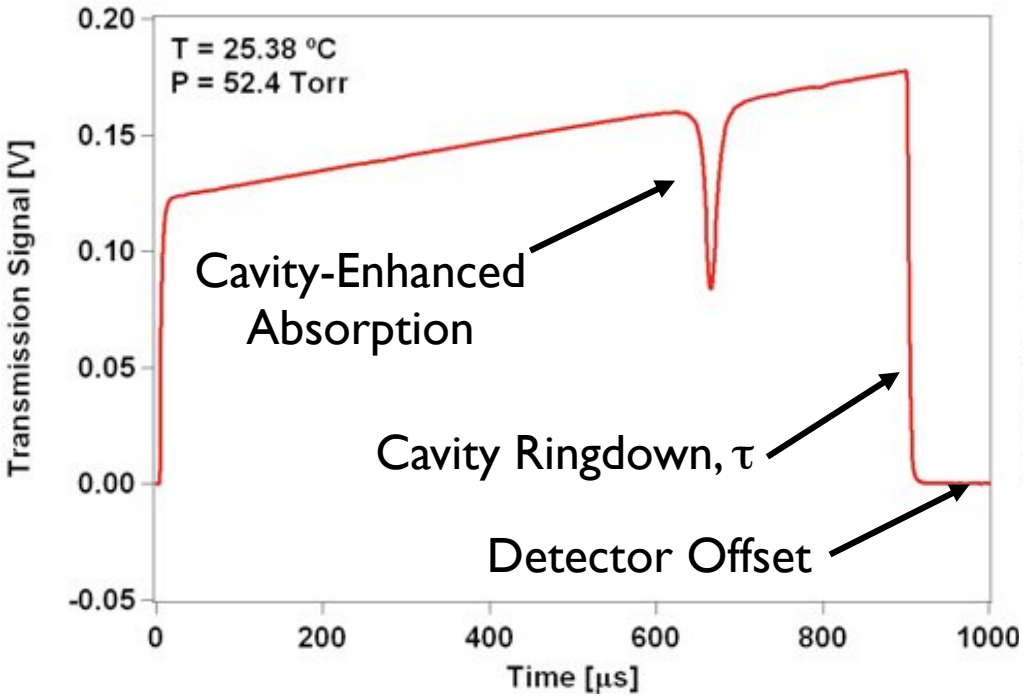
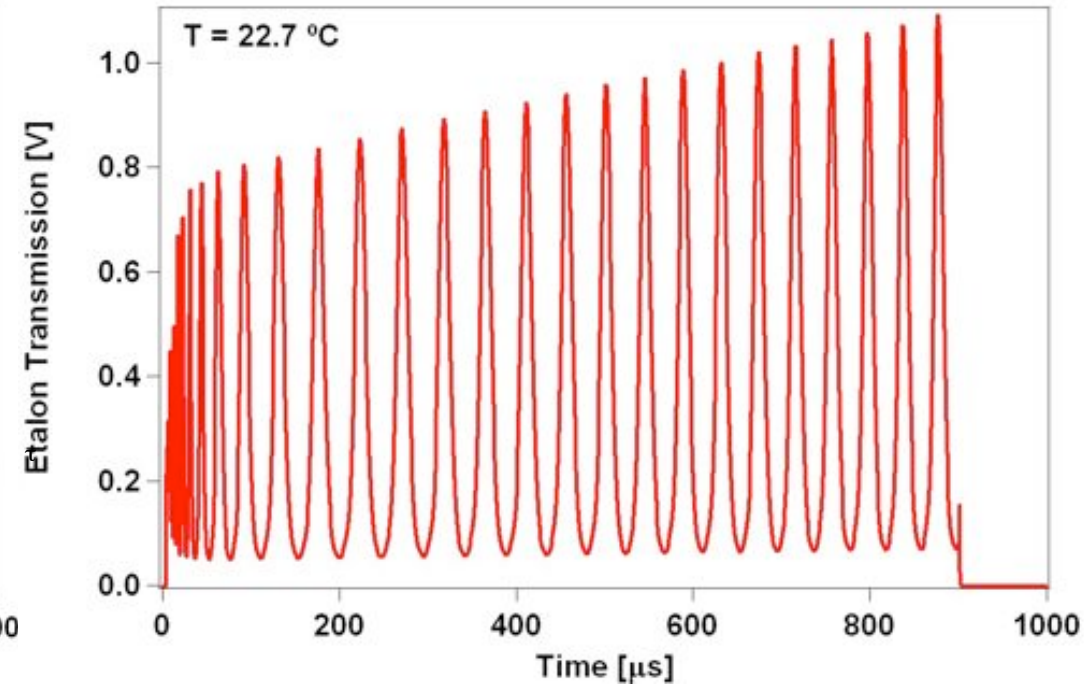
LGR's Off-Axis ICOS: typical raw data trace





Transmission Spectrum
- Measure absorption, baseline, detector offset
- Measured ringdown yields L_{eff}
- Measure/control gas flow parameters (T, P)
- Sweep laser wavelength at 100 – 1200 Hz

Measured Etalon Transmission
- Accurately determines laser tuning rate
- Solid etalon (SiO_2)
- Measure length and temperature
- Converts time to relative laser frequency

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Instrumentation Packaging: Options

rackmount (19” wide, 5U)

benchtop

- Portable rugged package allows field operation
- On-board computer provides real-time data analysis & storage
- 90-200 Watts, 27 kg
- Simple user interface; analog, digital (RS232), Ethernet outputs
- Fully autonomous operation
- Remote access via internet

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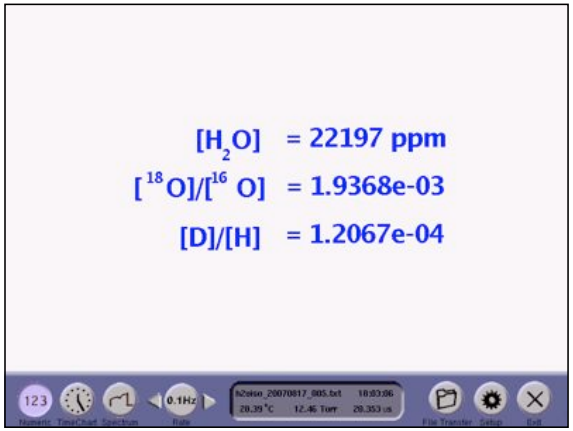
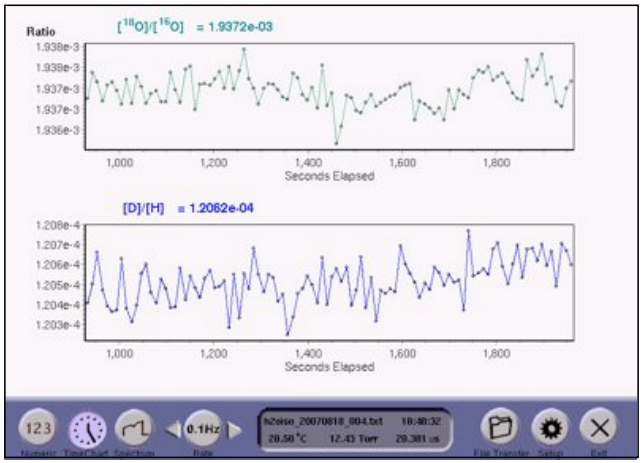
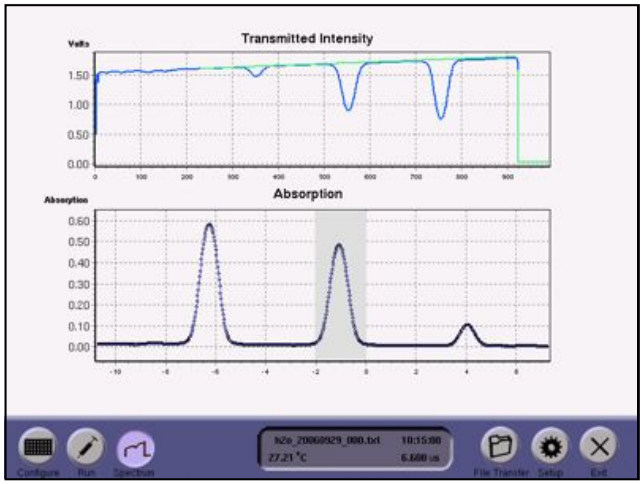
User Interface: Multiple Real-Time Displays



numeric

time chart





raw data / spectra

- Multiple display options: numeric, time chart, raw data/spectra
- Measurements of mixing ratio, gas temperature, pressure
- User-selectable data rate (0.01-20 Hz)
- Remote monitoring/control via internet

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Analyzer Options

- Multiport Inlet Unit: automatic sampling from 16 locations

- Manual injection of discrete samples

- Remote monitoring/control

- Dynamic dilution system (quantification of high concentrations)

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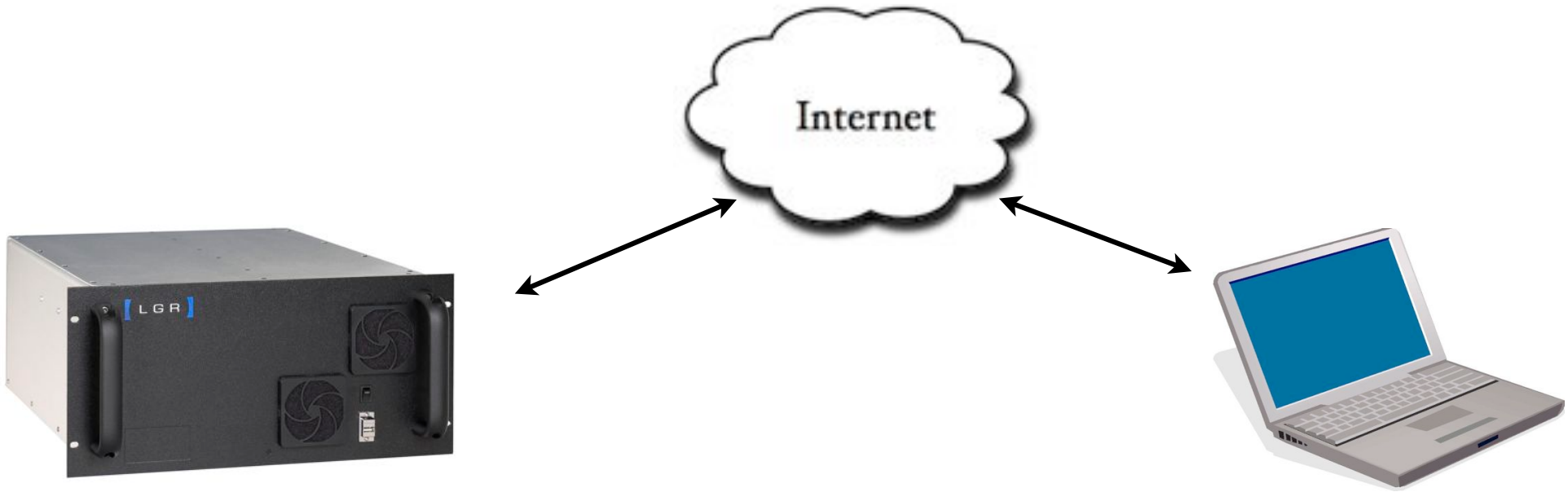
Remote Access and Control

- IP-KVM provides video, keyboard, mouse signals over Ethernet
- IP-KVM connects to LGR's power plane allowing BIOS control
- Allows remote access and control of LGR Analyzers (past and present)
- ~$1k (*includes* separate external computer)



- May be applied with all LGR Analyzers: past and present

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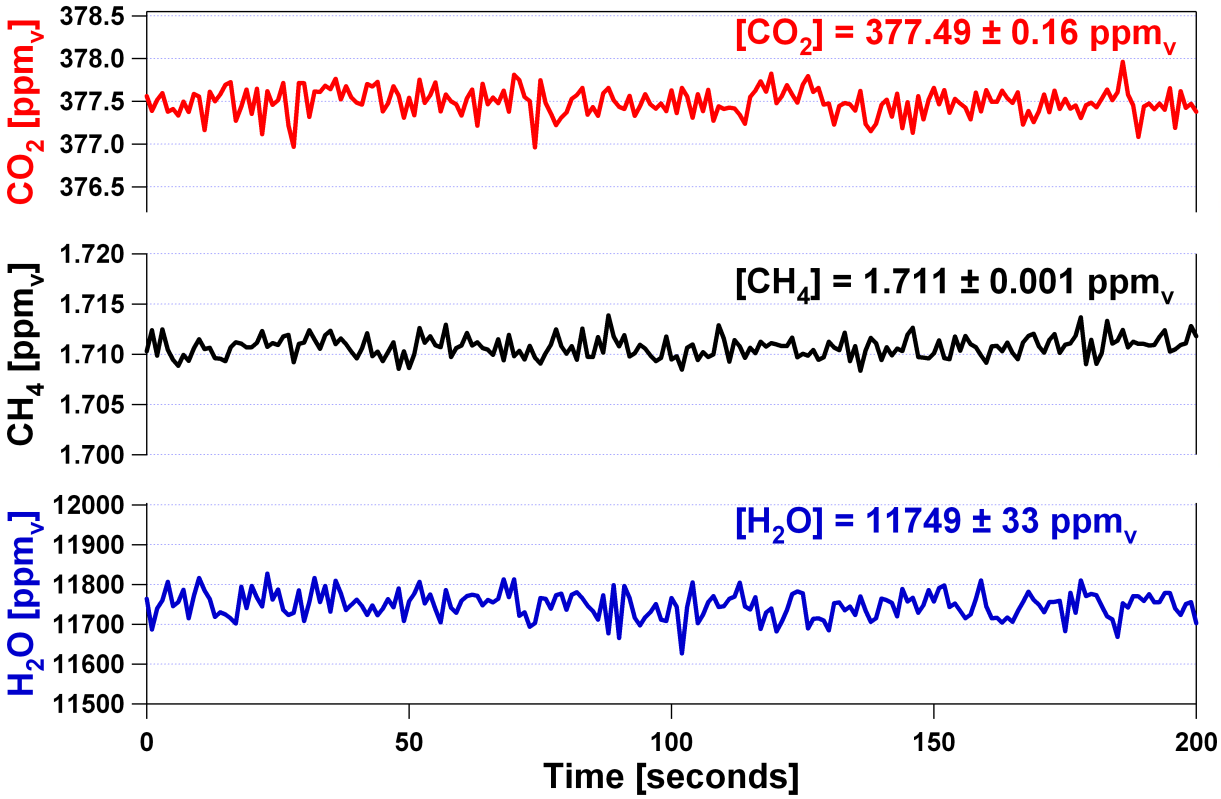
Fast Greenhouse Gas Analyzer: 1-Hz raw data



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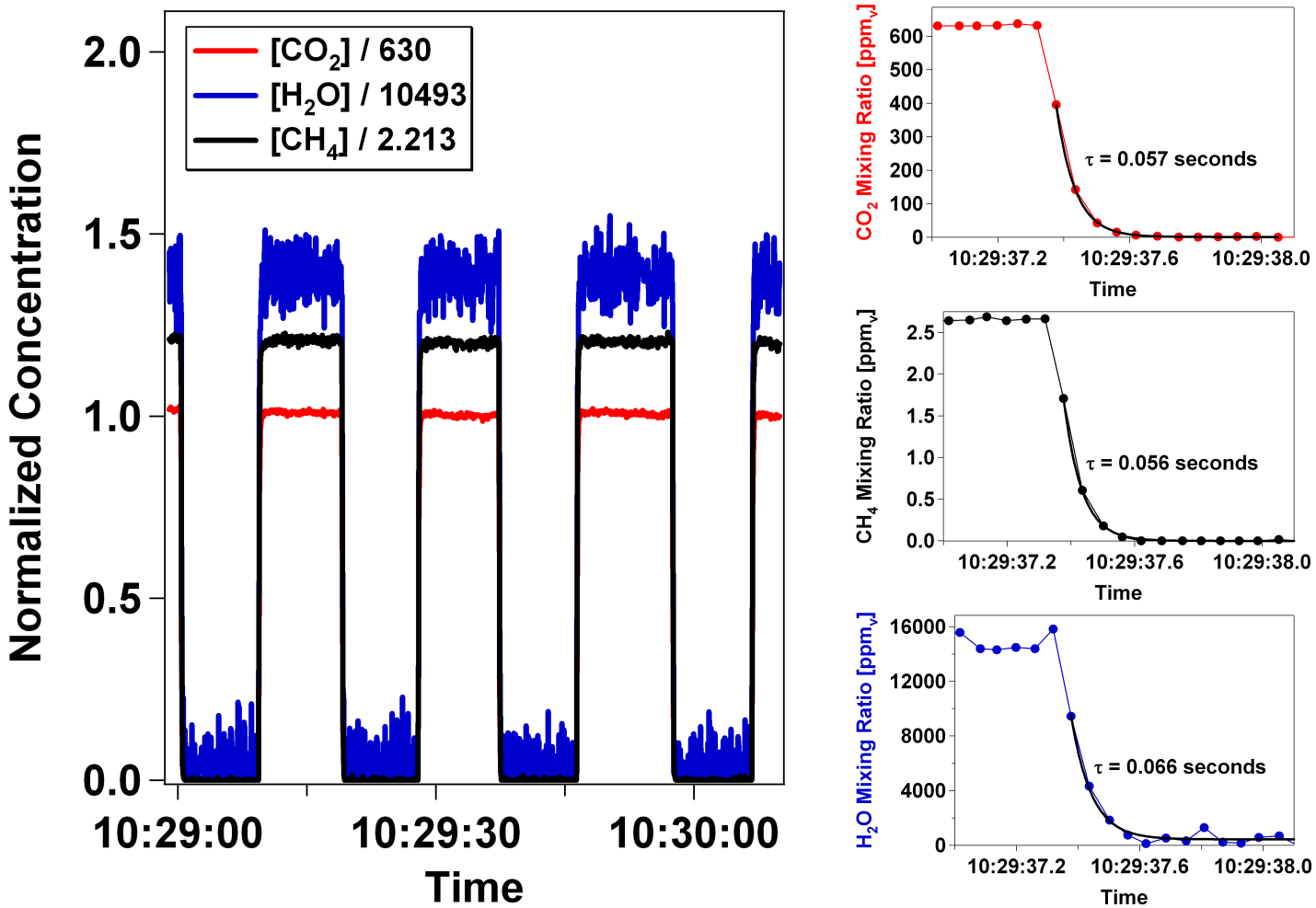
Fast Greenhouse Gas Analyzer: Time Response



- Inlet switched from air to "zero air" to determine flow response
- Concentration decays yield time constants sufficient for eddy flux

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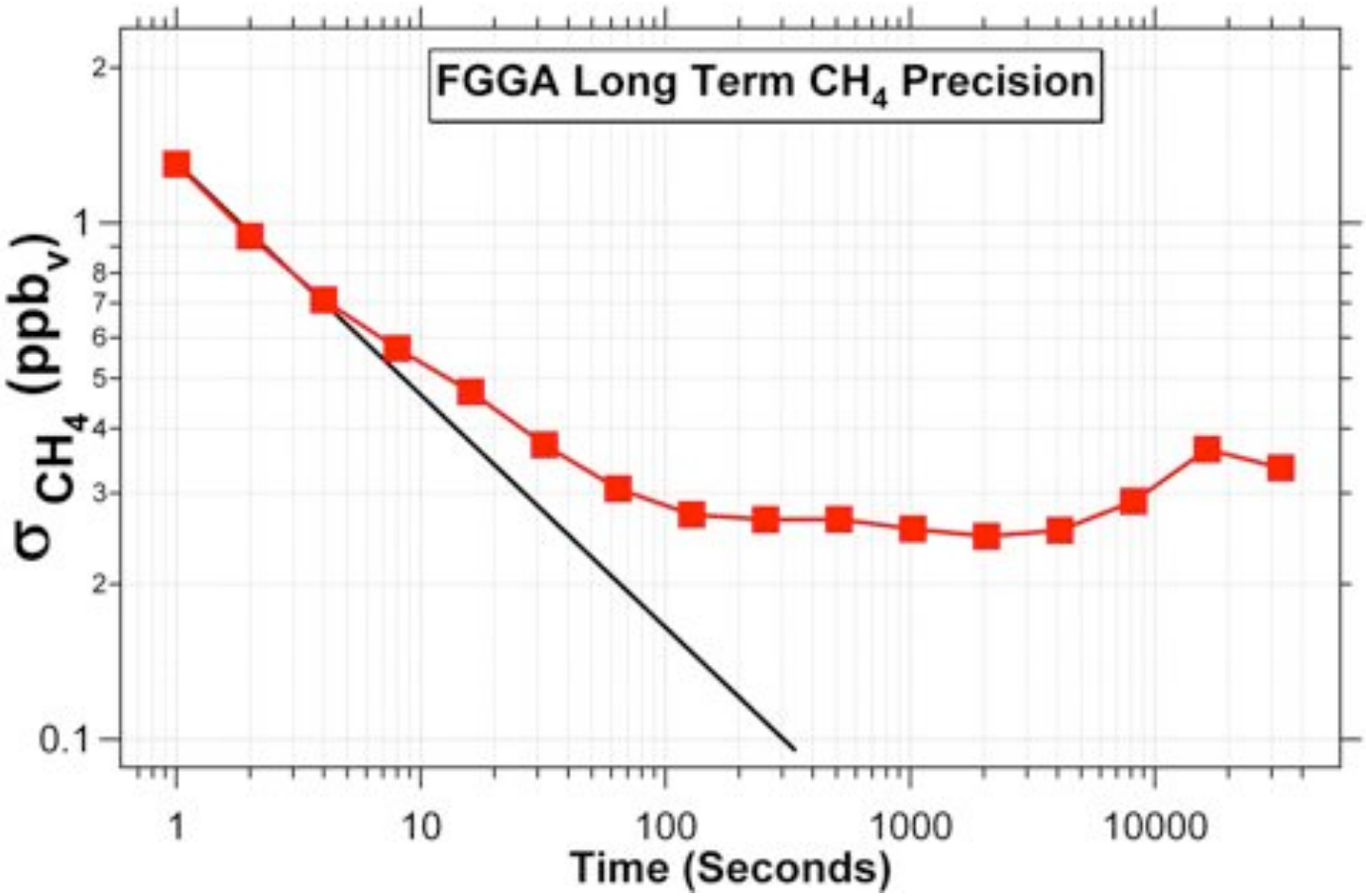
Fast Greenhouse Gas Analyzer: Stability



- CH_4 and CO_2 in air measured at a 1-Hz sampling rate (10-Hz possible)
- Data and associated Allan plots show stable operation and ability to reliably average over long times to improve precision

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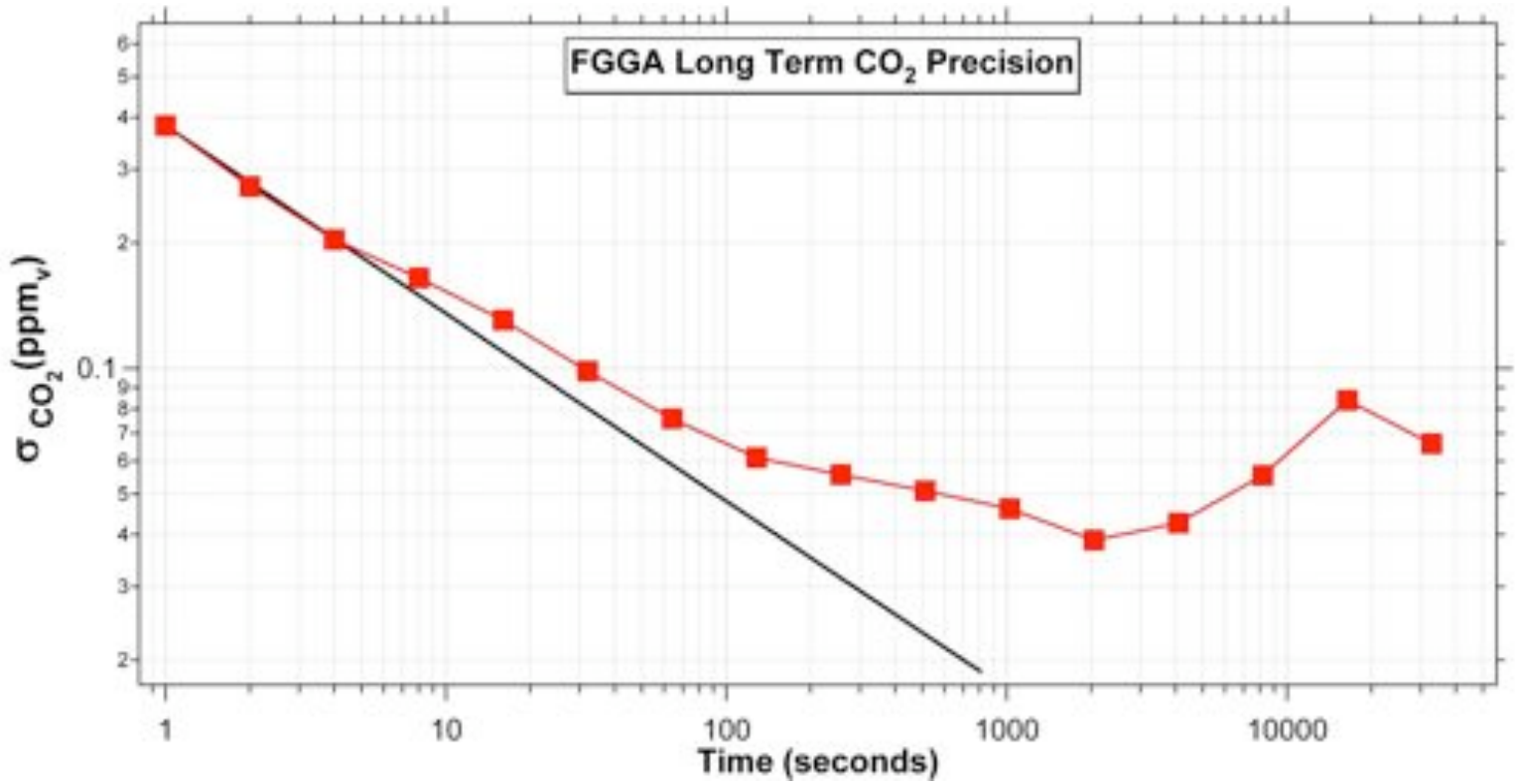
Fast Greenhouse Gas Analyzer: Stability



- CH_4 and CO_2 in air measured at a 1-Hz sampling rate (10-Hz possible)
- Data and associated Allan plots show stable operation and ability to reliably average over long times to improve precision.

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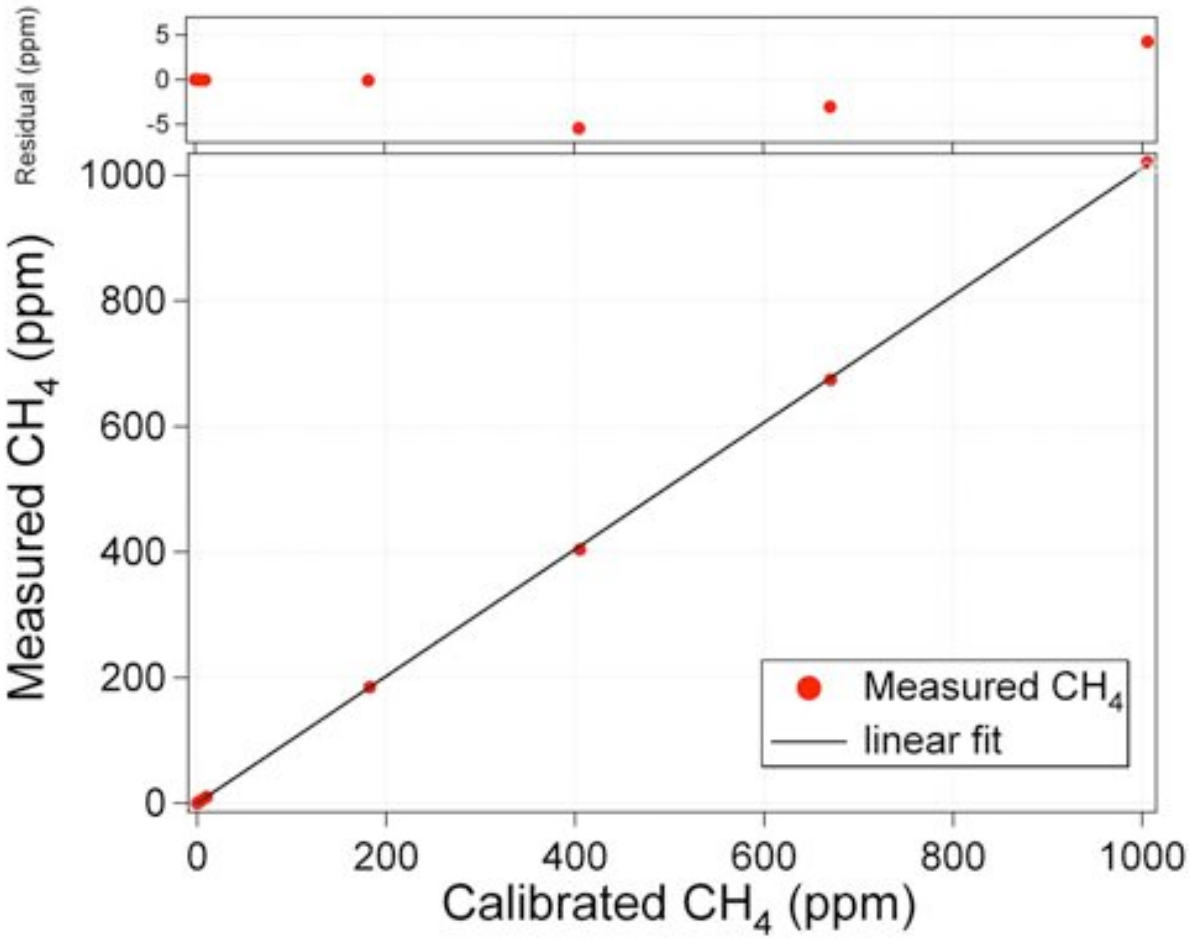
Accurate CH_4 measurements from 0.01 to 1000 ppmv



- Measurements agree with ref values to better than 1% (up to 1000 ppmv)
- High CH_4 levels recorded in field (rice, peat, landfills)
- Off-Axis ICOS reports mixing ratios with extremely high optical depth

Approved for public release; distribution is unlimited.

Fast, Accurate N_2O/CO Analyzer for Flux

- Real-time continuous measurements of CO and N_2O
- Extremely wide dynamic range
- Fast (20 Hz) allows eddy covariance flux
- Low power (200 watts) facilitates field operation
- No sample prep - direct measurements in air

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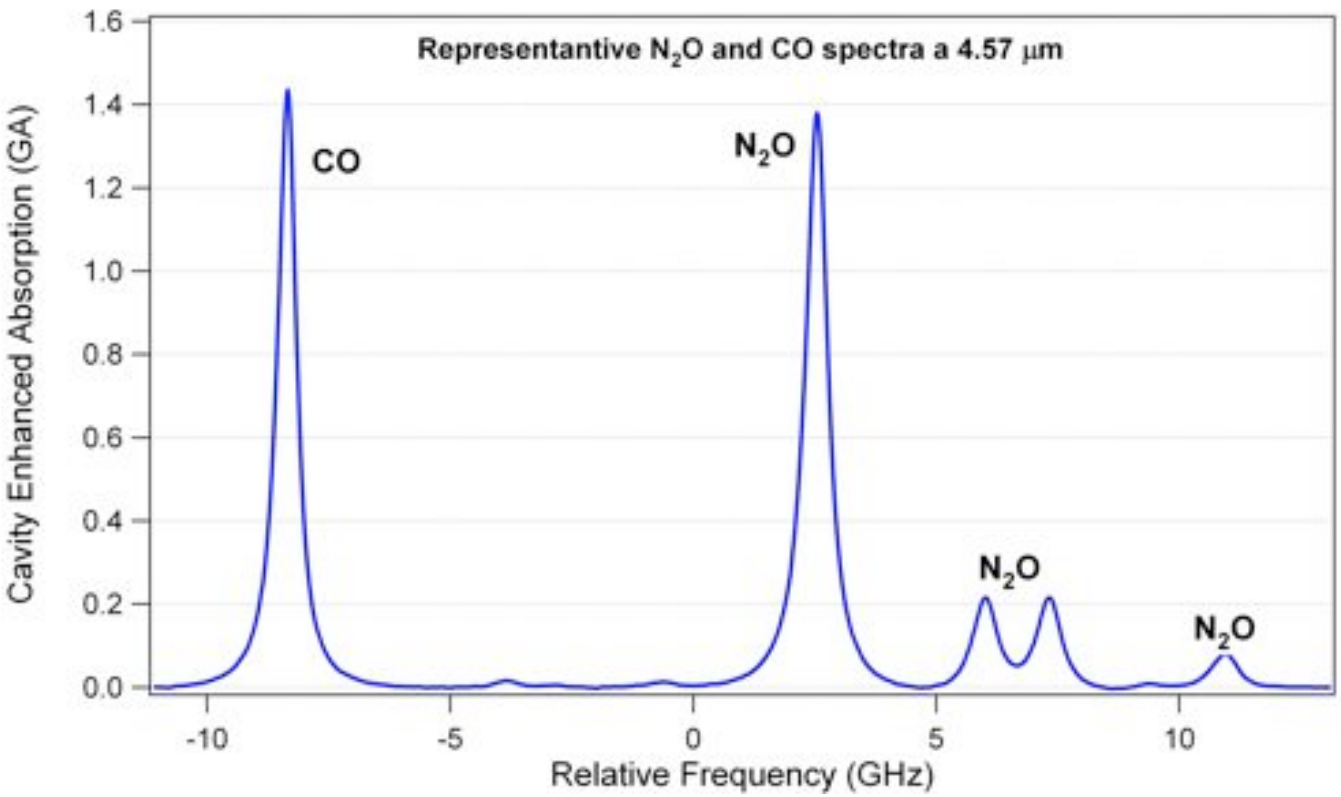
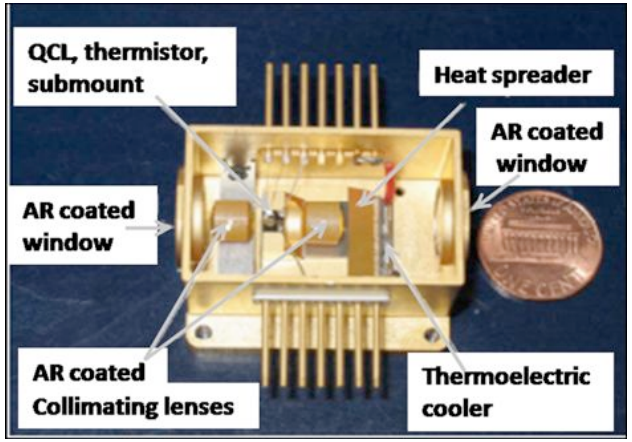
CO + N_2O Analyzer: No cryogenic requirements





- Simultaneous, rapid, accurate measurements of CO and N_2O
- 0.3-ppbv precision in < 1 second (or better)

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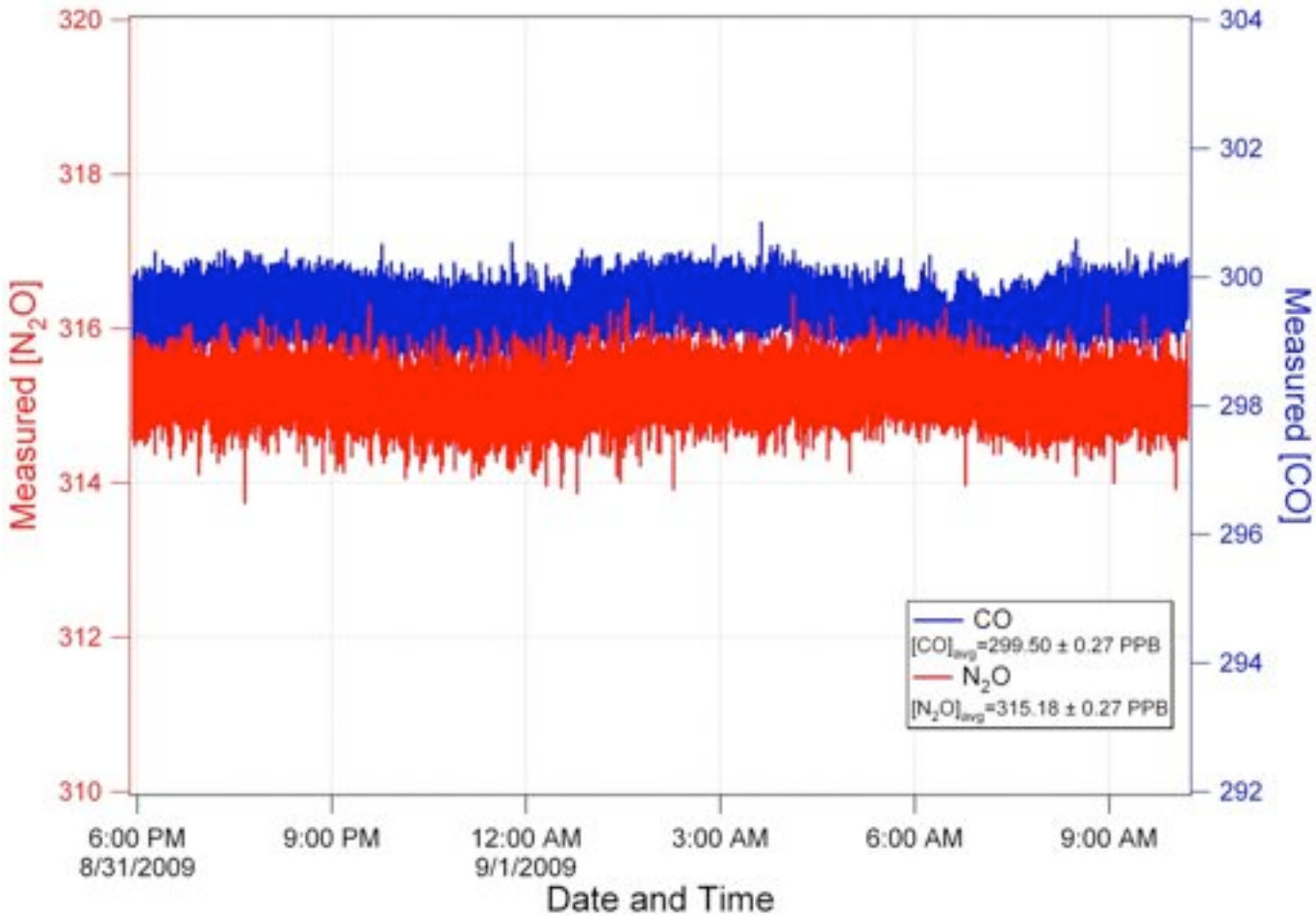
CO + N_2O Analyzer: real-time measurements in air



- Simultaneous measurements of CO and N_2O
- CO precision: < 0.3 ppbv in 1 second
- N_2O precision: < 0.3 ppbv in 1 second

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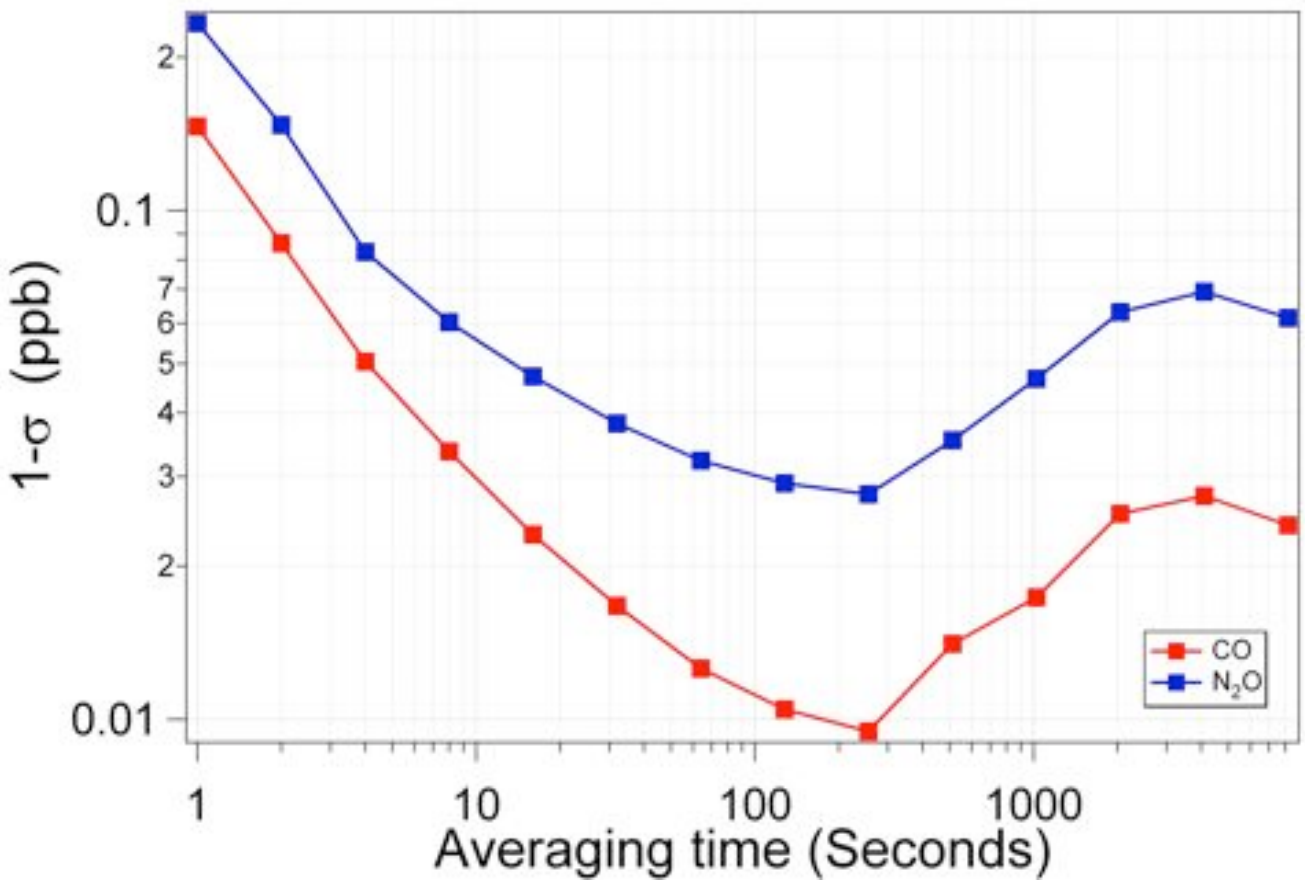
CO + N_2O Analyzer: Long-Term Performance

- Measurement precision vs integration time for dry air measurements



- Precision (1 second): 0.21 ppbv for N_2O, 0.14 ppbv for CO
- Long term precision: 0.070 ppbv for both gases (without calibration)

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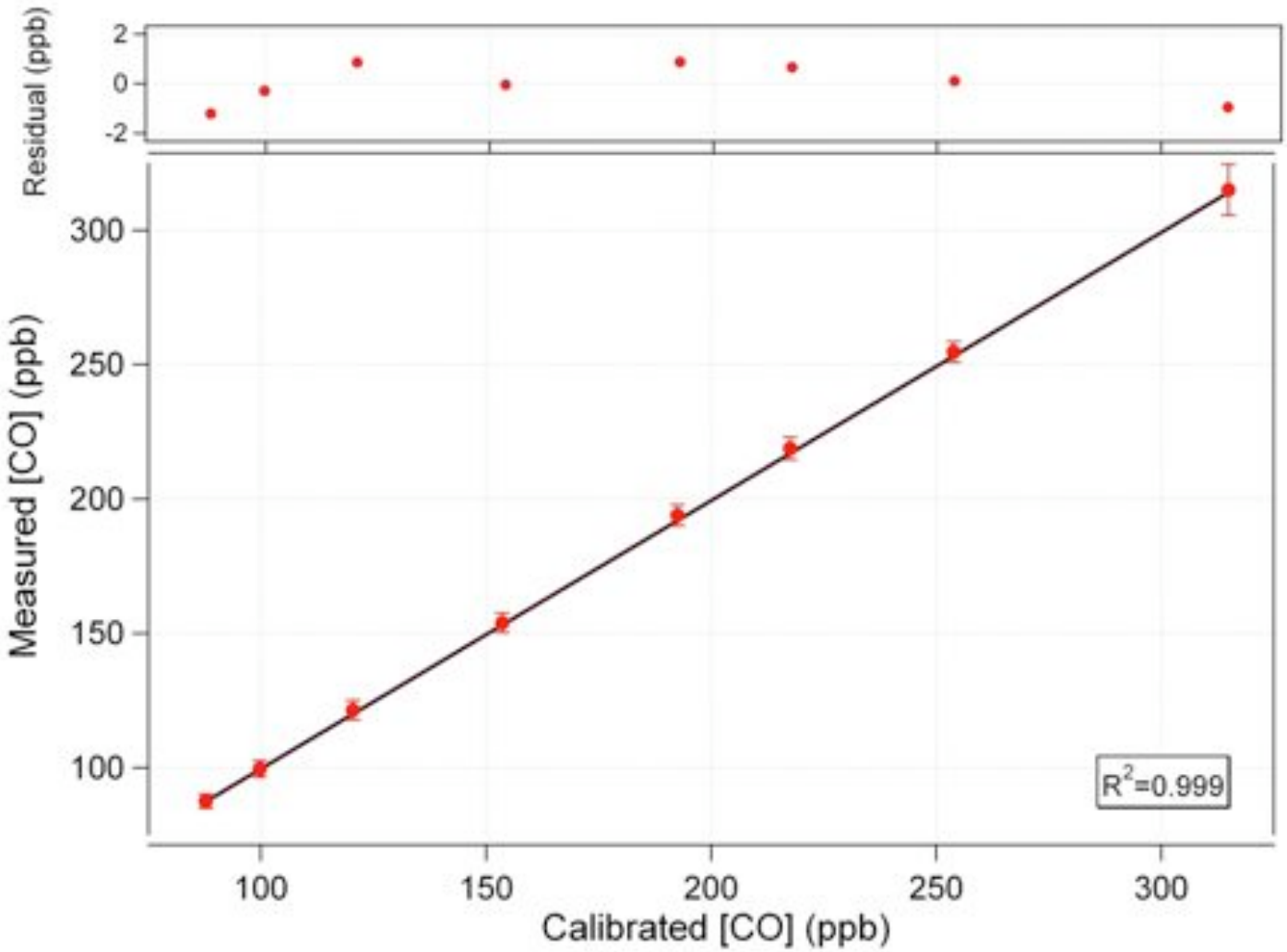
Fast CO + N_2O Analyzer: CO Accuracy



- Linear measurements over wide range
- Agree with mixture values (to within tank uncertainty)

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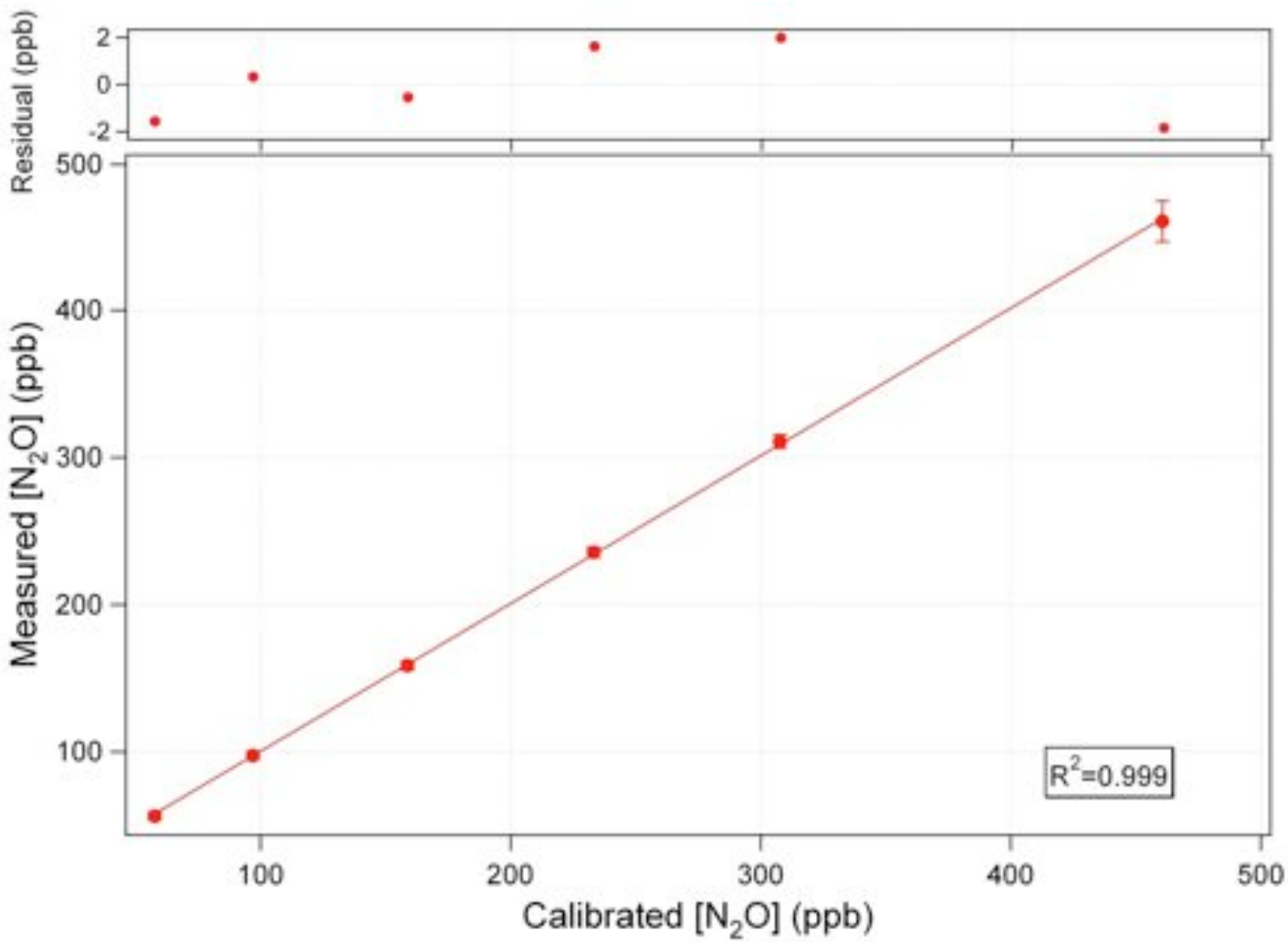
Fast CO + N_2O Analyzer: N_2O Accuracy



- <u>Linear</u> measurements over wide range
- Agree with mixture values (to within tank uncertainty)

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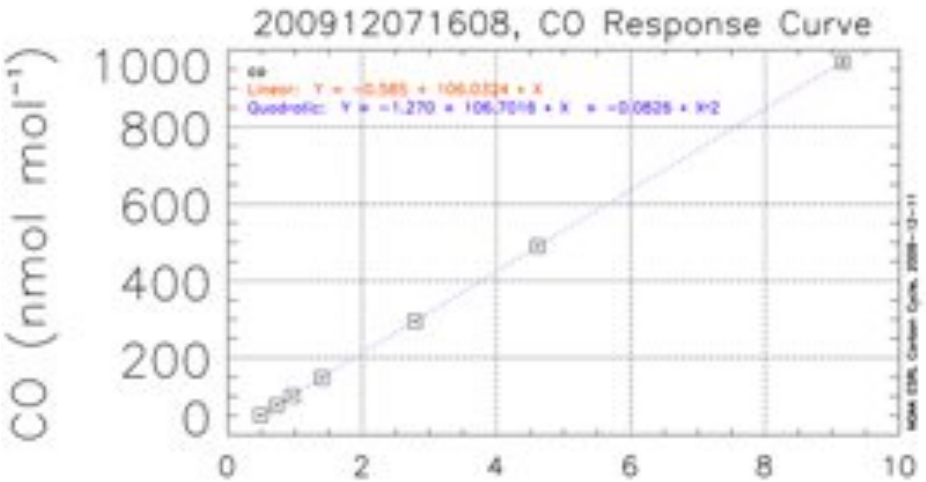
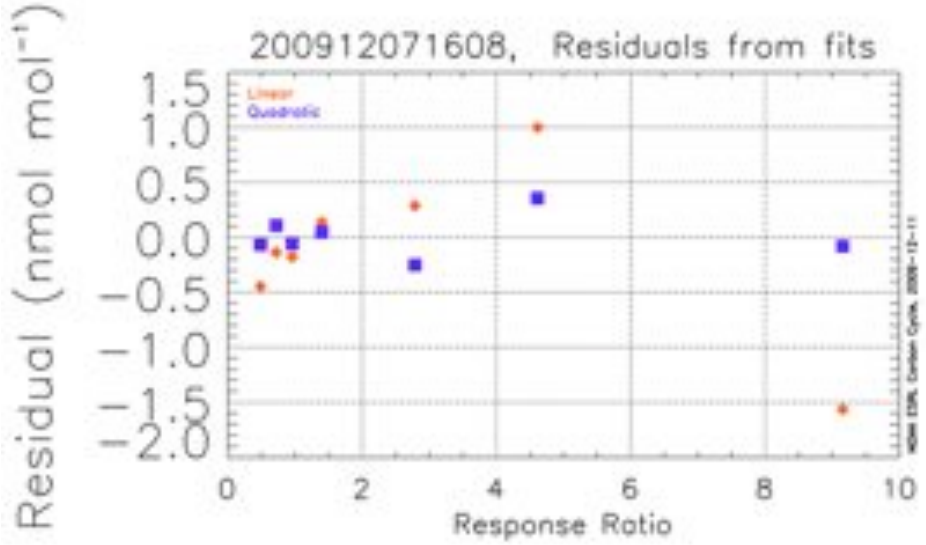
CO + N_2O Analyzer: High Accuracy





Measurements at NOAA ESRL
(A. Crotwell, E. Dlugokencky, P. Novelli)

Agreement with NOAA standards:
better than 0.5 ppbv over 50-1000 ppbv

- Linearity measured using multiple NOAA air cylinders at NOAA ESRL (Boulder)

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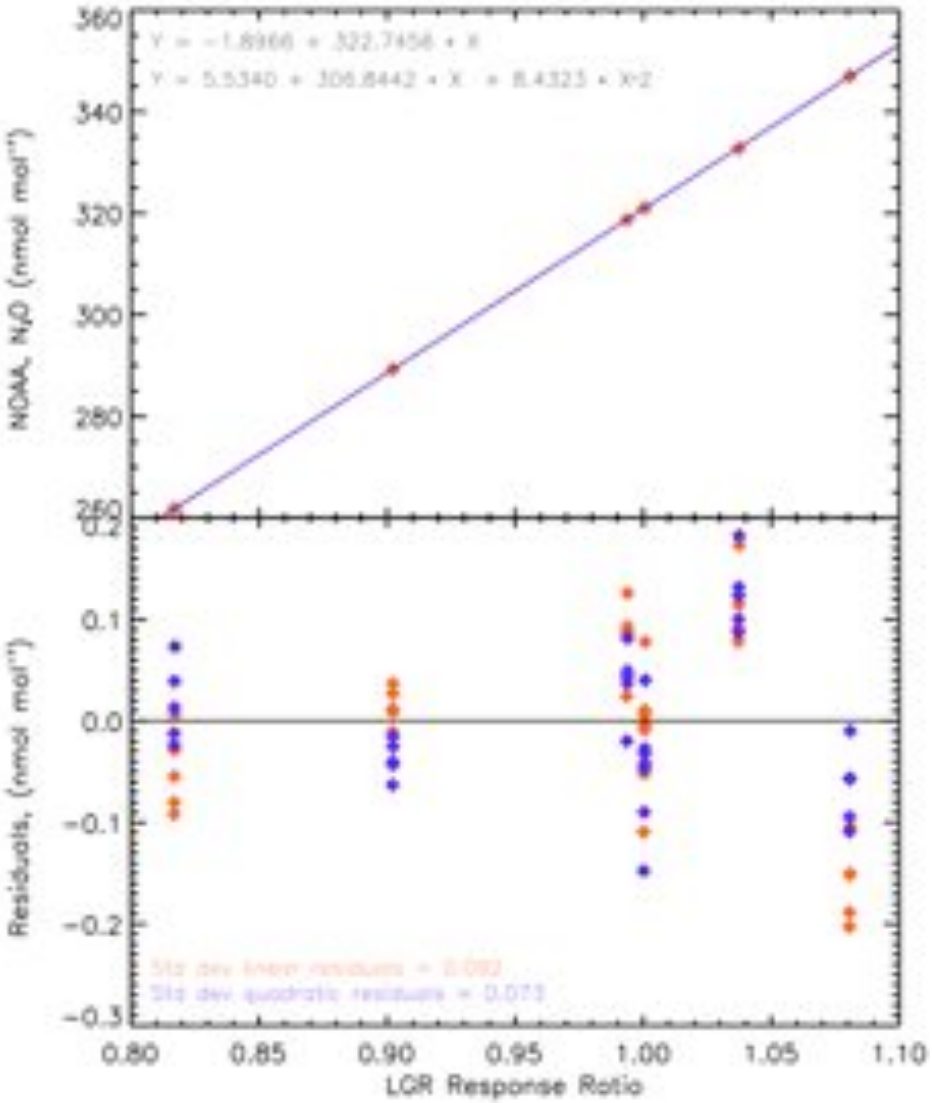
CO + N_2O Analyzer: High Accuracy



Measurements at NOAA ESRL
(A. Crotwell, E. Dlugokencky, P. Novelli)

Agreement with NOAA standards
better than 0.2 ppbv over 260-340 ppbv

- Linearity measured using multiple NOAA air cylinders at NOAA ESRL (Boulder)

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N_2O/CO Analyzer

- Continuous N_2O <u>and</u> CO with precision better than 0.3 ppbv (1 sec)
- Long term precision better than 0.1 ppbv
- Only 200 watts
- Multiport-Inlet Unit allows spatial multiplexing
- Independent verification of performance by NOAA ESRL

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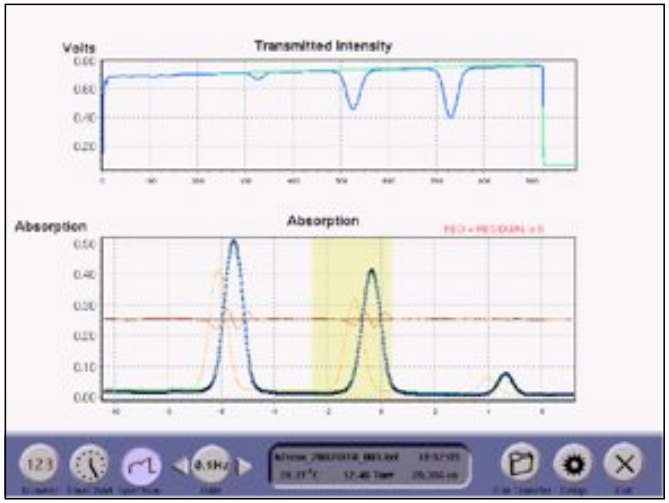
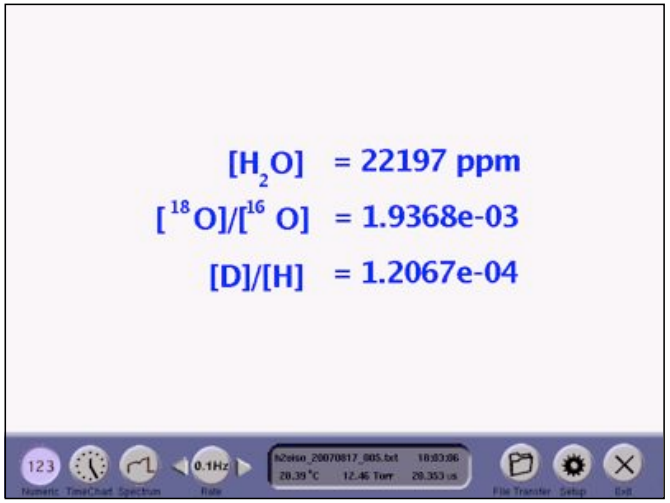
Water Vapor Isotope Flux: δ^2H, $\delta^{18}O$ and H_2O





- Fast, continuous measurements in air (non-condensing)
- Fast (2 Hz) allows (relaxed) eddy covariance flux
- Low power (180 watts) facilitates field operation
- No sample prep - direct measurements in air
- Simple to use

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Water Vapor Isotopic Standard Source (WVISS)

model 908-0004-9001 (accessory to Water Vapor Isotope Analyzer)

- Quantitative evaporation of (isotopic) liquid water reference yields stable air flow with controllable humidity (H_2O: 500-30000 ppmv)
- Automatically provides reference source to WVIA at user-selectable intervals
- Large reservoir provides liquid reference for months under routine operation
- WVIA with WVISS: *dual-inlet operation* - automatic switch between unknown and reference

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WVIA and WVISS provides absolute $\delta^{18}O$ and $\delta^{2}H$ measurements in Vapor and Liquid

Water Vapor Isotope Analyzer (WVIA, 2nd Generation)

- rapidly (>2 Hz) quantifies $\delta^{2}H$ and $\delta^{18}O$ in air
- insensitive to temperature (5-40 ℃)
- 180 watts
- resolves dynamic changes quickly (<0.5 sec) and over long time scales (months)
- unattended continuous measurements in field
- enables studies of ecohydrological processes and atmospheric mixing dynamics

Water Vapor Isotope Standard Source (WVISS)

- provides reference source $\delta^{2}H$, $\delta^{18}O$ for H_2O: 500-30000 ppmv (extended range available upon request)
- demonstrated long-term stability, repeatability
- unattended automatic operation and validation of WVIA

Measurement System (WVIA+WVISS) provides:

- dual inlet mode of operation provides absolute $\delta^{2}H$ and $\delta^{18}O$ measurements in air for H_2O: 500-30000 ppmv
- unattended long-term automatic operation
- opportunity for continuous δ measurements of liquid samples

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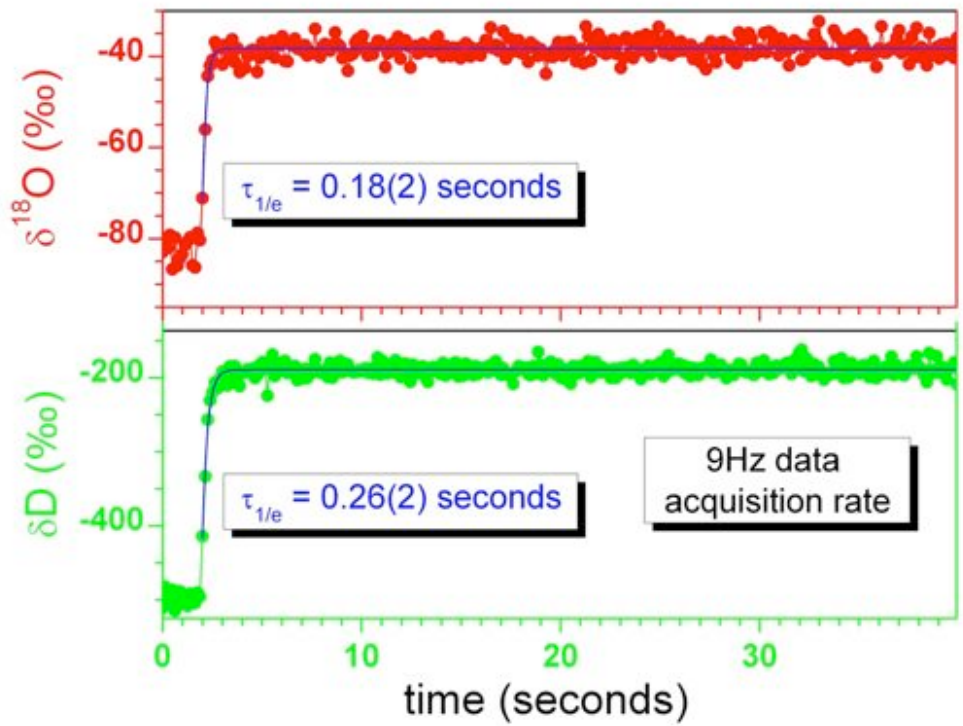
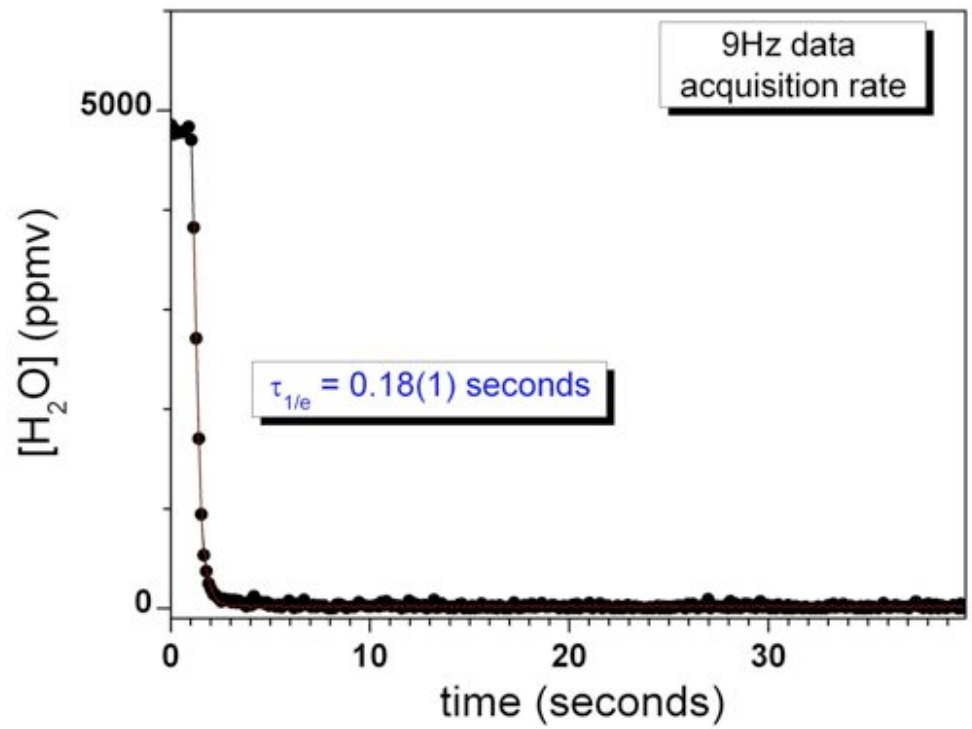
Fast measurement response





- ***5-Hz response*** demonstrated for $\delta^{18}O$ and $[H_2O]$ by switching between samples of different δ and $[H_2O]$
- ***4-Hz response*** demonstrated for $\delta^{2}H$

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Methane Carbon Isotope Analyzer: CH_4 and $\delta^{13}CH_4$

- $\delta^{13}C$ precision: better than 1‰
- CH_4: ambient levels - 100%
- No consumables, no cryogens
- Direct measurements
- Applications: bio-gas, methane sourcing
- Low power: 120 watts

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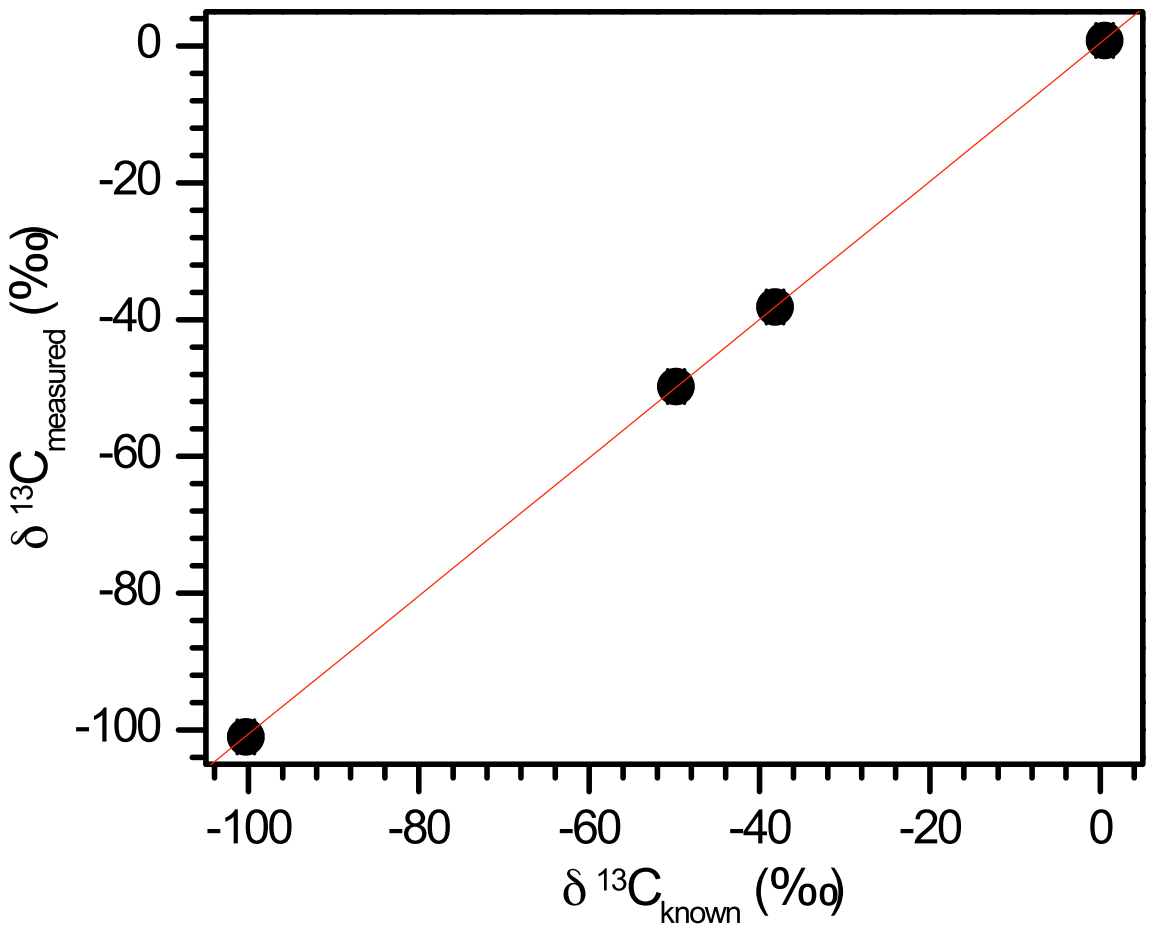
Methane Carbon Isotope Analyzer: Accuracy



- Comparison with IRMS-characterized methane/air gas mixtures
- $\delta^{13}C$ accurate to better than ± 0.2 ‰

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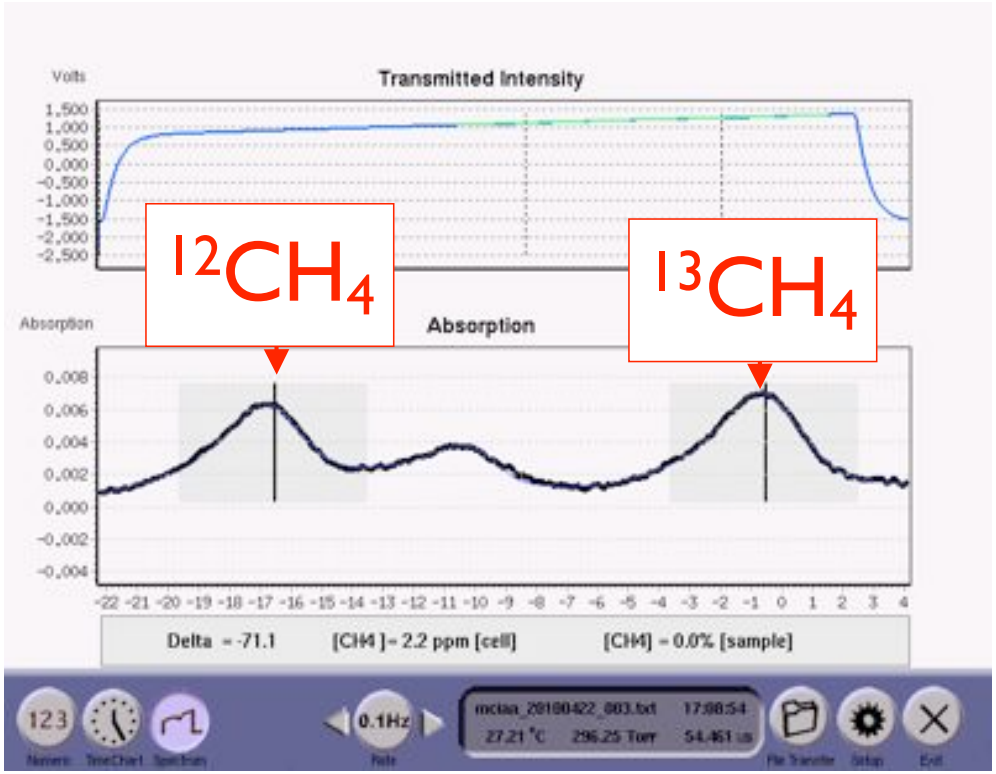
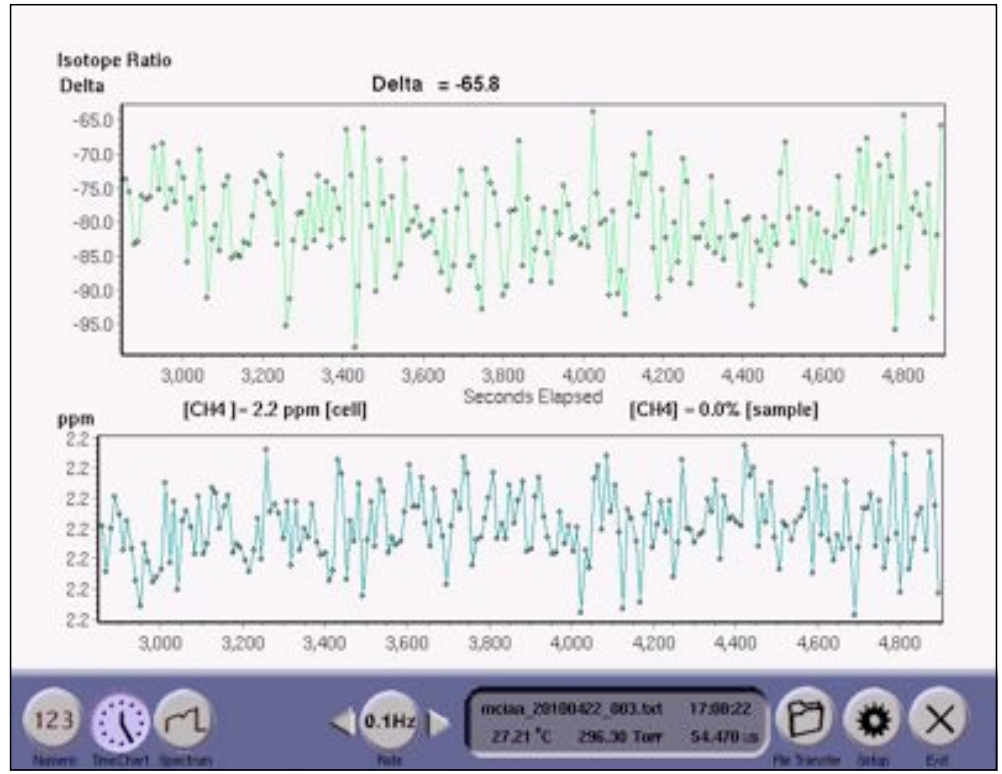
Methane Carbon Isotope Analyzer: ambient air





Raw data absorption spectra: $[CH_4] < 500$ppm

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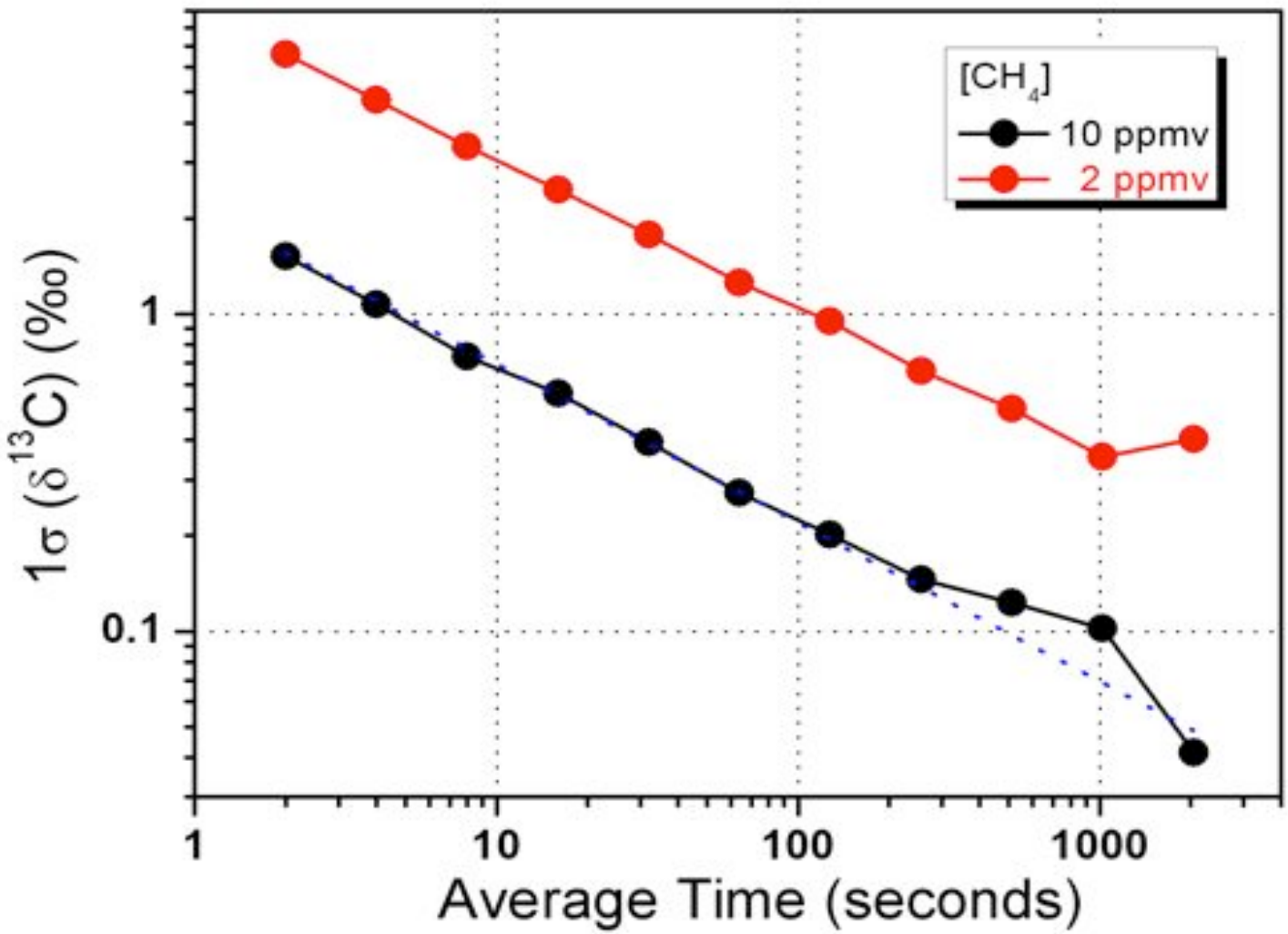
Methane Carbon Isotope Analyzer: precision



- $\delta^{13}C$ precision (2 ppmv): 1 ‰ (100 sec)
- $\delta^{13}C$ precision (10 ppmv): 0.2 ‰ (100 sec)

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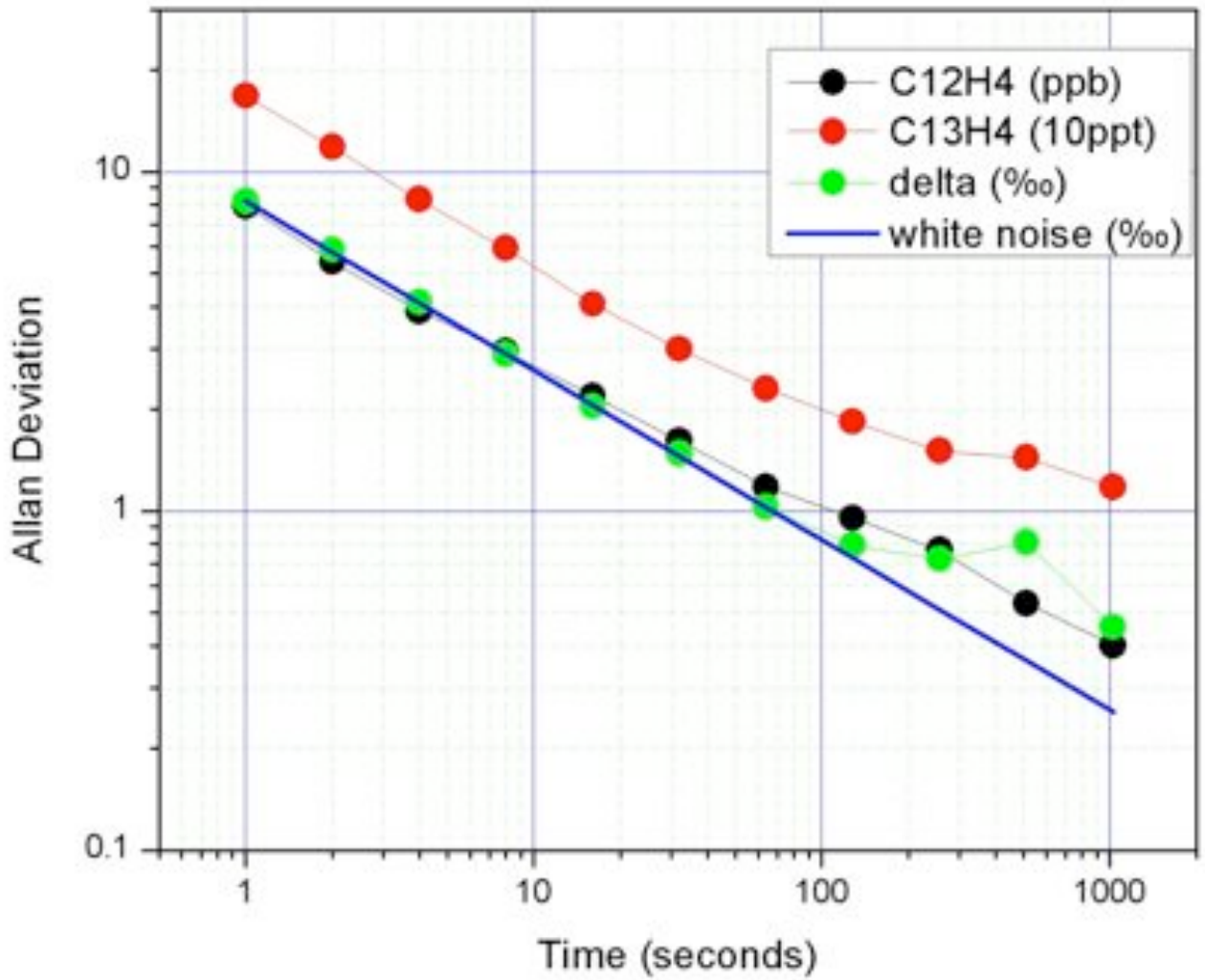
Measurement precision (CH_4 = 2 ppmv)



- $\delta^{13}C$ precision (2 ppmv): < 1 ‰ (100 sec)
- $\delta^{13}C$ precision (2 ppmv): < 0.5 ‰ (100 sec)

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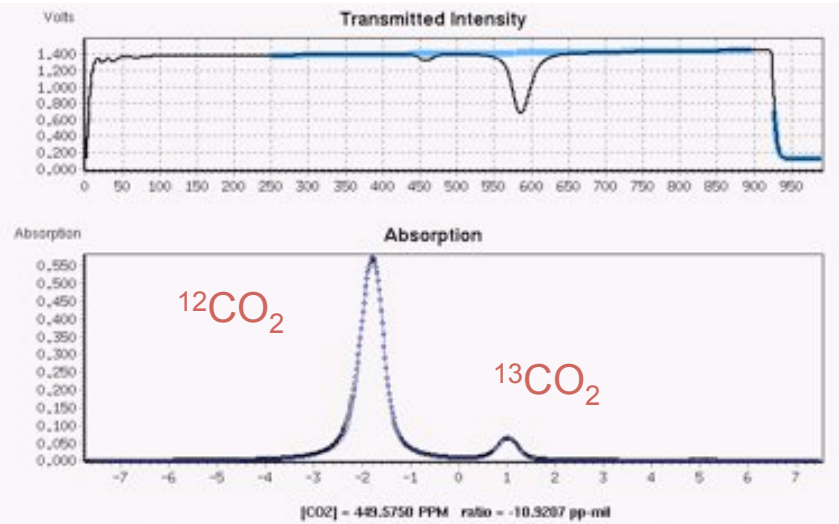
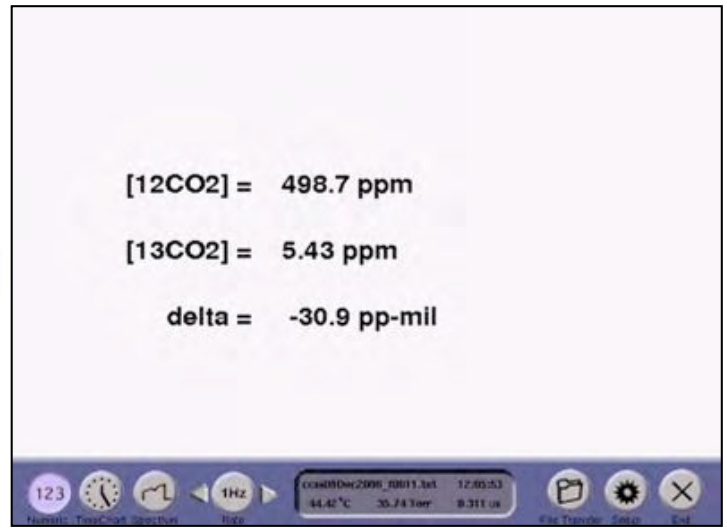
Carbon Dioxide Isotope Flux: $\delta^{13}C$ and CO_2





- Continuous measurements of ($^{13}CO_2/^{12}CO_2$) $\delta^{13}CO_2$ and CO_2
- Fast (2 Hz) allows eddy covariance flux
- Low power (150 watts) facilitates field operation
- No sample prep - direct measurements in air
- Precise: $\delta^{13}CO_2 < 0.2$ per mil (60 seconds)

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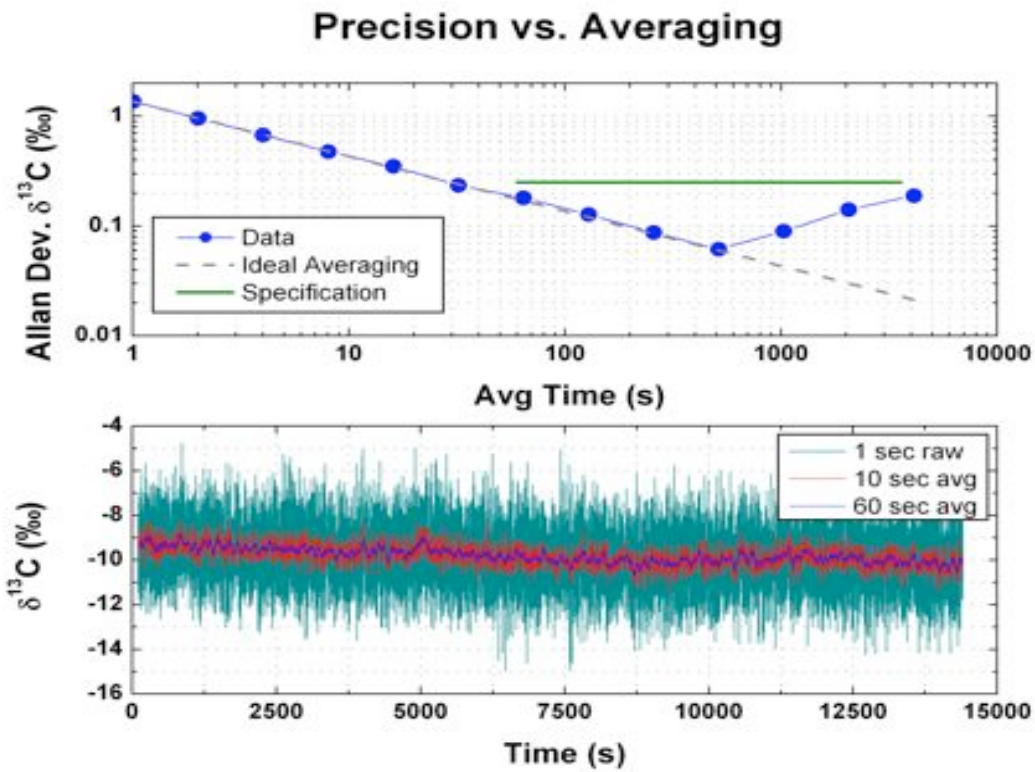
Carbon Dioxide Isotope Flux: $\delta^{13}C$ and CO_2



- Continuous $\delta^{13}CO_2$ and CO_2
- Fast (1 Hz) measurements provide rapid response
- Low power (150 watts) facilitates field operation
- No sample prep - direct measurements in air
- Precise: $\delta^{13}CO_2 < 0.2$ per mil (60 seconds)

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Field Deployment: CO_2 Isotope Analyzer at ZERT (Carbon Sequestration Study)

Zero Emissions Research and Technology (ZERT) test site
Bozeman, Montana (July 2009)

Month-long release of CO_2 (buried pipe) for testing various schemes of leak detection in carbon sequestering

- Quantify $\delta^{13}C$ signature of CO_2 release
- Measure $\delta^{13}C$ from ambient sources (plant respiration)
- Record spatial profile transverse to pipe
- Record temporal variations at several locations
- Compare co-located measurements with other groups

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ZERT: 0.2 tons/day release of CO_2

Month-long release of CO_2 (buried pipe) for testing various schemes of leak detection in carbon sequestering

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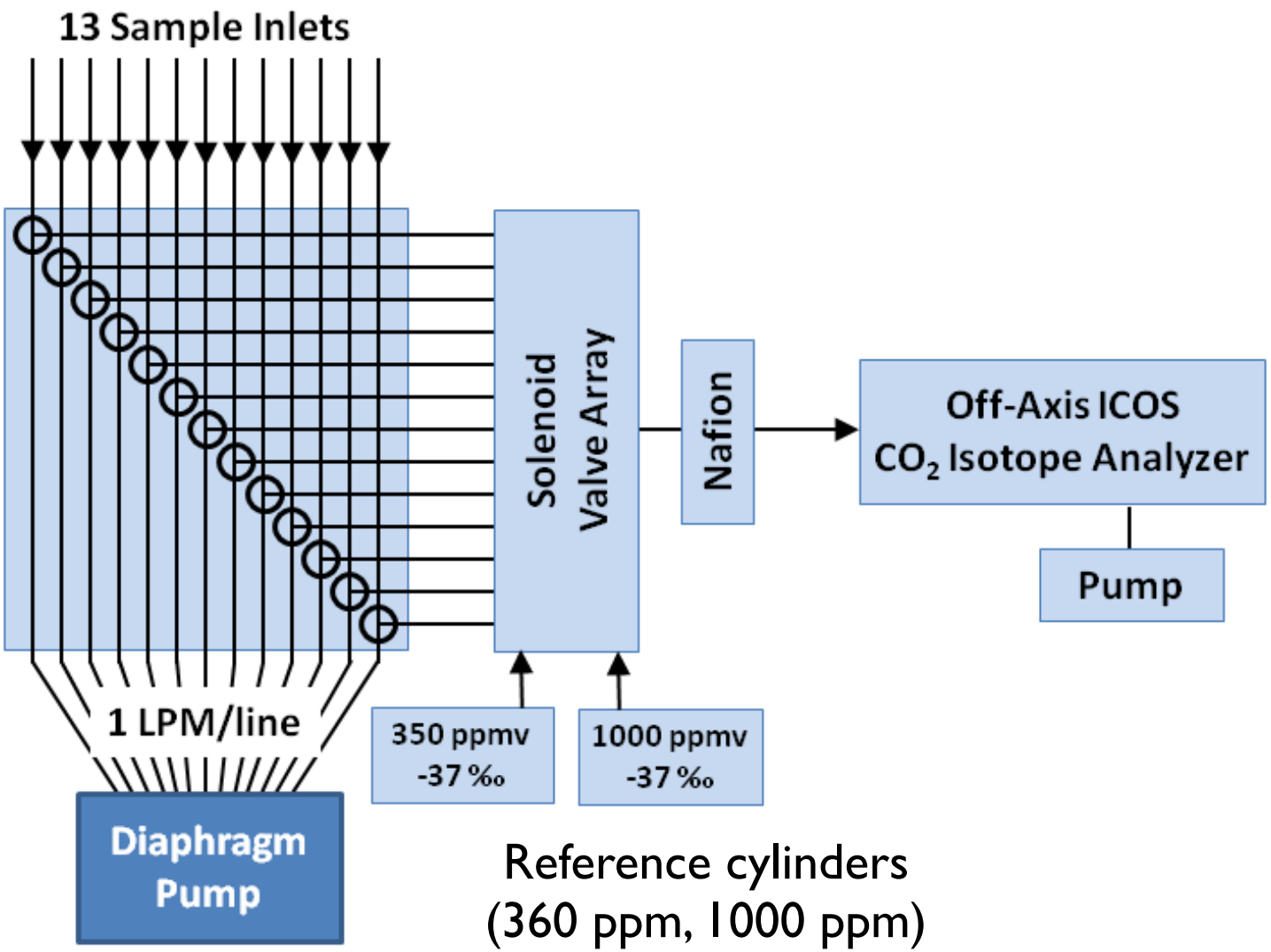
Multi-location continuous $\delta^{13}CO_2$ and CO_2



Reference cylinders
(360 ppm, 1000 ppm)

second pump provides fast flow through all sampling lines

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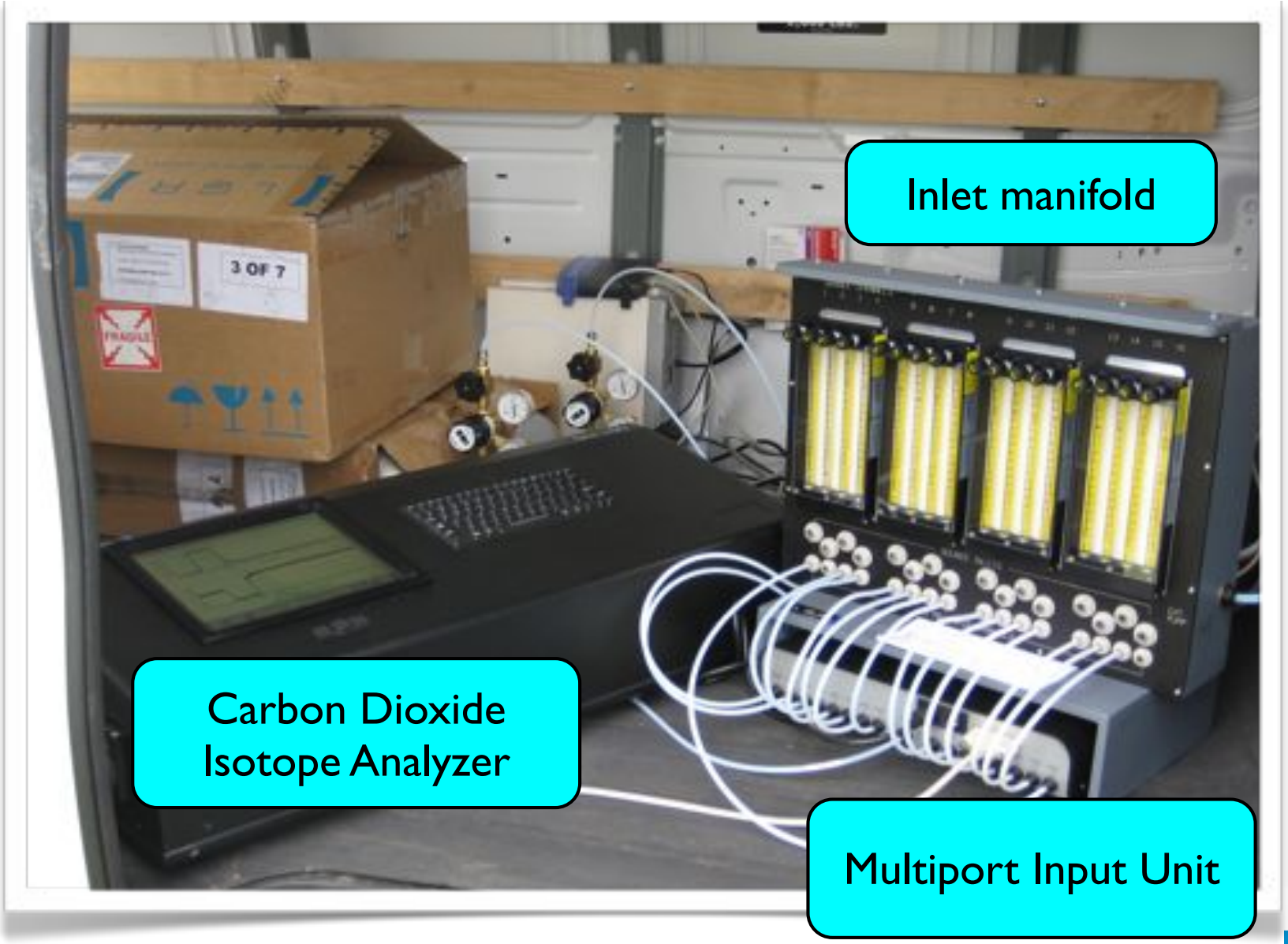
Carbon Dioxide Isotope Analyzer w/ Multiport Input Unit (16 valve array)



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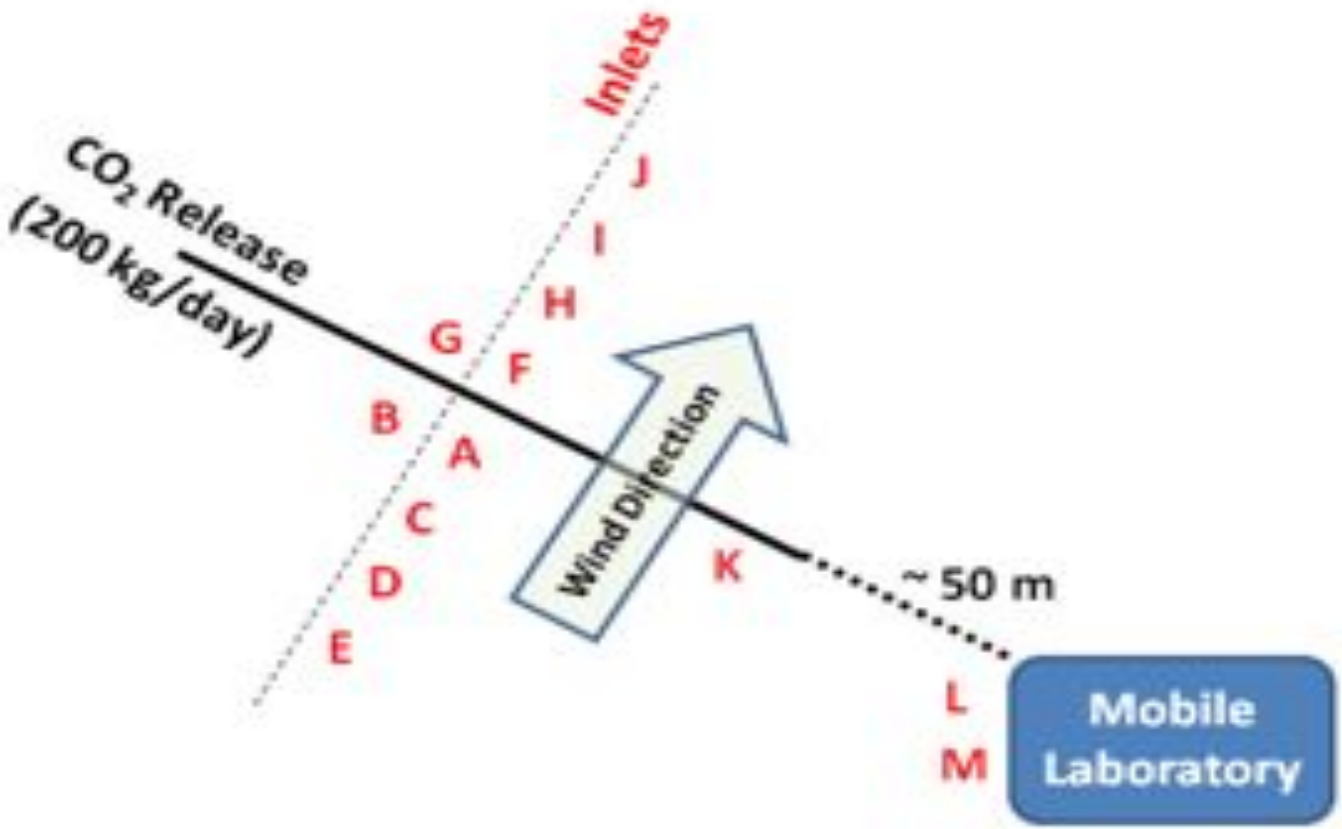
Layout of Sample Inlets: Grid, Tower, Controls



- Inlets A, C, D, E, F, H, I, J spaced by 0.5 m intervals
- Inlets B, G are 30 cm above A, F
- Inlet K is located with tower shared by other teams
- Inlets L, M are controls located 50 m away

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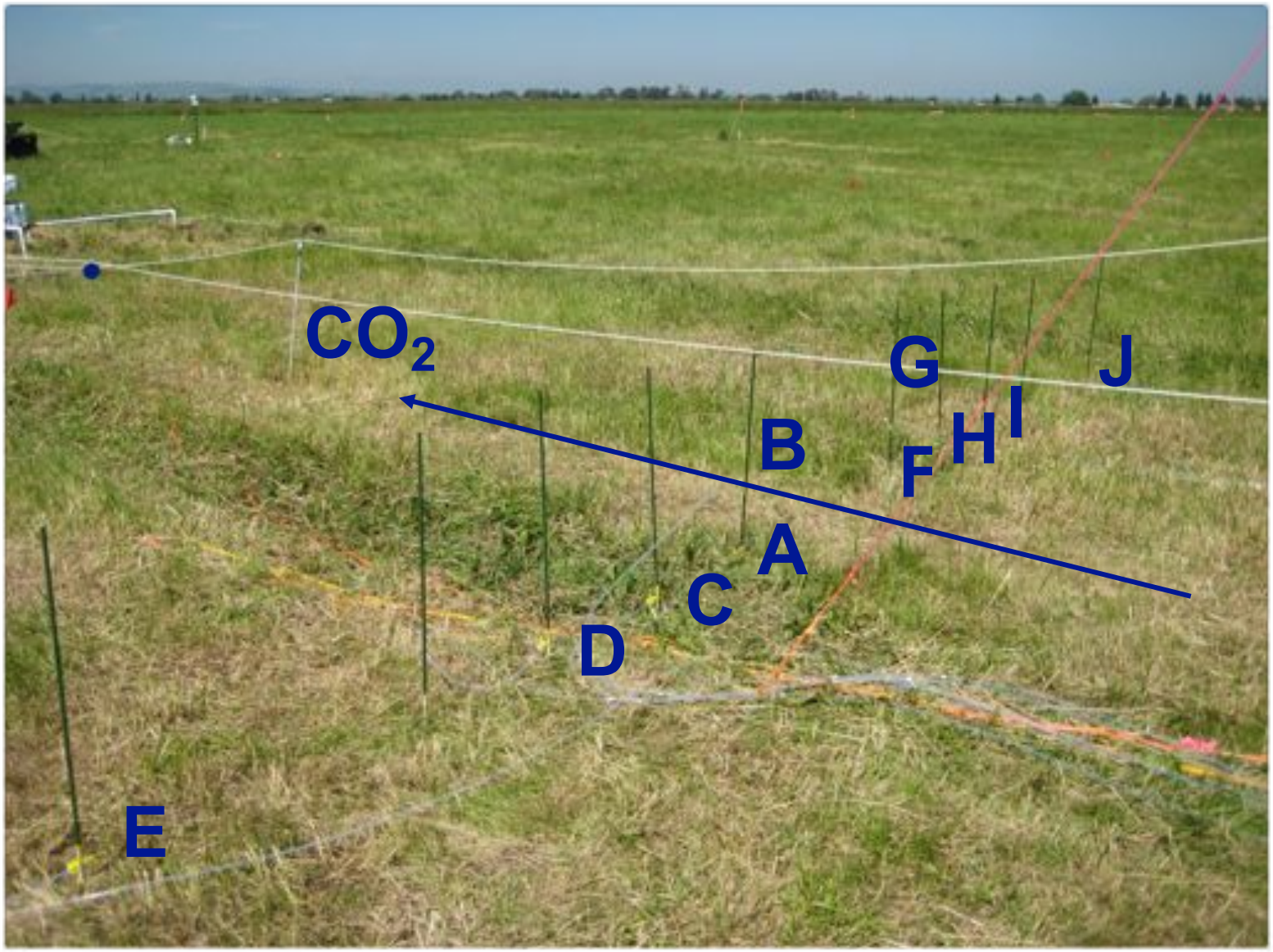
Transverse Grid



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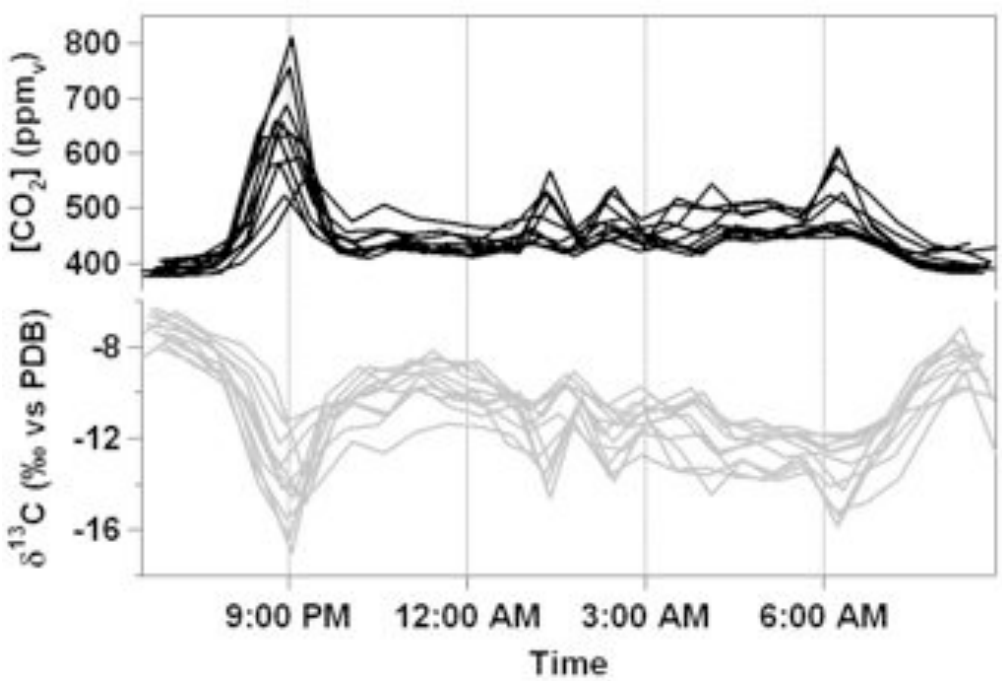
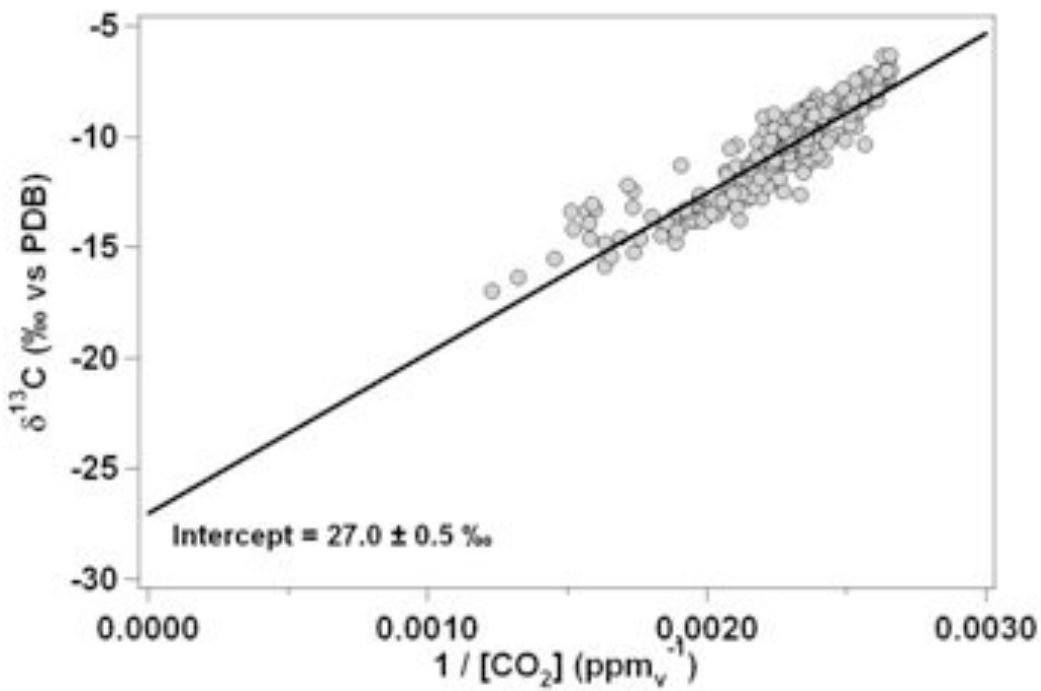
Inlets A-K *before* release: observation of plant respiration





- Measurements of ambient CO_2 and $\delta^{13}C$ for Inlets A–K prior to CO_2 release.
- Measurements span 14 hrs (14-15 July 2009) at ZERT during light precipitation.
- Overlaid traces show small spatial variation (delocalized C source = vegetation)
- Keeling plot indicates $\delta^{13}CO_2$ consistent with site's latitude and C_3 vegetation

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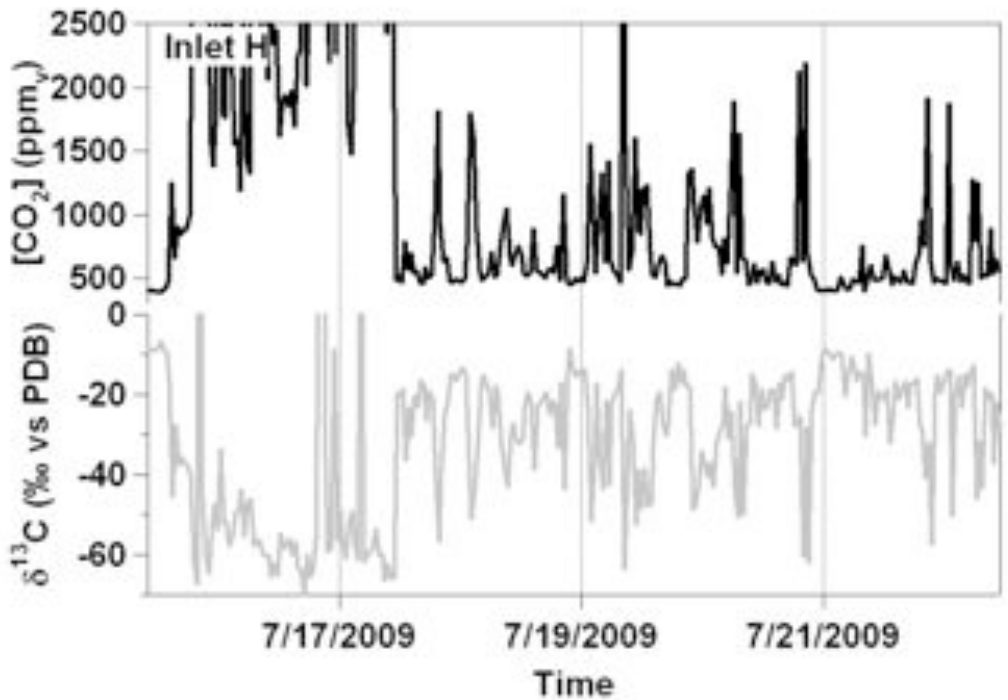
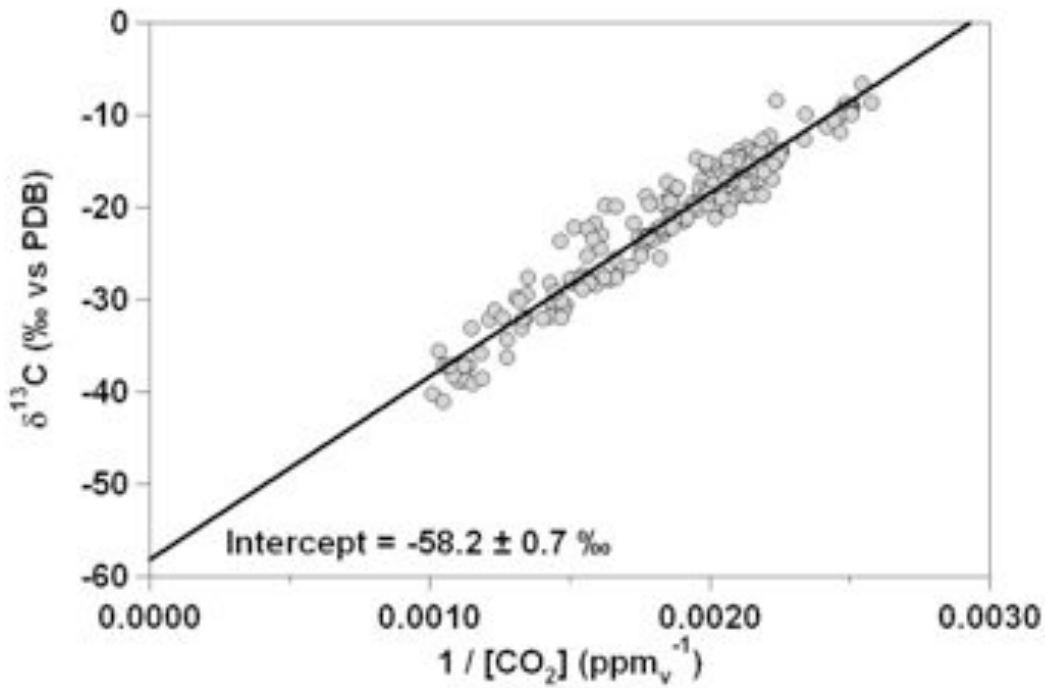
CO_2 and $\delta^{13}C$ *during* release: near leak (inlet H)





- Measurements near leak demonstrate ability to record real-time Keeling plots in field
- Keeling plot intercept yields $\delta^{13}C$ (-58‰ vs PDB) of released CO_2 at ZERT

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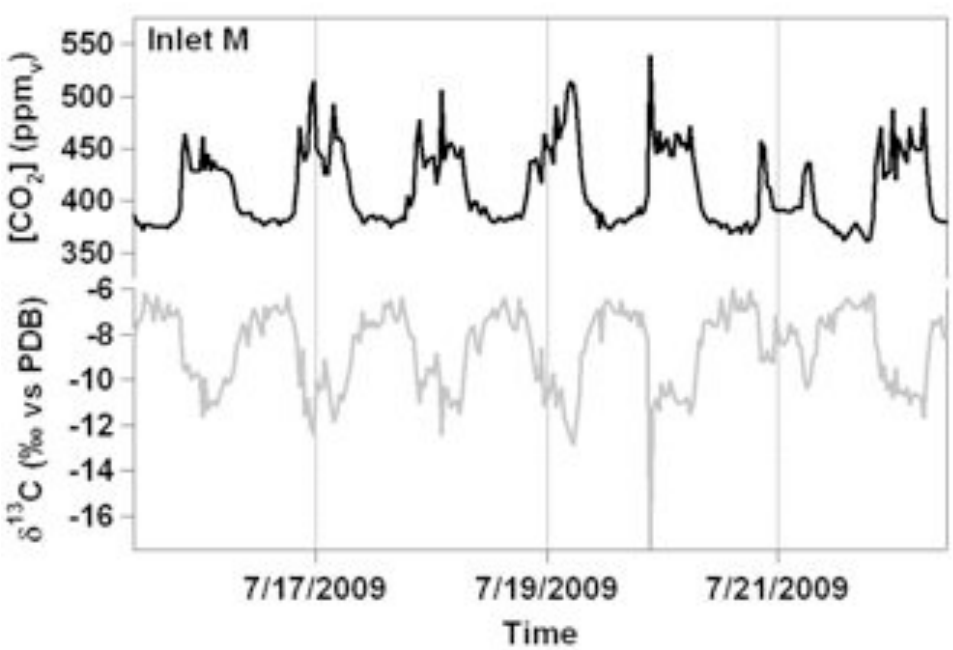
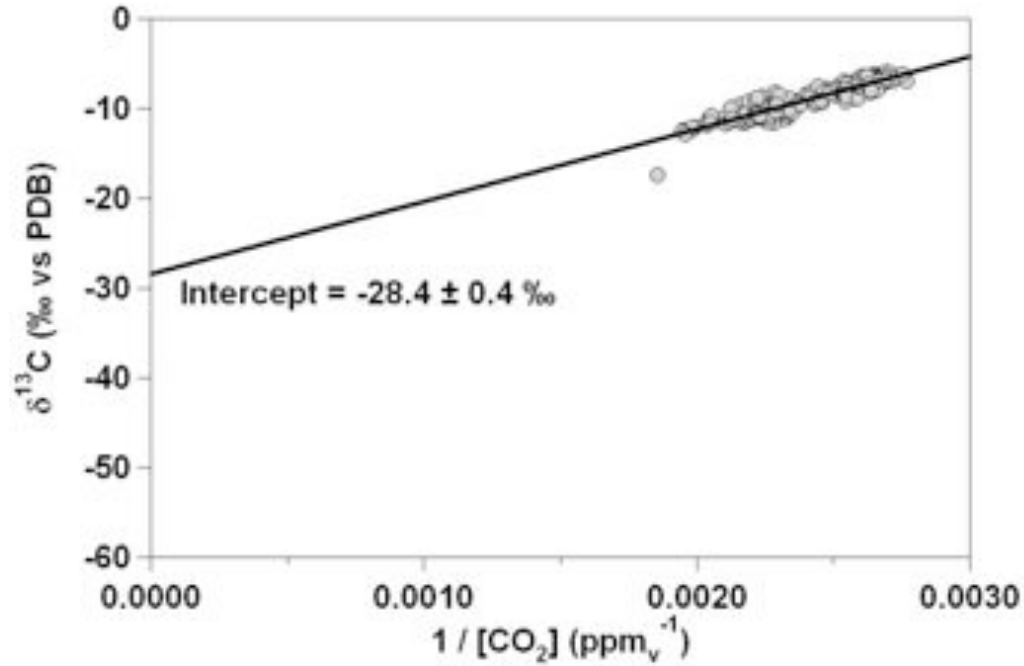
CO_2, $\delta^{13}C$ during release: far from leak (inlet M)





- Measurements generate real-time Keeling plots due to plant respiration.
- Keeling plot intercept yields $\delta^{13}C$ (-28‰ vs PDB) of the C3 plants.

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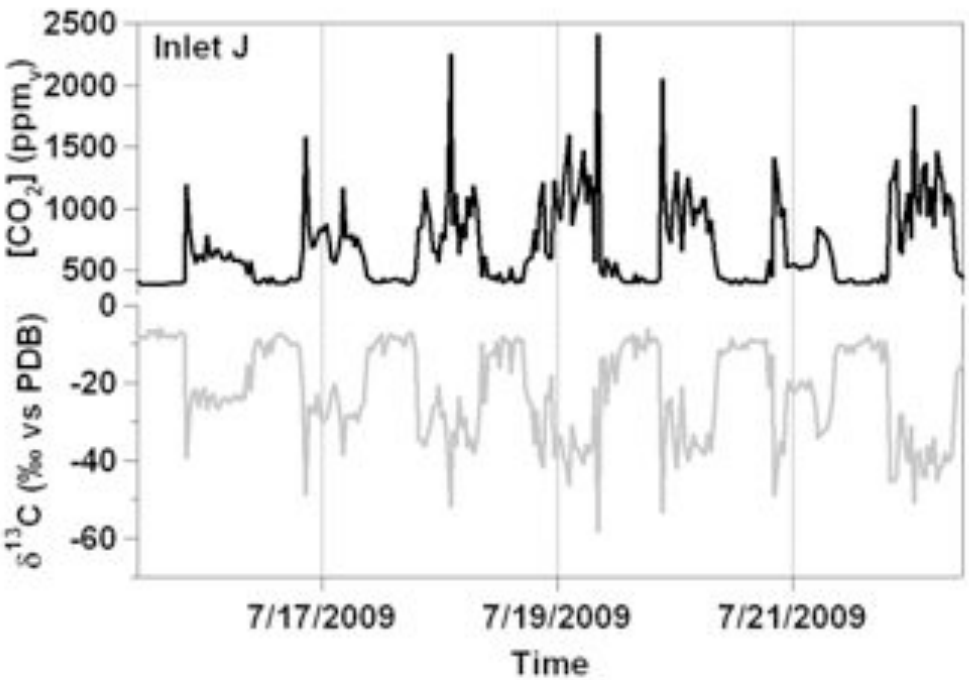
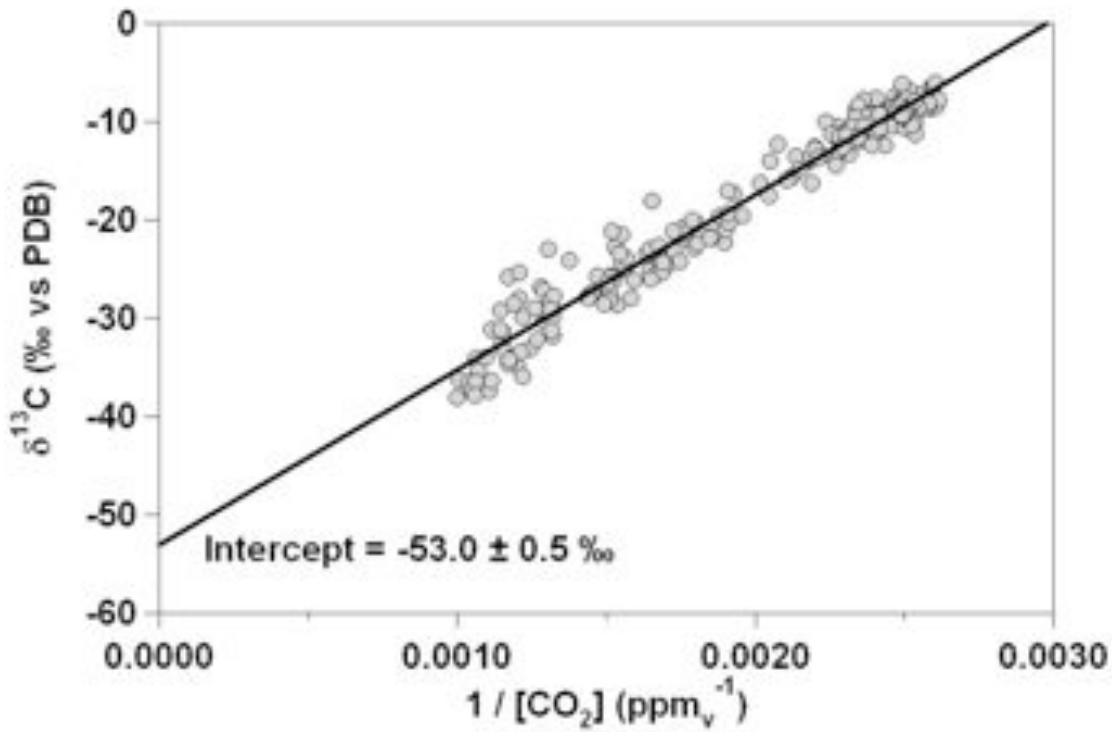
CO_2 and $\delta^{13}C$ during release (inlet J; downwind)





- Measurements of ambient CO_2 and $\delta^{13}C$ during CO_2 release and Keeling plots
- Inlet J located 1.5 m downwind of release
- $\delta^{13}C$ indicates that the measured CO_2 was due to leak
- Diurnal cycles suggest CO_2 might be trapped in soil and released due to fluctuations in temperature or soil moisture

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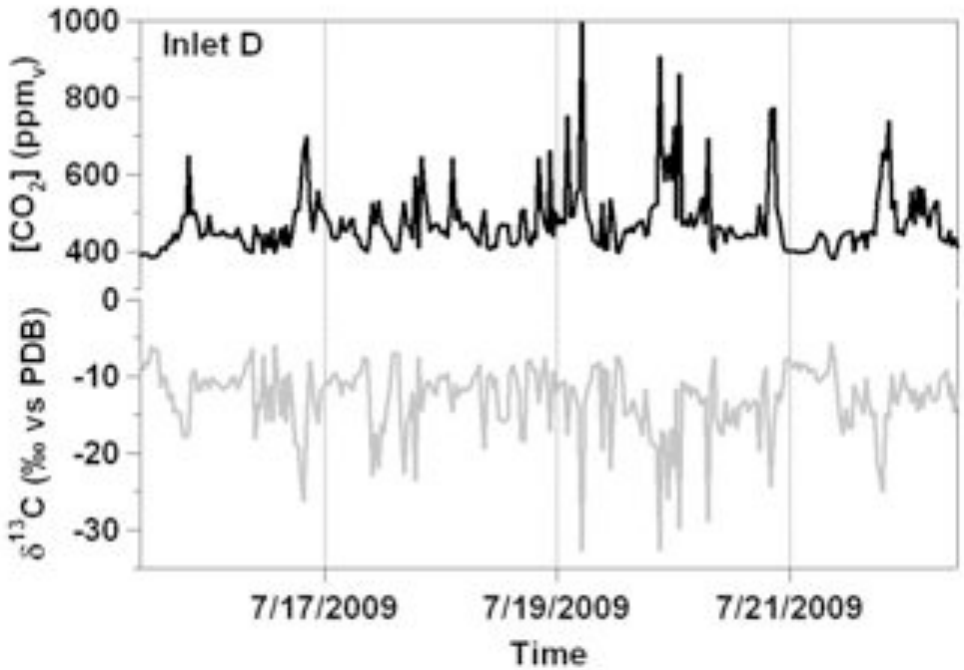
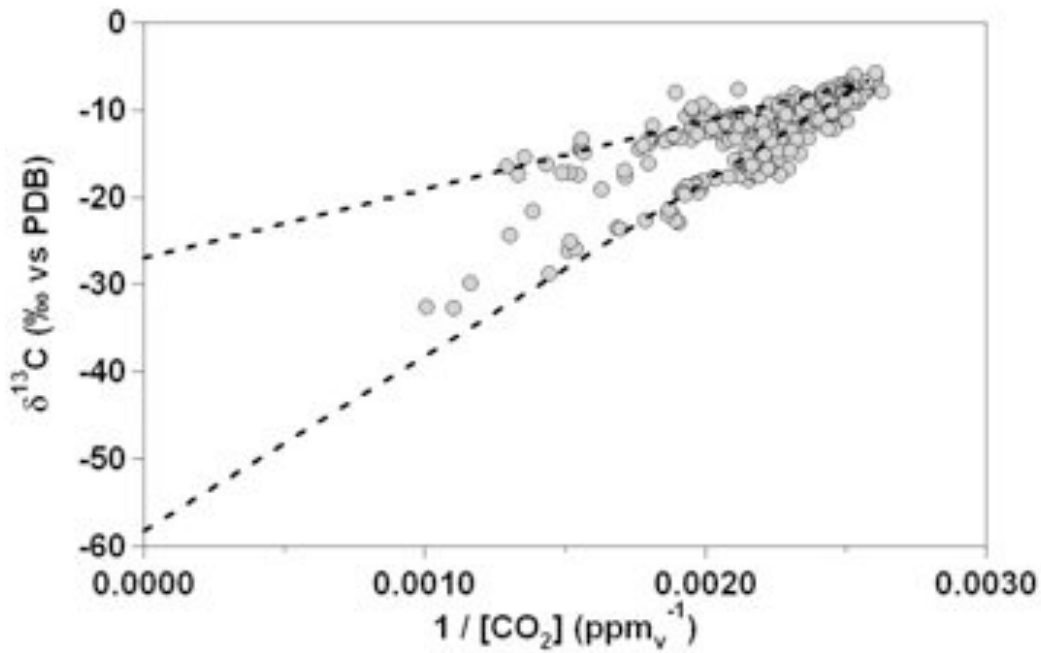
CO_2 and $\delta^{13}C$ during release (inlet D; upwind)





- Measurements of ambient CO_2 and $\delta^{13}C$ during CO_2 release upwind of release
- Keeling plot shows mixing between biogenic and released CO_2
- Dashes: linear, 2-member mixing for biogenic (-27 ‰) and released (-58 ‰) CO_2

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Deployment of CCIA for Carbon Sequestration

Real-time spatial, temporal measurements of
$\delta^{13}C$ and CO_2 at ZERT test site

- Ability to record distinct $\delta^{13}C$ signature of CO_2 release
- Measures $\delta^{13}C$ from ambient sources (plant respiration)
- Records physical map of release (leak) transverse to pipe
- Records temporal variations ($\delta^{13}C$, CO_2) at several locations
- Multi-port unit: spatial & temporal measurements in real time

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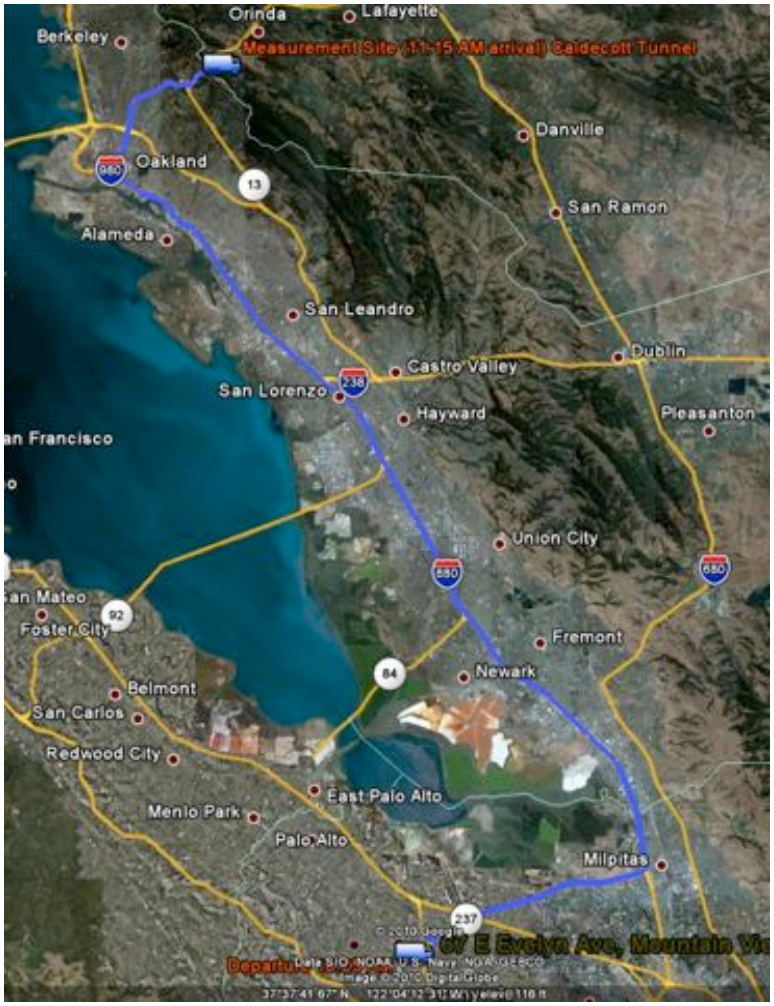
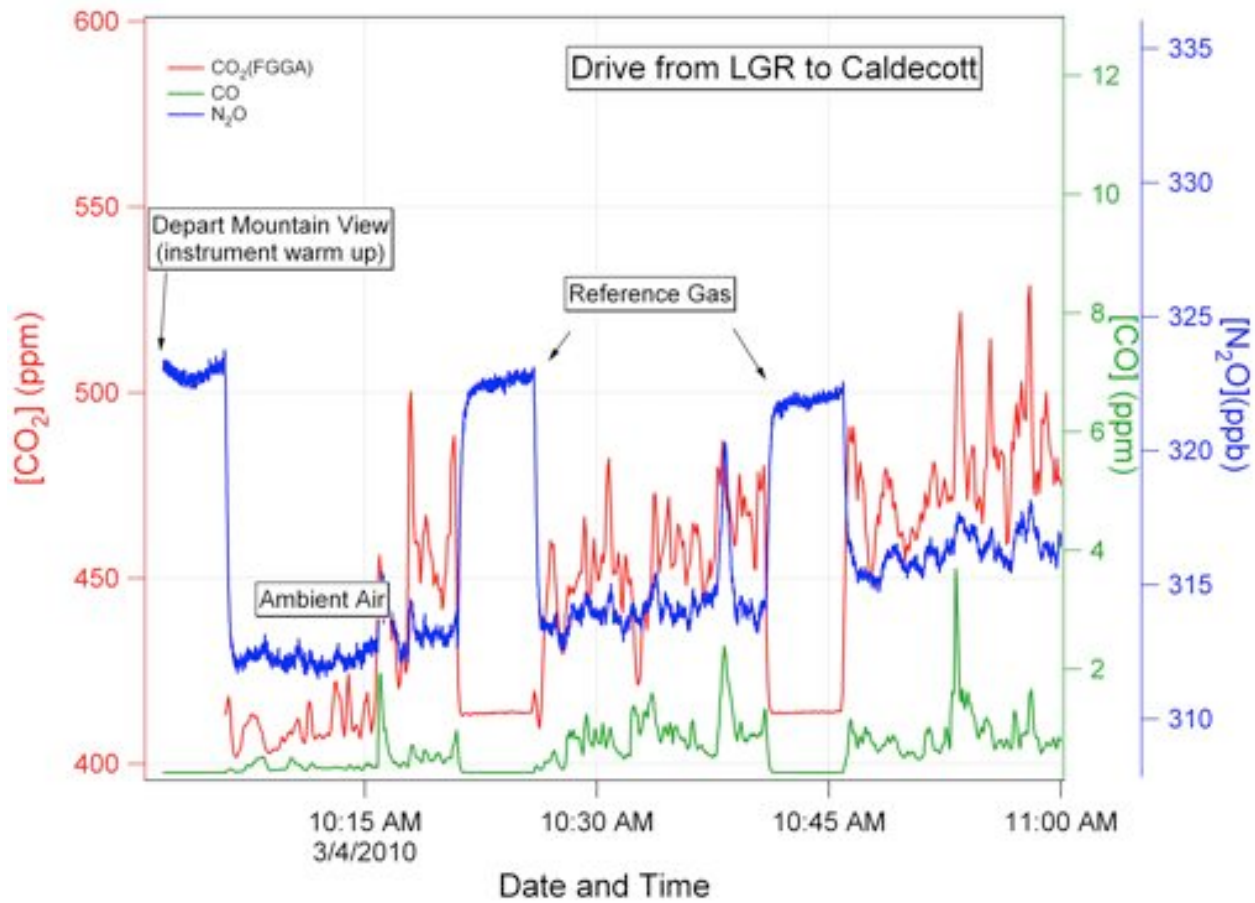
Field Deployment:
Measurements during journey to Caldecott Tunnel





- CO_2, CO, N_2O recorded by Mobile Lab from LGR to Caldecott Tunnel
- Reference cylinders in Mobile Lab sampled periodically as validation checks
- Provides sensitive measurements while driving or stationery.

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Field Deployment: Measurements at Caldecott Tunnel

- Teflon lines sample air inside tunnel, 10 cm above grate outside tunnel and far from Tunnel along with reference gases.

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Field Deployment: Measurements at Caldecott Tunnel

- Teflon lines sample air inside tunnel, 10 cm above grate outside tunnel and far from Tunnel along with reference gases.

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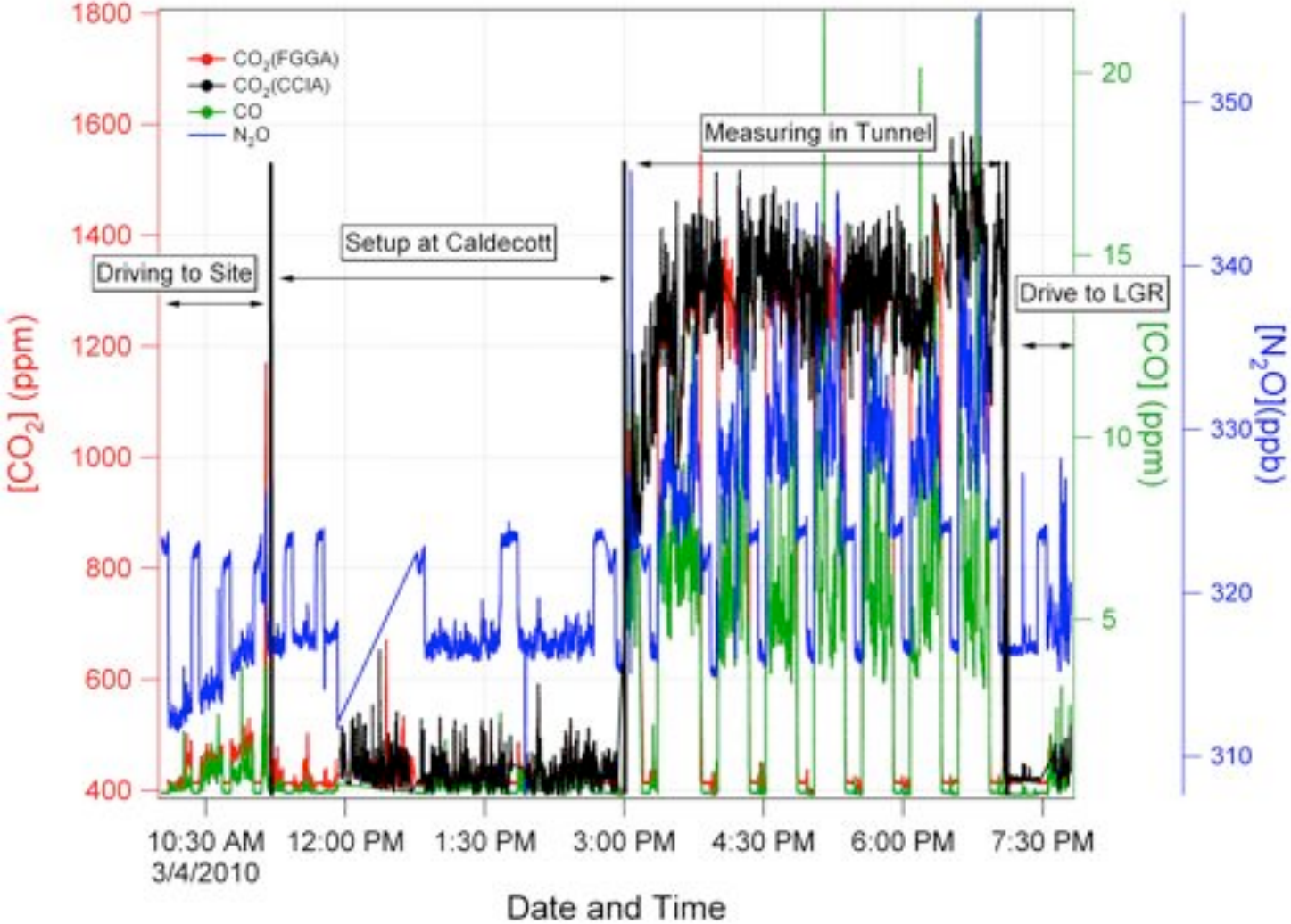
Measurements on site (Tunnel) using Mobile Lab

CO_2, CO, N_2O during journey to Caldecott Tunnel, setup, deployment, and return LGR



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas.
- Mobile Lab provides sensitive measurements while driving or stationery

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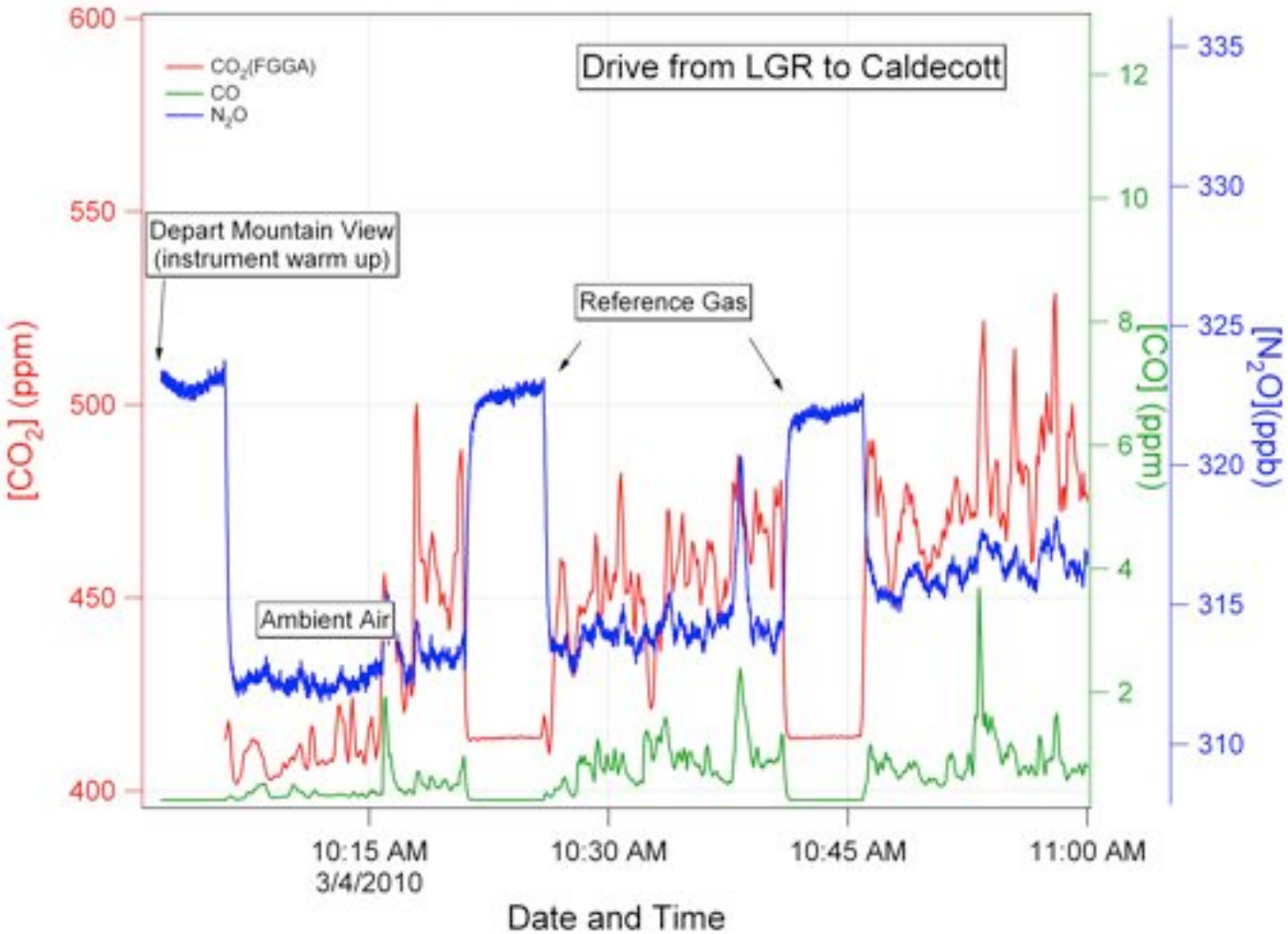
Measurements on site (Tunnel) using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas.
- Mobile Lab provides sensitive measurements while driving or stationery.
- Reference Gas cylinders in Mobile Lab sampled periodically as validation checks.

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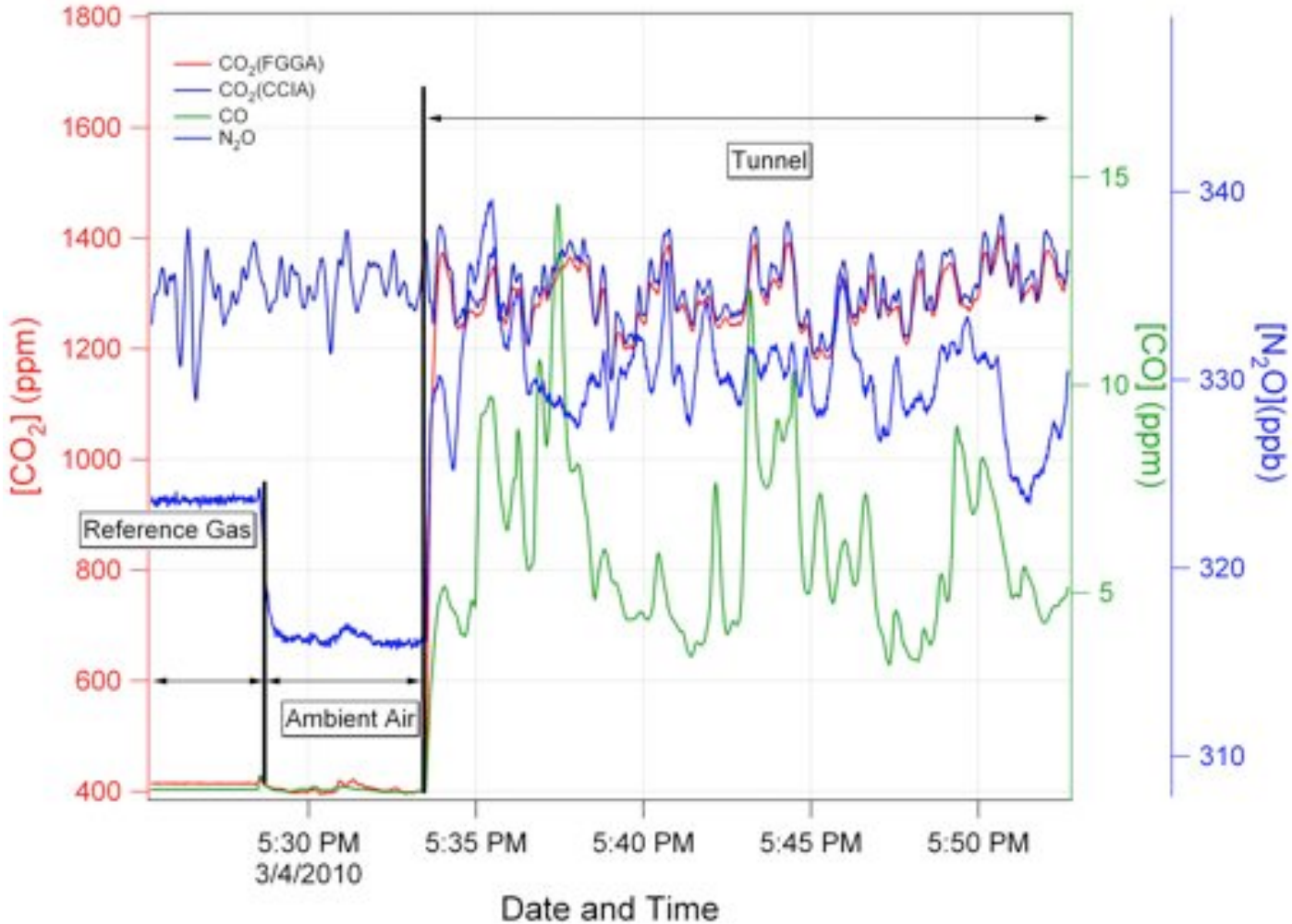
Measurements on site using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas.
- Measurements during commute hour of ambient and inside tunnel ("Tunnel")

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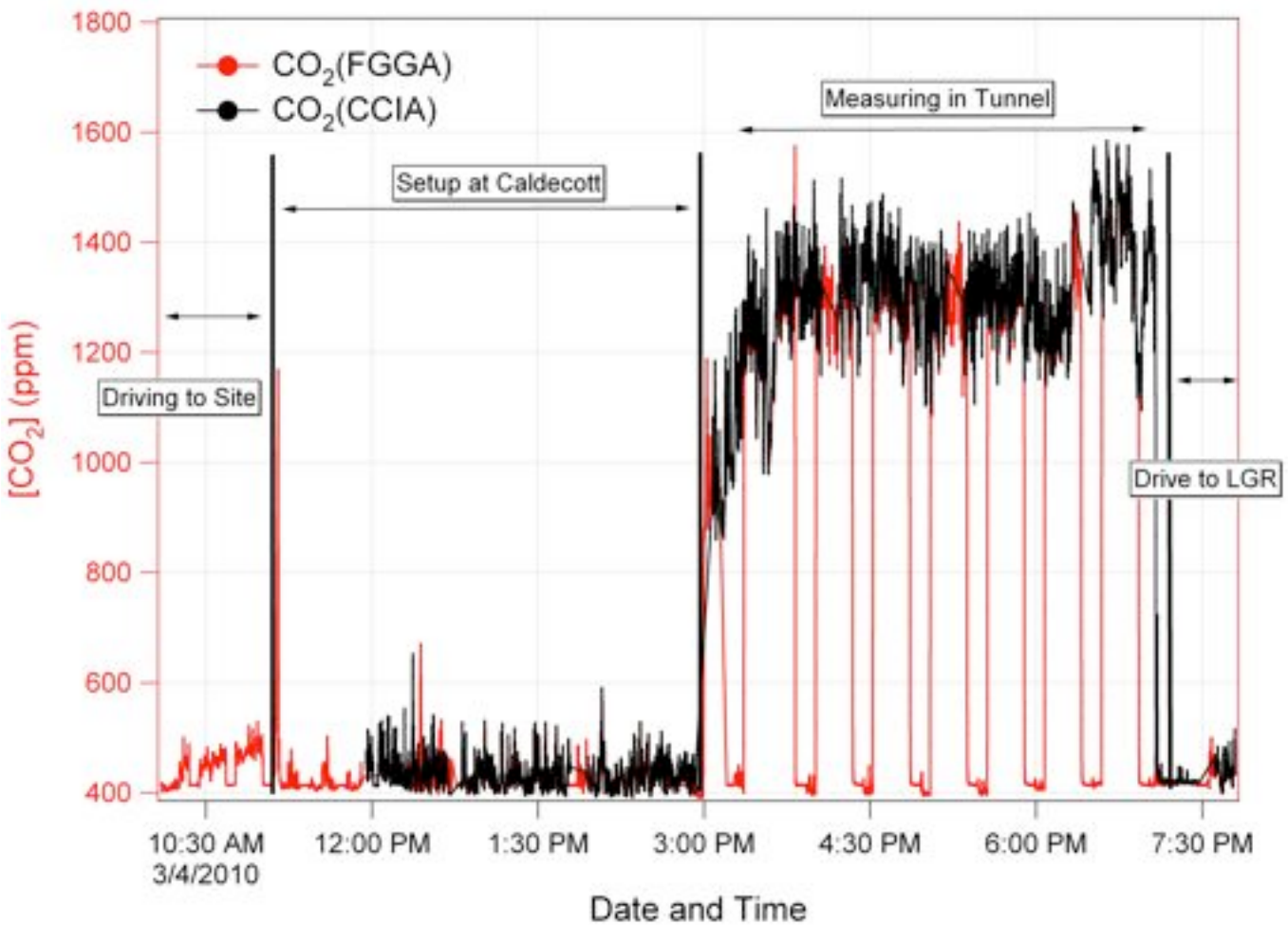
Measurements on site using Mobile Lab



- CO_2 recorded by Carbon Dioxide Isotope Analyzer and Fast Greenhouse Gas Analyzer
- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas.
- Measurements during commute hour of ambient and inside tunnel ("Tunnel")

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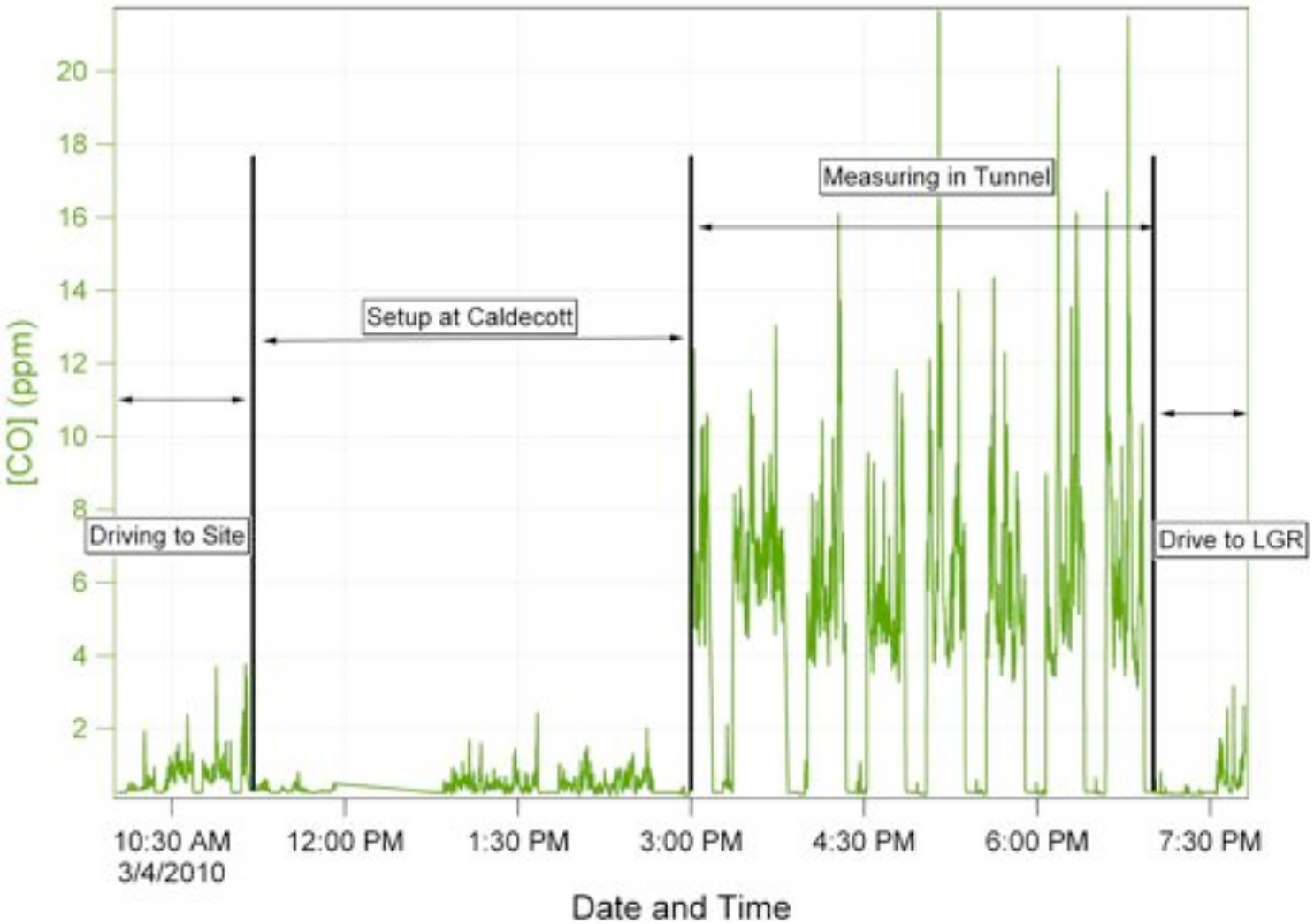
Measurements on site using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas
- Mobile Lab provides sensitive measurements while driving or stationery

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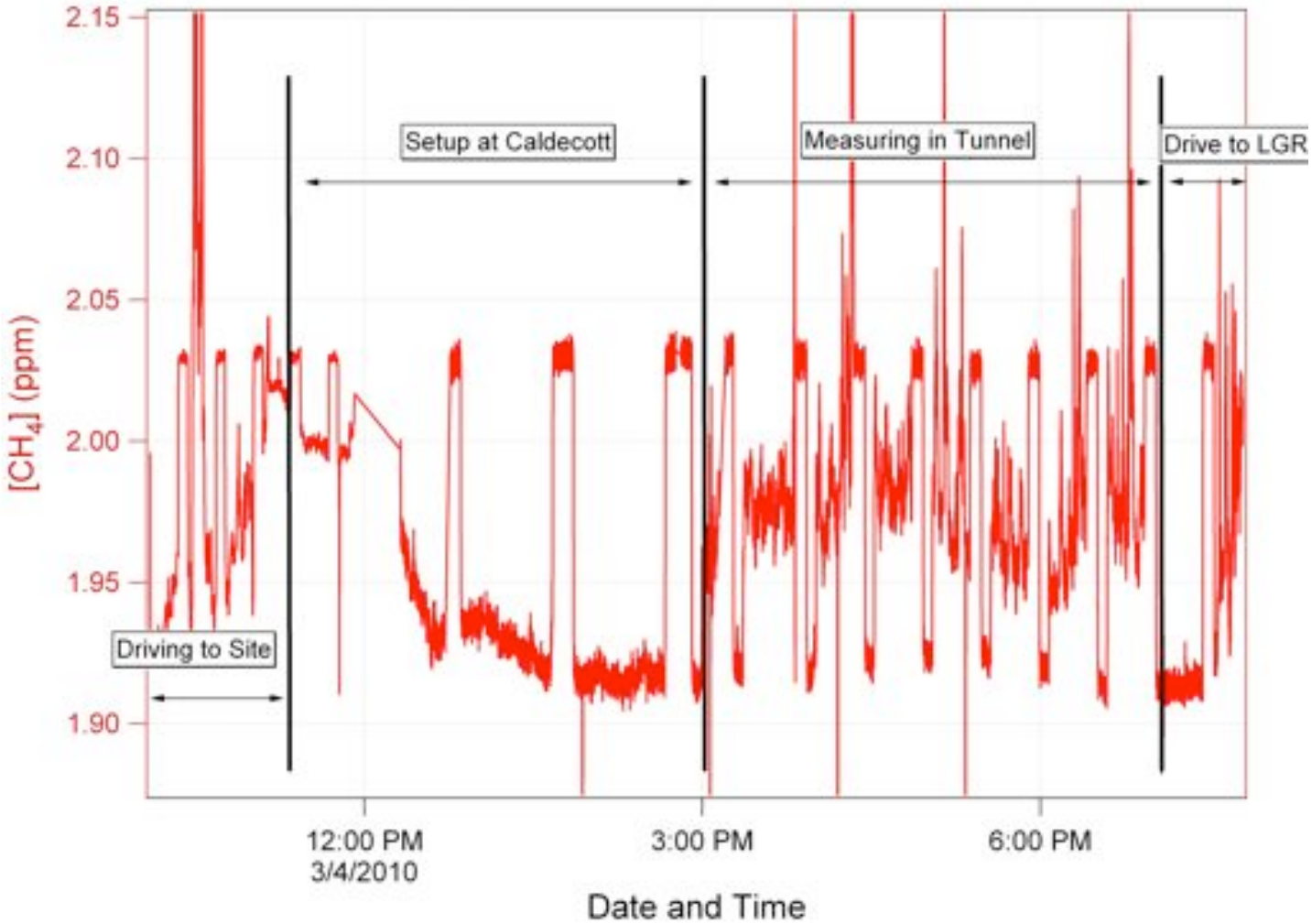
Methane measurements on site using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas
- Mobile Lab provides sensitive measurements while driving or stationery

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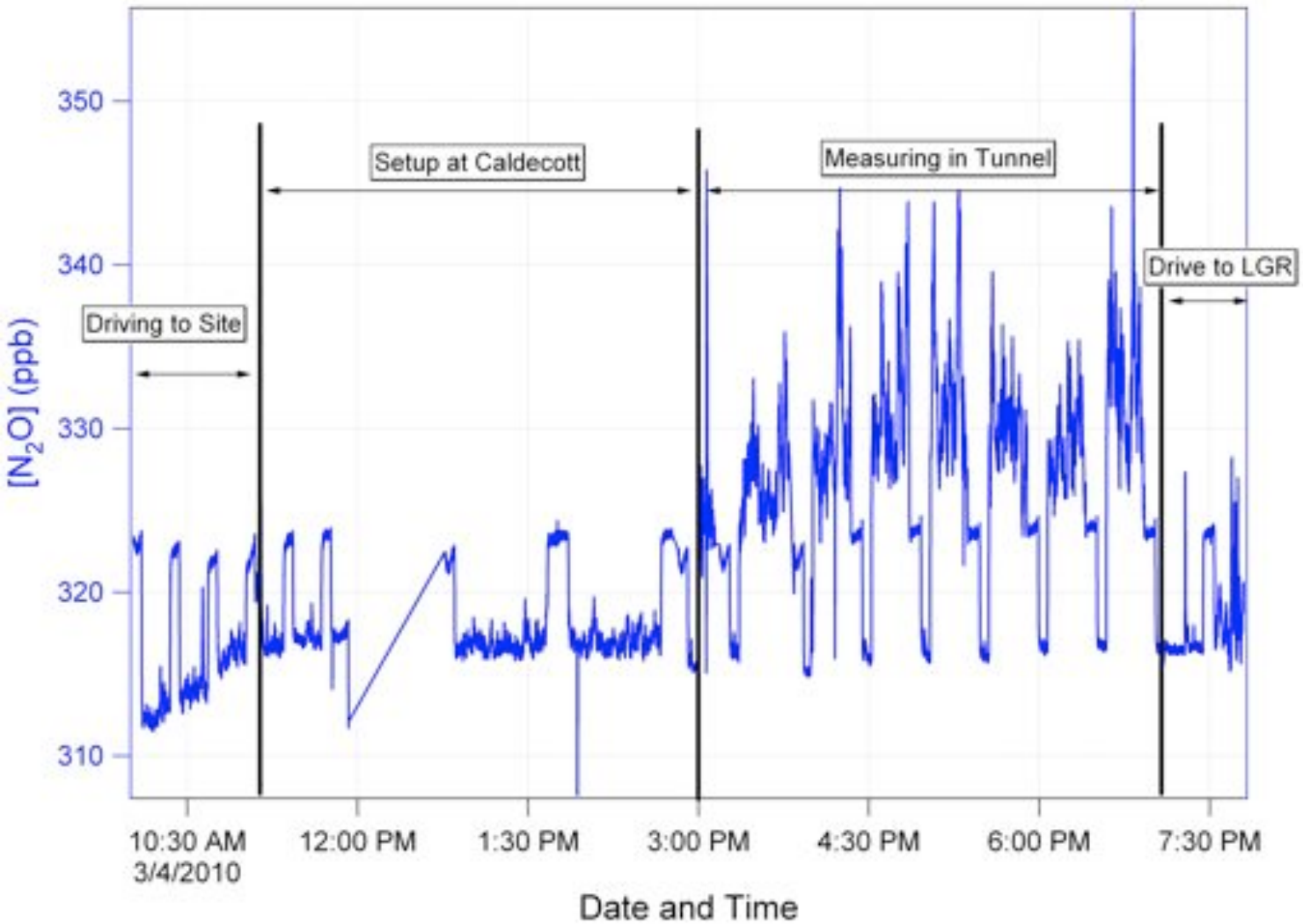
N_2O measurements on site using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas
- Mobile Lab provides sensitive measurements while driving or stationery

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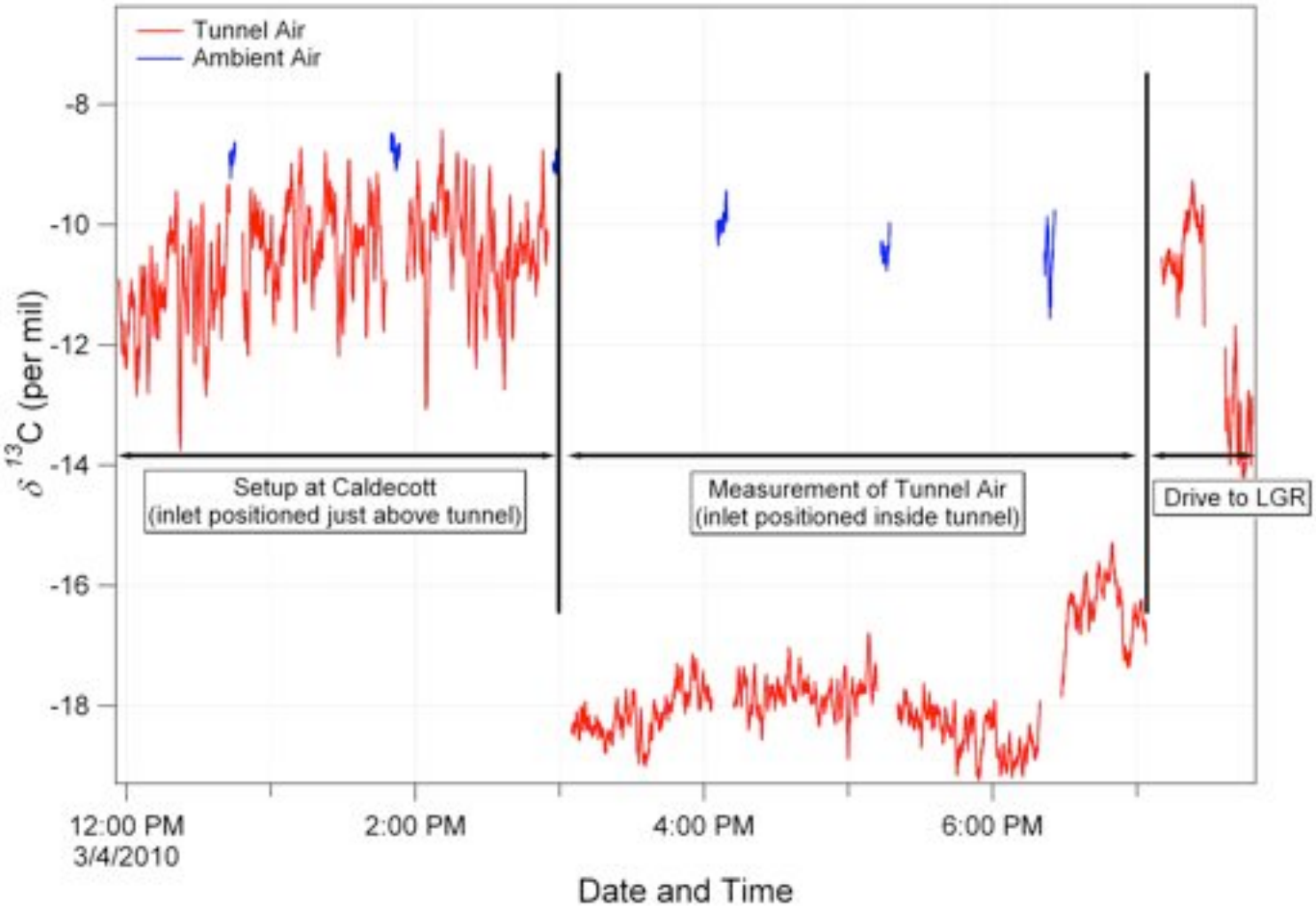
CO_2 and $\delta^{13}CO_2$ measurements on site using Mobile Lab



- "Measuring in Tunnel" includes ambient air near tunnel and of reference gas
- Mobile Lab provides sensitive measurements while driving or stationery

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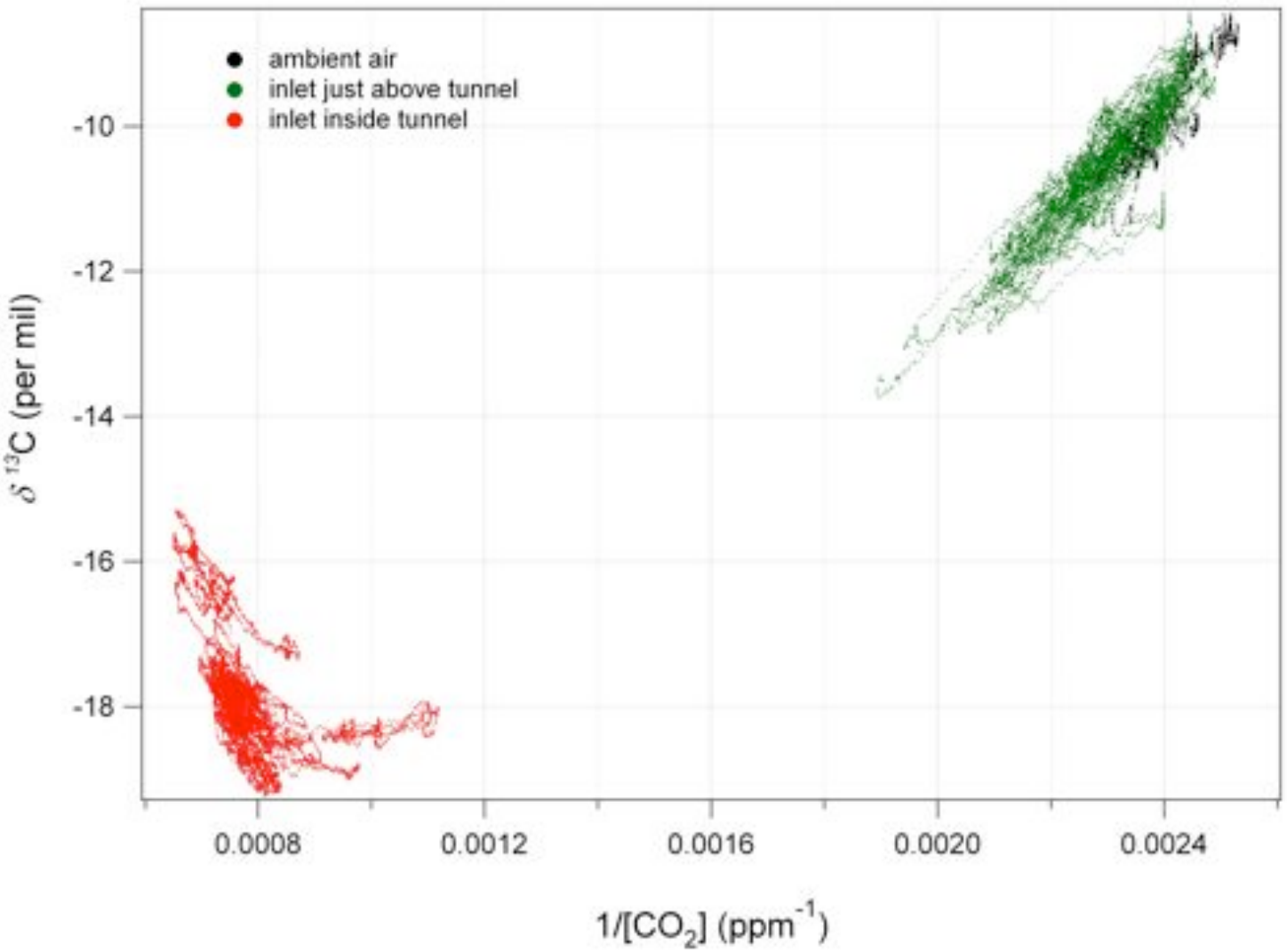
CO_2 and $\delta^{13}CO_2$ measurements on site using Mobile Lab



- Keeling plot ($\delta^{13}C$ vs $1/[CO_2]$) 25 meters from tunnel, in air near tunnel inlet and in tunnel air using data shown in previous plot

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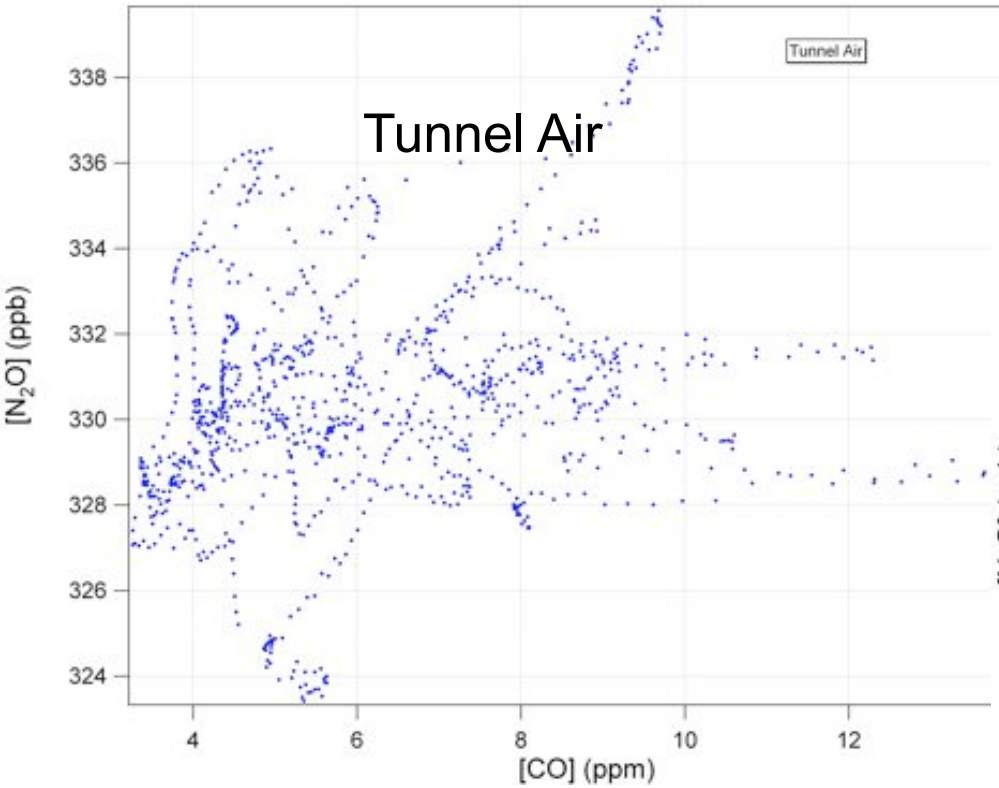
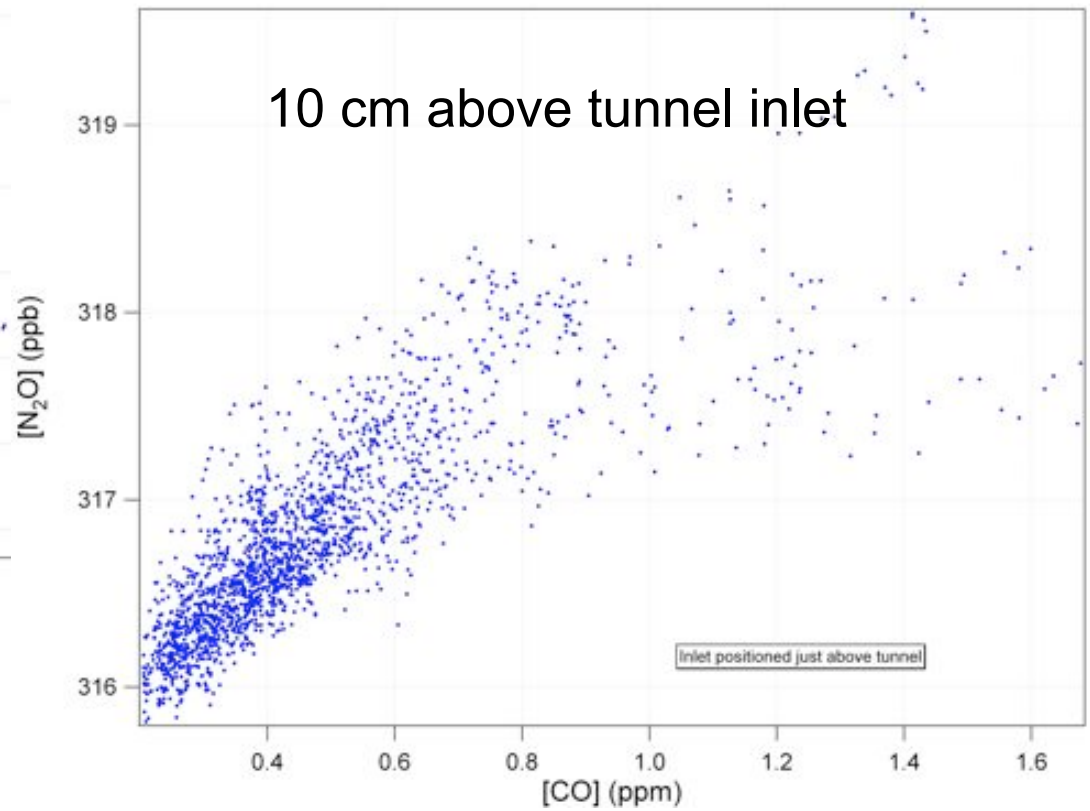
N_2O and CO measurements on site using Mobile Lab





- Measurements of N_2O and CO in tunnel air recorded by N_2O/CO Analyzer
- Correlations of N_2O and CO in tunnel and 10 cm above tunnel inlet

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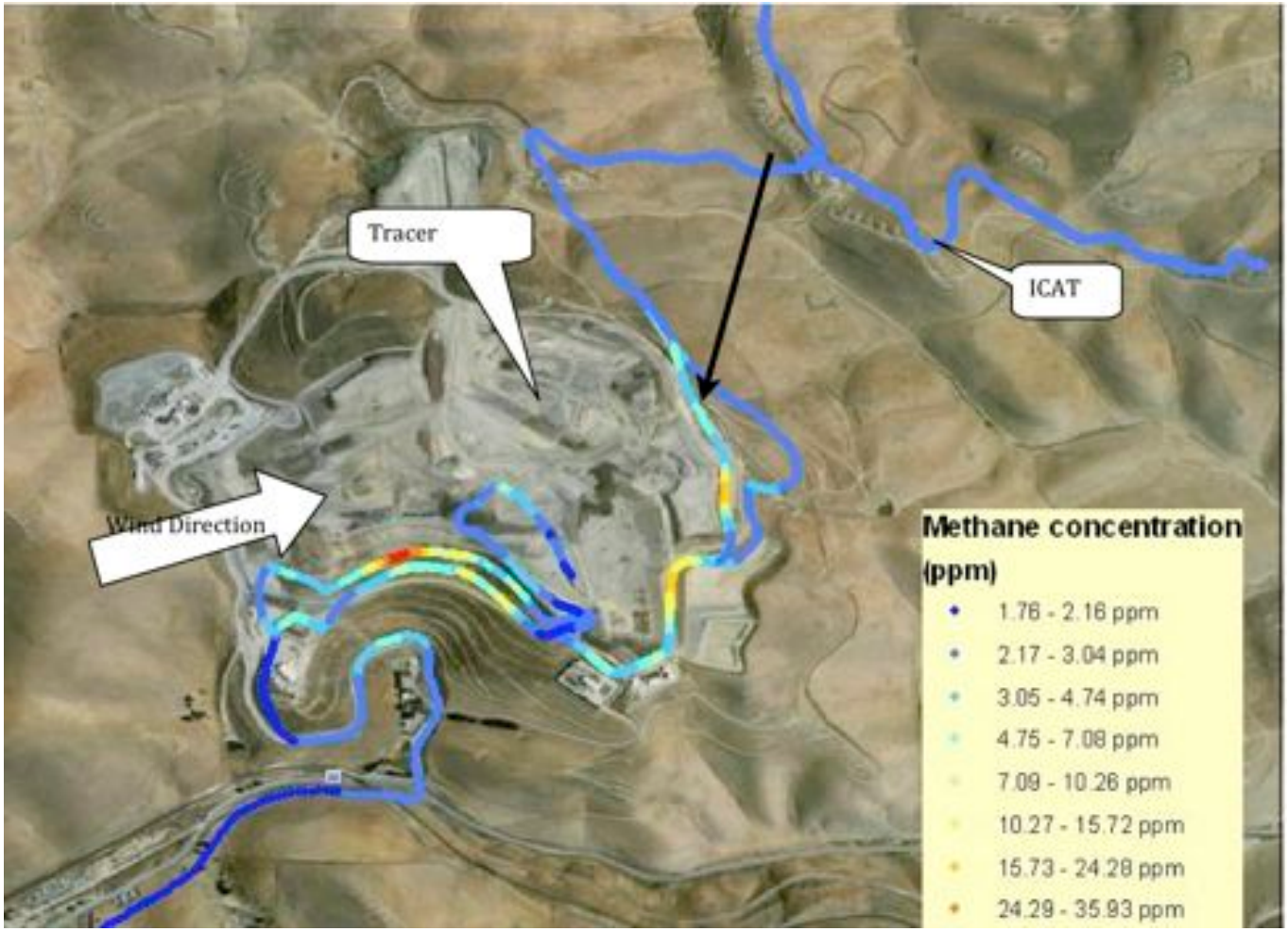
Field Deployment:
Altamont Landfill (Oct 2009; Livermore, CA)



- Deployment (collaboration with CARB) characterized variations of methane fugitive emissions and acetylene tracers due to intentional (plume) release

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Deployment at Altamont Landfill Site

- Deployment (collaboration with CARB) characterized variations of methane fugitive emissions and acetylene tracers due to intentional (plume) release

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Deployment at Altamont Landfill Site

- Deployment (collaboration with CARB) characterized variations of methane fugitive emissions and acetylene tracers due to intentional (plume) release

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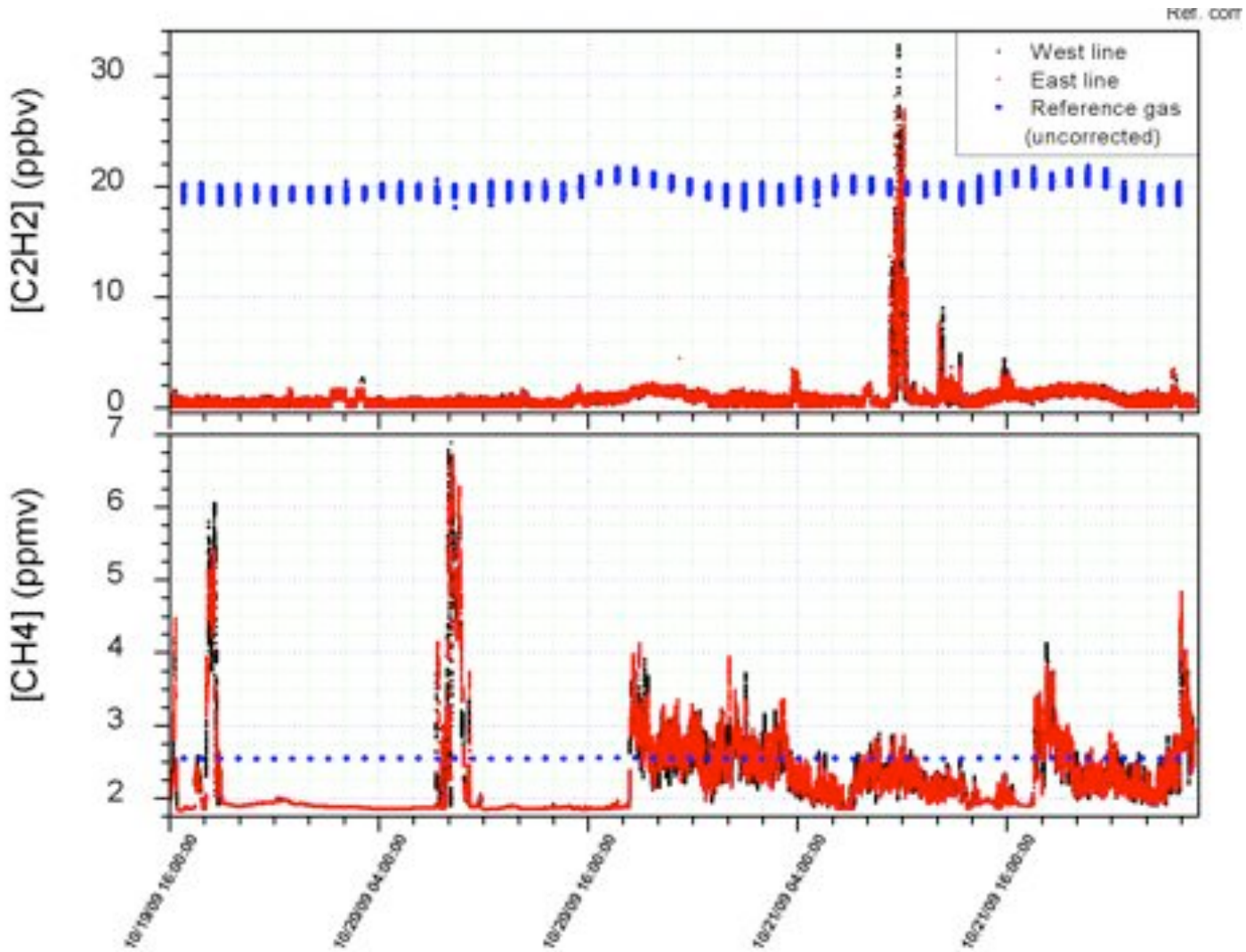
Deployment at Altamont Landfill Site (using LGR's Fast Methane and Acetylene Analyzer)



- Measurements of methane and acetylene recorded from locations 100 feet apart
- Autonomous measurements at multiple locations using a single analyzer

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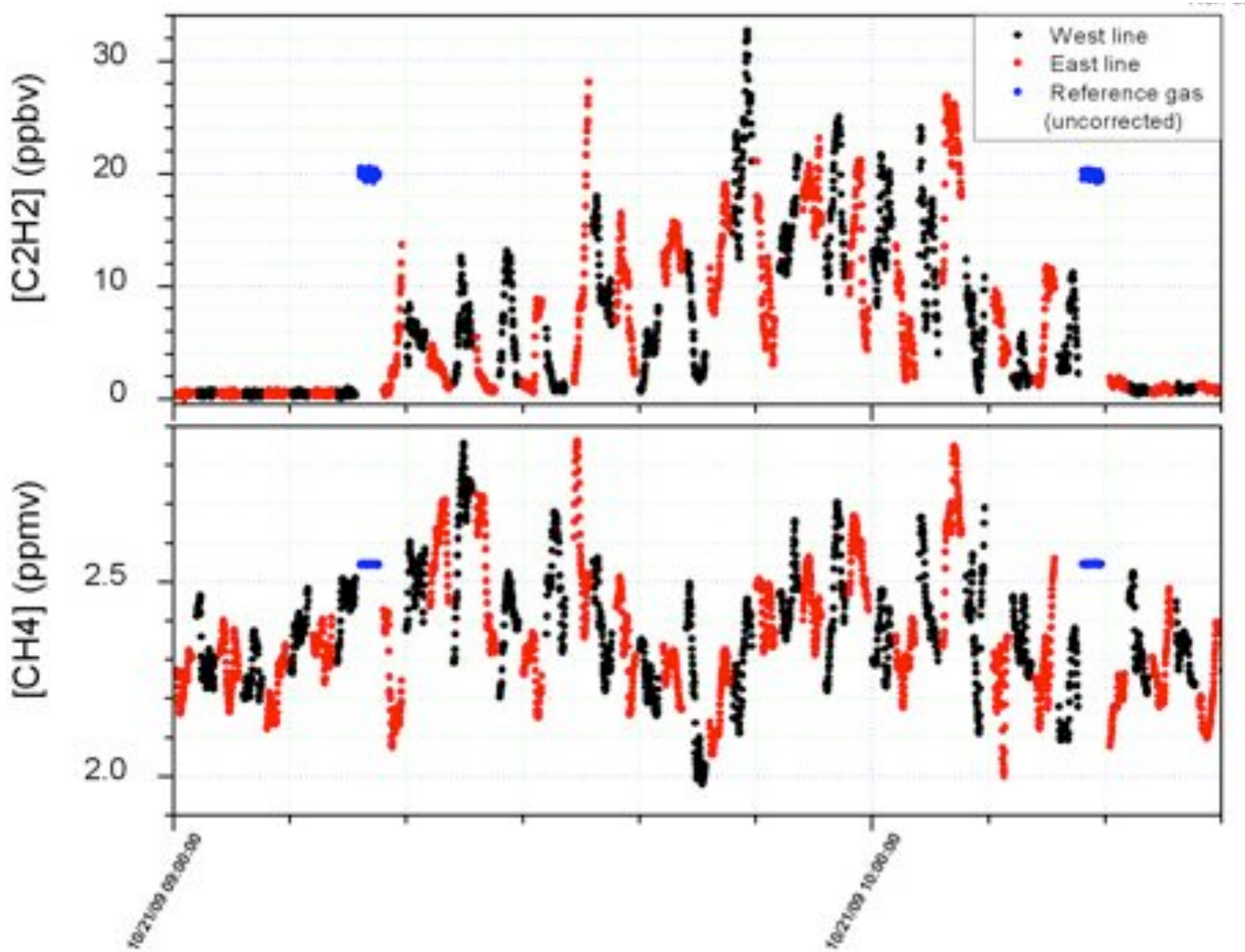
Deployment at Altamont Landfill Site (using LGR's Fast Methane and Acetylene Analyzer)



- Measurements of methane and acetylene recorded from locations 100 feet apart
- Autonomous measurements at multiple locations using a single analyzer

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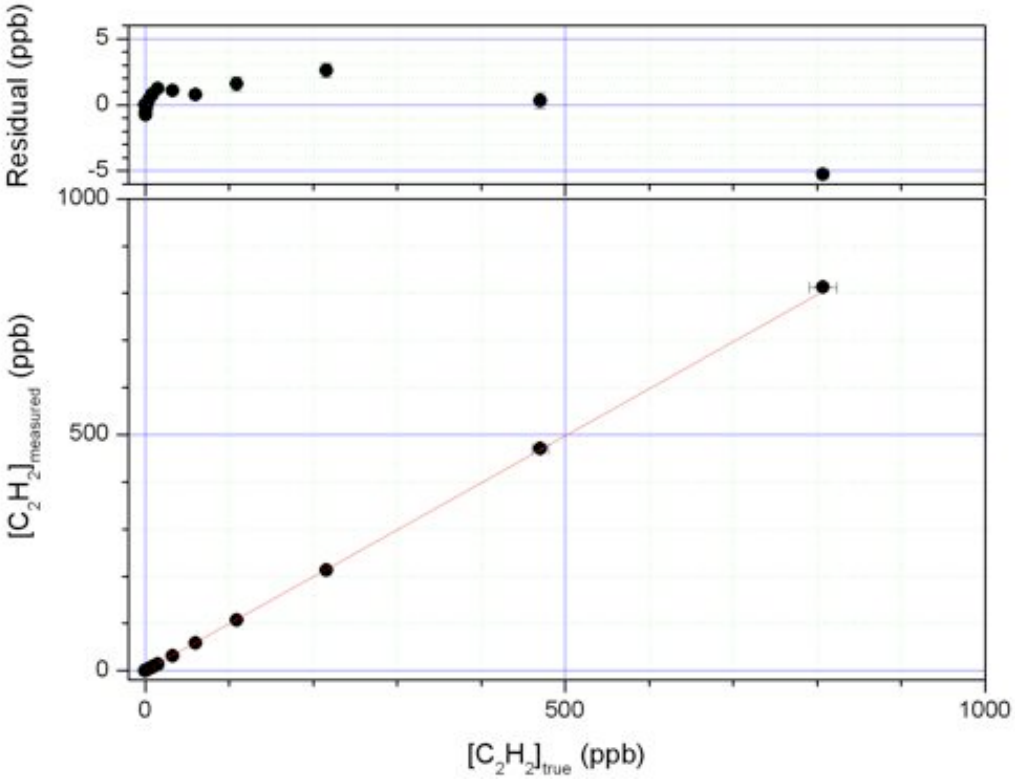
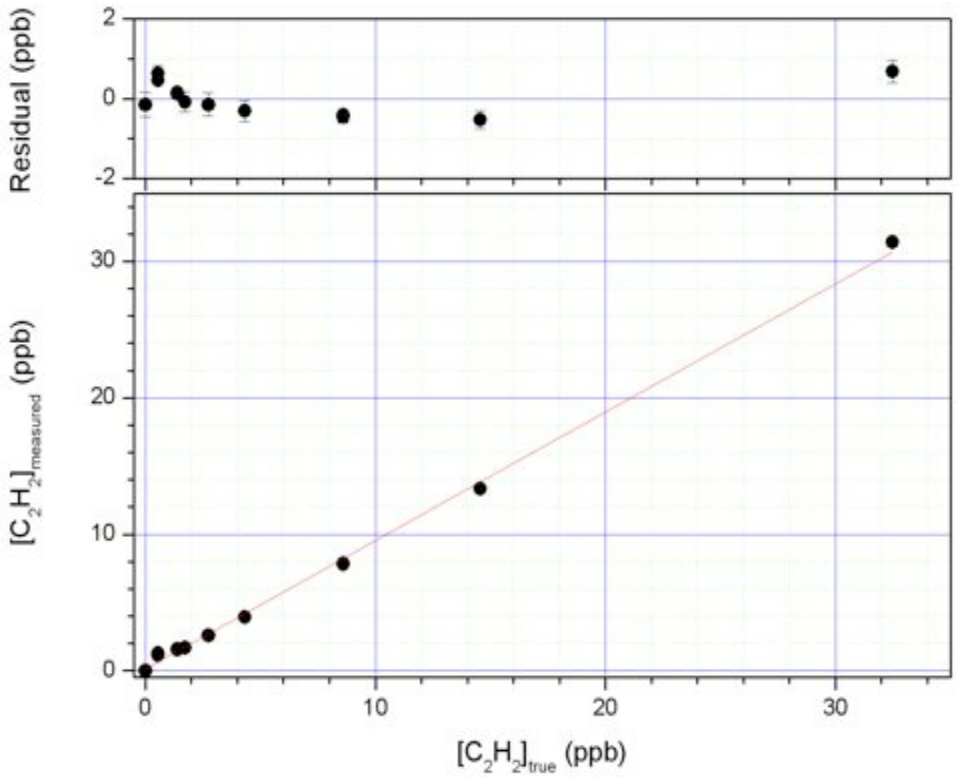
Deployment at Altamont Landfill Site (LGR's Fast Methane and Acetylene Analyzer)





- FAMA: fast, sensitive, linear measurements over extremely wide range
- σ_{CH4} = 0.6 ppbv and σ_{C2H2} = 0.2 ppbv in 2 seconds measurement time
- High sensitivity (< 0.1 ppbv in 10-seconds measurement time)

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Field Deployment: Twitchell Island with UC Berkeley

measurements in rice paddy fields
using LGR's FGGA (methane, carbon dioxide and water)

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Field Deployment: Twitchell Island with UC Berkeley

measurements in rice paddy fields
using LGR's FGGA (methane, carbon dioxide and water)

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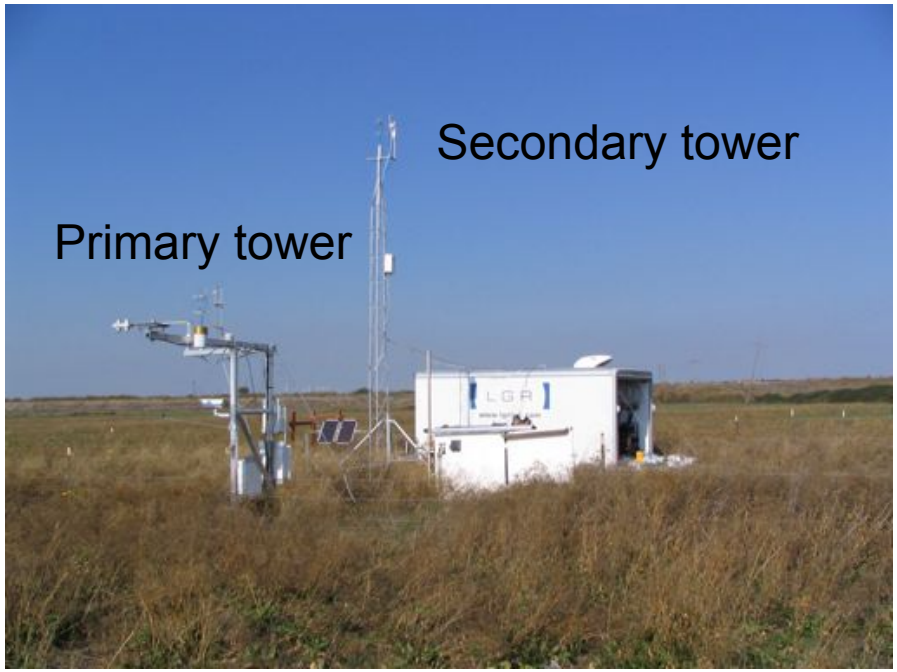
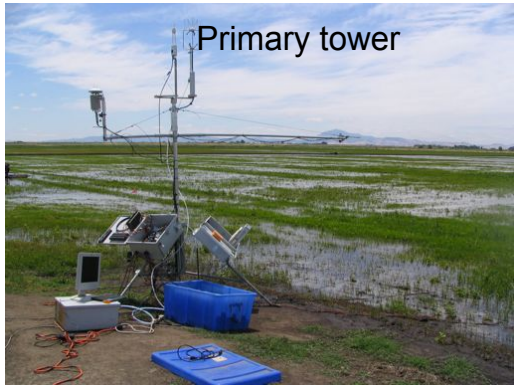
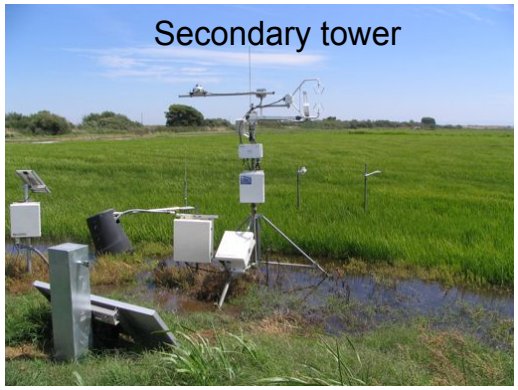
Mobile Lab at Twitchell Island
with UC Berkeley Profs Baldocchi, Goldstein, Cohen



Sherman Island:
Land use: Semi-abandoned field
Land cover: invasive pepperweed
(*Lepidium Latifolium)*
Water table: 50 cm below ground

Twitchell Island:
Land use: agricultural crop
Land cover: rice
Water table: 10 cm above ground





measurements in rice paddy fields
using LGR's FGGA (methane, carbon dioxide and water vapor)

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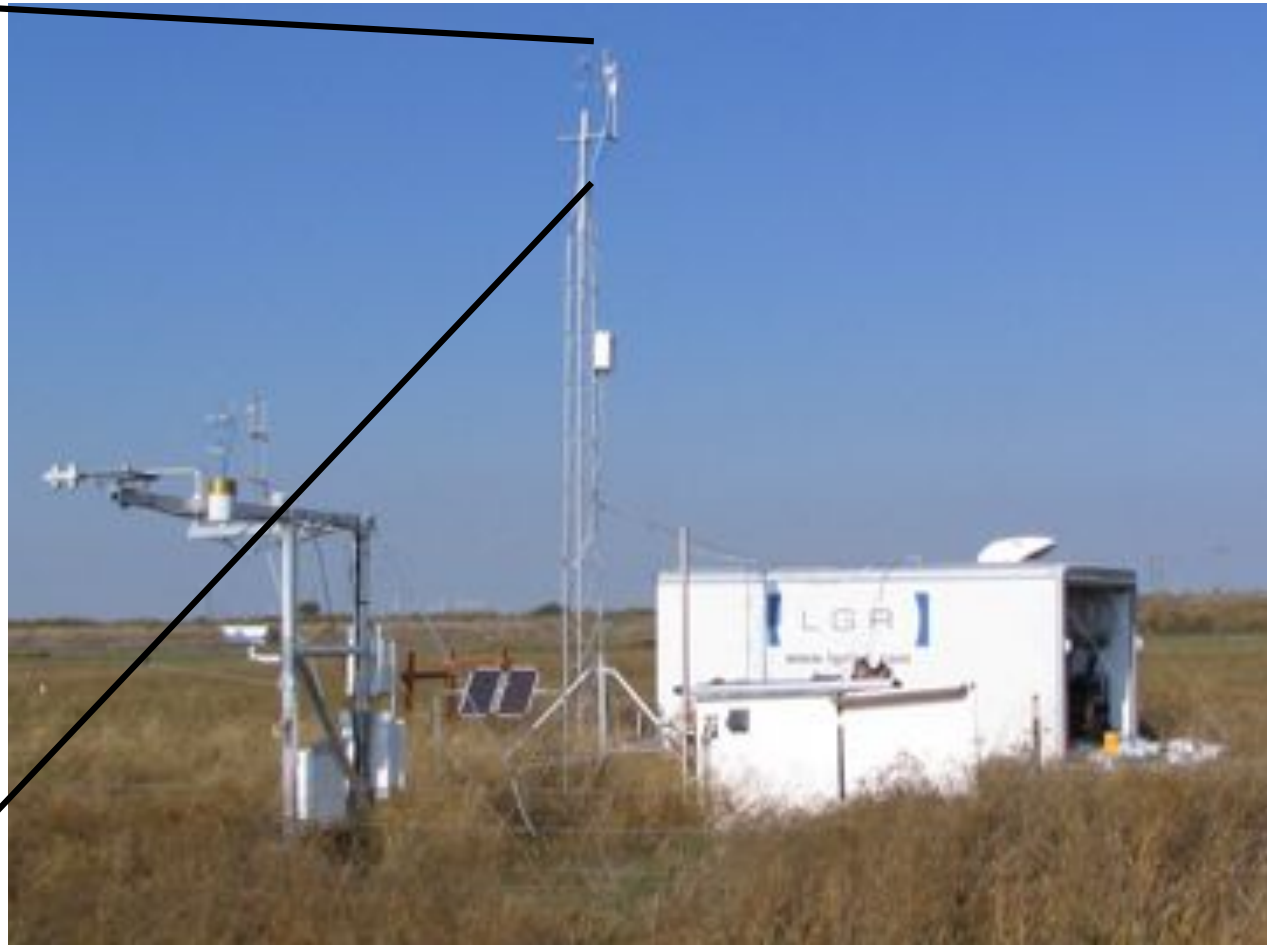
δ^2H, $\delta^{18}O$ and H_2O measurements at flux tower site



- WVIA and WVISS provide continuous unattended calibrated measurements in the field
- Fast, continuous measurements (2-Hz data rate) of δ^2H, $\delta^{18}O$ and H_2O in air
- Dual flux tower setup in a semi-abandoned field near invasive pepperweed

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Rapid δ^2H, $\delta^{18}O$ and H_2O measurements at flux site

- photo from balloon above dual tower flux site

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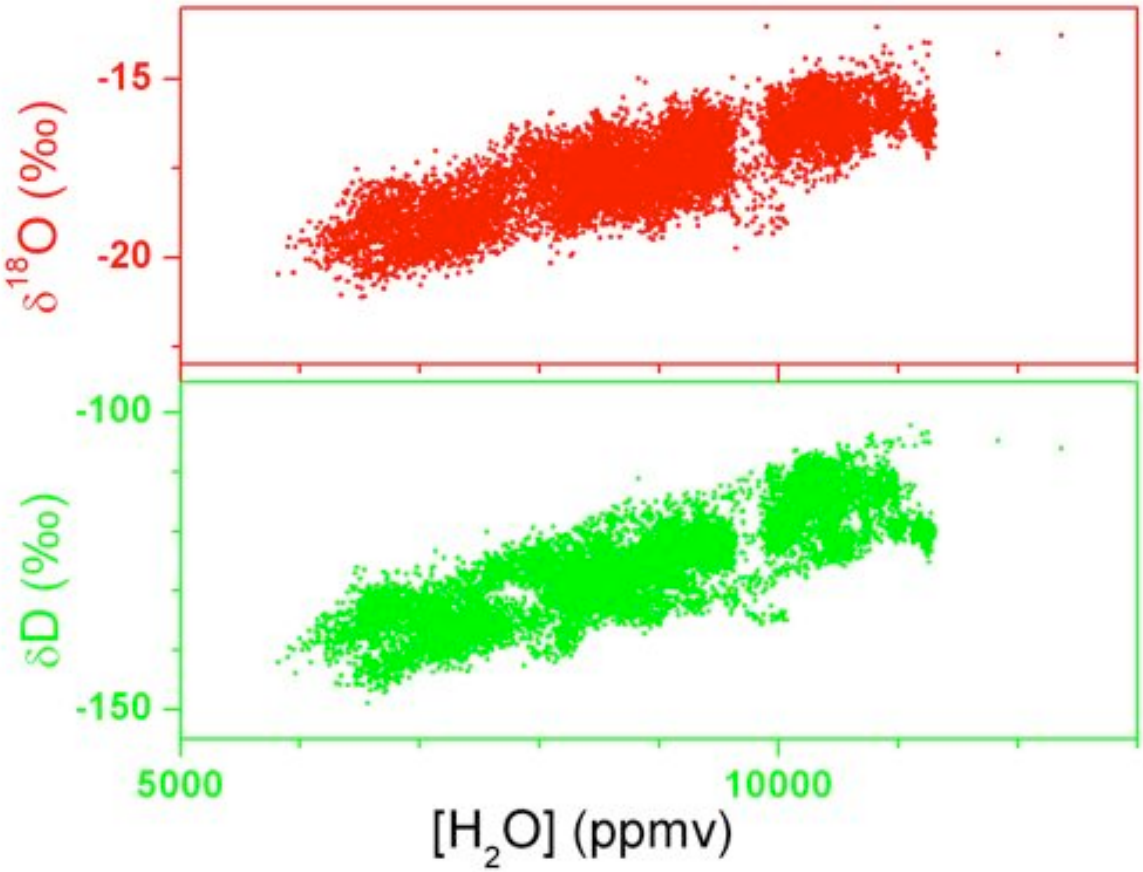
Continuous δ^2H, $\delta^{18}O$, H_2O recorded at dual flux tower site



- Continuous unattended calibrated measurements in real time
- Fast, continuous measurements (2-Hz rate) of δ^2H, $\delta^{18}O$ and H_2O in air
- Dual flux tower in a semi-abandoned field near invasive pepperweed

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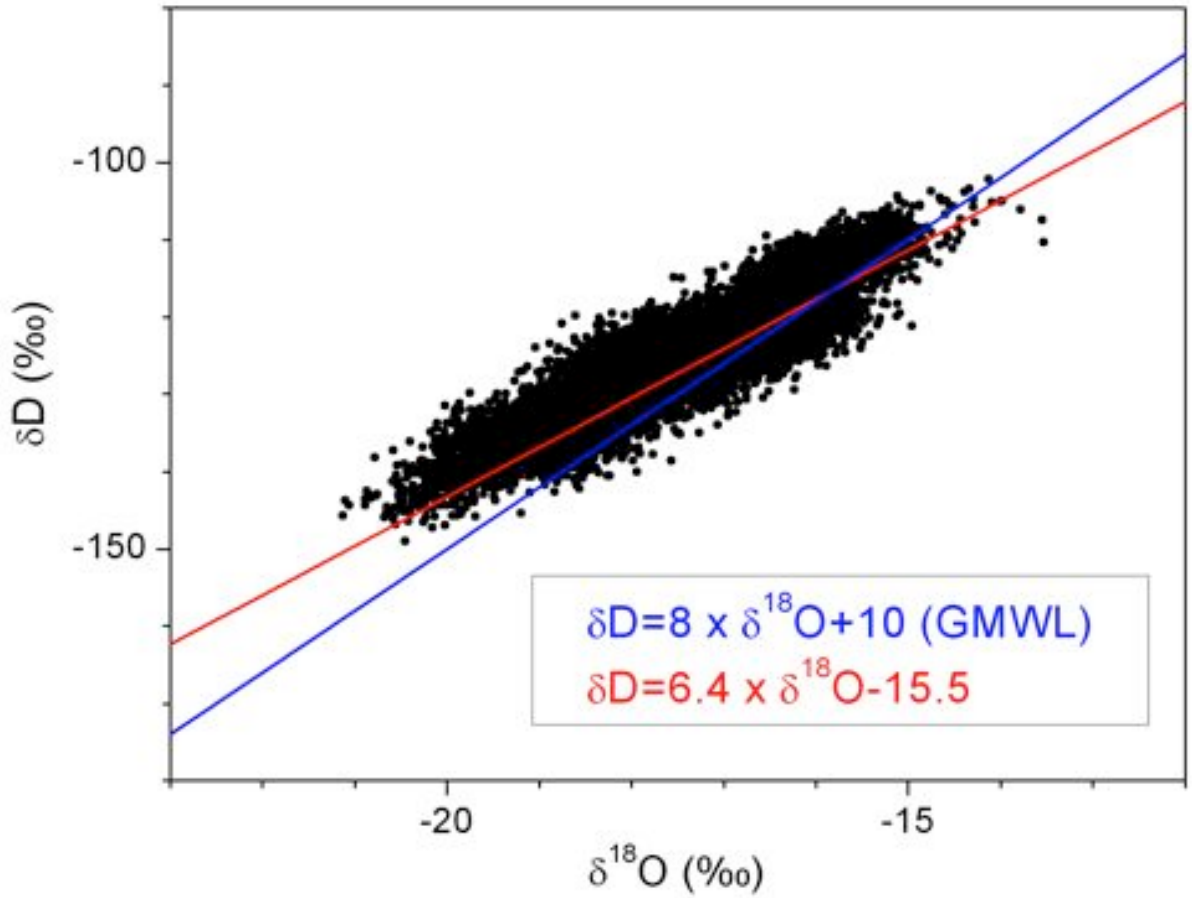
δ^2H, $\delta^{18}O$ and H_2O measurements in the field



- Continuous unattended absolute measurements at Sherman Island, CA
- Fast, continuous measurements (2-Hz rate) of δ^2H, $\delta^{18}O$ and H_2O in air
- Local $\delta^{18}O$, δ^2H measurements compared with Global Meteoric Water Line

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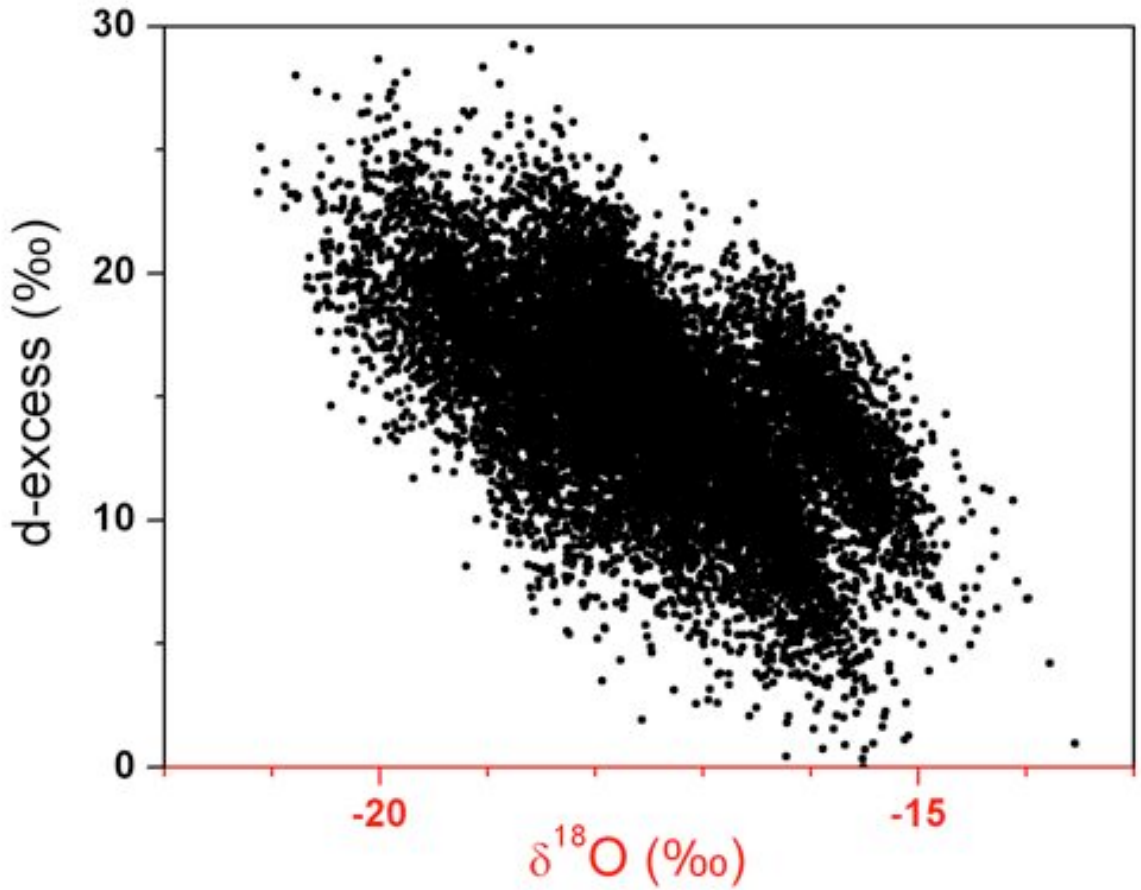
2-Hz measurements of δ^2H, $\delta^{18}O$, H_2O at dual flux site



- Continuous unattended absolute measurements at Sherman Island, CA
- Fast, continuous measurements (2-Hz rate) of δ^2H, $\delta^{18}O$ and H_2O in air
- Local $\delta^{18}O$, δ^2H provides d-excess measurements

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Measurements at CalNEX (June 2010) with CARB/NOAA/UCB

- N_2O, CO, CO_2, CH4 recorded by Mobile Lab from LGR
- Reference cylinders in Mobile Lab sampled periodically as validation checks
- Provides sensitive measurements while driving or stationery

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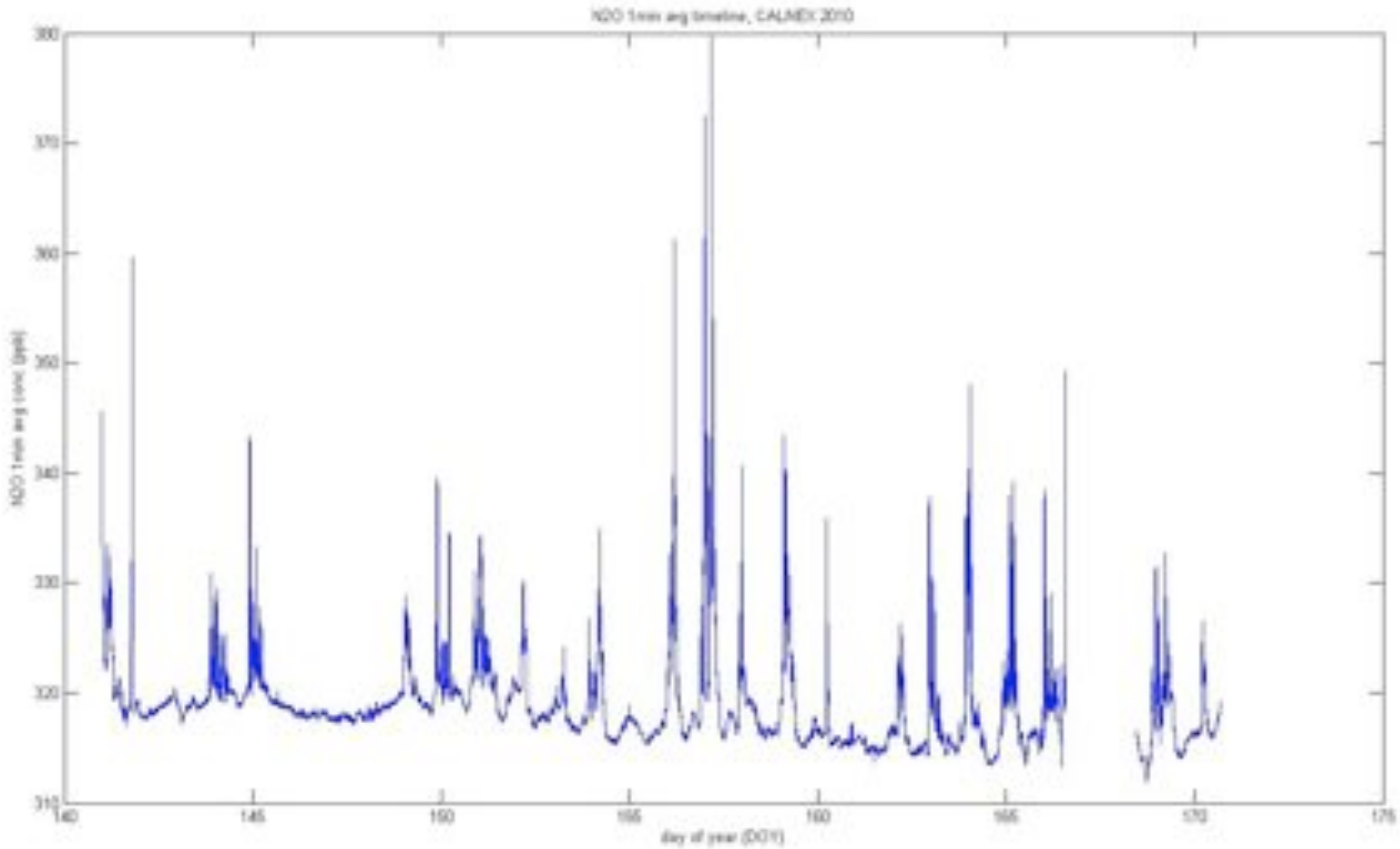
Measurements at CalNEX (June 2010) with CARB/NOAA/UCB



- N_2O recorded by LGR Mobile Lab

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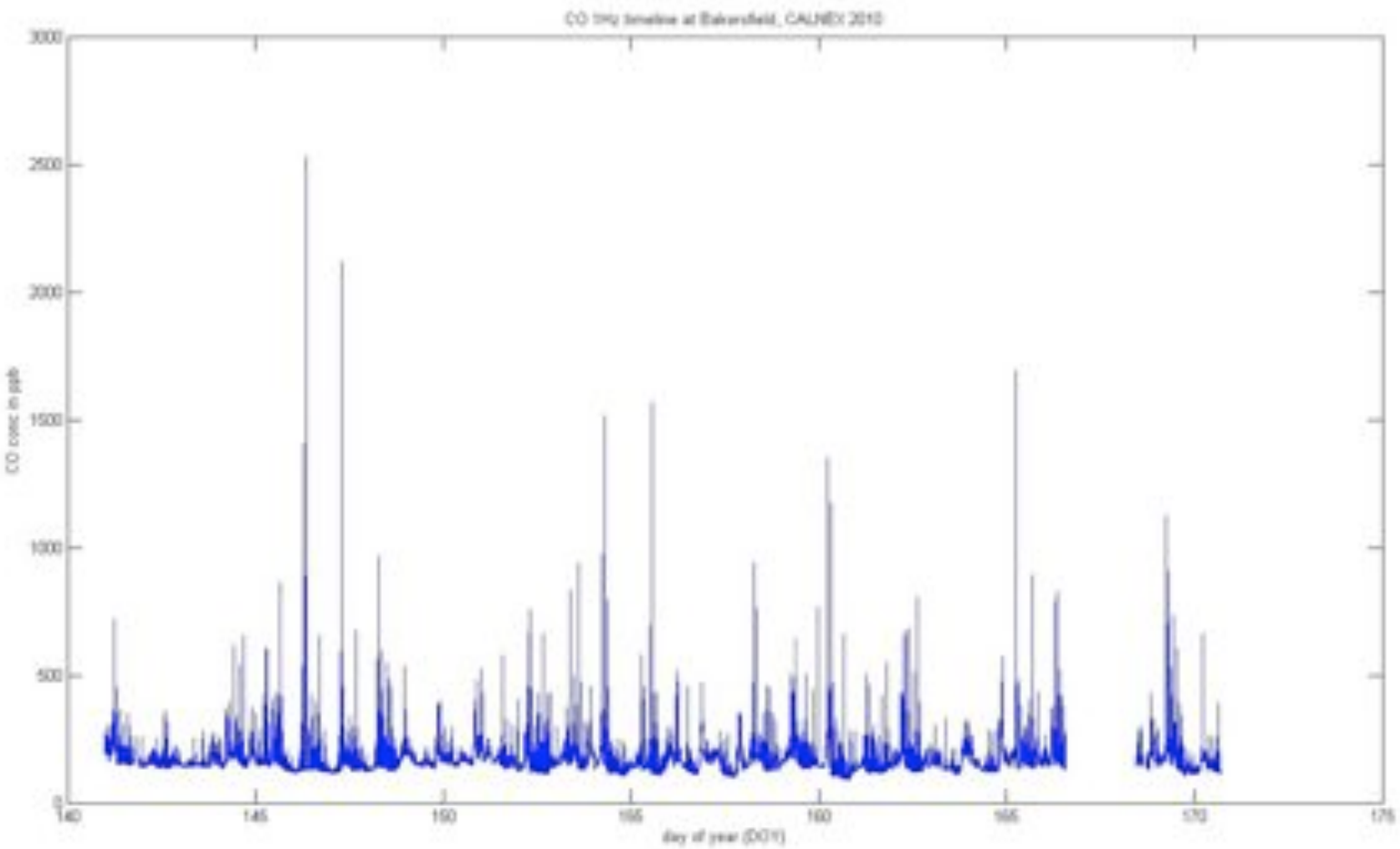
Measurements at CalNEX (June 2010) with CARB/NOAA/UCB



- CO recorded by LGR Mobile Lab

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Summary:
Novel Instruments Provide New Opportunities

- Fast, accurate, continuous real-time data in the field
- Measurements up to 20 Hz (fluxes)
- Precise, accurate over wide concentration ranges
- Measurements of discrete samples (via syringe)
- Measurements of $\delta^{13}C$, $\delta^{2}H$, $\delta^{18}O$ at 2 Hz
- Low power requirements

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Results and Conclusions

LGR's Mobile Emissions Lab provides:

- continuous measurements of multiple gases -- anywhere.
- measurements while stationary and moving.
- CH_4 (and $\delta^{13}CH_4$), CO_2 (and $\delta^{13}CO_2$), N_2O, CO, H_2O (and $\delta^{2}H$, $\delta^{18}O$), C_2H_2, NH_3, H_2S, …
- on-board batteries allow operation without external power.
- measurements of many species simultaneously

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Results and Conclusions

LGR's Mobile Emissions Lab provides:

- long-term monitoring of mobile and fixed-location (fugitive) emissions and pollutant sources with high accuracy, precision, sensitivity in real time.

- ability of regulatory agencies, monitoring stations, scientists to report temporally and spatially resolved measurements of GHG and pollutants for compliance monitoring, as well as cap and trade, at any location.

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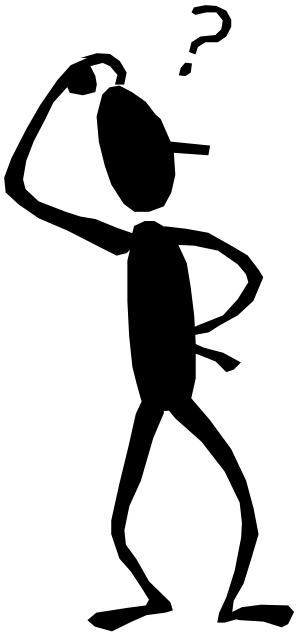
Questions/Comments



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