NOAA research to understand and quantify the atmospheric impacts of oil and natural gas operations

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Overview of NOAA research about O&G impacts

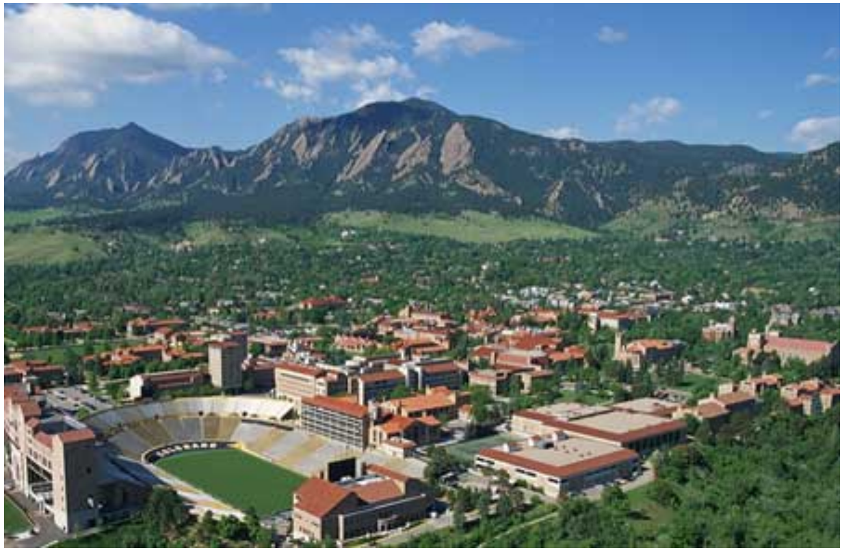
NOAA measurements and analysis focus on two O&G basins

- Denver-Julesburg Basin (DJB), Colorado :
  - Monitoring and source sampling since 2008
  - Field intensives in 2008, 2011, 2012
- Uintah Basin, Utah:
  - Intensives in 2012, 2013

Note that the data shown, unless otherwise indicated, are preliminary.

➢ Please do not cite unless you have received explicit permission of the relevant investigators.

NOAA Research Facilities and Partners
Boulder, Colorado



National Oceanic & Atmospheric Administration (NOAA)

- Earth System Research Laboratory (ESRL), http://www.esrl.noaa.gov
  - Chemical Sciences Division (CSD), http://www.esrl.noaa.gov/csd/
  - Global Monitoring Division (GMD), http://www.esrl.noaa.gov/gmd/
  - Physical Sciences Division (PSD), http://www.esrl.noaa.gov/psd/

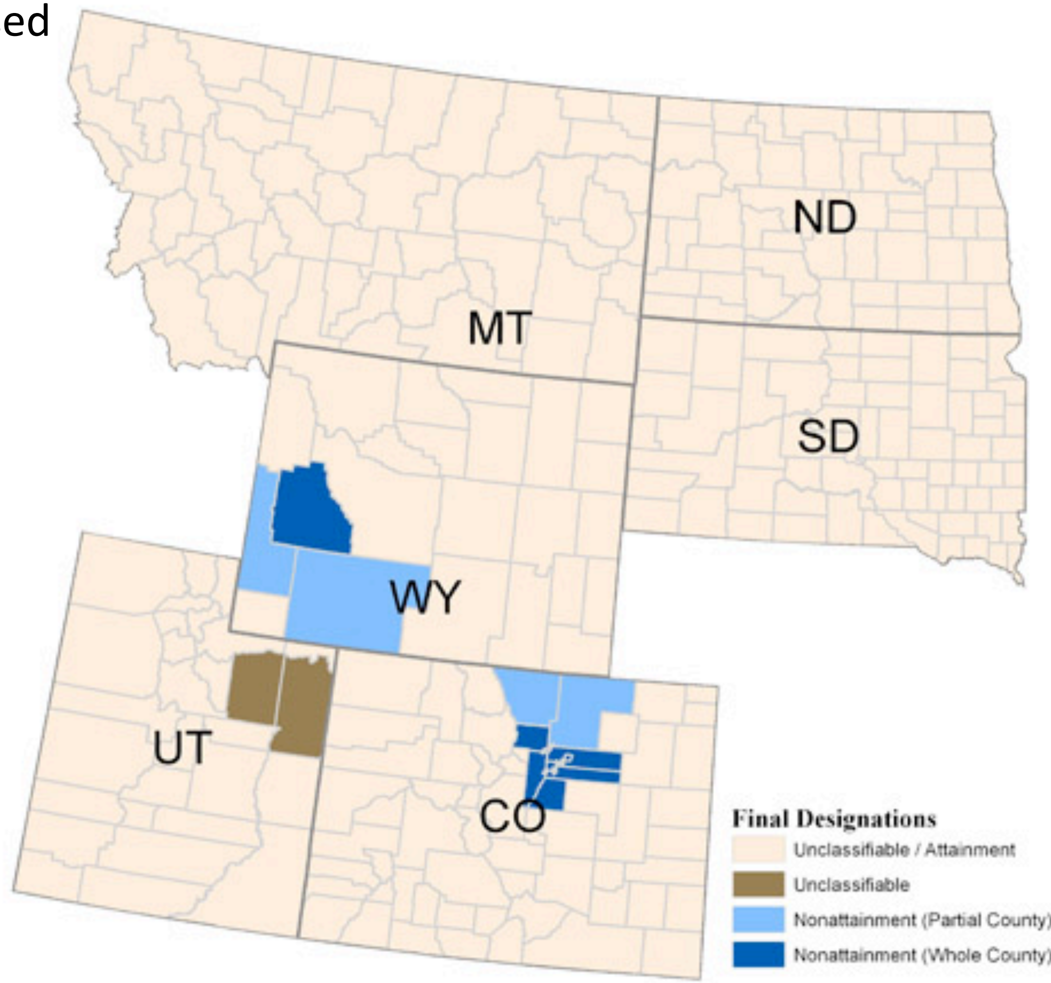
University of Colorado (CU)

- Cooperative Institute for Research in Environmental Sciences (CIRES), http://cires.colorado.edu/
- Institute for Arctic and Alpine Research (INSTAAR), http://instaar.colorado.edu/

Where is air quality impacted?

EPA Region 8 Final Designations for Surface Ozone

Released April 2012



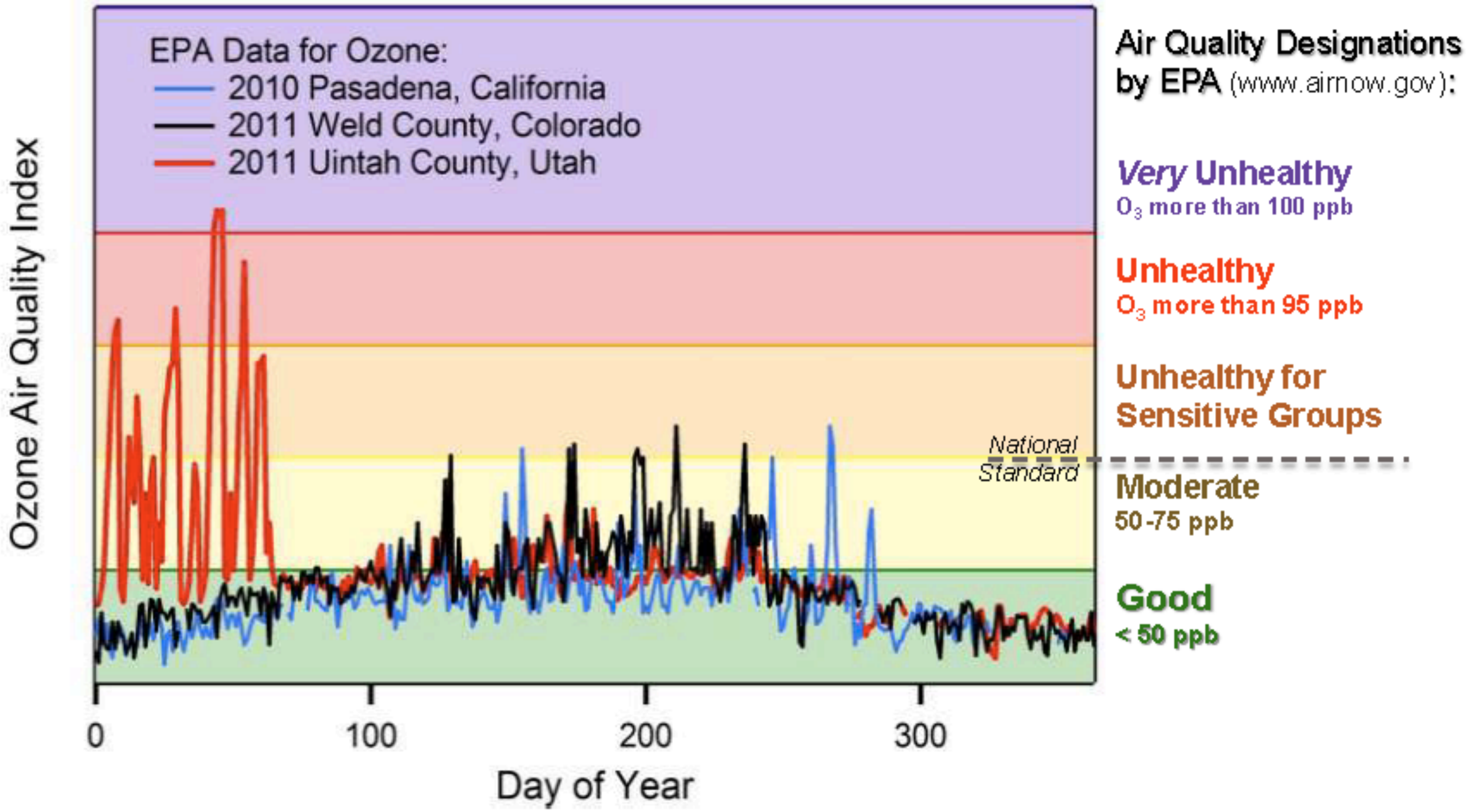
Wyoming:
Lincoln, Sublette, Sweetwater are non-attainment in the winter time

Colorado:
Adams, Arapahoe, Boulder, Broomfield, Denver, Douglas, Jefferson, Larimer, Weld are non-attainment in the summer time

Utah:
Uintah and Duchesne unclassifiable

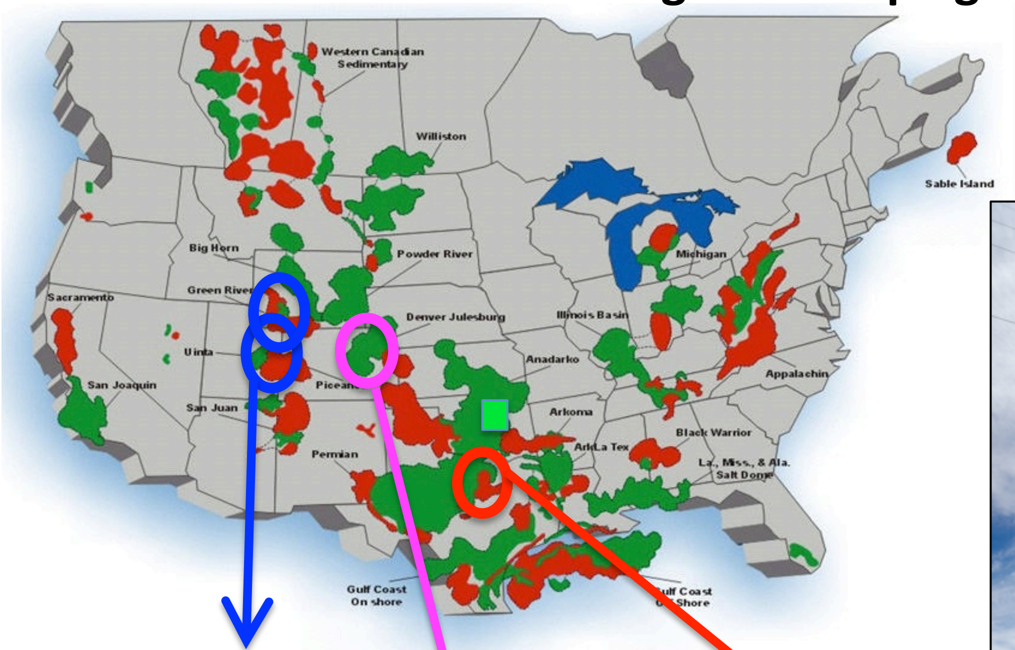
Photochemical formation of ozone (O_3)

Ground-level O_3 degrades air quality



Past and Ongoing NOAA Monitoring & Field Intensives

Map of unconventional oil and gas reservoirs and dates of NOAA monitoring and campaigns



Wyoming 2008
Utah 2012, 2013

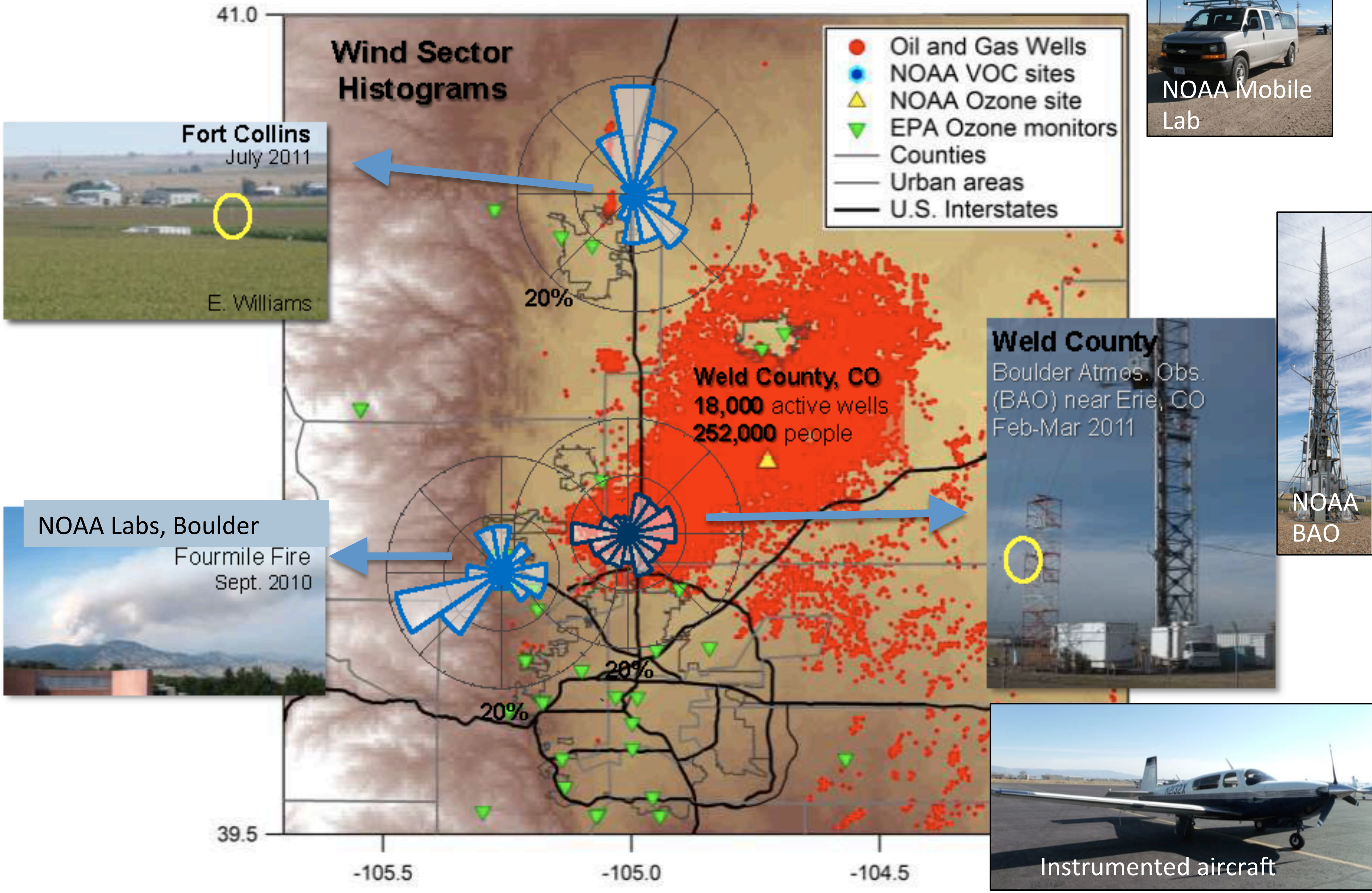
Colorado
2008-present

Texas
2013

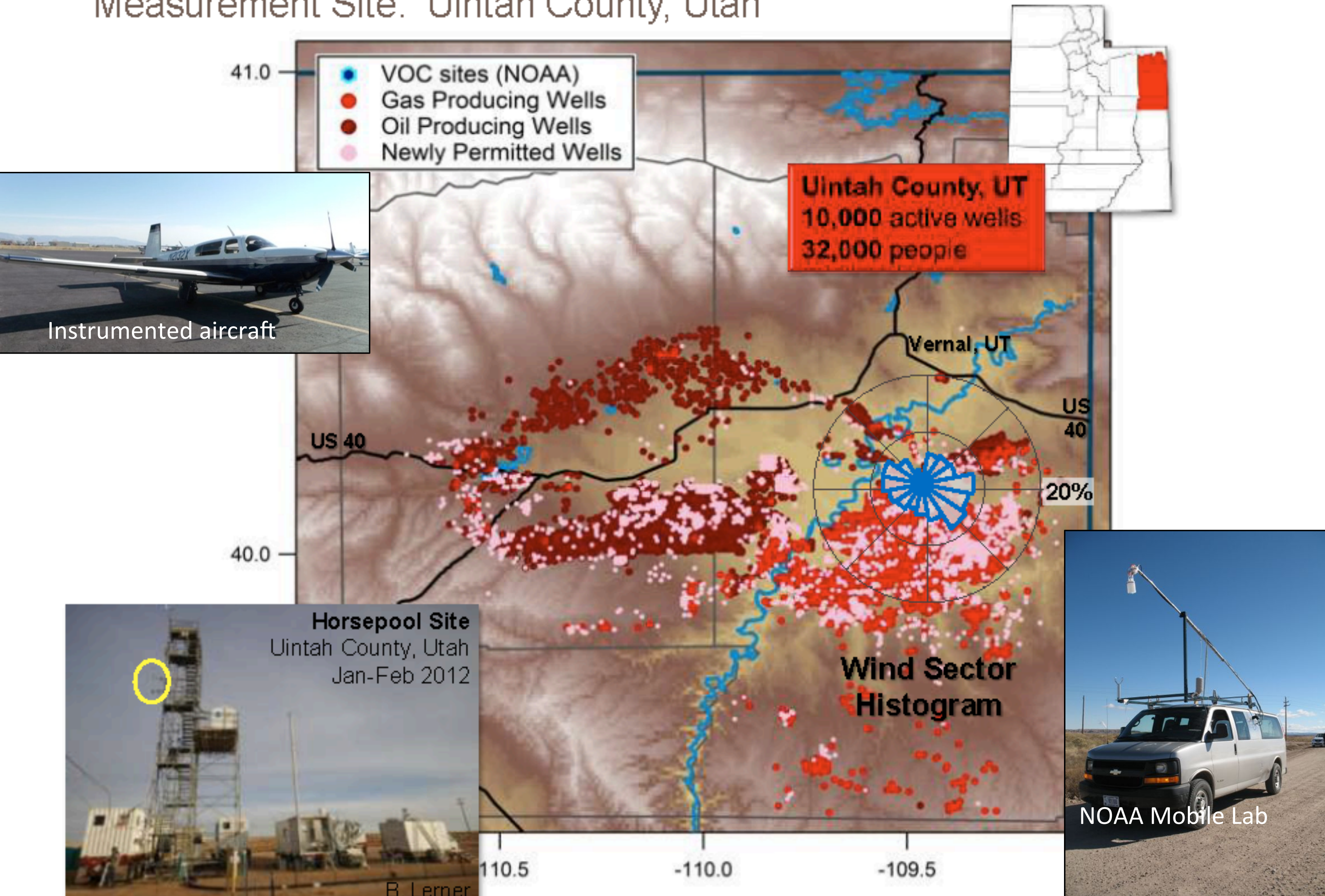
Research Questions:

- Emissions from O&G
- Summertime ozone drivers
- Wintertime ozone drivers

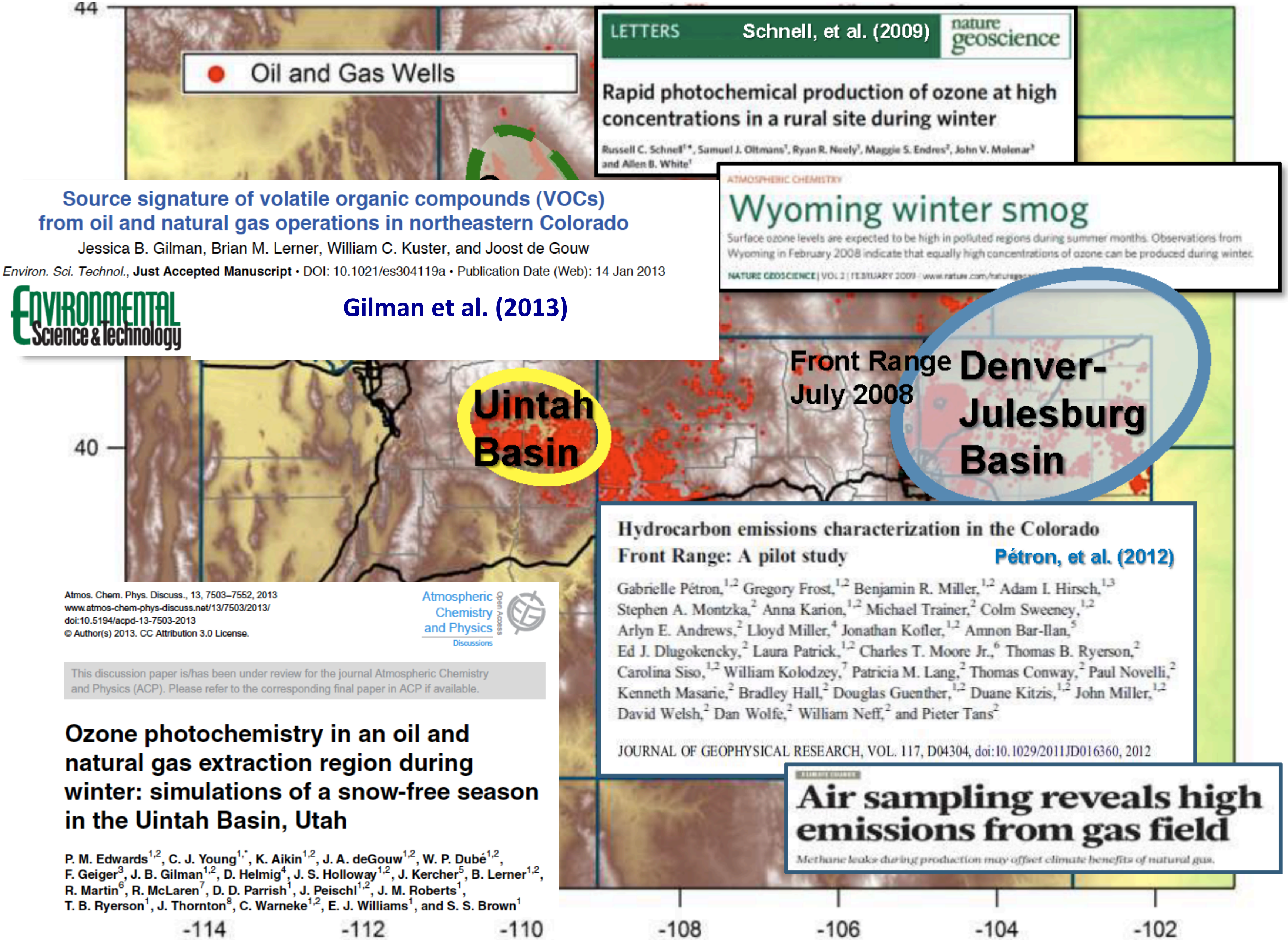
Measurement Sites: Colorado Front Range



Measurement Site: Uintah County, Utah



CO/UT NOAA O&G Studies in Literature



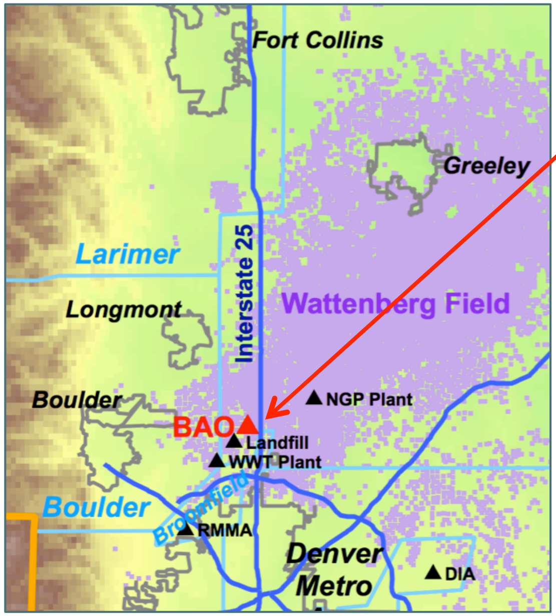
NOAA measurements and analysis of emissions in O&G regions

Some examples of top-down emissions analysis

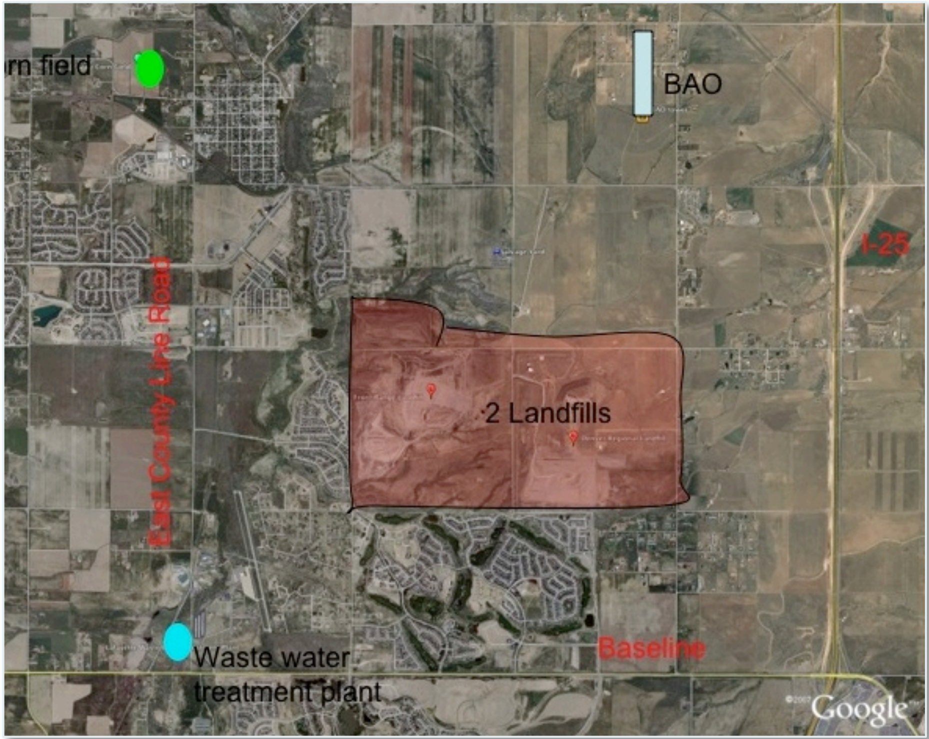
- Top-down = based on atmospheric observations
- Use coincident measurements of multiple species
- Regional emissions quantification
- Source identification and characterization

1. Tall tower + mobile sampling, DJB summer 2008 (Pétron et al., 2012)
2. Surface sampling, DJB winter 2011/ Uintah winter 2012 (Gilman et al., 2013)
3. Aircraft sampling, Uintah winter 2012/ DJB spring 2012 (Karion, Sweeney, et al.)
4. Mobile sampling, Uintah winter 2012 (Pétron, Geiger/ Warneke, Edwards, et al.)

Boulder Atmospheric Observatory

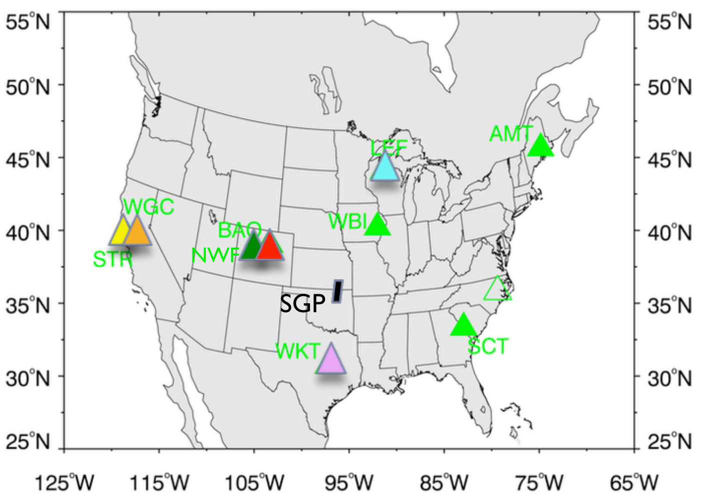


- 300 meter tall tower
- located in Erie, Weld County
- Instrumented with LICOR (CO_2) and TECO (CO) in April 2007: sampling from 3 intake heights (22m, 100m, 300m)
- 30 sec- Met Data at three levels
- Equipped to collect discrete air samples from 300 meter level in August 2007. Analyses performed in NOAA Boulder lab.

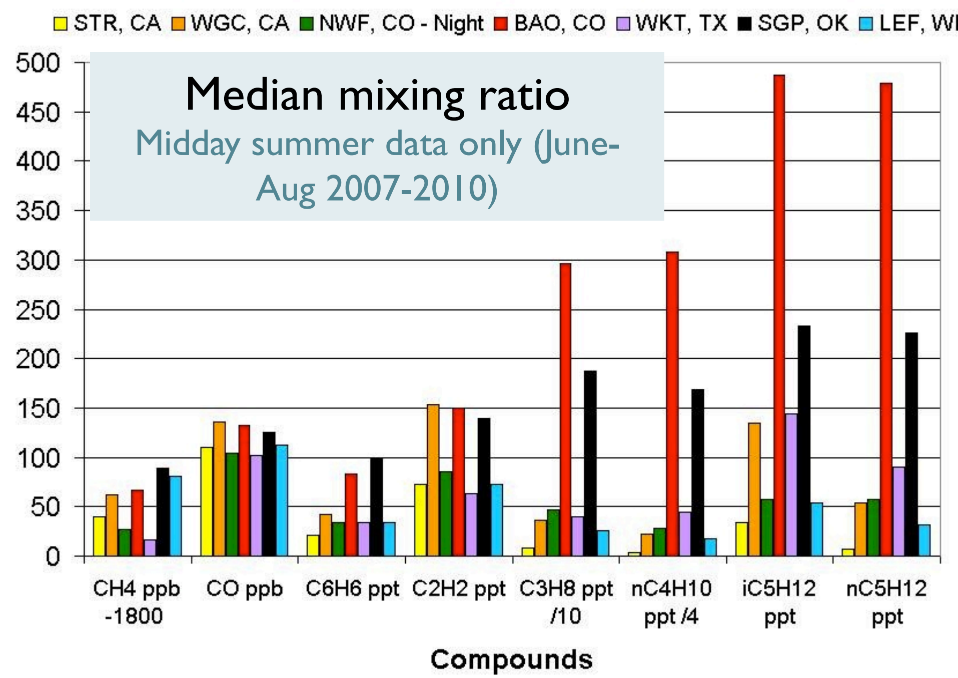


http://www.esrl.noaa.gov/gmd/ccgg/towers/index.html

BAO: Distinct Alkane Signature



NOAA Tall Tower Measurement and Sampling Network (PI Arlyn Andrews)

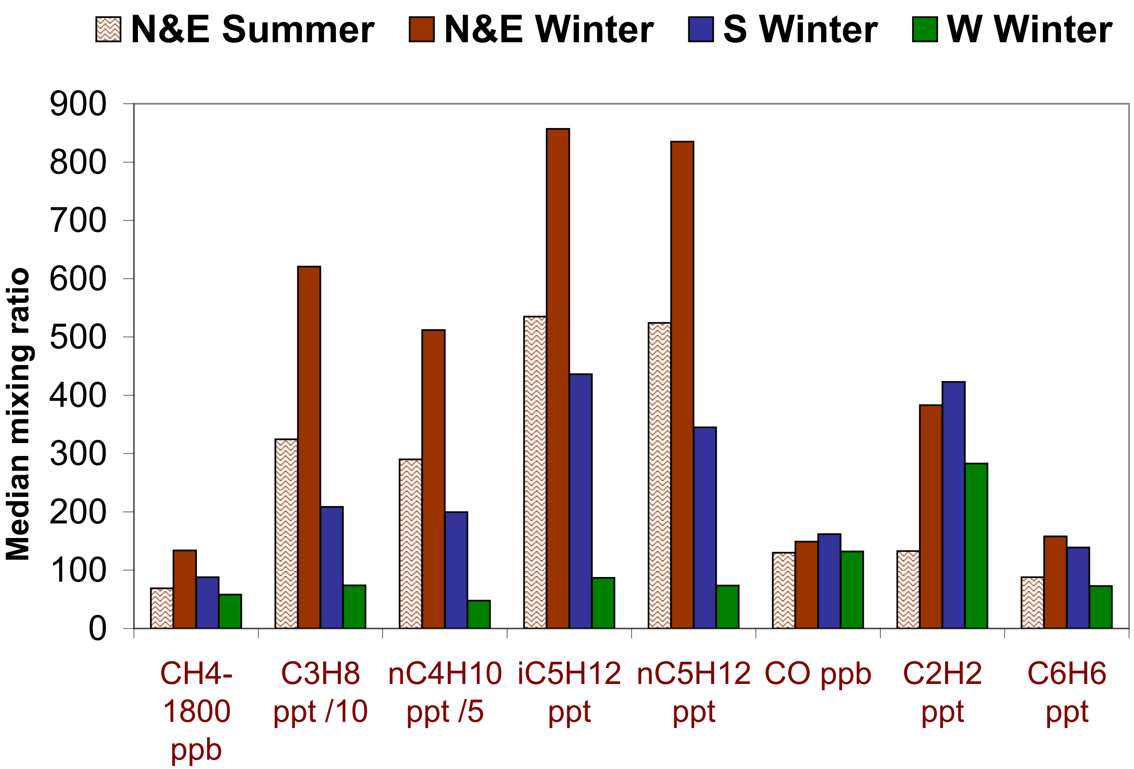


Air samples collected at the BAO and SGP* have a strong alkane signature.

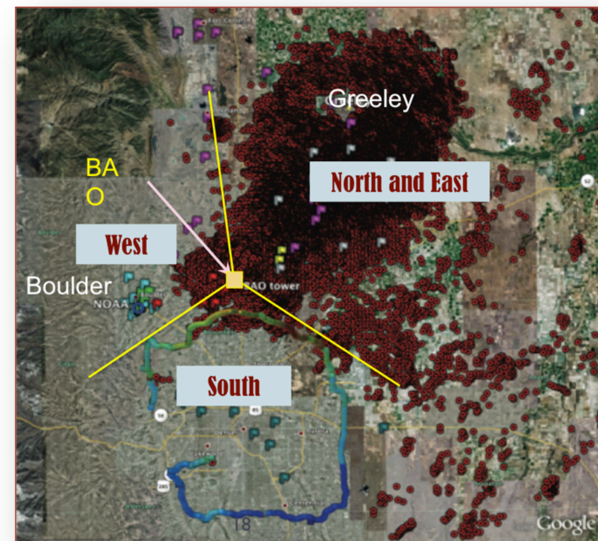
* SGP is a NOAA aircraft site in Northern Oklahoma. Samples collected below 650 meters were used for this analysis.

Pétron et al., JGR-Atmospheres, doi:10.1029/2011JD016360, *2012*

BAO: Data Filtered By Wind Sector



Strongest alkane signature in North & East wind sector



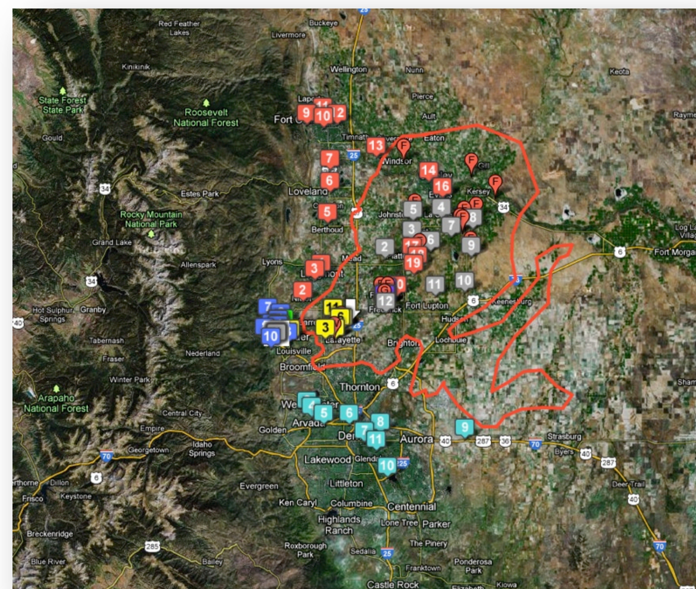
Midday Data from the BAO (August 2007-April 2010).
Wind sector designation based on 30-min average (prior to sample collection) wind direction and wind speed (data retained if |w.speed|> 2.5 m/s).

Pétron et al., JGR-Atmospheres, doi:10.1029/2011JD016360, *2012*

Mobile lab sampling of methane source chemical signatures in the Front Range

- Mobile Platform to sample close to sources
- High-frequency stable analyzers to detect plumes and target flask sampling
- Discrete air sampling for multi-species chemical analyses in the NOAA lab

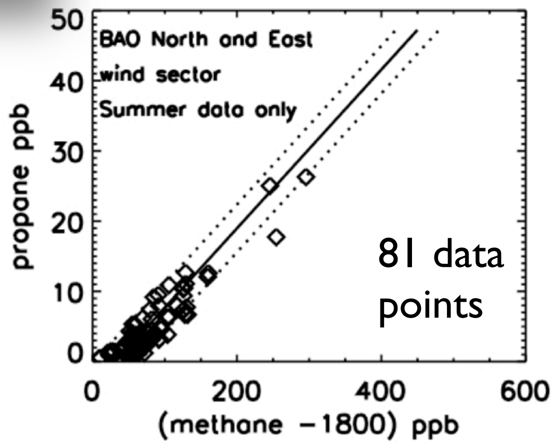
Toyota Prius equipped with:
○ Fast response CO_2 and CH_4 analyzer (Picarro)
○ Real Time Display of Measurements
○ GPS
○ Programmable Flask Package (PFP with 12 sampling glass flasks) and Programmable Compressor Package (PCP) with GPS



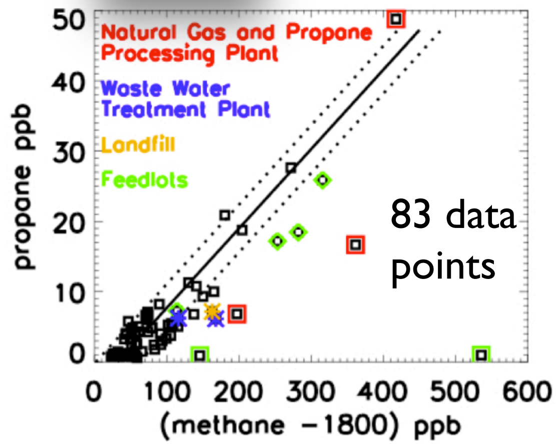
Pétron et al., JGR-Atmospheres, doi:10.1029/2011JD016360, *2012*

Very Strong Correlations between Alkanes

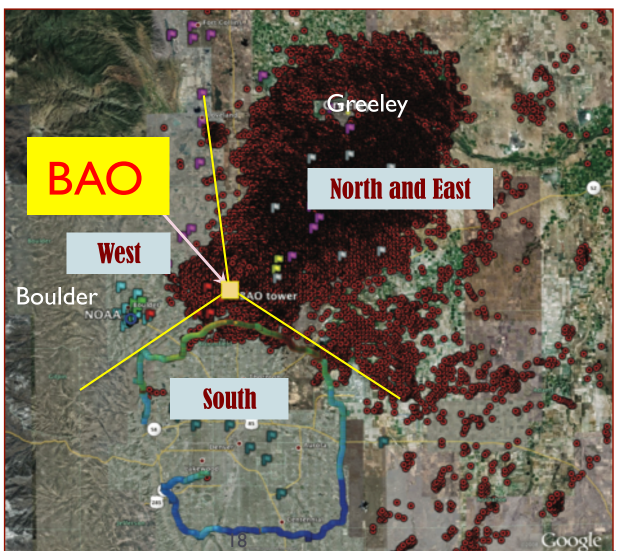
BAO N&E wind sector



Mobile Lab



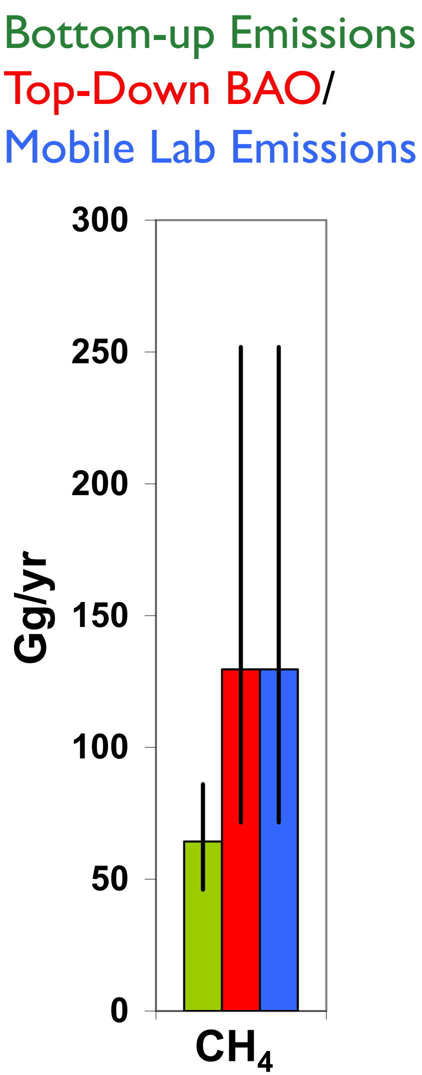
- Methane is strongly correlated with propane.
- Samples collected close to feedlots, a landfill, a waste water treatment plant have enhanced methane compared to the other samples.



We use the measured atmospheric propane-to-methane enhancement ratios observed at the BAO tall tower and at the surface across the Front Range to evaluate the proportion of flashing and venting emissions.

Pétron et al., JGR-Atmospheres, doi:10.1029/2011JD016360, *2012*

Top-down Estimates versus Inventory



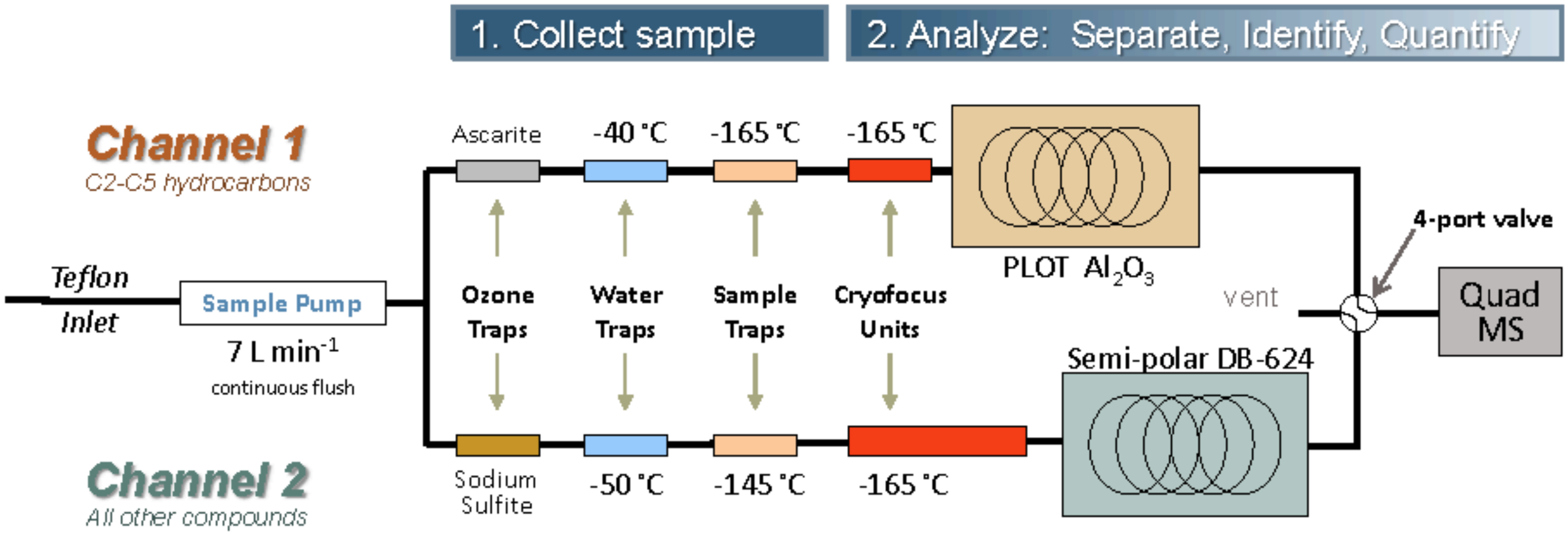
Constraints	Bottom-up inventory	Enhancement Ratio Method
Flash emissions for tanks (total VOC mass) in O_3 NAA	X	X
Fugitive emissions estimates (volume of raw gas)	X	
Raw gas composition profile (average or subset)	X	X
Flash emissions composition profile	X	X
Atmospheric enhancement ratios		X

- Fugitives emissions of CH_4 are likely underestimated in bottom-up inventory for 2008.
- Still very large uncertainties on top-down estimates.
- **We need truly independent methods to evaluate inventories!**

Pétron et al., JGR-Atmospheres, doi:10.1029/2011JD016360, *2012*

VOC instrumentation

Gas chromatograph-Mass Spectrometer (GC-MS)



- **5 min sample collection**
  - 350 mL sample for each channel
- **25 min sample analysis**
- **60+ species** analyzed
  - C2 to C11 hydrocarbons
  - C6-C9 aromatics, OVOCs, etc.

GC-MS Instrument Specs*:

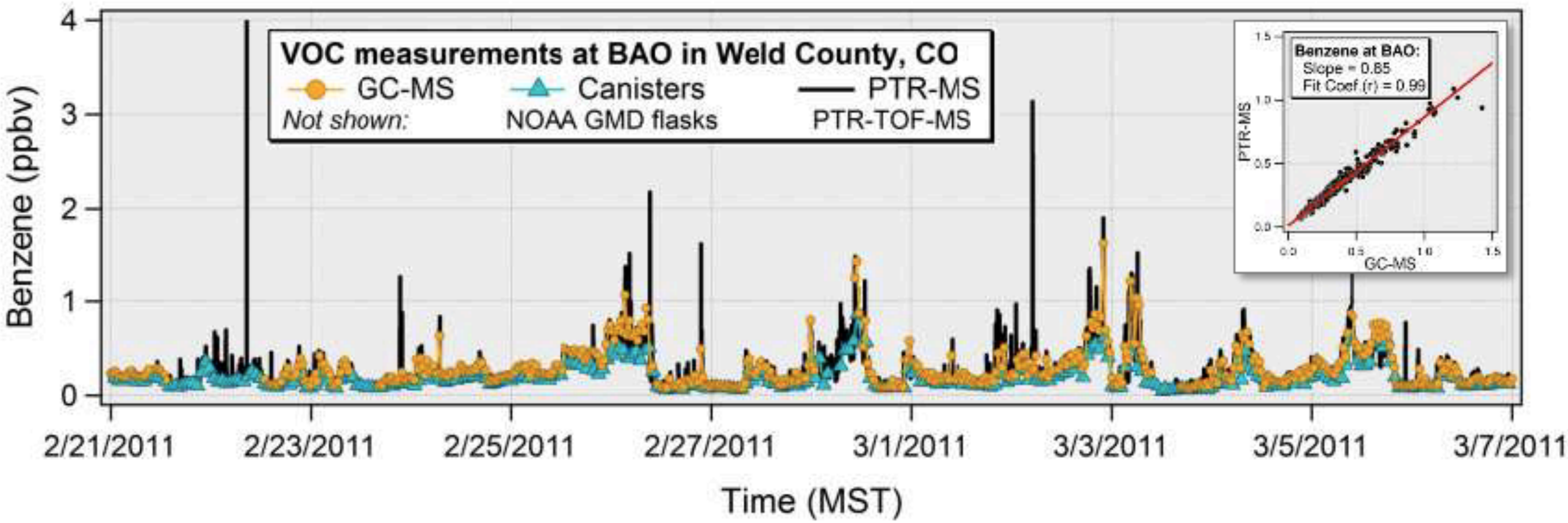
Limit of detection	< 1 - 10 ppt
Precision	**0.5% to 5%**
Accuracy	**15 to 25%**

* values are compound dependent

Instrument Details: Gilman, et al. (2010), *Atmos. Chem. Phys.*, **10** (21), p. 10223

VOC instrumentation in Colorado study

Summary of all measurement techniques



Discrete measurements: In-situ Gas Chromatograph-Mass Spectrometer (GC-MS) — *NOAA CSD*
Whole air canister samples analyzed by GC-MS — *Appalachian St. Univ.*

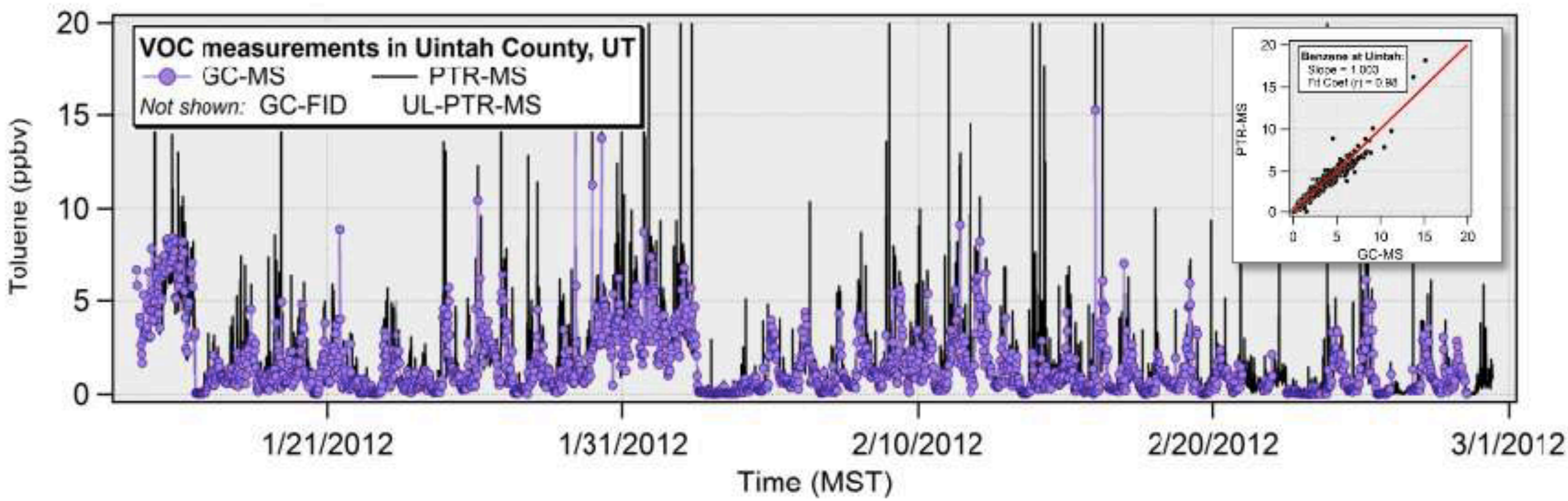
Continuous measurements: Proton Transfer Reaction-Mass Spectrometer (PTR-MS) — *NOAA CSD*
Proton Transfer Reaction-Time of Flight (TOF)-Mass Spec. — *NCAR*

Long-term measurements: Programmable Flask Package (PFP) at 300m at BAO — *NOAA GMD*

Gilman et al., Environ. Sci.Technol., 2013

VOC instrumentation in Utah Study

Summary of all measurement techniques



Discrete measurements:	In-situ Gas Chromatograph-Mass Spectrometer (GC-MS)	*NOAA CSD*
	Whole air canister samples analyzed by GC-MS	*Appalachian St. Univ.*
	In-situ GC-FID and Total Non-Methane Hydrocarbons	*Utah State Univ.*
	Programmable Flask Package (PFP) in mobile lab	*NOAA GMD*
Continuous measurements:	Proton Transfer Reaction-Mass Spectrometer (PTR-MS)	*NOAA CSD*
	Proton Transfer Reaction-Time of Flight (TOF)-Mass Spec.	*NCAR*
	Ultra Light Weight PTR-MS in mobile lab	*Karlsruhe Inst. Tech.*
Long-term measurements:	Programmable Flask Package (PFP) at 300m at BAO	*NOAA GMD*

Gilman et al., Environ. Sci. Technol., 2013

Emission Source Comparison

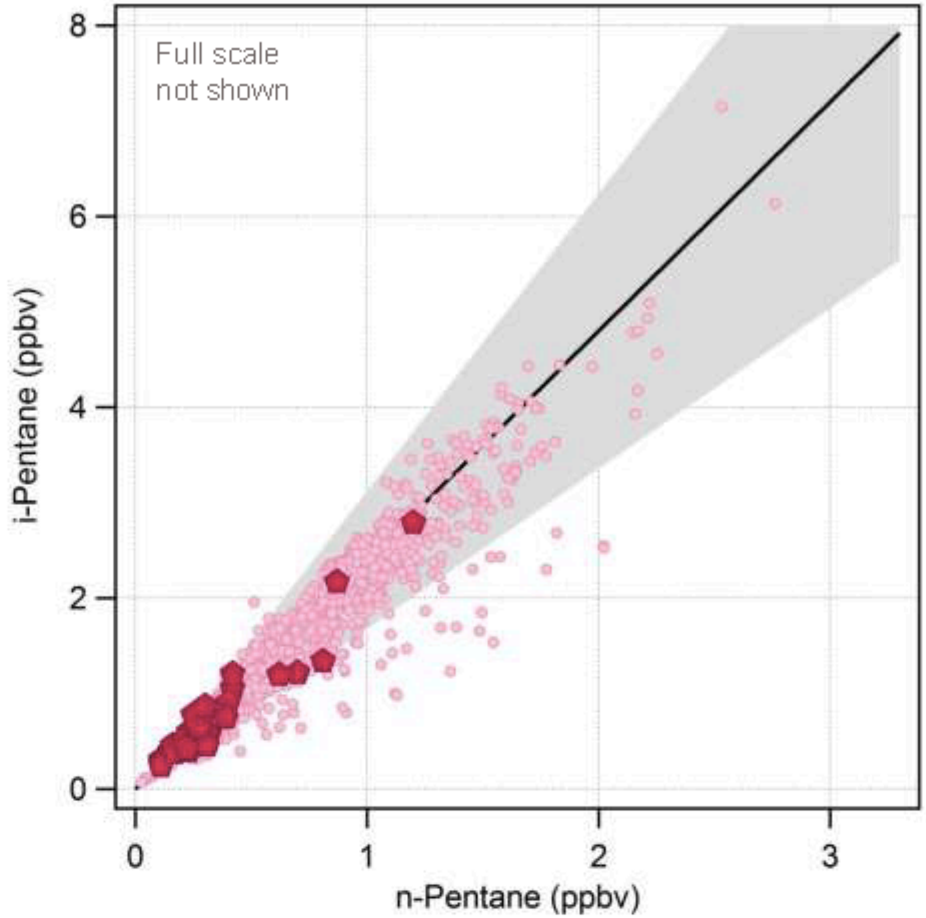
Gasoline combustion vs. Natural gas production

Enhancement Ratios (ER)
i-Pentane vs. n-Pentane:

Urban ER = 2.4 ± 20%
Pasadena, CA 2010
25 US Cities (Baker, 2008)

Enhancement Ratios

- ER is determined from <u>slope</u> of 2-sided linear regression
- Can be used to:
  - **Identify emission sources**
  - Examine oxidation chemistry
- Ratios minimize the effects of air mass mixing and dilution



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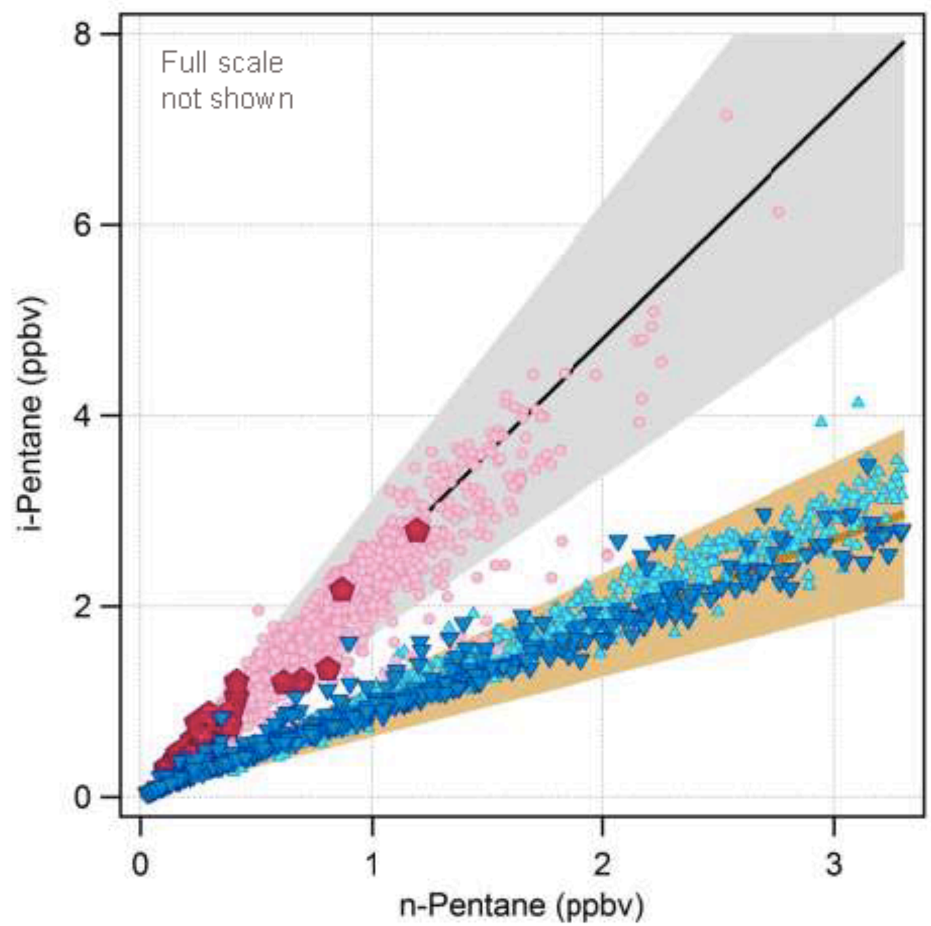
Natural Gas ER = 0.88 ± 20%
- Uintah Co., UT 2012
- Weld Co., CO 2011

i-Pentane is the most abundant hydrocarbon in gasoline

Source	iso- to n-Pentane Ratio*
Liquid gasoline	2.9
Tailpipe emissions	**2.6**

*Literature values from:
Schauer, et al. (2002) *Environ. Sci. Tech.*
Gentner, et al. (2009) *Environ. Sci. Tech.*



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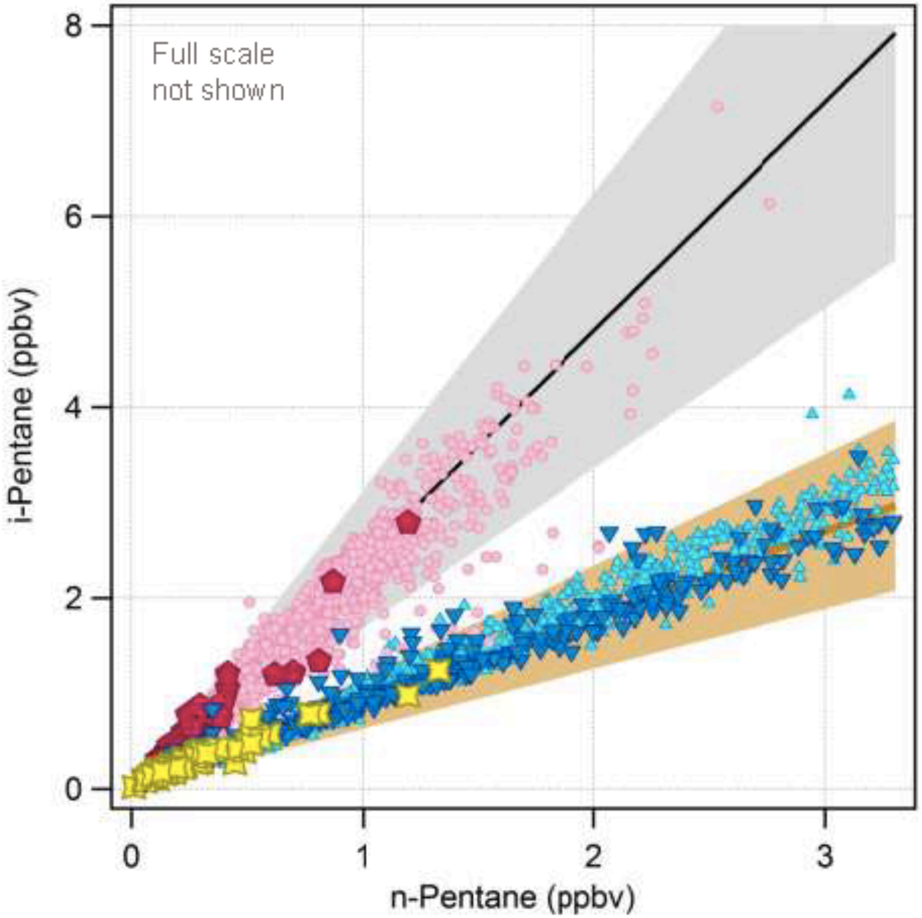
Natural Gas ER = 0.88 ± 20%
- Uintah Co., UT 2012
- Weld Co., CO 2011
- *Raw Natural Gas (COGCC rep.)*

Raw natural gas composition:

- Report for Colorado Oil and Gas Conservation Commission (COGCC)
- "Greater Wattenberg Area (GWA) Baseline Study" by LT Environmental
  - 77 natural gas wells
  - 6 different production zones
- *Values for natural gas samples are in plotted in mole percent (mol/mol × 100%)*
- *1 ppbv = nmol/mol = mol/mol × 10^{-9}*



http://cogcc.state.co.us/Library/DenverBasin/Greater_Wattenberg_Baseline_Study_Report_062007.pdf

Gilman et al., Environ. Sci. Technol., 2013

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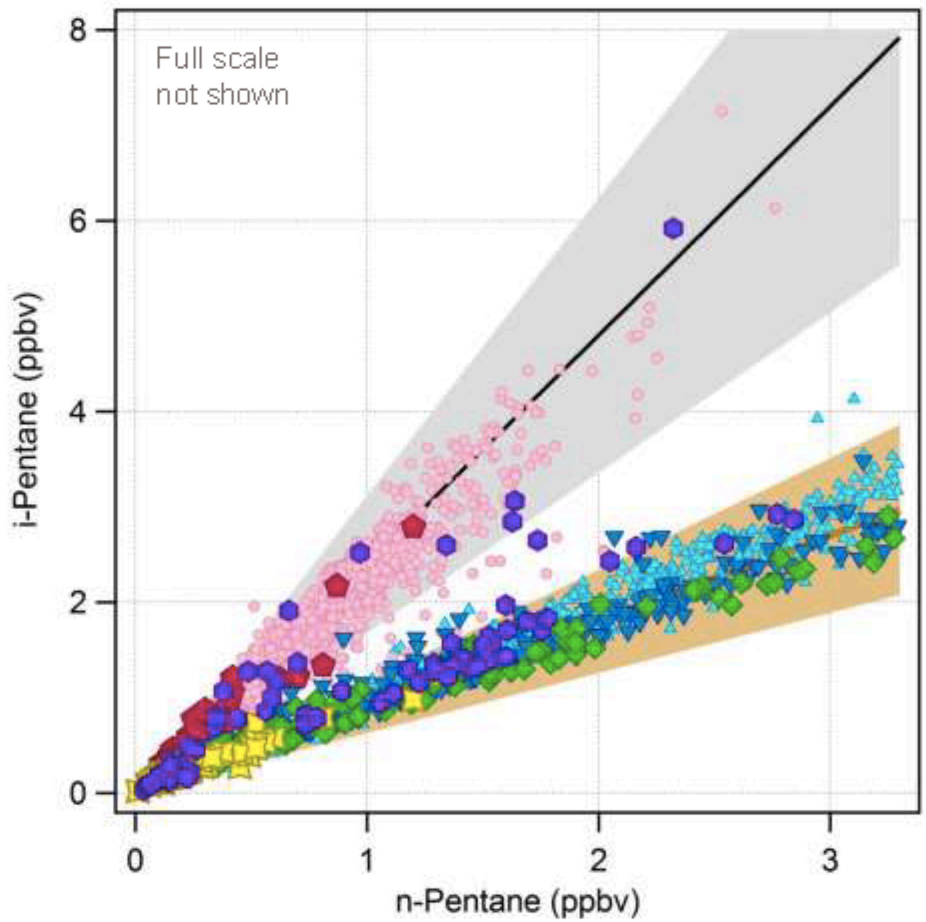
- Fort Collins, CO 2011
- Boulder, CO 2010



All of the measurement sites in Colorado's Front Range were influenced by emissions from oil and gas operations in the area

Gilman et al., Environ. Sci. Technol., 2013

Emission Source Comparison

Gasoline combustion vs. Natural gas production

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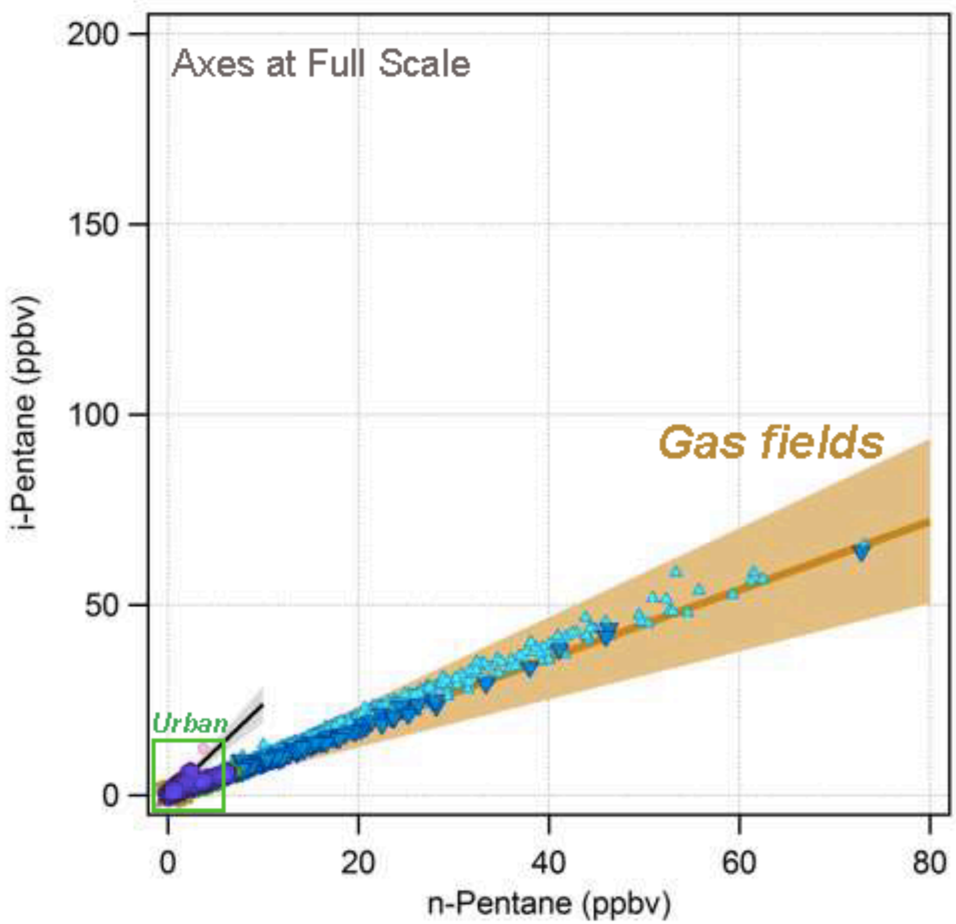
- Fort Collins, CO 2011
- Boulder, CO 2010

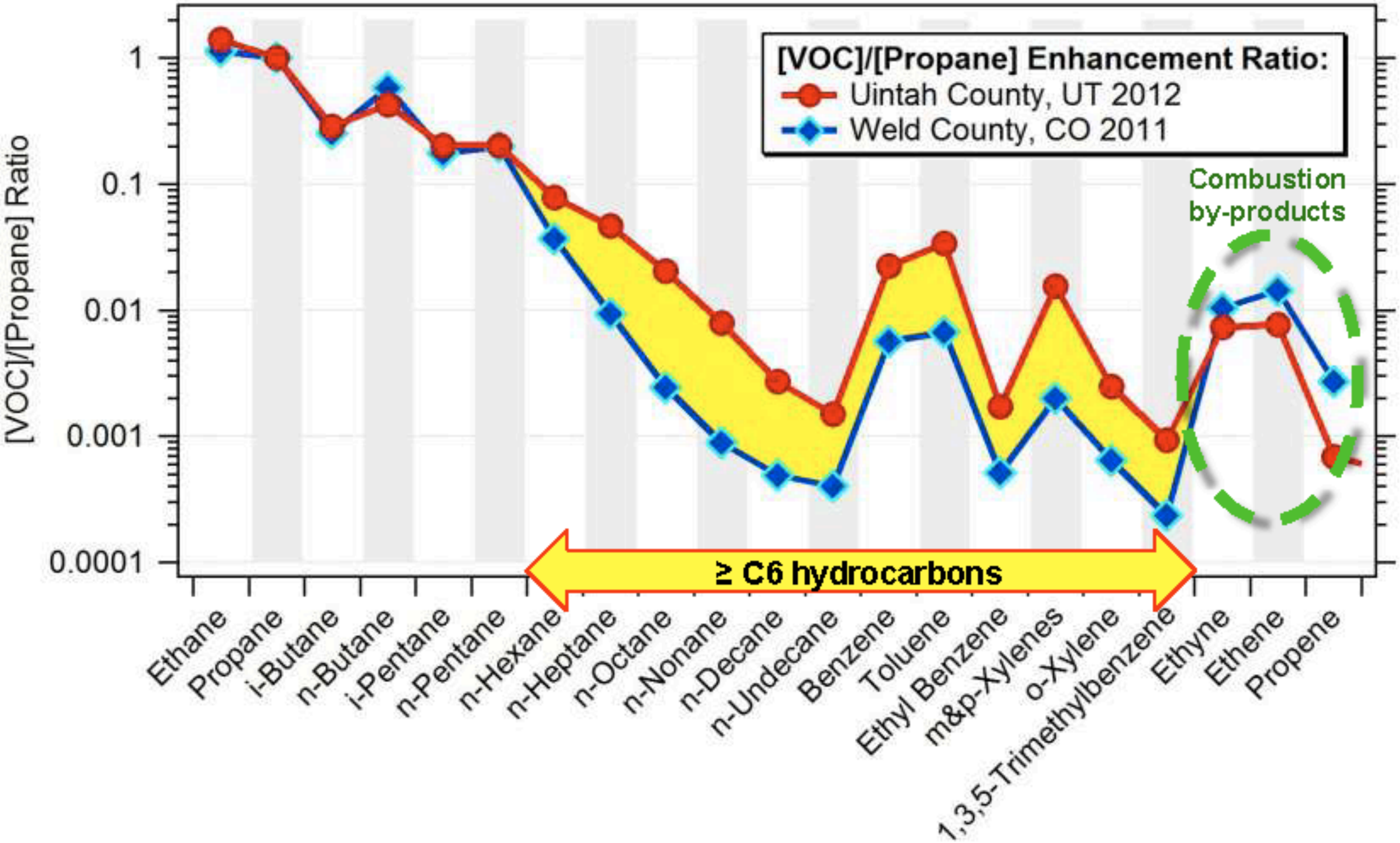
Very large enhancements of alkanes near the areas of oil and gas operations



Gilman et al., Environ. Sci. Technol., 2013

Emission Source Comparison

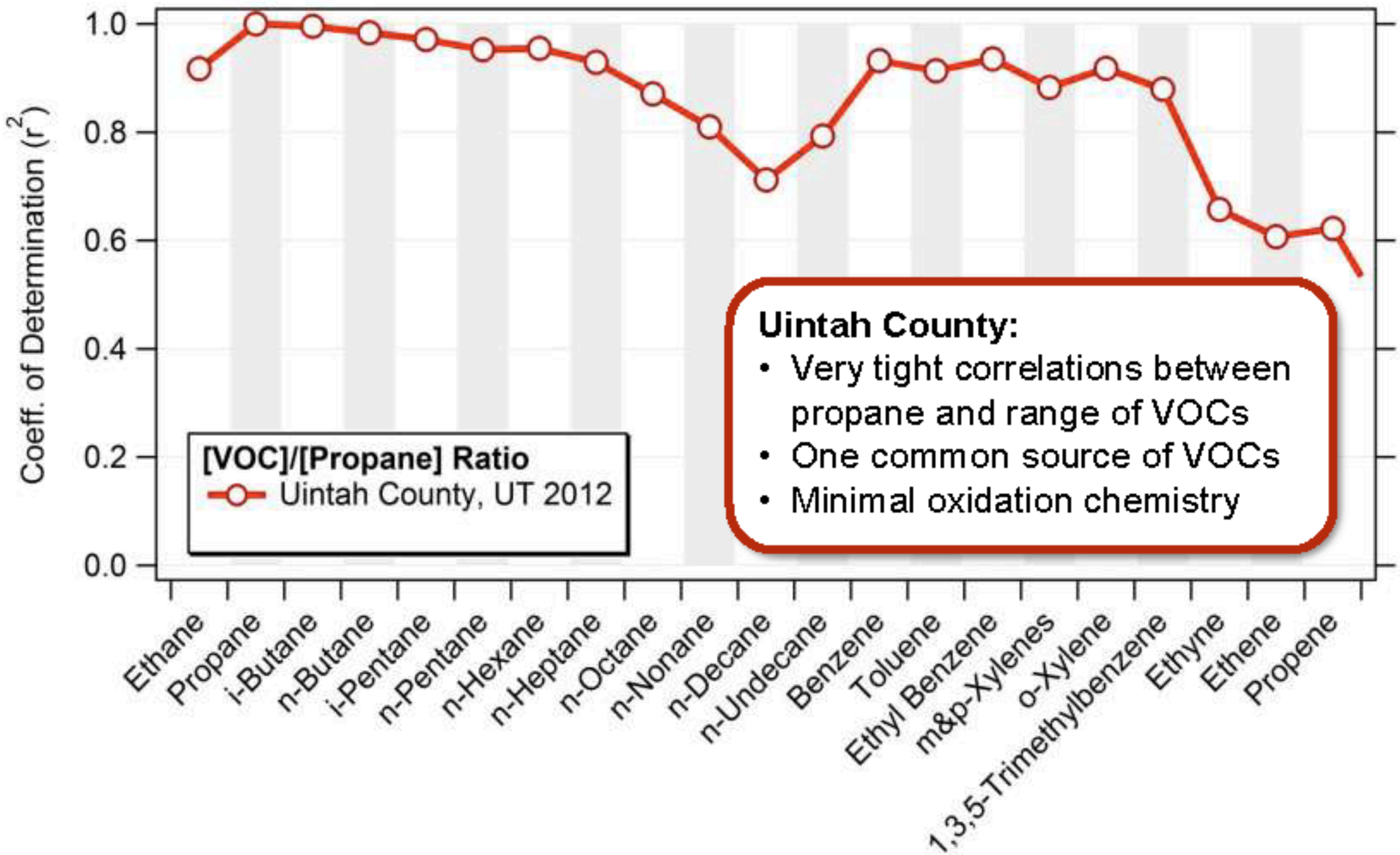
Colorado vs. Utah Composition



Gilman et al., Environ. Sci. Technol., 2013

Emission Source Comparison

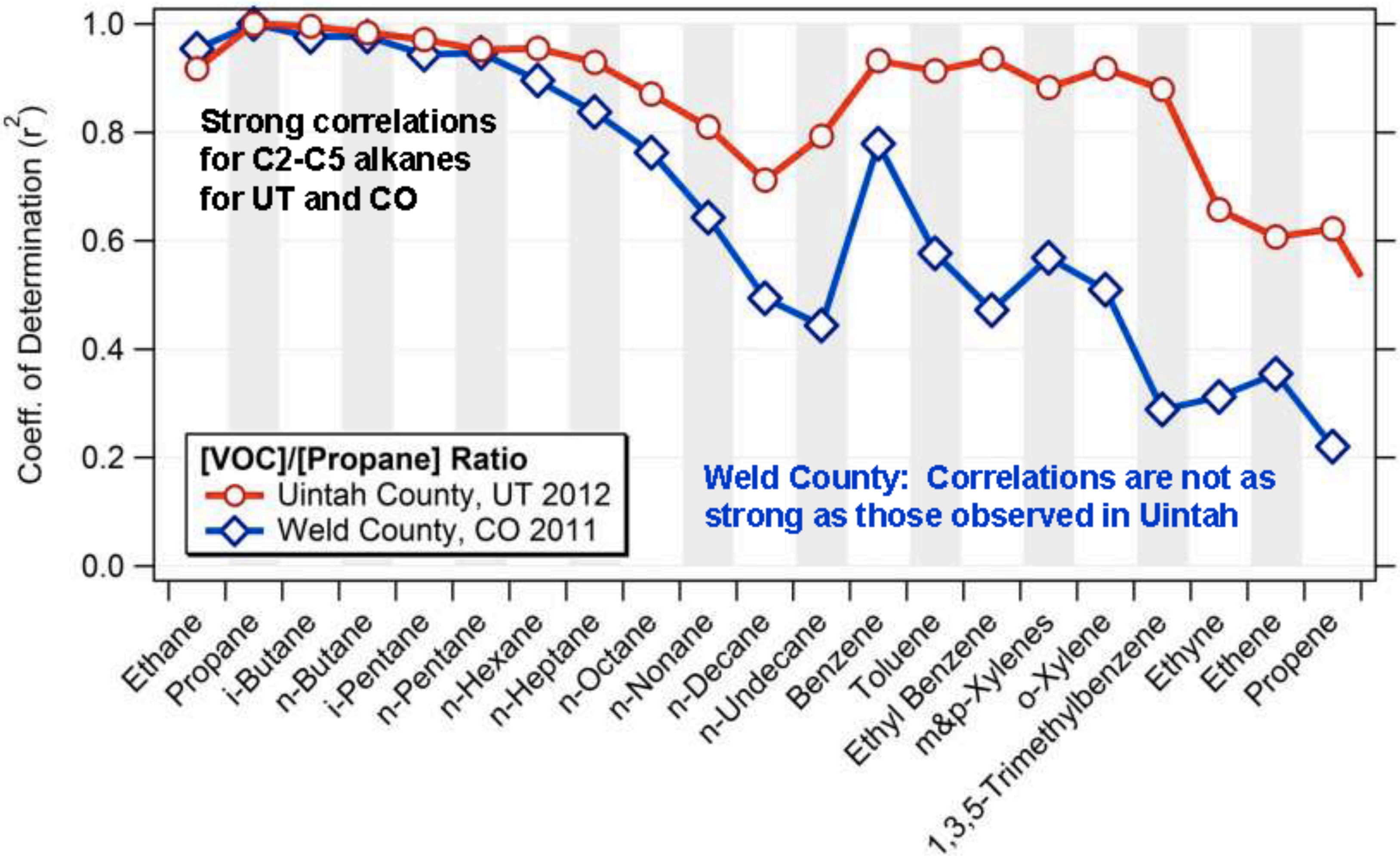
Colorado vs. Utah Source Variability



Gilman et al., Environ. Sci. Technol., 2013

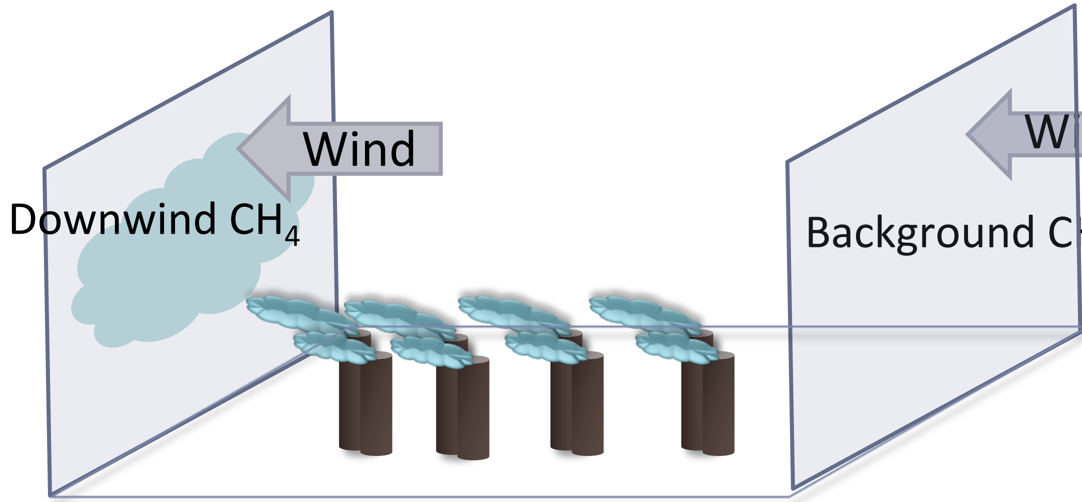
Emission Source Comparison

Colorado vs. Utah Source Variability



Gilman et al., Environ. Sci. Technol., 2013

Integral Equation of Mass Conservation



mass of CH_4 out of box — mass of CH_4 into box

$$m_{CH_4} = \iint_{CS} \rho_{CH_4} V_n dA_{out} - \iint_{CS} \rho_{CH_4} V_n dA_{in}$$

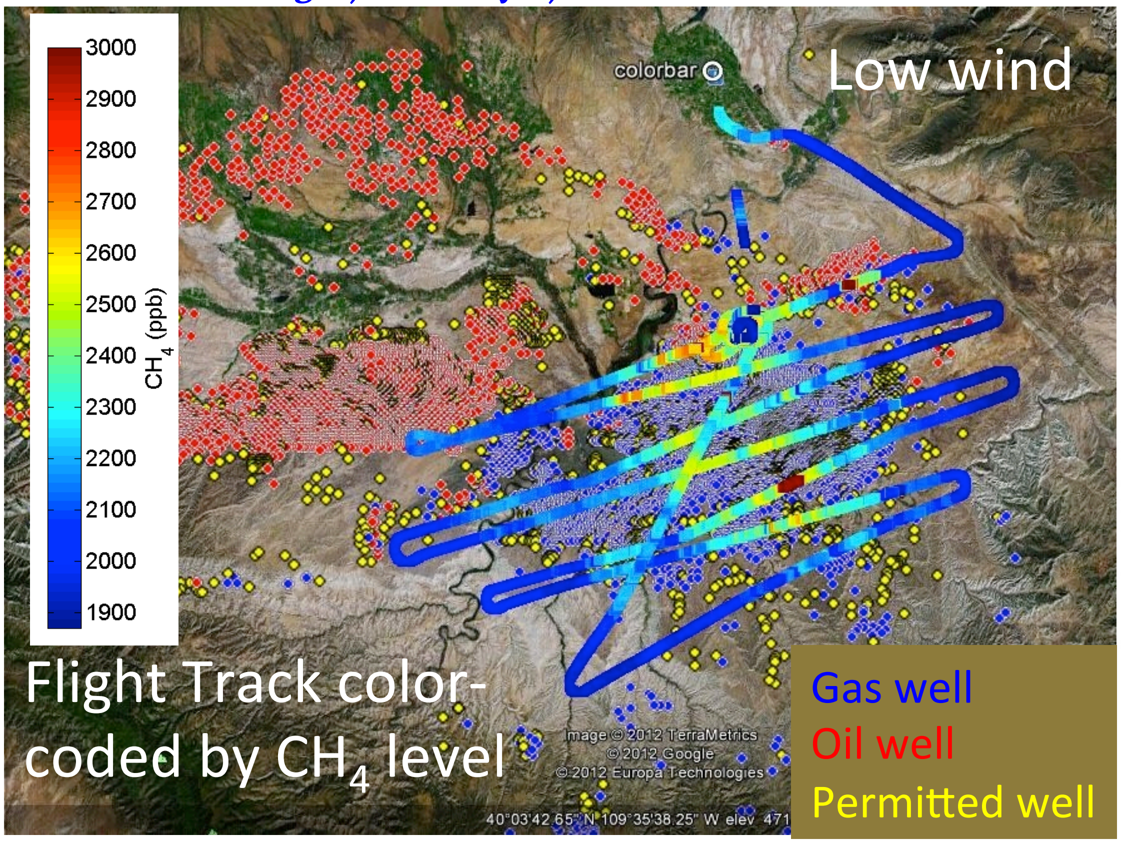
Density of methane (CH4)

Wind direction normal to plane

A. Karion, C. Sweeney

Uinta Basin - February 2012
13 flights with in-situ measurements and flask sampling

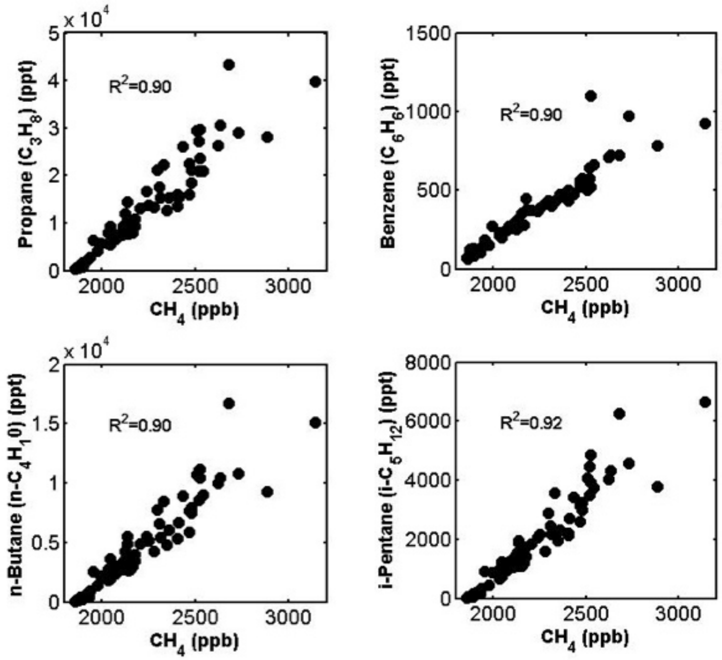
Uintah Basin Flight, February 7, 2012



Oil and Gas production is the main activity in the Uintah Basin.

Multi-laboratory campaign coordinated by EPA region 8 and State of Utah

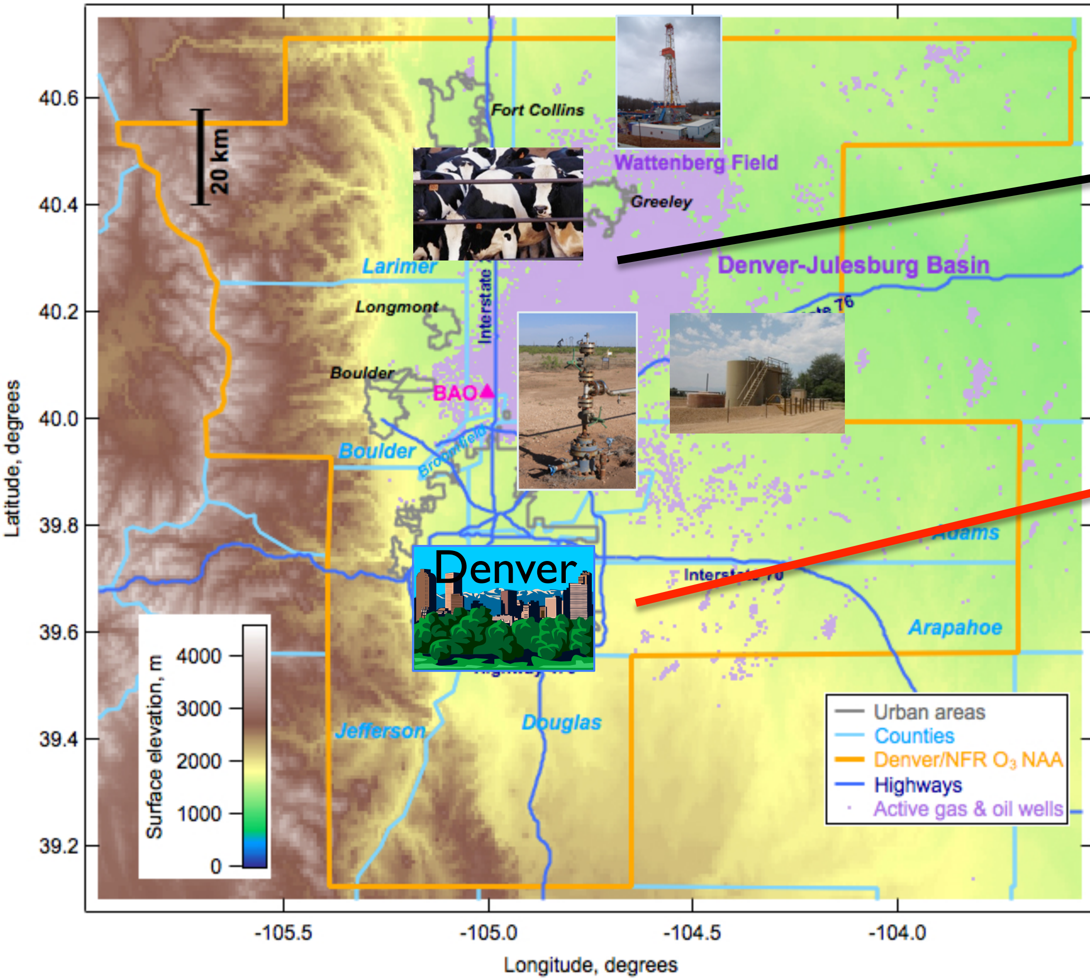
Uintah Basin - February 2012
Aircraft discrete samples data

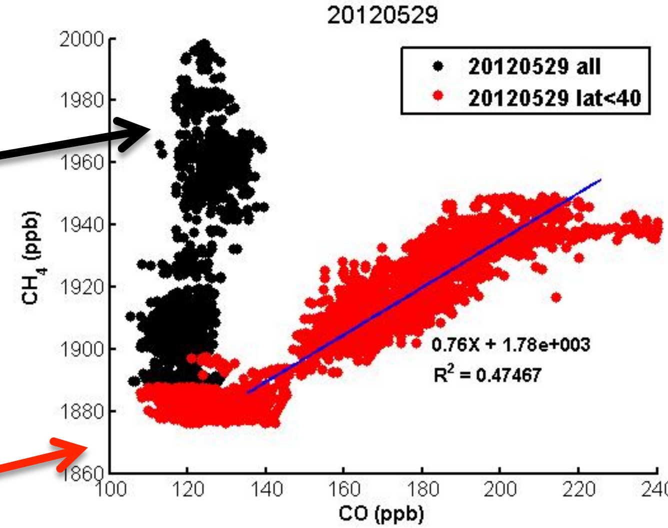


Strong correlation between methane, the light alkanes and benzene in samples collected in the Uintah Basin in 2012.

A. Karion, C. Sweeney

Can we use multiple species measurements to do source attribution?





Different sources of CH_4 have different chemical signatures. Here flight measurements in the Denver Basin in May 2012 show two distinct plumes downwind of Denver (red) and downwind of the oil and gas field (black).

Denver Basin, home to > 20,000 oil and gas wells.

*A **multi-species** approach is needed to determine the significance of different CH_4 sources as well as to separate different emission processes within the NG industry.*

G. Pétron

Mobile Lab, Uintah 2012

Displays of measurements

- In situ CH_4: natural gas marker
- In situ CO_2, CO: combustion markers
- In situ NO/NO_2: combustion markers, ozone precursors
- In situ VOCs (aromatics, oxygenates): ozone precursors
- In situ O_3 (two instruments)
- 240 discrete air samples: CH_4, CO, CO_2, C_2-C_8 alkanes, aromatics

NO, NO_2

flasks

VOCs

CH_4, CO, CO_2

On-going Mobile Lab Data Analysis, Uintah 2012

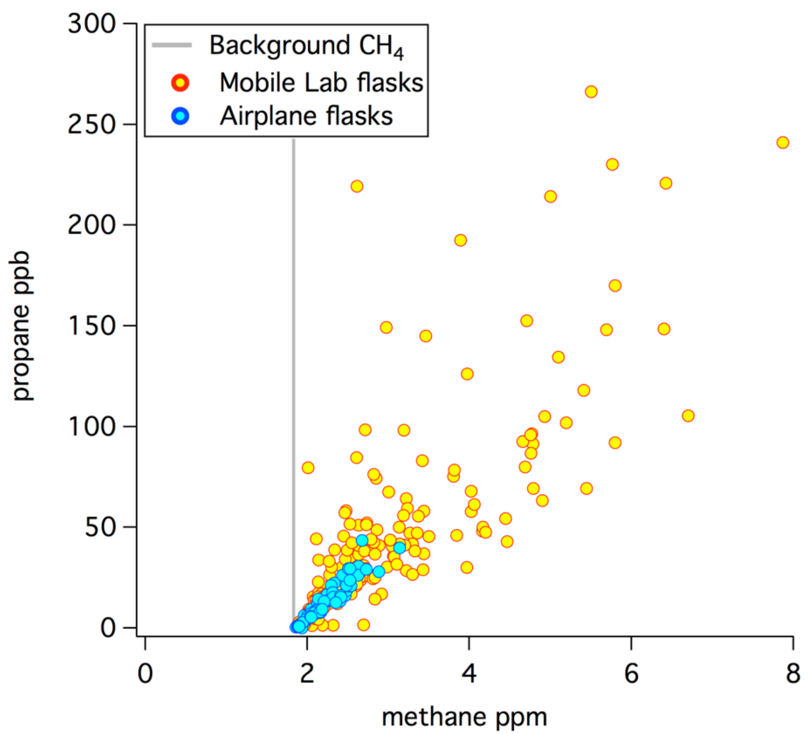
Source identification and characterization

- Tracking chemical signatures of different sources
  - VOCs speciation (Alkanes, Aromatics, Alkenes)
  - VOCs/CH_4 ratios: fugitive emissions versus condensate/oil vapors
  - NOx/CO_2 ratios: emission factors for various point sources in the Basin
  - CO/CO_2, (CO+CO_2)/(CO+CO_2+CH_4): combustion efficiencies
- Comparison with WRAP upstream O&G emissions inventory
  - Using measured emission factors and activity data
  - Using airplane and NOAA HRDL met fields measurements

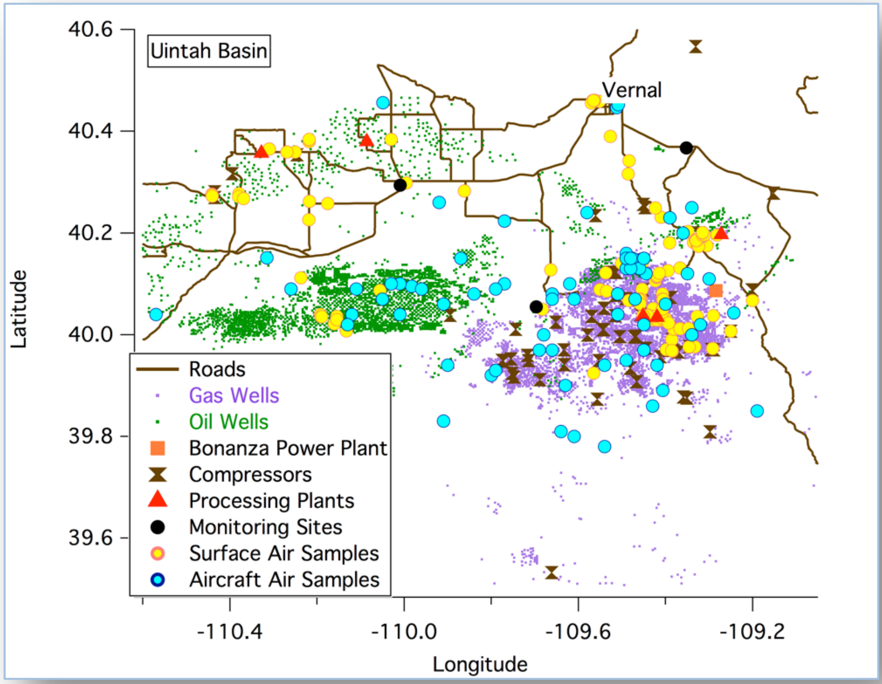
Note: Similar studies carried out in DJB over past several years

G. Pétron

Discrete Air Samples: CH_4, CO, CO_2 and VOCs



- Mobile Lab: 240 flasks
- Airplane: 81 flasks
- Collected in various areas in the oil and gas Basin

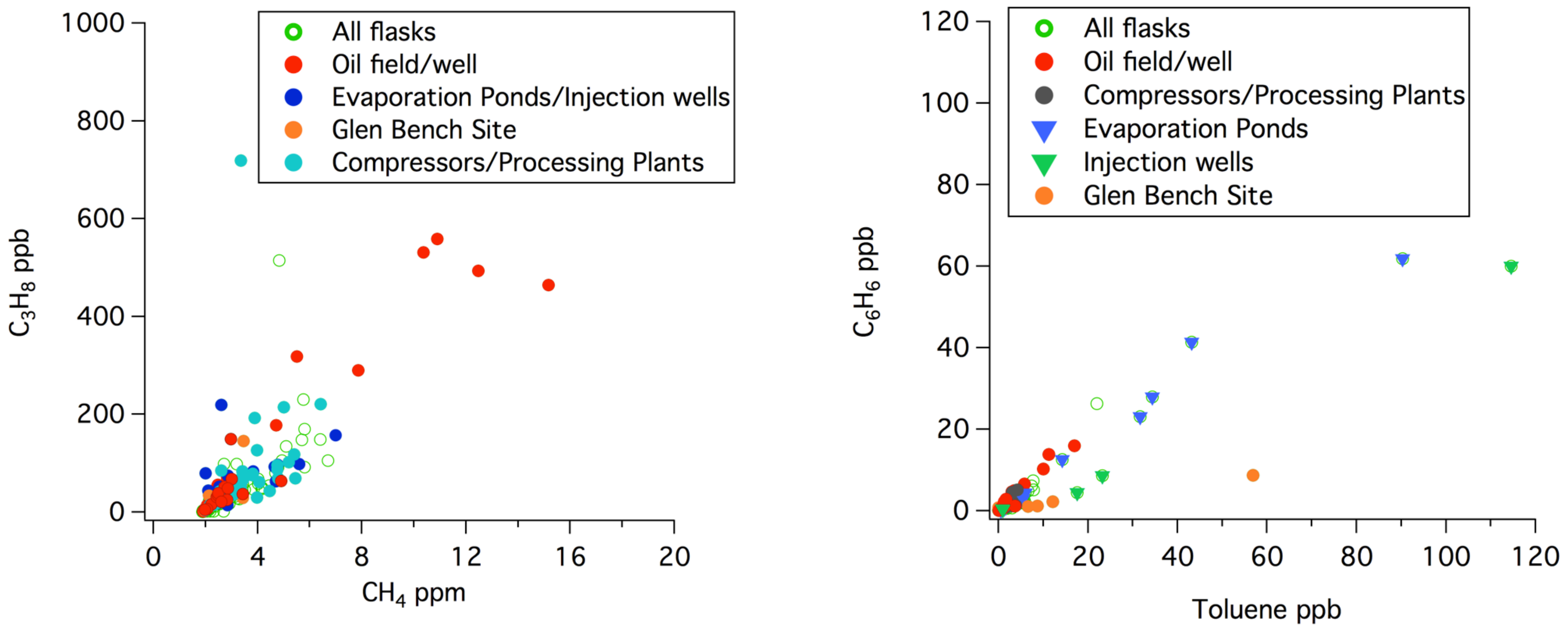


Example of flasks data:
The airplane sampled the mixture of emissions while the Mobile Lab was able to target specific sources.

G. Pétron

NOAA GMD & University of Colorado CIRES & INSTAAR

Mobile Lab flasks data shows a mixture of VOCs

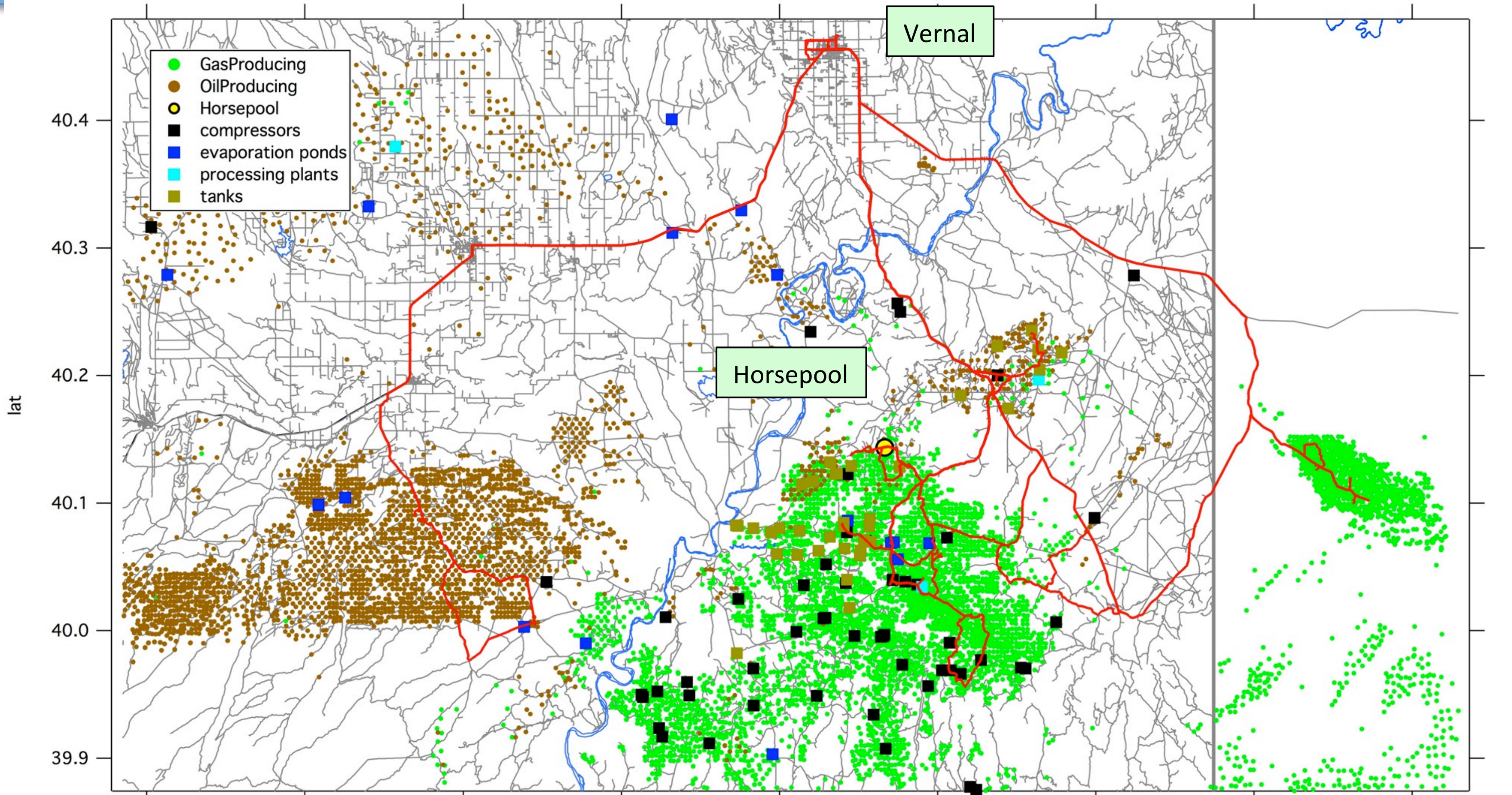


Targeted sampling of regional enhancements and specific point sources plumes show that alkanes are the most enhanced VOCs.
Aromatics levels are also elevated (> 2 ppb) downwind of certain sources.
Measured levels are dependent on proximity to the center of the plume and dispersion conditions when the sample was collected.

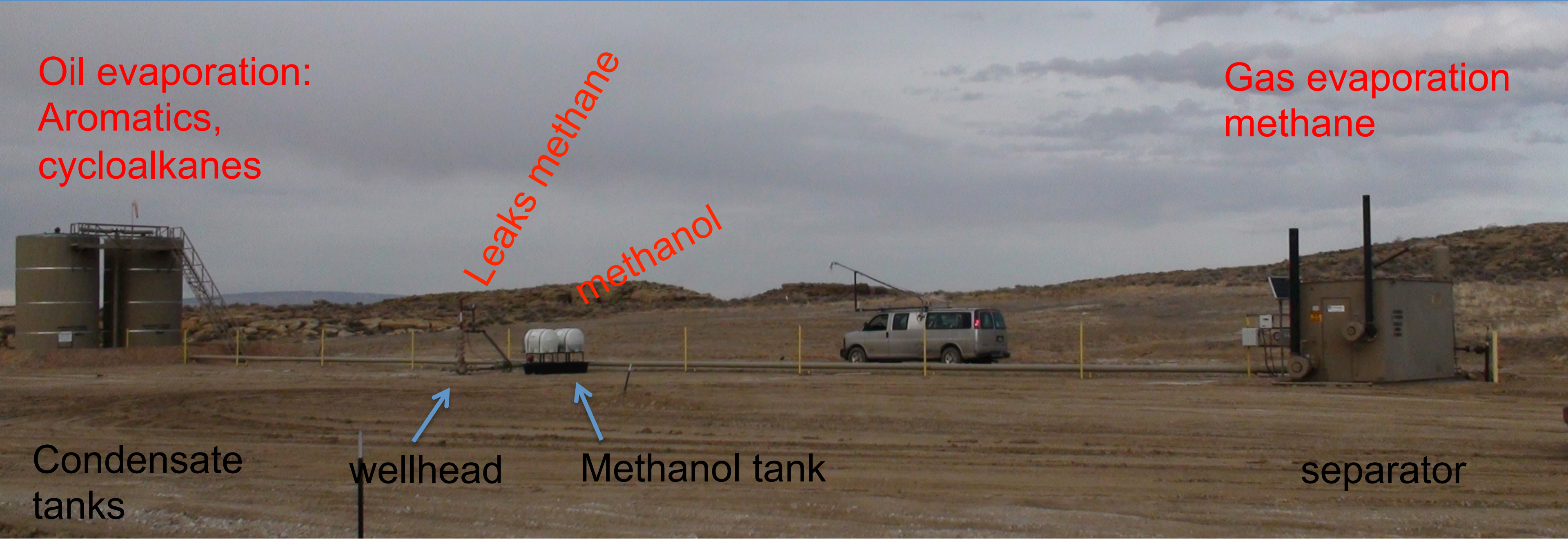
G. Pétron

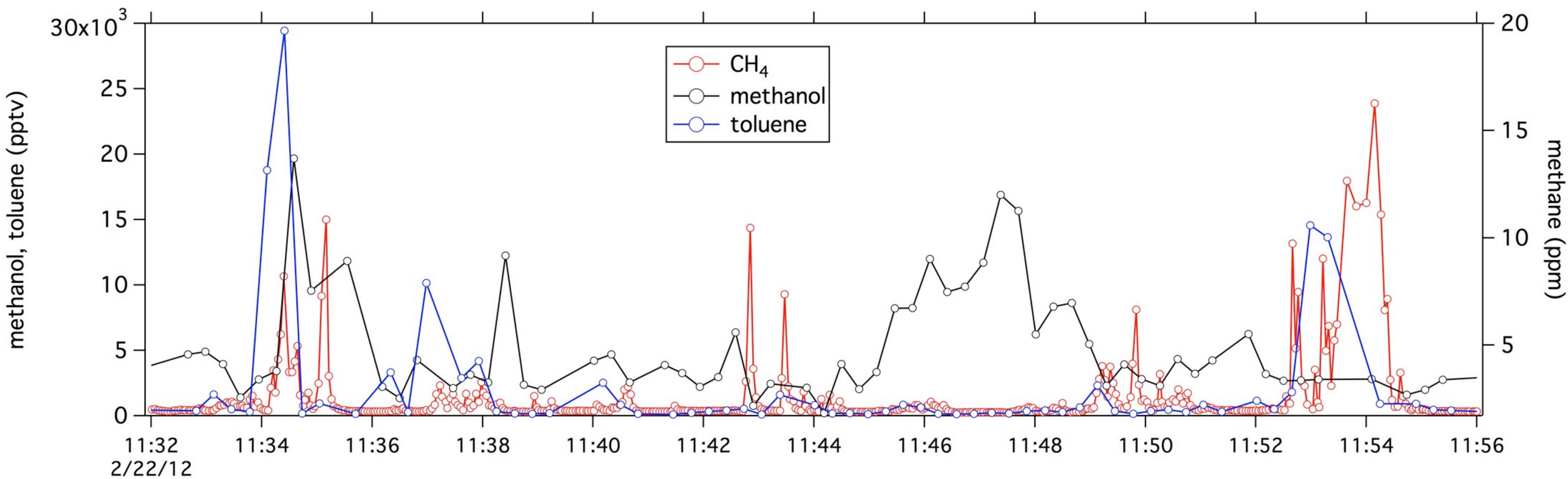
Fast-Response VOC Measurements

Felix Geiger, Carsten Warneke



- Mobile Laboratory sampled wide variety of individual sources
- Source composition useful for comparison with data from Horsepool and aircraft: which sources are the largest contributors?





Fast-response VOC measurements allow separate measurements of individual sources

F. Geiger, C. Warneke

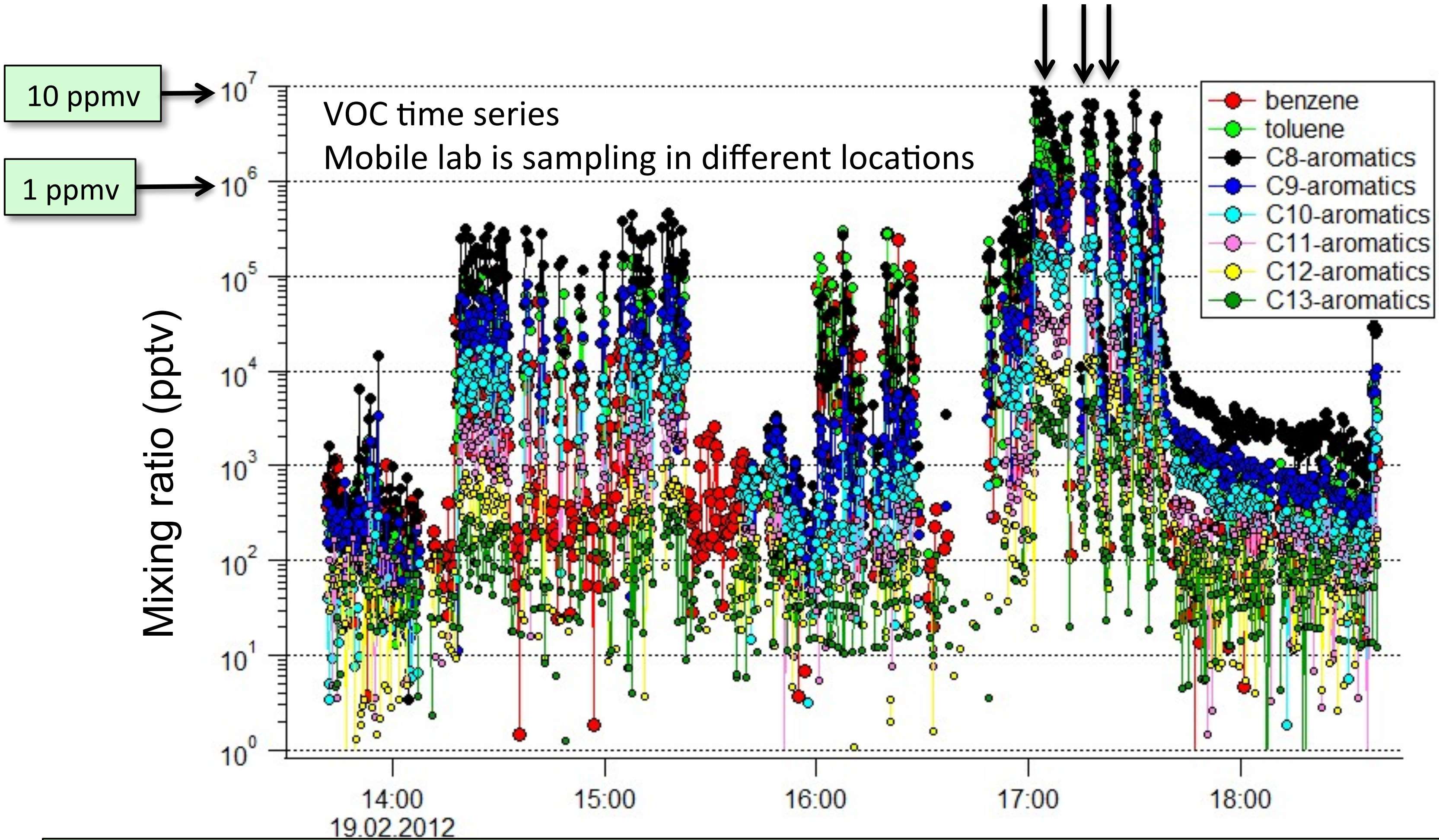
<u>Measurements at new well near Glen Bench:</u>

1. Well was fracked on February 8-9
2. Flow-back pond was used
3. Mobile laboratory went by 11 times between Feb 14-28; well not completed on Feb 28

F. Geiger, C. Warneke

Results from one day (Feb 19):

- Repeated drive-by of the new well site
- Aromatics concentrations peak to 1-10 ppmv **every time** the mobile lab is downwind from flow-back pond

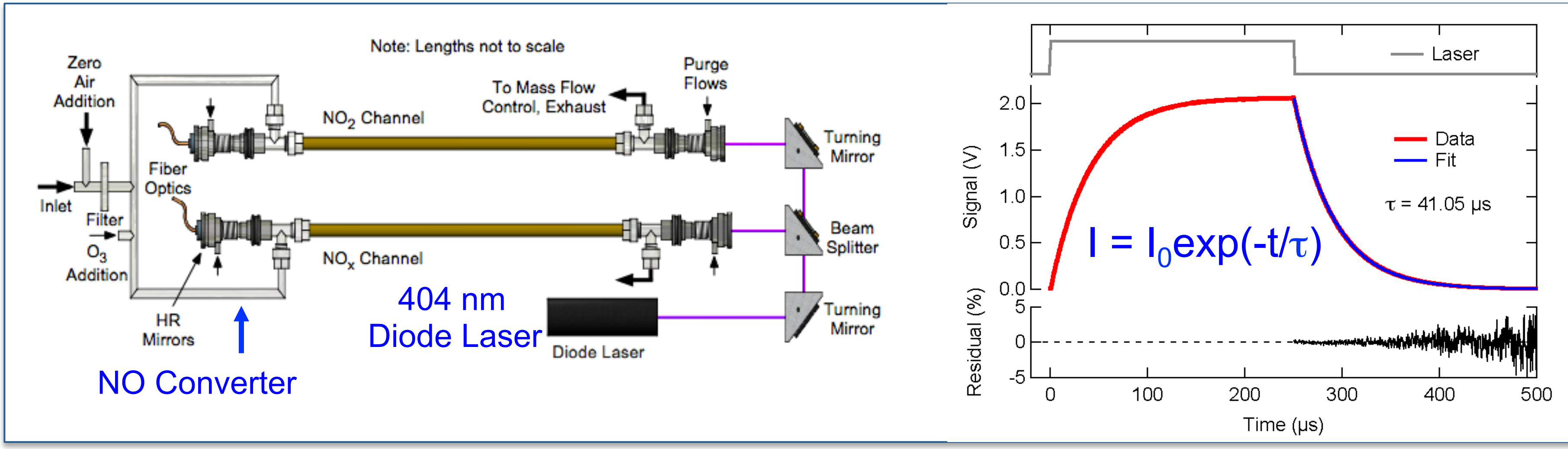


High concentration of aromatics (1-10 ppmv) near flow-back pond

F. Geiger, C. Warneke

Cavity Ring-Down NO_2, NO_x (O_3) Measurements

Peter Edwards, Steven Brown, William Dubé



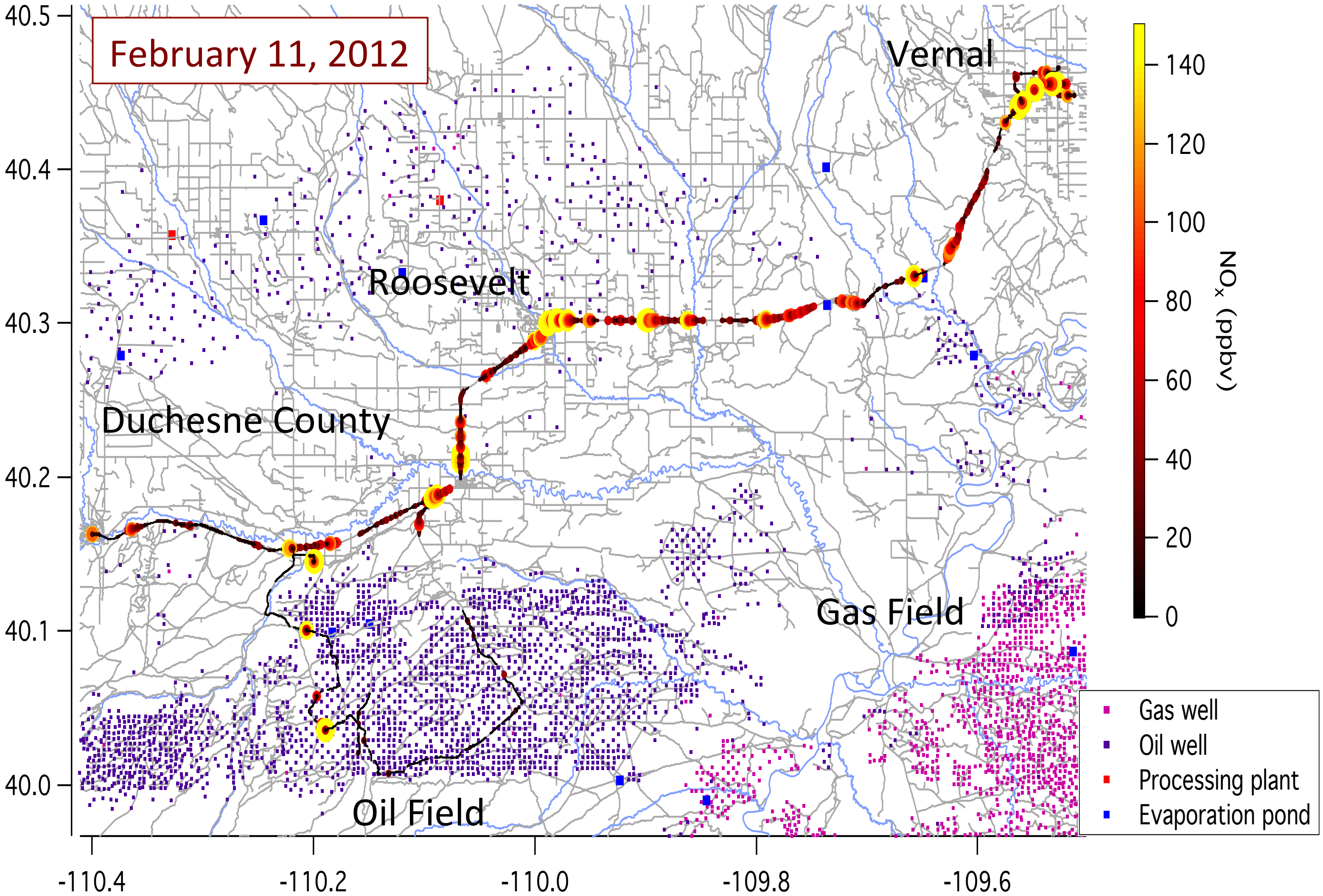
- Measure NO_2 directly by 404 nm optical absorption
  Effective path length = 12 km; Sensitivity to NO_2 = 0.05 ppb, 1 s; Accuracy = 5%
  Measured signal in Utah: 0.5 – 10 ppb background, 10-1000 ppb in plumes

- Measure NO_x (=$NO+NO_2$) by quantitative (98%) conversion of NO to NO_2 in excess O_3
  $NO + O_3 \rightarrow NO_2 + O_2$
  Simple and accurate scheme for measurement of total NO_x

- For some drives, measure O_3,rather than NO_x, by addition of excess NO to second channel

- Instrument weight, power consumption, size appropriate for deployment in Mobile Lab

Example Mobile lab NO_2, NO_x and O_3 data for single drive



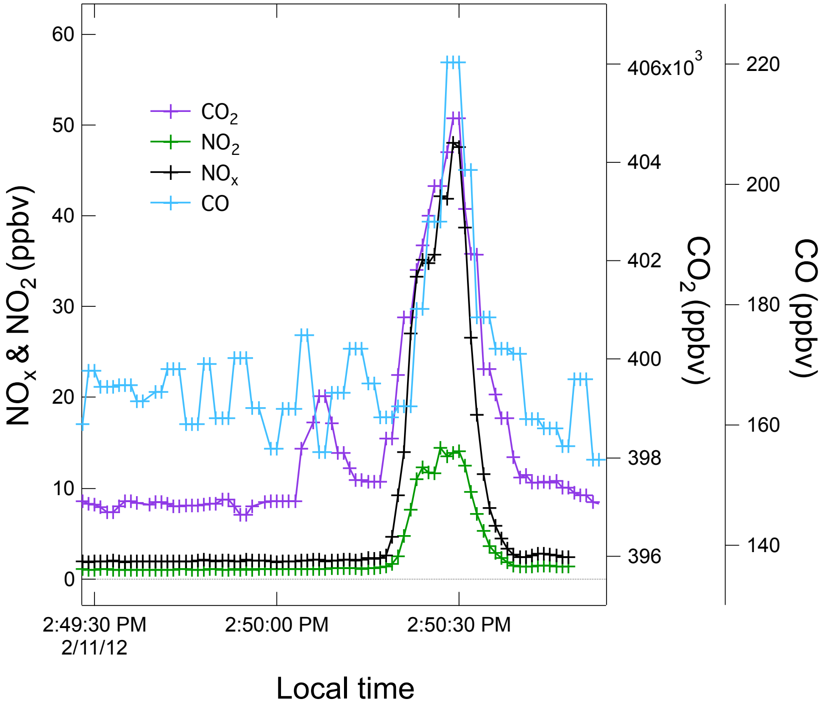
10 drives measuring NO_2 and NO_x

9 drives measuring NO_2 and O_3

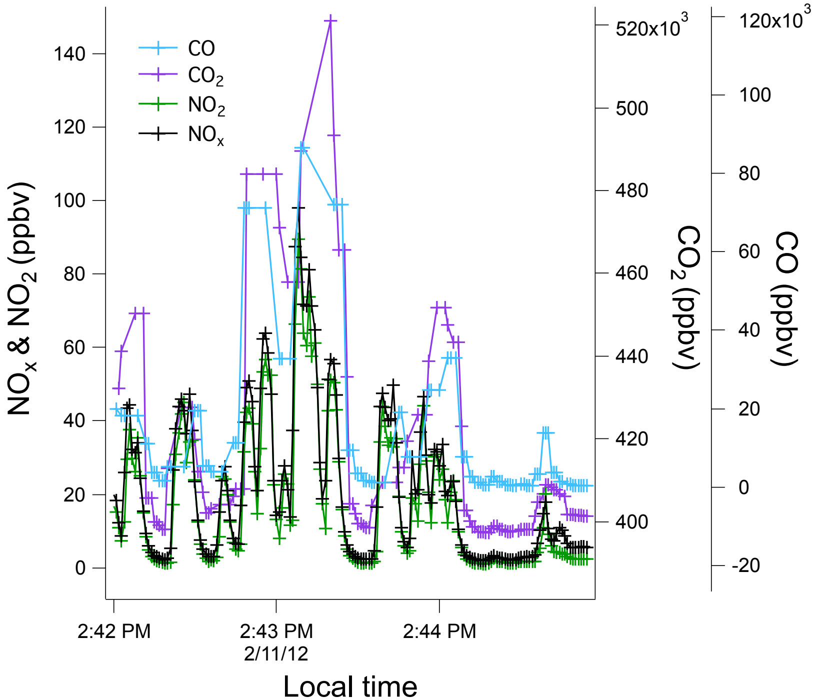
P. Edwards et al.

Example NO_x Emissions Factors Calculation

Truck Plume



Pumpjack Plume



NO_x vs CO_2 useful for NO_x emissions vs. fuel use or number of engines / vehicles

NO_x vs CO useful for comparison to emissions inventories (e.g., CO often used as urban tracer)

NO_2 vs NO_x useful for direct NO_2 emission *if* this ratio is large

P. Edwards et al.

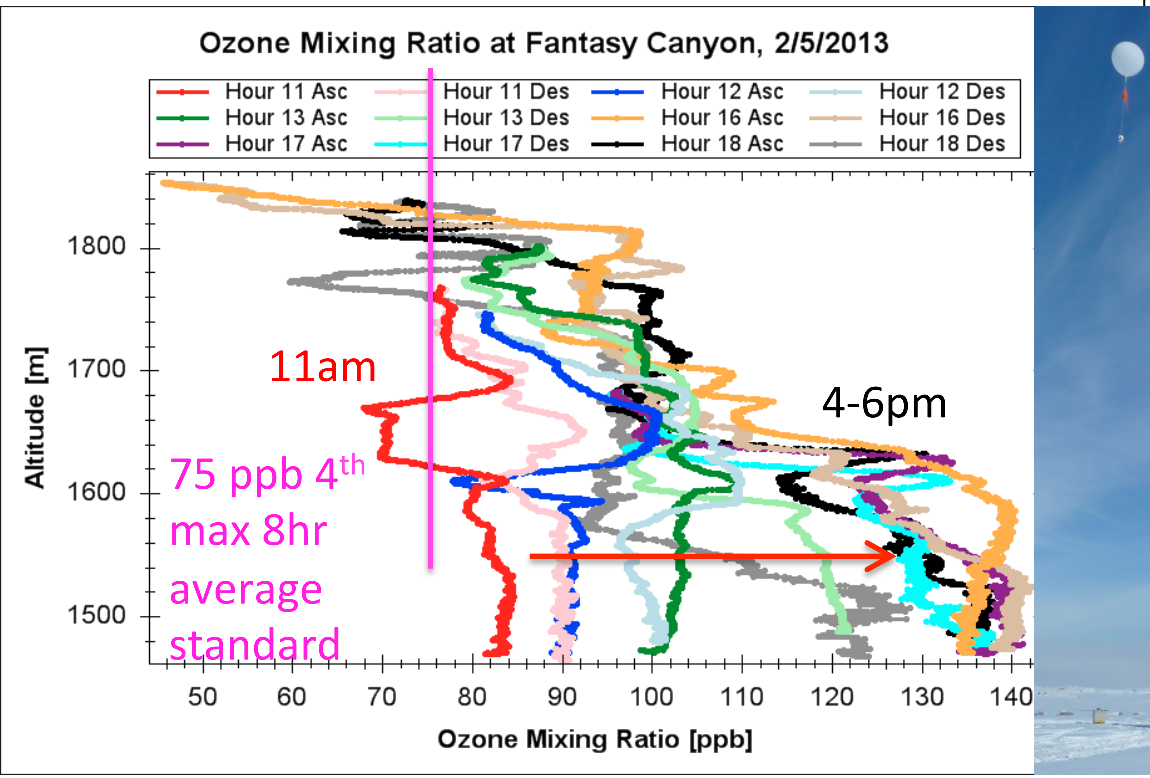
NOAA measurements & analysis of chemical and meteorological processes in O&G regions

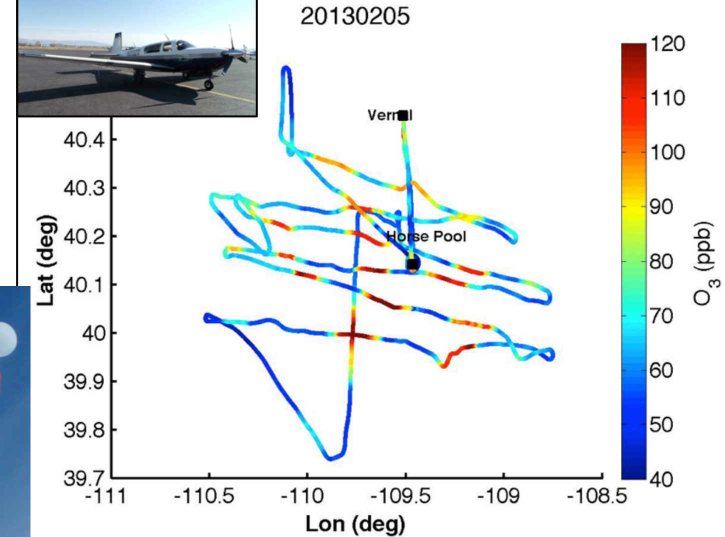
Examples of measurements that probe…

- Ozone distributions
- Chemical processes leading to ozone formation
- Basin flows and boundary layer dynamics

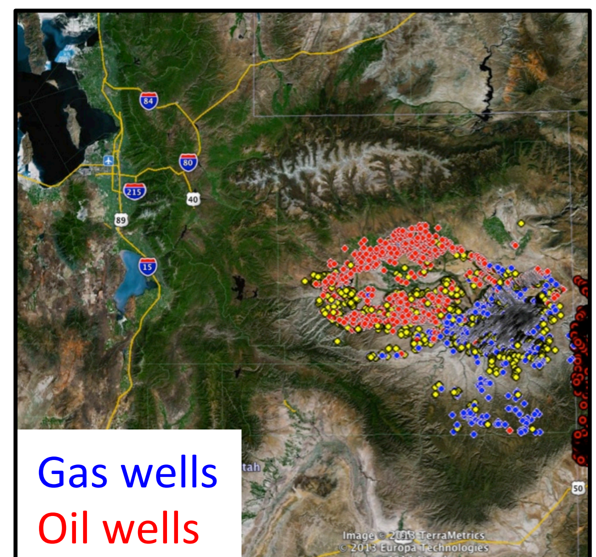
1. Tethered sondes + aircraft, Uintah winter 2013 (Schnell et al.)
2. Surface sampling, Uintah winter 2012/2013 (Roberts et al.)
3. Surface sampling, DJB winter 2011/ Uintah winter 2012 (Gilman et al., 2013)
4. Surface remote sensing, Uintah winter 2012/2013 (Senff, Brewer, Banta, et al.)

Wintertime Ozone in NE Utah



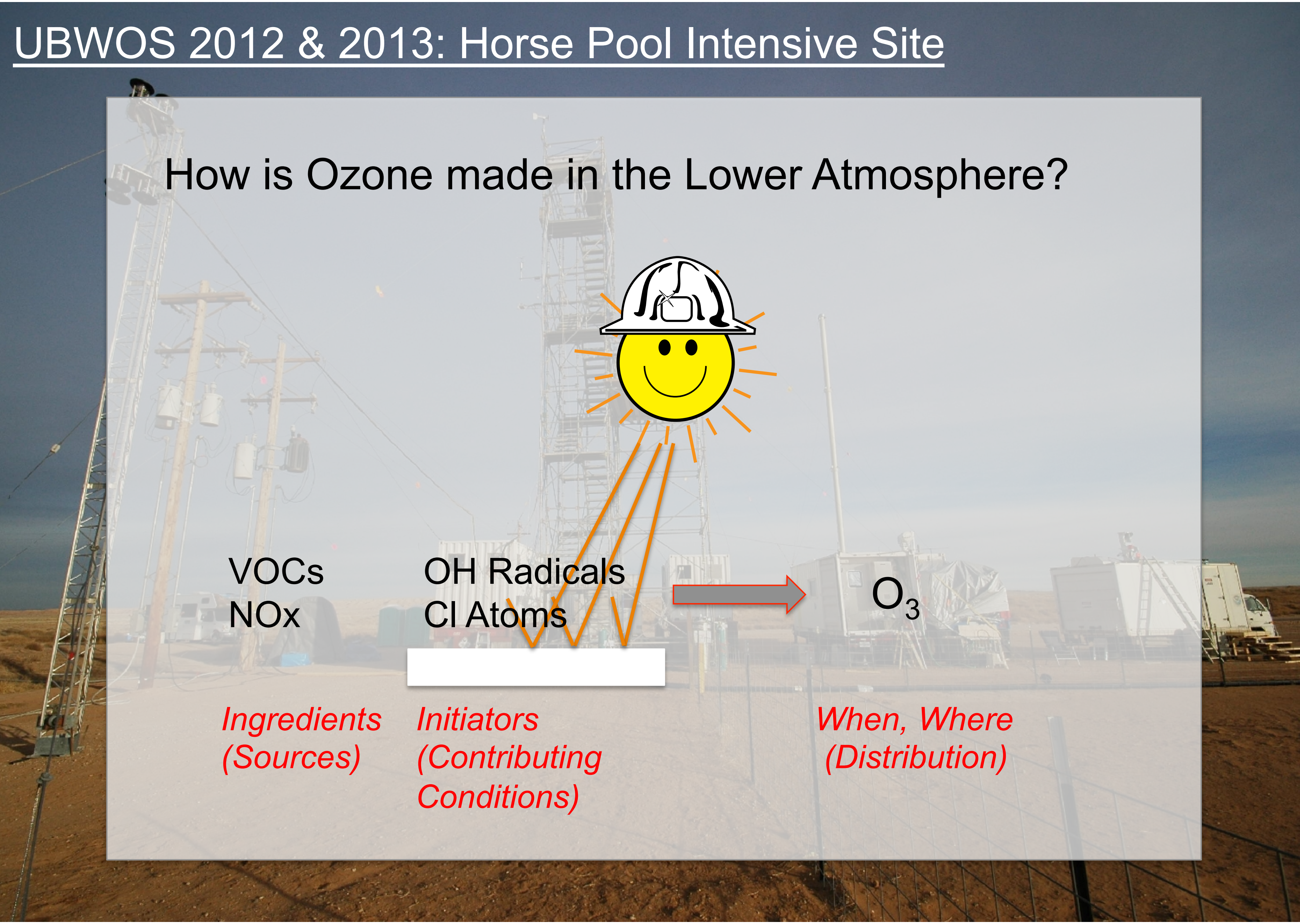


Tethered sonde and airborne O_3 measurements in the Uinta Basin in February 2013



Under very strong temperature inversion, the Uinta Basin experienced very high ozone levels at the surface (up to 150 ppb). Oil and gas operations are the main activity in the region.

R. Schnell et al.

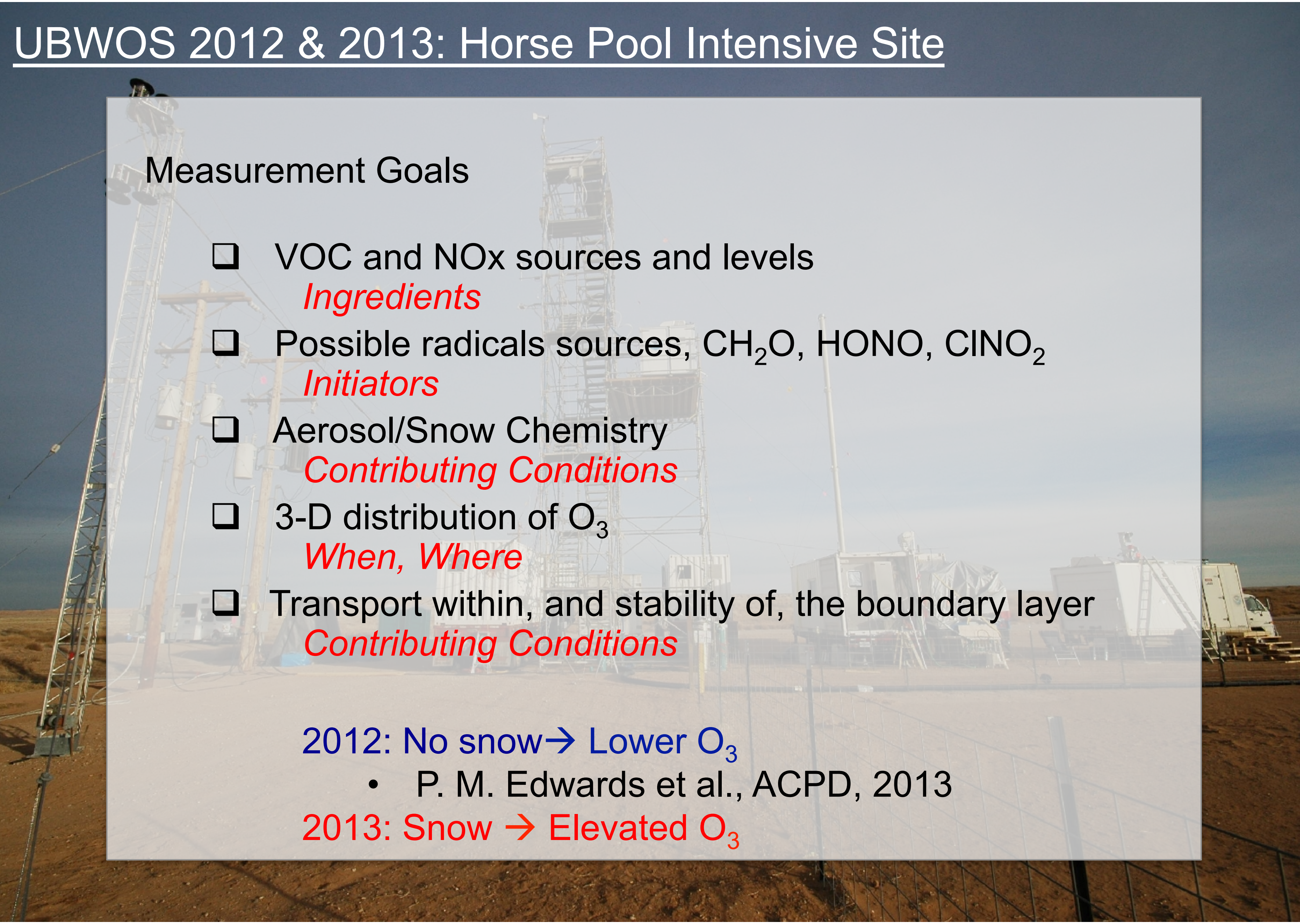


J. Roberts et al.

UBWOS 2012 & 2013: Horse Pool Intensive Site

J. Roberts et al.

UBWOS 2012 & 2013: Horse Pool Intensive Site



Measurement Goals

- ❑ VOC and NOx sources and levels
  Ingredients
- ❑ Possible radicals sources, CH_2O, HONO, $ClNO_2$
  Initiators
- ❑ Aerosol/Snow Chemistry
  Contributing Conditions
- ❑ 3-D distribution of O_3
  When, Where
- ❑ Transport within, and stability of, the boundary layer
  Contributing Conditions

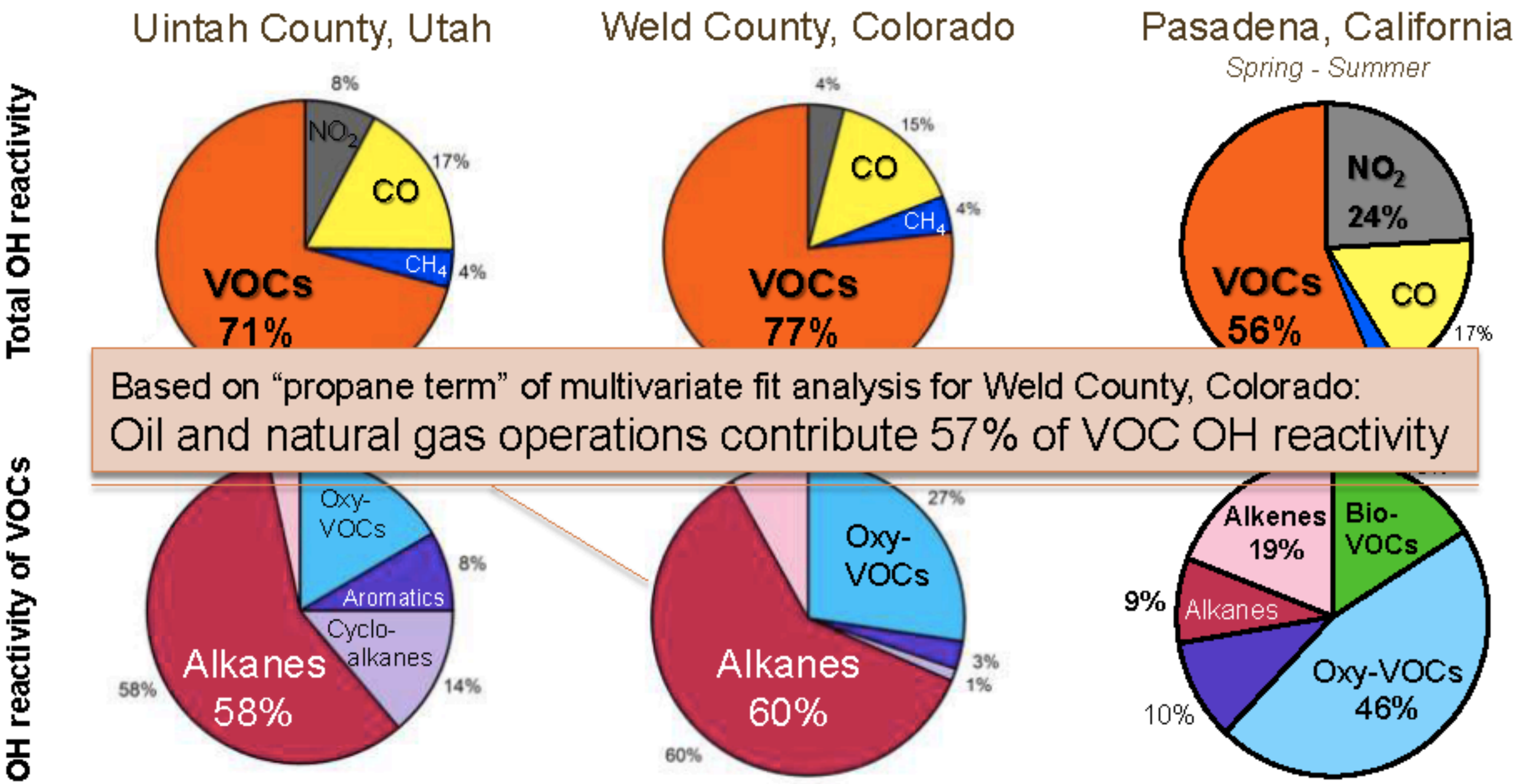
2012: No snow→ Lower O_3

- P. M. Edwards et al., ACPD, 2013

2013: Snow → Elevated O_3

J. Roberts et al.

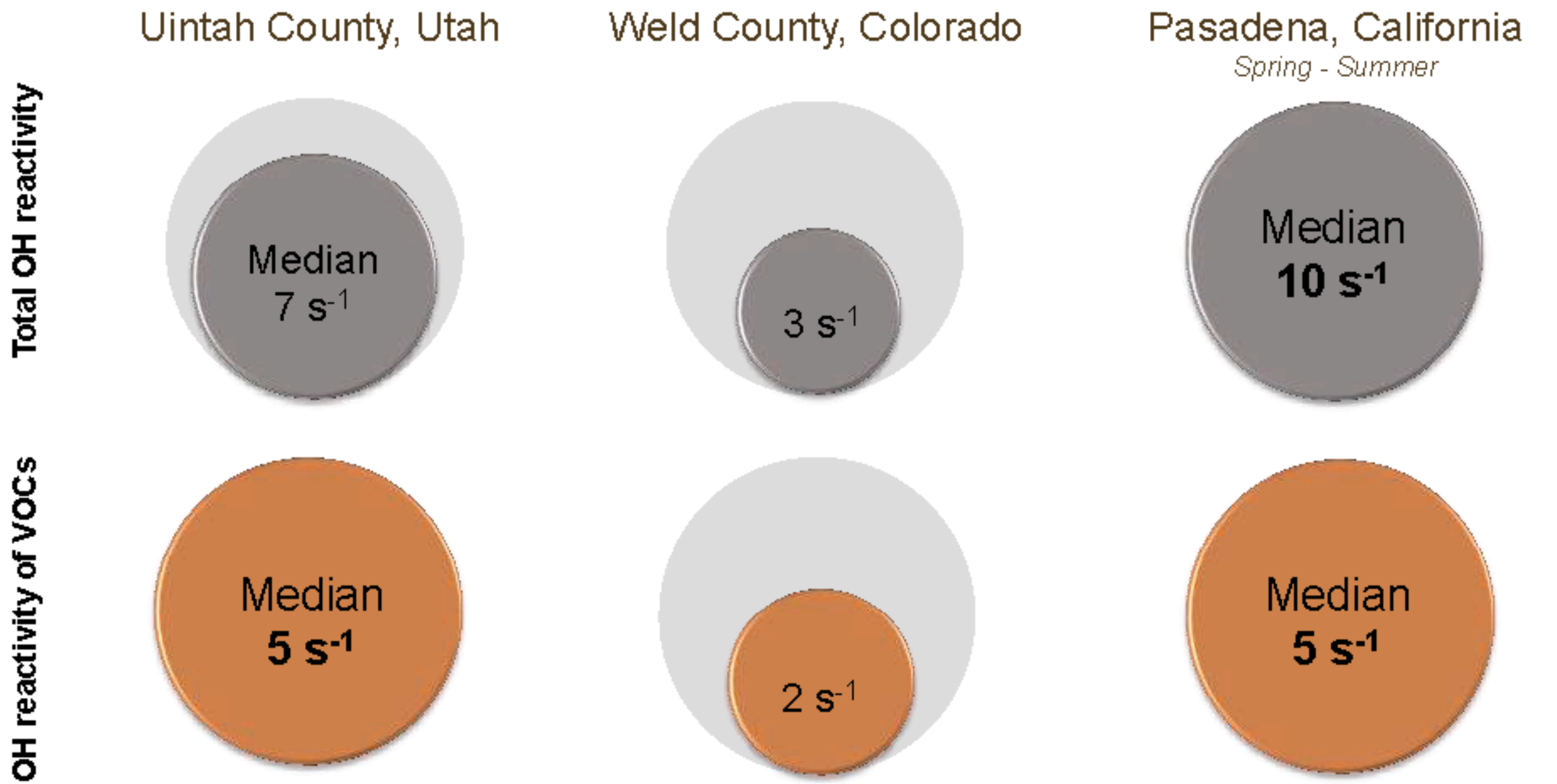
OH reactivity:
Comparison of composition



Based on "propane term" of multivariate fit analysis for Weld County, Colorado:
Oil and natural gas operations contribute 57% of VOC OH reactivity

Gilman et al., Environ. Sci. Technol., 2013

OH reactivity: Comparison of median values



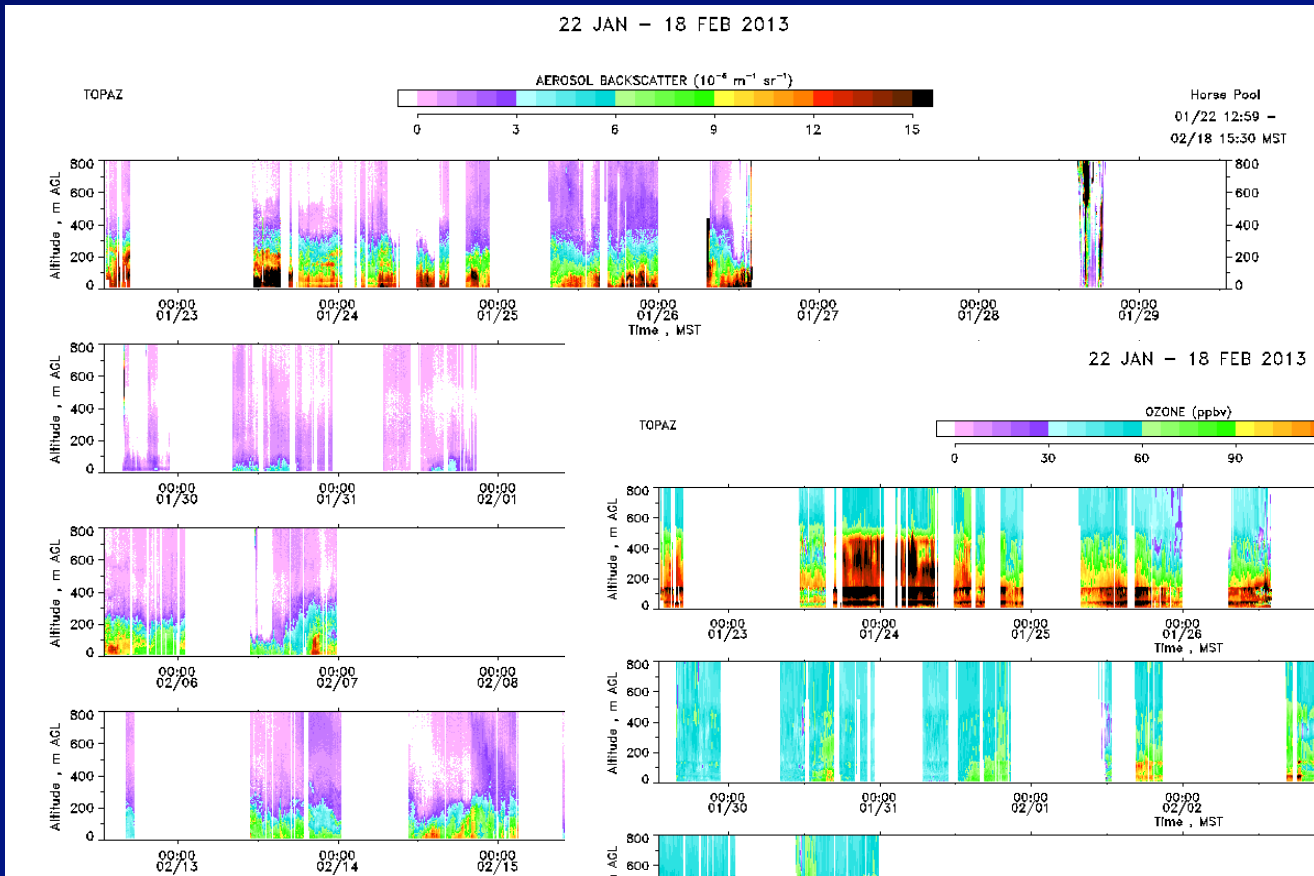
Areas scaled by median OH reactivities

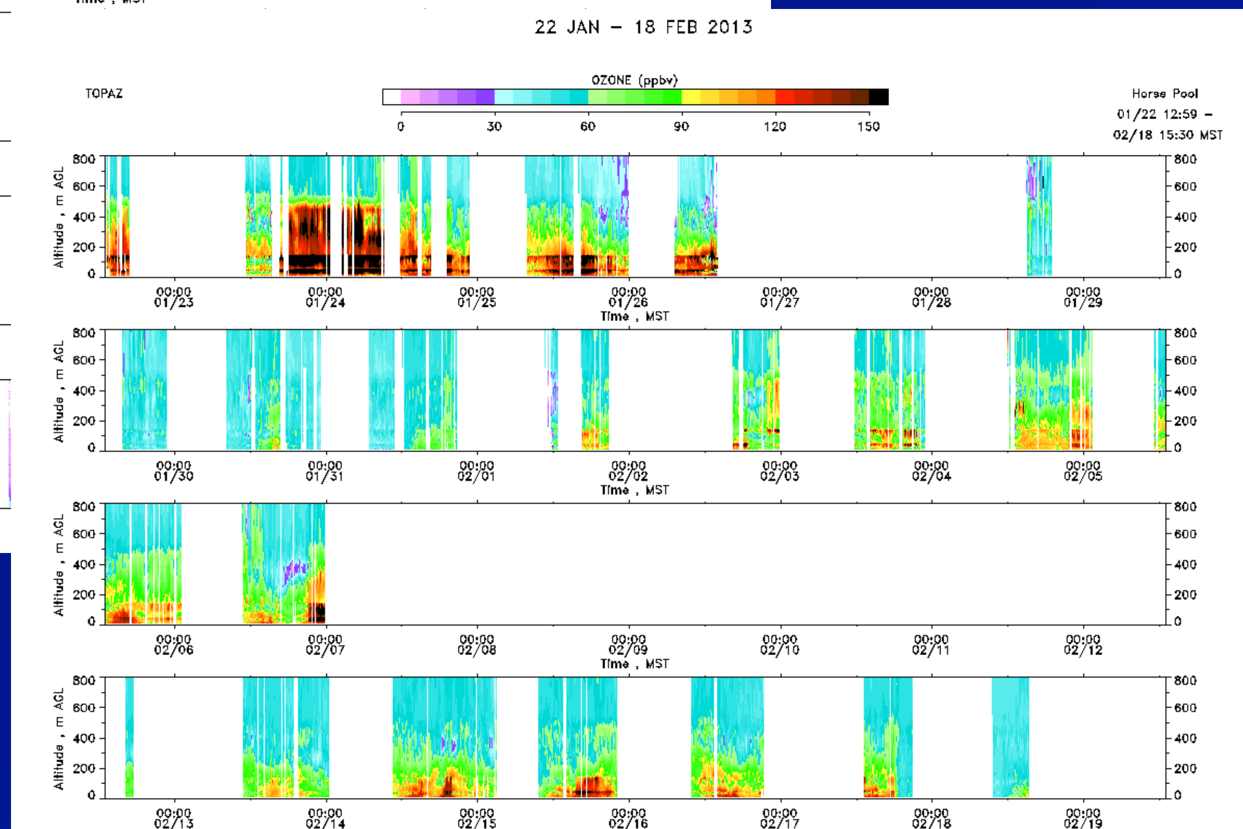
Gilman et al., Environ. Sci. Technol., 2013

Dynamics studies: Uintah 2013

- Depth of pollution layer
- Spatial variability of source activity
- Mixing mechanisms
- Patterns of horizontal flow evolution
- Horizontal spread of pollutants

Time-height O_3, aerosol – Uintah 2013



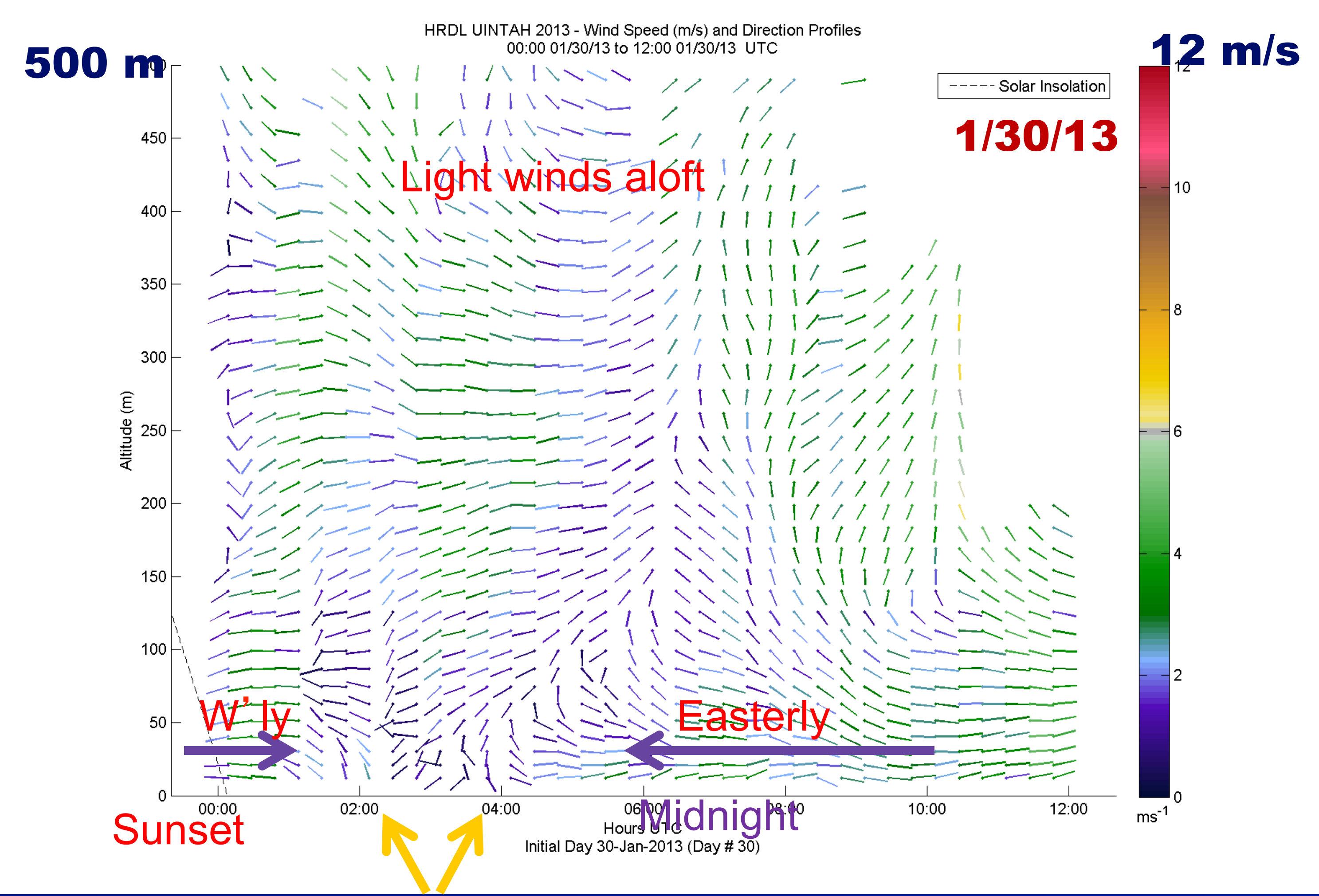


Depth of polluted layer – deepens thru event

C. Senff et al.

HRDL profiles – nighttime, Uintah 2013

Time vs. height – wind speed color coded

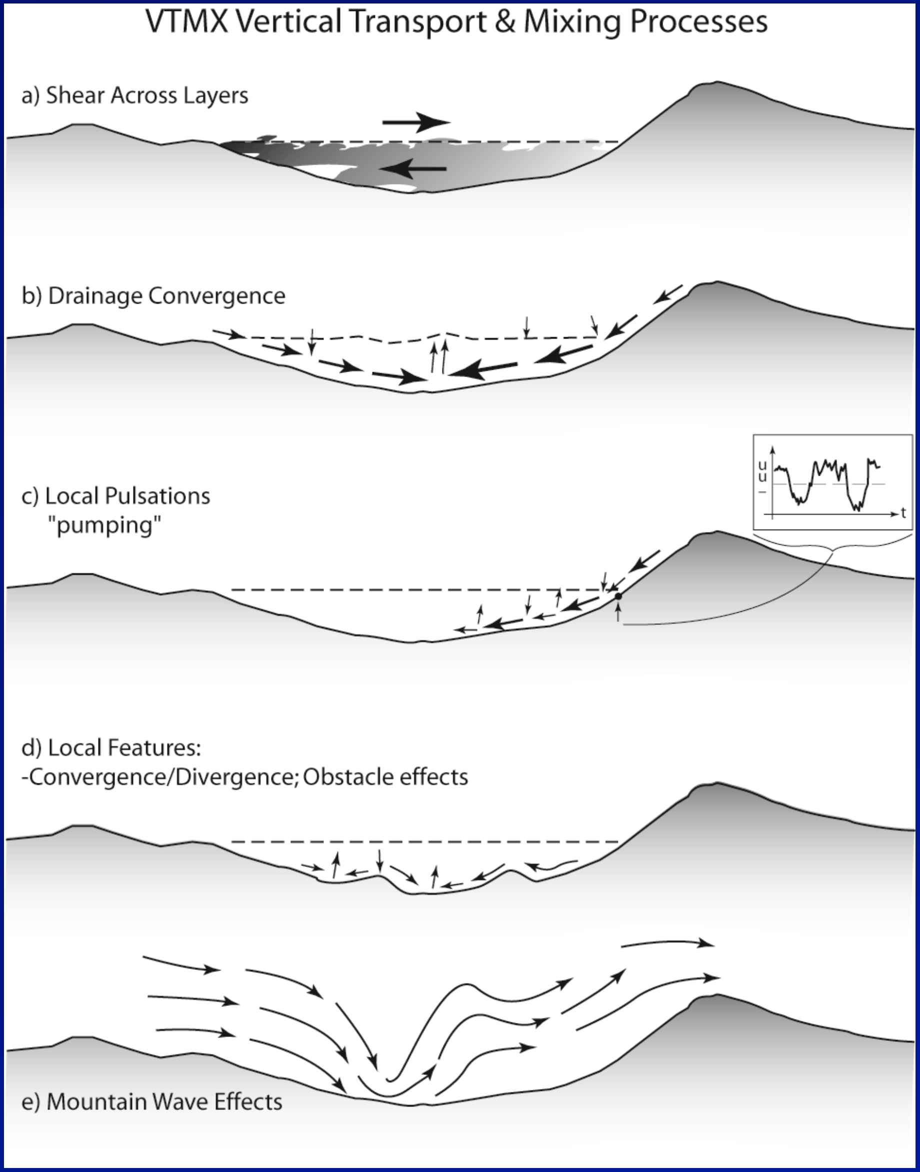


Evening transition – *to easterly nocturnal flow near surface*

A. Brewer et al.

COLD SURFACE

How can we get O_3 (e.g.) up to 300 - 400 m when stable?



Vertical mixing processes in a basin cold pool: *complex, for a cold surface*

Banta, R.M., et al., 2004: Nocturnal low-level jet in a mountain basin complex. Part I: Evolution and implications to other flow features. ***J. Appl. Meteor.*****, 43, 1348-1365.**

NOAA modeling to understand emissions and impacts on air quality in O&G regions

Use meteorological/chemical models in forward and inverse approaches to test understanding of emissions, chemical processes, and atmospheric dynamics

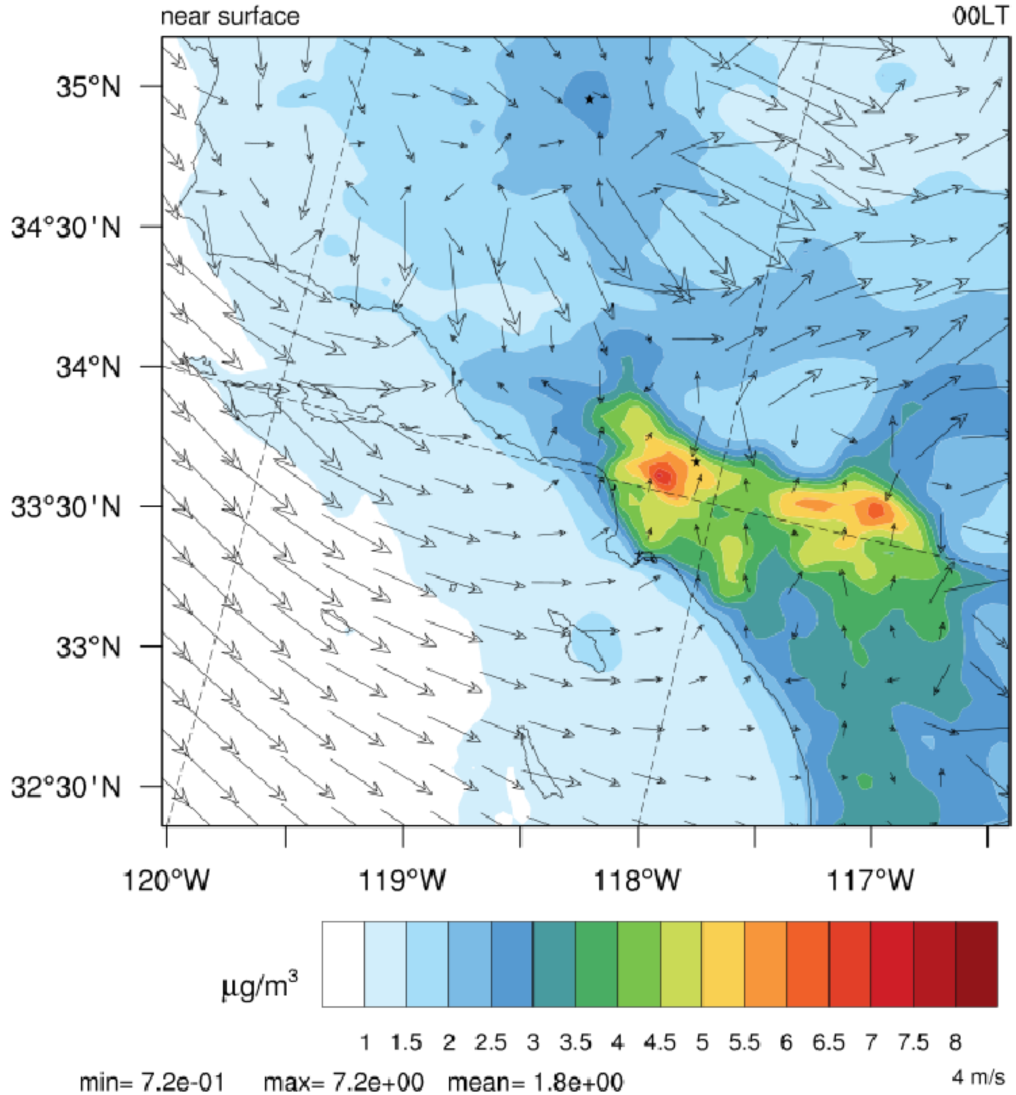
1. Regional chemical-transport model, California/Utah/Colorado (Ahmadov and McKeen)
2. Inverse model, California (Brioude)

WRF-Chem Modeling

Stuart McKeen and Ravan Ahmadov

- Coupled meteorology-chemistry modeling for CalNex-2010, UBWOS-2012 and 2013 studies
- Secondary organic aerosol simulations by using volatility basis set approach in WRF-CHEM
- Evaluation of different anthropogenic emission inventories
- Extensive evaluations against aircraft and surface measurements

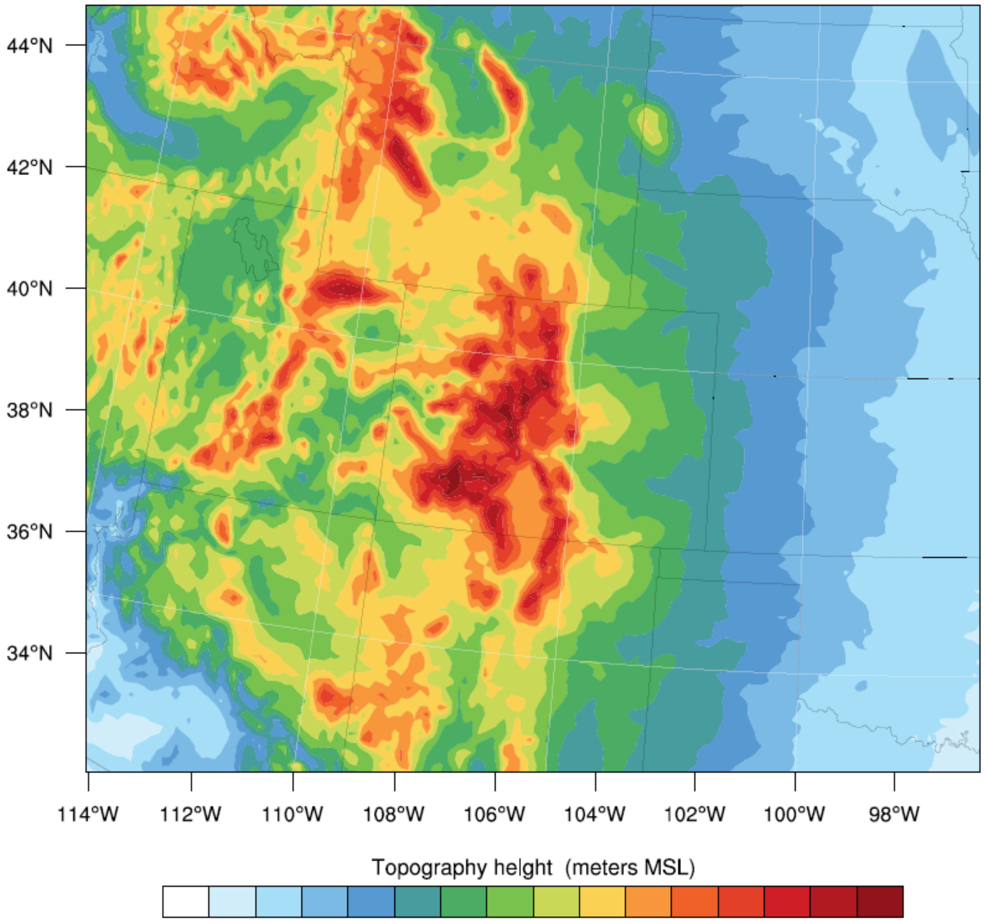
Organic aerosol simulations for LA Basin
Averaged fields 05/14-06/15, 2010



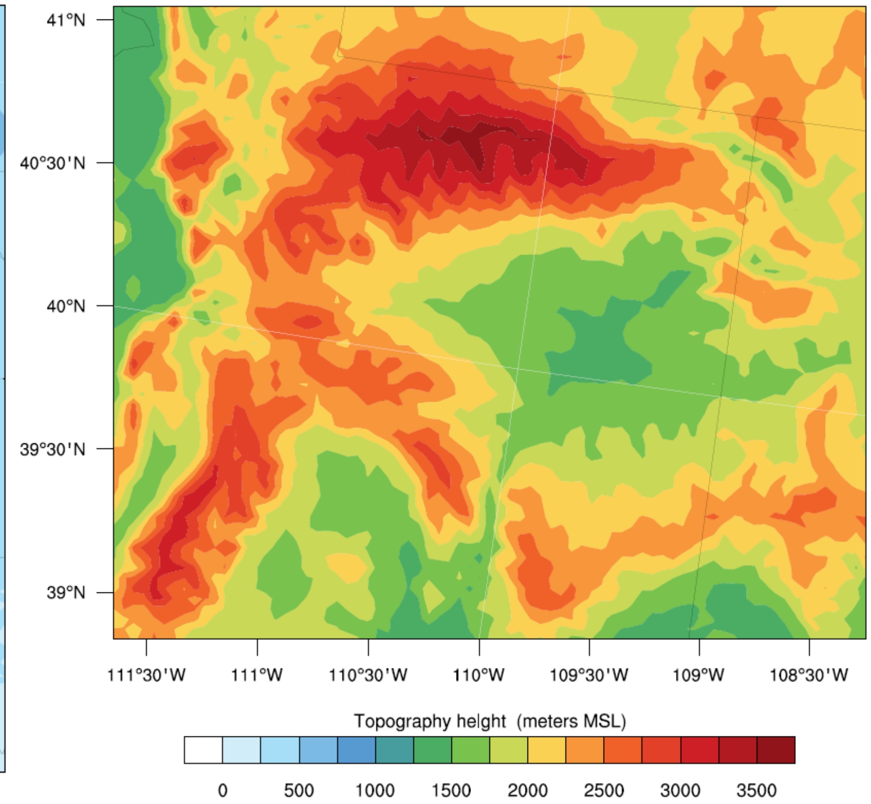
R. Ahmadov, S. McKeen

Nested WRF-CHEM domains for the UBWOS modeling study

12km resolution

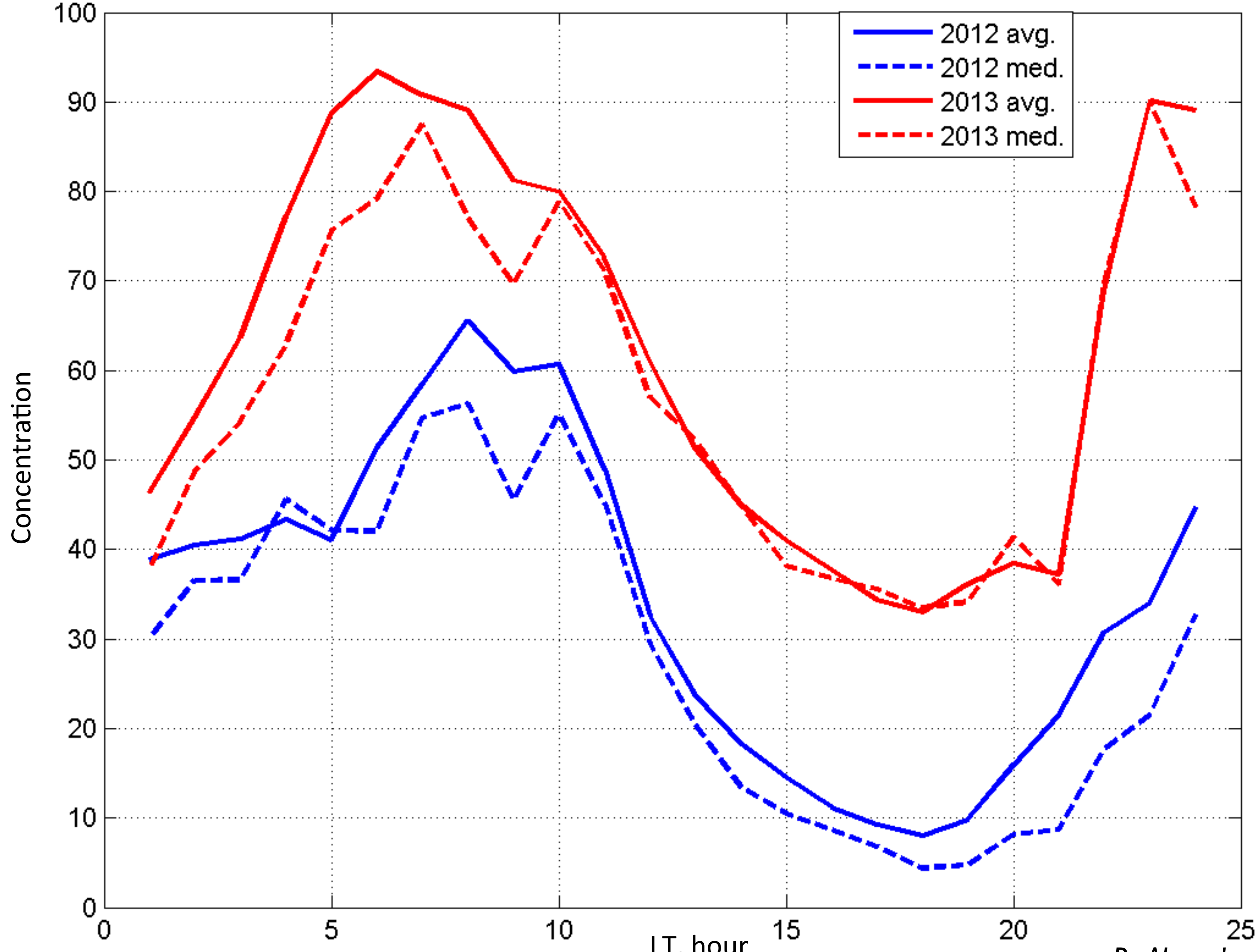


4km resolution



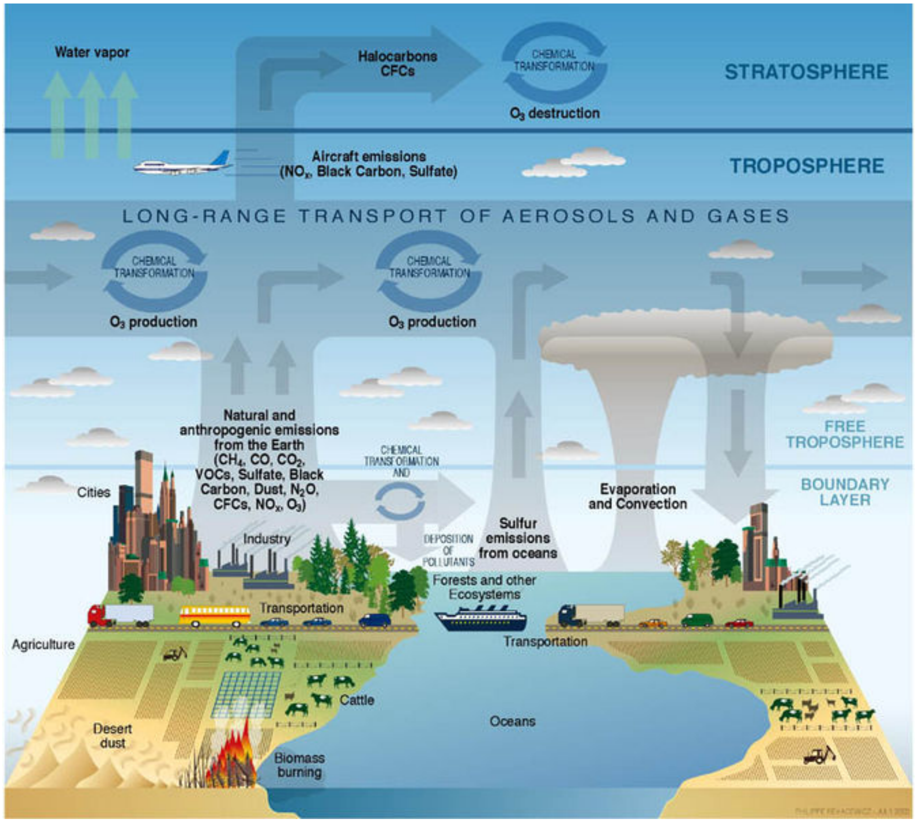
R. Ahmadov, S. McKeen

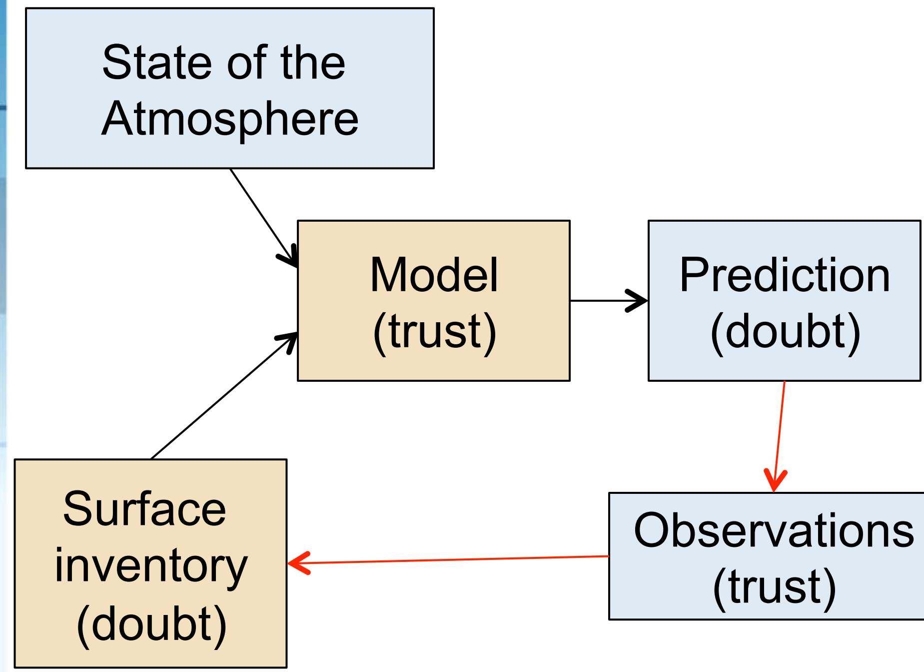
Diurnal cycle of a modeled passive tracer at the HorsePool site, February, 2012, and 2013



R. Ahmadov, S. McKeen

Mesoscale inverse modeling to evaluate emissions

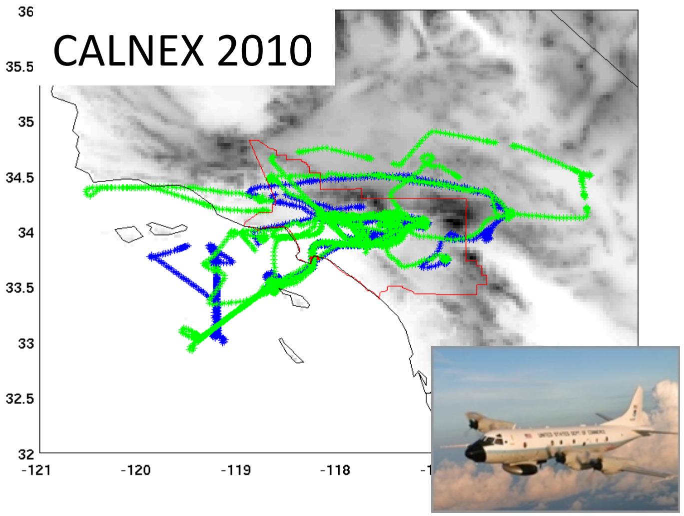




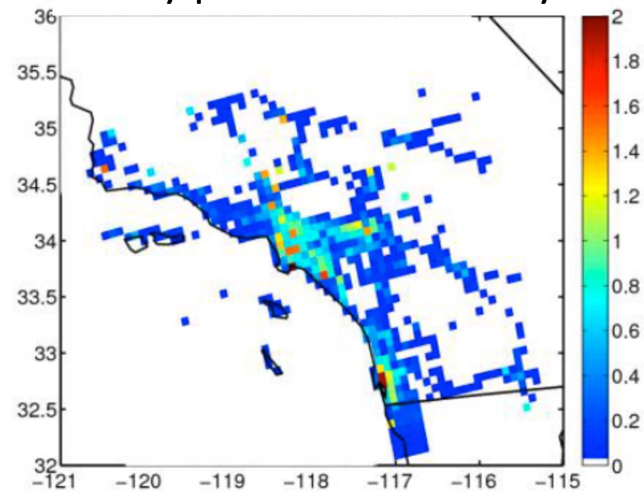
Inverse modeling applied to aircraft measurements helps to improve existing emission inventories at mesoscale

J. Brioude

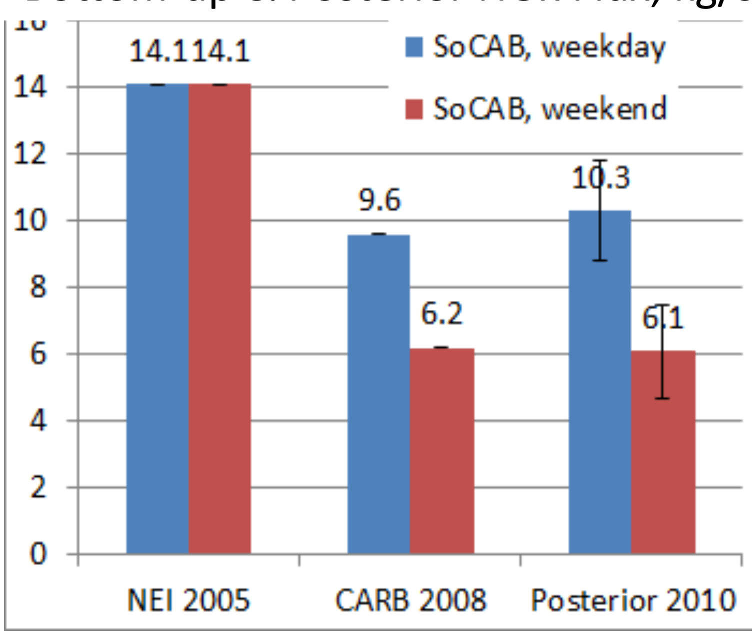
Exmaple: Inverse Modeling of Urban NOx Emissions



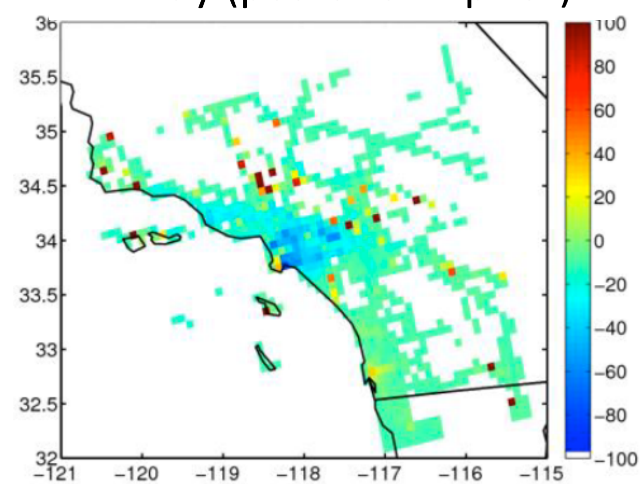
NOy posterior weekday



Bottom-up & Posterior NOx Flux, kg/s



ΔNOy (posterior – prior)



- Aircraft observations lower inverse modeled emissions compared with NEI2005 prior
- Observations show weekend-weekday variations
- CARB inventory captures emissions magnitude & day-of-week cycle better than NEI

J. Brioude et al., Atmos. Chem. Phys., 2013

Concluding remarks

- NOAA's atmospheric measurements can be used to quantitatively assess emissions from oil and gas upstream and midstream activities
  - Our top-down emission estimates are
    - for a specific location and time
    - integrated fluxes from various O&G operations
- These types of studies provide objective evaluations of bottom-up inventories
  - Specifically can be used to assess at the regional scale
    - new inventory methodologies
    - impact of new regulations/practices
- NOAA measurements and modeling probe the processes that lead to air quality and climate impacts
  - We examine ozone distributions, chemical reactivity, basin/boundary layer dynamics, long-range transport, etc.
- Results from several on-going analyses should be available later this year

Planned work for CU NSF SRN

- Field work this summer in the DJB in connection with water sampling study
- Website portal with links to existing studies and public data sets
- National and regional meetings

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