Estimating Biomass Burning Emissions with Toronto FTIR Data

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U of Toronto Atmospheric Observatory (TAO)

Location:
43.66N, 79.40W,
174 masl

↓ Solar tracker

- Primary instrument:
  - ABB Bomem DA8 Fourier transform infrared (FTIR) spectrometer
- Measurements started in 2002
- Urban site (downtown Toronto)

Scientific Objectives

- To assess biomass burning emissions and quantify their emission factors
  - Use TAO FTIR measurements to identify enhancements due transport of biomass burning emissions
  - Use models and satellite data for source attribution
- Other scientific objectives include:
  - To investigate the daily, seasonal, and interannual trace gas variability and trends in an urban setting (downtown Toronto)
  - To characterize the origin of urban pollution events (local or long-range transport) with models (e.g., the GEOS-Chem chemical transport model)

Biomass Burning

- Biomass burning emissions can negatively affect air quality
- Emissions can have photochemical and radiative forcing effects, particularly when plumes are transported to Arctic regions (Amiro et al., 2001)
- Quantifying biomass burning emissions and understanding their transport poses a challenge
  - Types of vegetation burned, the combustion phase (smoldering vs. flaming), and atmospheric conditions at the time of the fire must all be accounted for when quantifying emissions
  - Emitted gases may also decay and undergo chemistry in the atmosphere during transport

Biomass Burning Enhancement Events

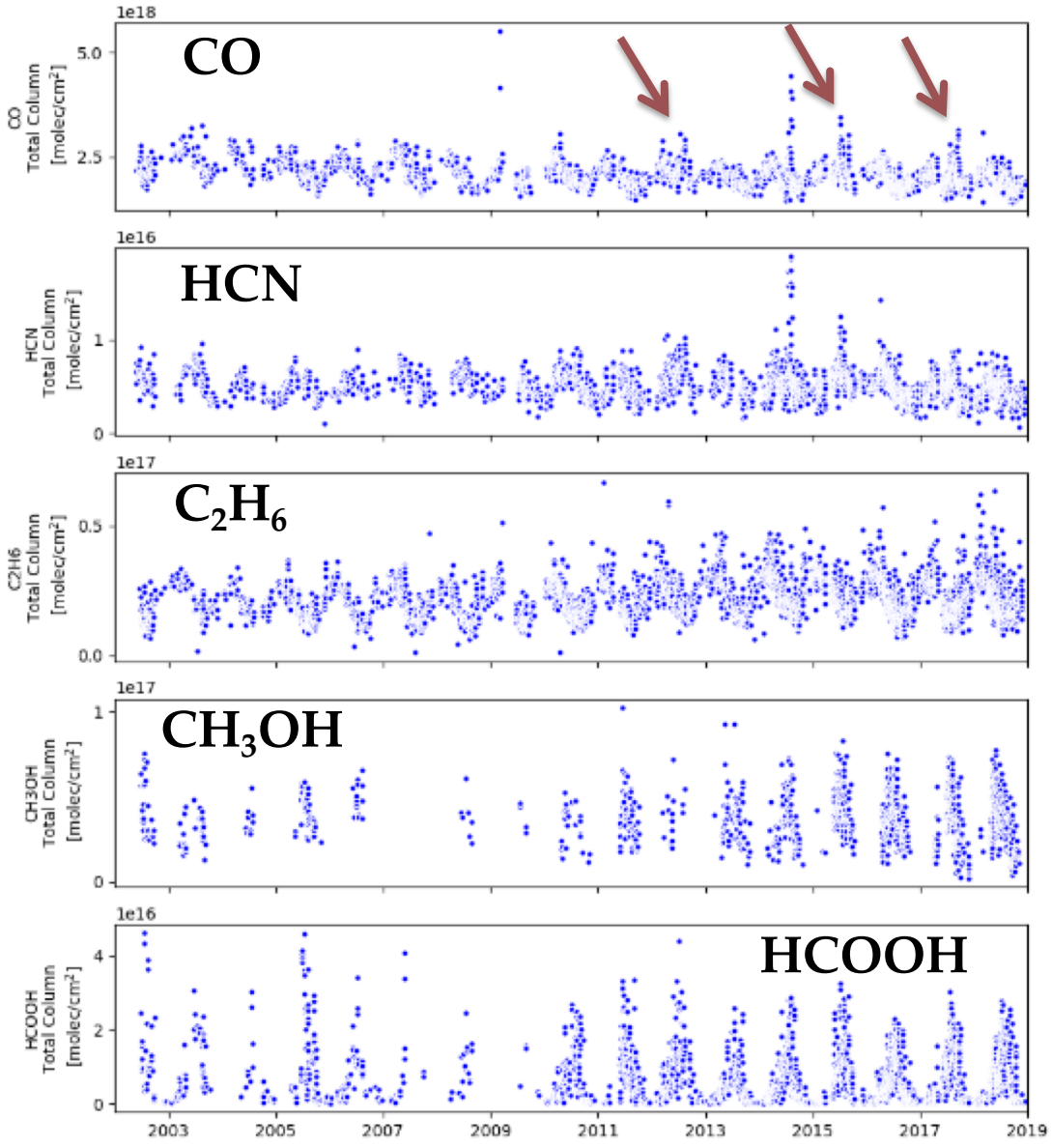
- Wildfires emit numerous chemical species including CO, C_2H_6, HCN, HCOOH, H_2CO, CH_3OH and others
- Given their long lifetimes, CO, HCN and C_2H_6 are good tracers of biomass burning events (e.g., Viatte et al., 2013)
- Formic acid, methanol and formaldehyde have shorter lifetimes
  - Typical lifetimes (τ) are: 61 days (CO), 45 days (C_2H_6), ~150 days (HCN), 4 days (HCOOH), 7.5 days (CH_3OH) and ~1 day (H_2CO)

(Viatte et al., 2013, Millet et al., 2015, Jacob et al., 2005, Pommier et al., 2017)

Biomass Burning Analysis

- Enhancements identified in August 2012, July 2015 and September 2017
  - Simultaneous enhancements of CO, HCN, C_2H_6
    - For 2015 and 2017, HCOOH and CH_3OH enhancements were also seen
- FLEXPART used for source attribution
- Travel-time estimated with HYSPLIT
  - Travel-time used to estimate decay in the atmosphere
- Emission factors and emission ratios reported
- 2015 event analysis will be presented in detail here

Biomass Burning Analysis



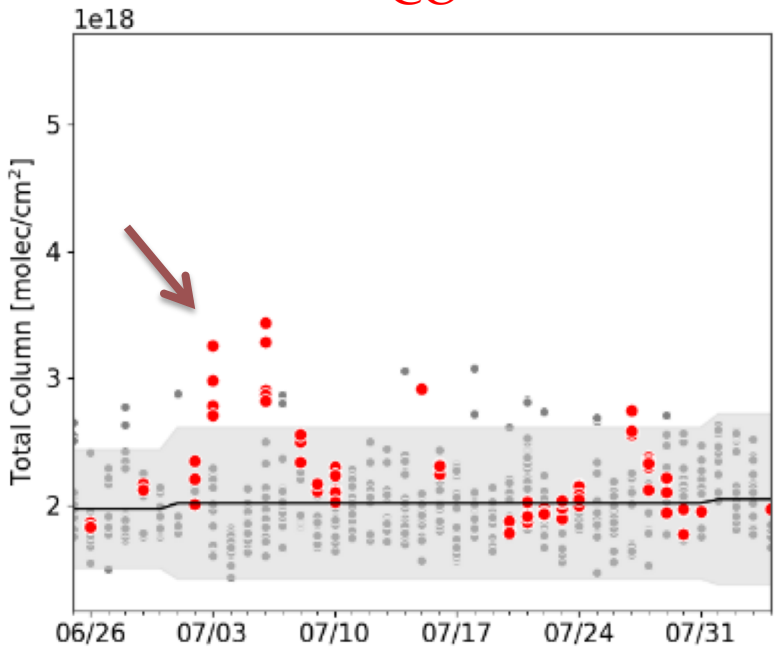
TAO timeseries for CO, HCN, C_2H_6, CH_3OH and HCOOH

- The arrows indicate the 2012, 2015 and 2017 events (in CO observations)
- Also of note is the clear enhancement in 2014 (Lutsch et al., 2016)

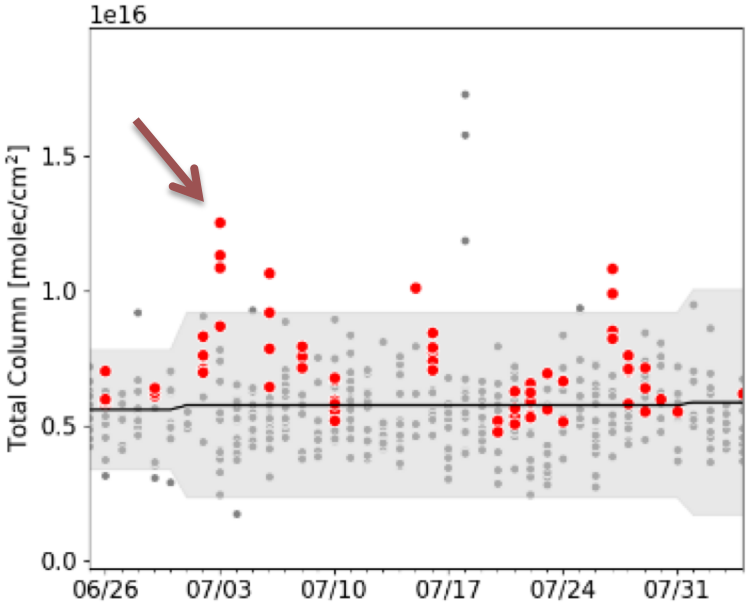
2015 Early July

- Sharp peak in HCN on July 3rd to 6th (indicated by arrows)
- Gray band indicates 2 standard deviations above and below the monthly mean

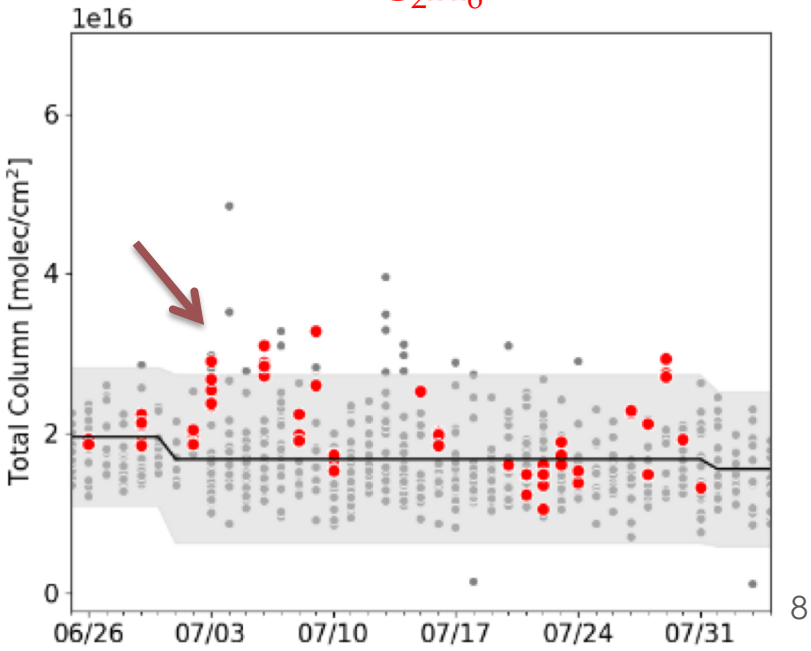
CO



HCN

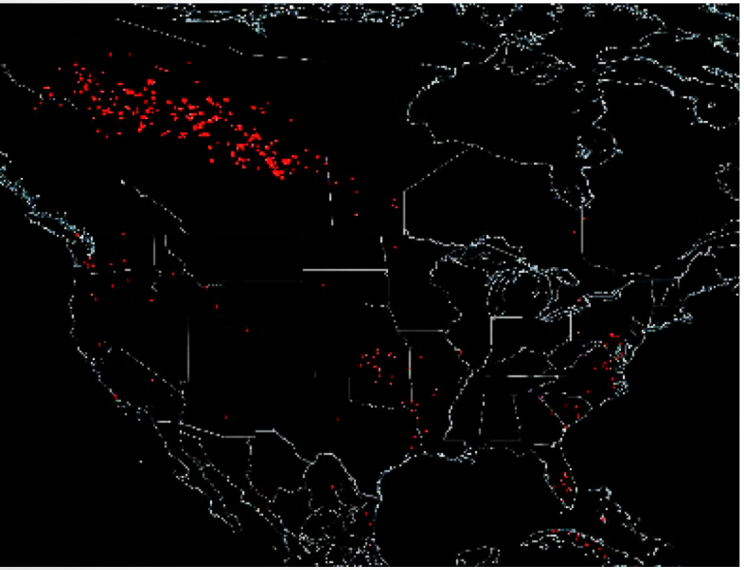


C_2H_6

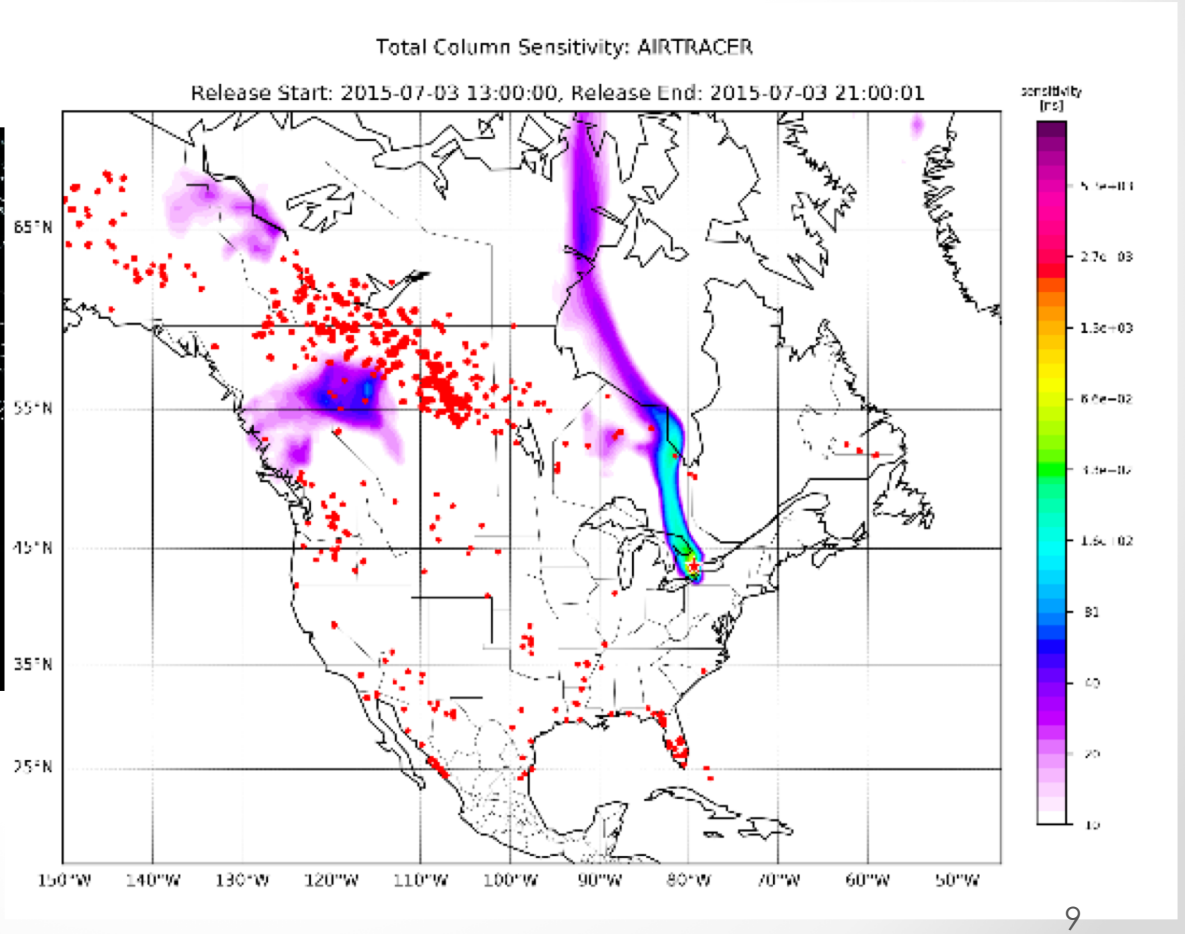


Source Attribution with FLEXPART: 2015

MODIS fire product data for June 29th



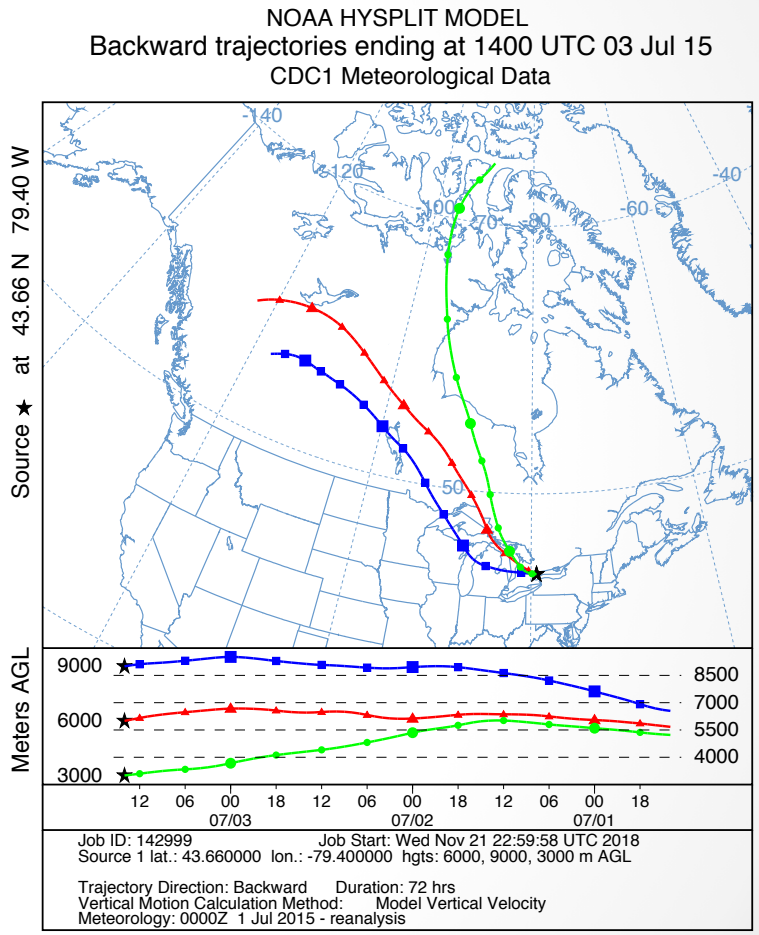
FLEXPART sensitivity plot for July 3rd



Travel Time and Decay

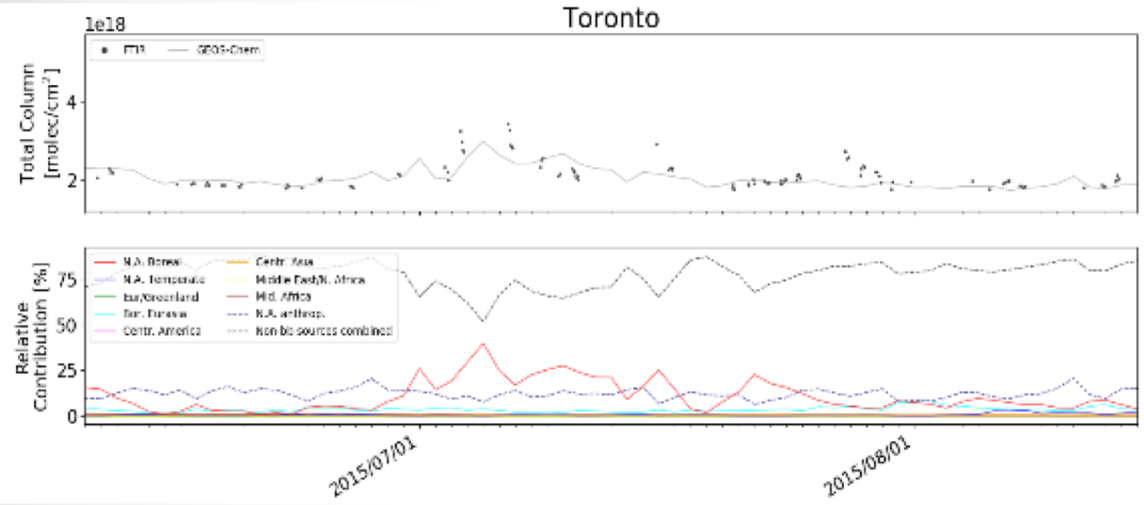
- Chemical species decay exponentially in the atmosphere
  - i.e. $[x]_t = [x]_0 e^{-t/\tau}$

  where τ denotes the lifetime
- HYSPLIT back trajectory was ran with reanalysis meteorological field for travel time estimation

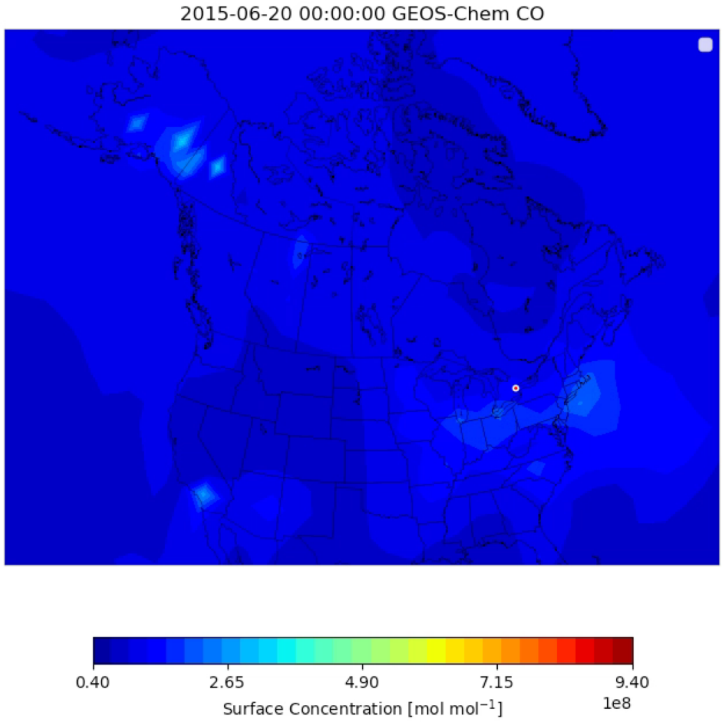


Tagged CO GEOS-Chem (v12)

GEOS-Chem tagged CO

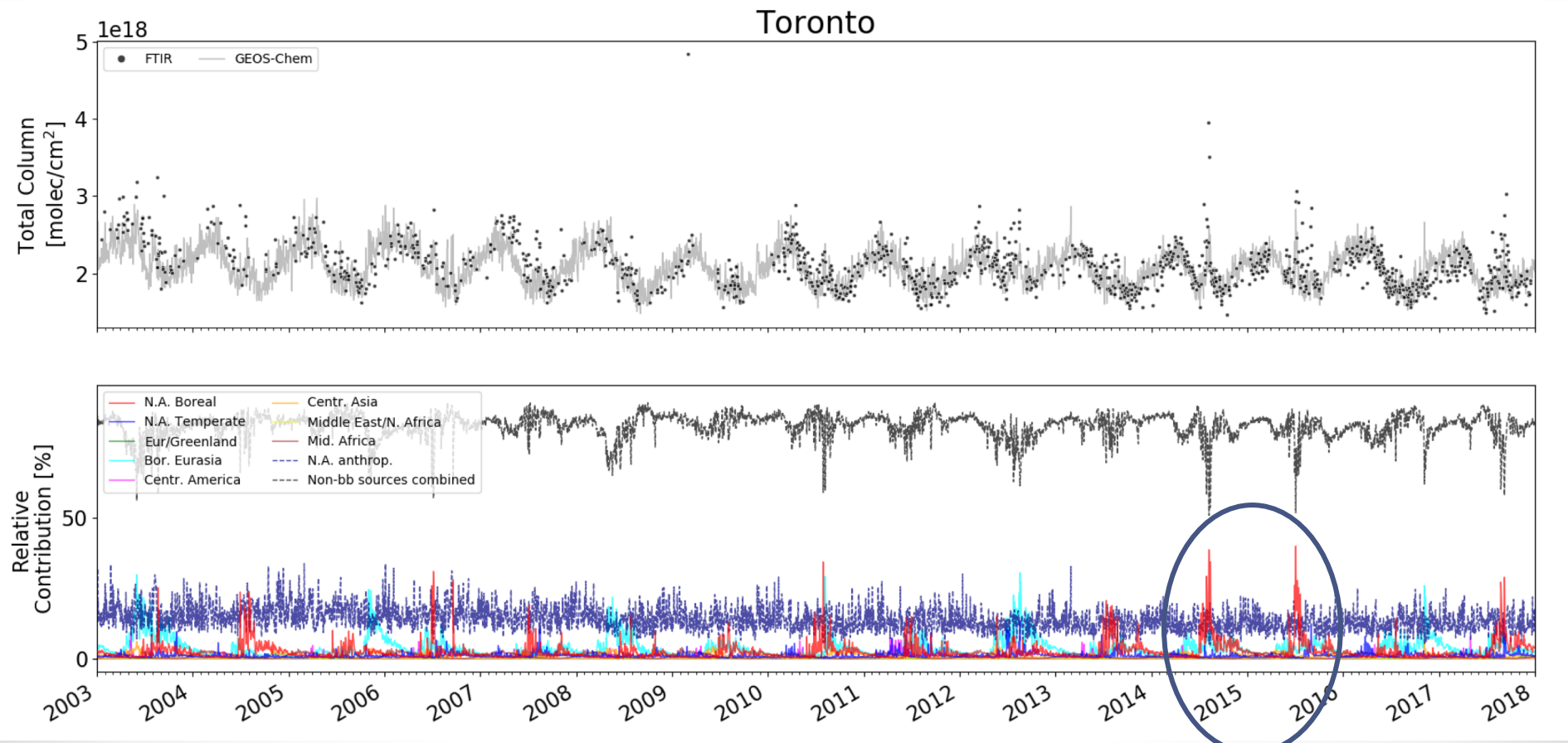


GEOS-Chem surface CO for 2015 July event



Not shown here are similar surface GEOS-Chem outputs for other biomass burning events

Tagged CO Full Timeseries



Note the red spike in 2015

FTIR and GEOS-Chem comparison (top)
Relative contribution (bottom)

Interpreting the Data

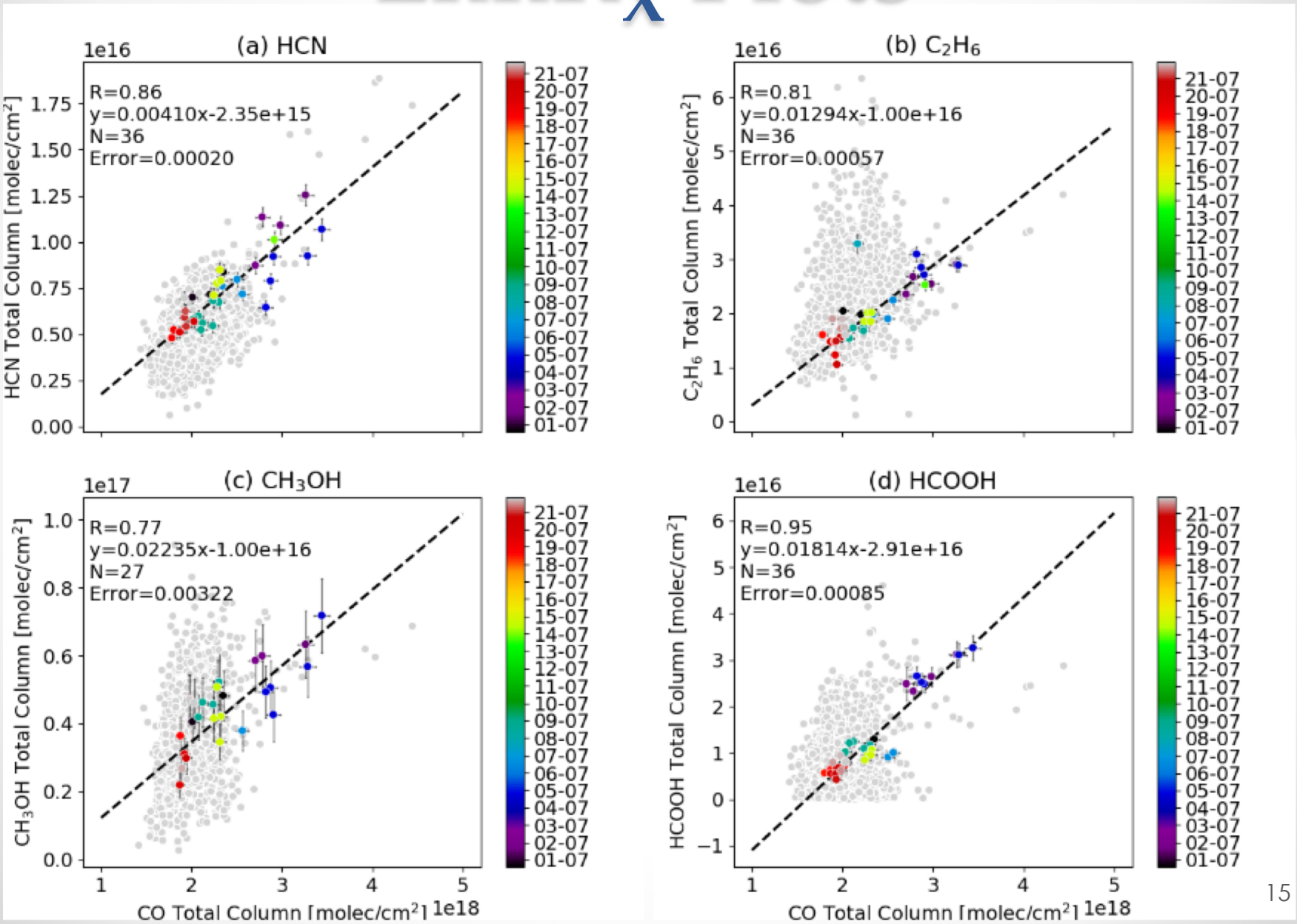
- Enhancement Ratio (EnhR)
  - Ratio of column of species of interest to column of CO
  - Calculated for HCN, C_2H_6, CH_3OH and HCOOH
  - EnhR is the slope of the linear regression when the two species are plotted against each other
  - To account for the travel time decay and to calculate the emission ratio (ER), the following equation is used:

$$ER_X = EnhR_X \left(\frac{e^{t/\tau_X}}{e^{t/\tau_{CO}}} \right)$$

ER and EF

- ER: Emission Ratio
  - Enhancement ratio with plume aging correction
  - Since travel times are accounted for, it is not location specific
- EF: Emission Factor
  - Defined by: $$EF_X = EF_{CO} \cdot ER_X \cdot \left(\frac{MW_X}{MW_{CO}} \right)$$
    - where EF_{CO} is the emission factor of CO, which is 127 ± 45 g/kg (Akagi et al., 2011), and MW are the molecular weights
  - Not location specific

$EnhR_X$ Plots


(a) HCN
1e16
R=0.86
y=0.00410x-2.35e+15
N=36
Error=0.00020
HCN Total Column [molec/cm²]
(b) C₂H₆
1e16
R=0.81
y=0.01294x-1.00e+16
N=36
Error=0.00057
C₂H₆ Total Column [molec/cm²]
(c) CH₃OH
1e17
R=0.77
y=0.02235x-1.00e+16
N=27
Error=0.00322
CH₃OH Total Column [molec/cm²]
CO Total Column [molec/cm²] 1e18
(d) HCOOH
1e16
R=0.95
y=0.01814x-2.91e+16
N=36
Error=0.00085
HCOOH Total Column [molec/cm²]
CO Total Column [molec/cm²] 1e18
21-07
20-07
19-07
18-07
17-07
16-07
15-07
14-07
13-07
12-07
11-07
10-07
09-07
08-07
07-07
06-07
05-07
04-07
03-07
02-07
01-07

Results: HCN

Source	Platform	EF_{co} (g/Kg)	ER	EF (g/Kg)
			HCN	
[a]2012 August	FTIR	89 ± 32	0.0038 ± 0.0006	0.33 ± 0.13
[b]2012 August	FTIR	89 ± 32	0.0038 ± 0.0006	0.32 ± 0.13
[a]2015 July	FTIR	127 ± 45	0.0040 ± 0.0002	0.49 ± 0.18
[b]2015 July	FTIR	127 ± 45	0.0039 ± 0.0002	0.48 ± 0.17
[a]2017 September	FTIR	127 ± 45	0.0038 ± 0.0003	0.47 ± 0.17
[b]2017 September	FTIR	127 ± 45	0.0037 ± 0.00027	0.45 ± 0.16
[a]2017 September	FTIR	89 ± 32	0.0038 ± 0.00027	0.33 ± 0.12
[b]2017 September	FTIR	89 ± 32	0.0037 ± 0.00027	0.32 ± 0.12
[e]Lutsch et al. (2016)	FTIR	127 ± 45	0.0068 ± 0.0003	0.84 ± 0.30
[c]Viatte et al. (2015)	FTIR	127 ± 45	0.00429 ± 0.00245	0.44 ± 0.25
[d]Viatte et al. (2015)	FTIR	127 ± 45	0.00343 ± 0.00094	0.36 ± 0.17
[f]Paton-Walsh et al. (2005)	FTIR	107 ± 37	0.0043 ± 0.0016	0.43 ± 0.22
Akagi et al. (2011)	Compilation	127 ± 45	-	1.52 ± 0.82
Akagi et al. (2011)	Compilation	89 ± 32	-	0.73 ± 0.19
[g]Goode et al. (2000)	Aircraft	88.8	0.0069	0.61

[a],[b]This study.
[a]HCN and CO lifetimes 150 and 61 days respectively.
[b]HCN and CO lifetimes 75 and 30 days respectively.
[c],[d]Values from Thule and Eureka respectively.
[e]Values from Toronto.
[f]Values from Australian fires.
[g]Values from Alaskan fires.

Results: C_2H_6

Source	Platform	EF_{co} (g/Kg)	ER C_2H_6	EF (g/Kg)
[a]2012 August	FTIR	89 ± 32	0.0091 ± 0.0012	0.87 ± 0.33
[b]2012 August	FTIR	89 ± 32	0.0089 ± 0.0012	0.85 ± 0.33
[a]2015 July	FTIR	127 ± 45	0.013 ± 0.0006	1.79 ± 0.64
[b]2015 July	FTIR	127 ± 45	0.013 ± 0.0006	1.72 ± 0.62
[a]2017 September	FTIR	127 ± 45	0.019 ± 0.0011	2.64 ± 0.95
[b]2017 September	FTIR	127 ± 45	0.019 ± 0.0011	2.53 ± 0.91
[a]2017 September	FTIR	89 ± 32	0.019 ± 0.0011	1.85 ± 0.67
[b]2017 September	FTIR	89 ± 32	0.019 ± 0.0011	1.77 ± 0.65
[e]Lutsch et al. (2016)	FTIR	127 ± 45	0.0101 ± 0.0005	1.38 ± 0.49
[c]Viatte et al. (2015)	FTIR	127 ± 45	0.01211 ± 0.00476	1.39 ± 0.68
[d]Viatte et al. (2015)	FTIR	127 ± 45	0.00956 ± 0.00319	1.09 ± 0.74
[f]Paton-Walsh et al. (2005)	FTIR	107 ± 37	0.0023 ± 0.0005	0.26 ± 0.11
Akagi et al. (2011)	Compilation	127 ± 45	-	1.79 ± 1.14
Akagi et al. (2011)	Compilation	89 ± 32	-	1.12 ± 0.67
[g]Goode et al. (2000)	Aircraft	88.8	0.0073	0.66

[a],[b]This study.
[a]HCN and CO lifetimes 150 and 61 days respectively.
[b]HCN and CO lifetimes 75 and 30 days respectively.
[c],[d]Values from Thule and Eureka respectively.
[e]Values from Toronto.
[f]Values from Australian fires.
[g]Values from Alaskan fires.

Results: CH_3OH

Source	Platform	EF_{co} (g/Kg)	ER	EF (g/Kg)
			CH_3OH	
[a]2012 August	FTIR	89 ± 32	-	-
[b]2012 August	FTIR	89 ± 32	-	-
[a]2015 July	FTIR	127 ± 45	0.030 ± 0.0064	4.35 ± 1.80
[b]2015 July	FTIR	127 ± 45	0.029 ± 0.0056	4.17 ± 1.68
[a]2017 September	FTIR	127 ± 45	0.045 ± 0.008	6.54 ± 2.60
[b]2017 September	FTIR	127 ± 45	0.043 ± 0.007	6.27 ± 2.43
[a]2017 September	FTIR	89 ± 32	0.045 ± 0.008	4.58 ± 1.85
[b]2017 September	FTIR	89 ± 32	0.043 ± 0.007	5.39 ± 1.72
[e]Lutsch et al. (2016)	FTIR	127 ± 45	-	-
[c]Viatte et al. (2015)	FTIR	127 ± 45	-	-
[d]Viatte et al. (2015)	FTIR	127 ± 45	0.02813 ± 0.01252	3.44 ± 1.68
[f]Paton-Walsh et al. (2005)	FTIR	107 ± 37	-	-
Akagi et al. (2011)	Compilation	127 ± 45	-	2.82 ± 1.62
Akagi et al. (2011)	Compilation	89 ± 32	-	1.93 ± 1.38
[g]Goode et al. (2000)	Aircraft	88.8	0.0135	1.41

[a],[b]This study.
[a]HCN and CO lifetimes 150 and 61 days respectively.
[b]HCN and CO lifetimes 75 and 30 days respectively.
[c],[d]Values from Thule and Eureka respectively.
[e]Values from Toronto.
[f]Values from Australian fires.
[g]Values from Alaskan fires.

Results: HCOOH

Source	Platform	EF_{co} (g/Kg)	ER	EF (g/Kg)
			HCOOH	
[a]2012 August	FTIR	89 ± 32	-	-
[b]2012 August	FTIR	89 ± 32	-	-
[a]2015 July	FTIR	127 ± 45	0.015 ± 0.0012	3.07 ± 1.12
[b]2015 July	FTIR	127 ± 45	0.014 ± 0.0012	2.94 ± 1.07
[a]2017 September	FTIR	127 ± 45	0.0071 ± 0.0034	1.48 ± 0.88
[b]2017 September	FTIR	127 ± 45	0.013 ± 0.008	2.64 ± 1.87
[a]2017 September	FTIR	89 ± 32	0.0071 ± 0.0034	1.03 ± 0.62
[b]2017 September	FTIR	89 ± 32	0.013 ± 0.008	1.85 ± 1.31
[e]Lutsch et al. (2016)	FTIR	127 ± 45	-	-
[c]Viatte et al. (2015)	FTIR	127 ± 45	0.01790 ±0.00937	3.14 ± 1.46
[d]Viatte et al. (2015)	FTIR	127 ± 45	0.01531 ± 0.00403	2.69 ± 1.14
[f]Paton-Walsh et al. (2005)	FTIR	107 ± 37	-	-
Akagi et al. (2011)	Compilation	127 ± 45	-	0.57 ± 0.46
Akagi et al. (2011)	Compilation	89 ± 32	-	0.35 ± 0.33
[g]Goode et al. (2000)	Aircraft	88.8	0.0062	0.99

[a],[b]This study.
[a]HCN and CO lifetimes 150 and 61 days respectively.
[b]HCN and CO lifetimes 75 and 30 days respectively.
[c],[d]Values from Thule and Eureka respectively.
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[f]Values from Australian fires.
[g]Values from Alaskan fires.

Data Analysis

- Trend analysis
  - Fitting a trended Fourier series
- Pollution events
  - Pollution events can be identified by using residuals of the fit (Zellweger et al., 2009)
- Statistical significance of trends can be examined using a method outlined by Weatherhead et al. (1998)
- Bootstrap resampling to find confidence intervals (Gardiner et al., 2008)

Long-term Trend Analysis

- Trend analysis:
  - Fitting a trended Fourier series:

$$F(t) = a_0 + \frac{a \cdot t}{T} + \sum_{\omega \in \Omega} \alpha_\omega \sin\left(\frac{\omega t}{T}\right) + \beta_\omega \cos\left(\frac{\omega t}{T}\right)$$

where T is the total time of the data

- Pollution events are defined by:
  - Finding residuals, ie. Observation minus $F(t)$
  - Mirroring negative residuals as well as calculating the standard deviation of the residuals
  - Any measurements that are 2 standard deviations or higher than the fit and residuals are considered pollution events

(Zellweger et al., 2009; Gardiner et al., 2008)

Trend Analysis and Pollution Event Identification

- For both trend analysis and identifying pollution events (by looking at residuals), fitting a trended Fourier series is needed
- How do we know if we are fitting the data correctly?
  - Try several fits of different orders and varying frequencies
  - Look for correlation between the fit and the data
  - Look at the residuals and test for normality (using Kolmogorov-Smirnov test)
    - If the fit captures all of the natural variability and trend in the data, then the residuals will tend towards a normal distribution given enough sample size due to central limit theorem
    - If residuals exhibit normality at low order but not in higher orders, it means an artificial pattern was added to the data i.e. overfitting

Fitting TAO Data: *Preliminary Results*

- Even at high orders (6+) over fitting was not observed
- Higher order fits led to marginally better correlation and lower RMS in the residuals
- Higher order fits did not change the number of years needed for trend detection
  - E.g. CH_4: ~12 years, N_2O: ~17 years, C_2H_6: ~26 years
- Currently working on
  - Utilizing other tests of normality (Lilliefors and Jarque–Bera)
  - Allowing the amplitude of the annual cycle to change over time (linearly)
  - Effects of varying the Q-value (for bootstrap reanalysis)
  - Applying similar analyses to other NDACC data

Summary and Discussion

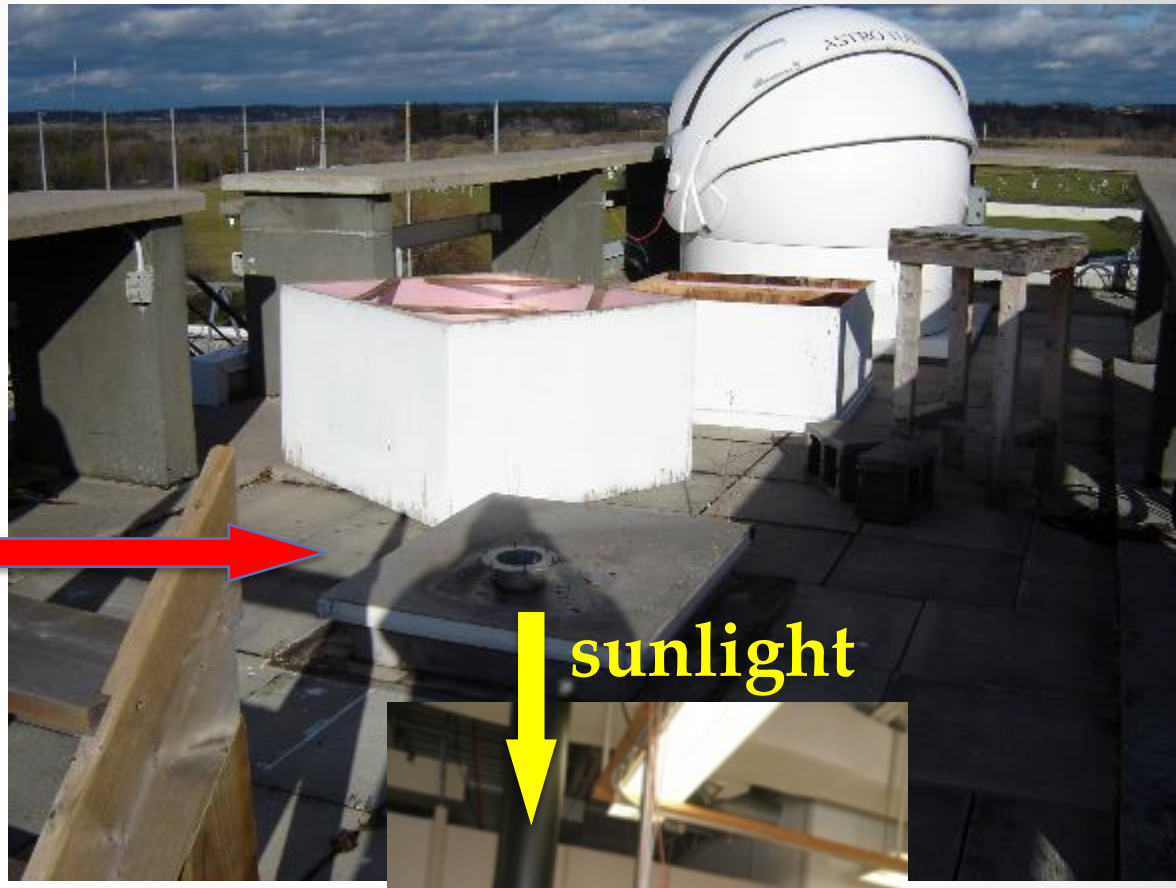
- The 15+ years of data collected at TAO were analyzed:
  - Analysis of several biomass burning events
  - Examination of trends and pollution events
- Emission factors of several biomass burning events seen in Toronto were calculated
  - FLEXPART in conjunction with MODIS Fire Product was used for source attribution
  - GEOS-Chem tagged CO simulation was used to supplement the FTIR observations
- Statistics for trend analysis methods were examined in the context of TAO data

Future Work

- Trend analysis of hydrocarbons and pollutants
- Analysis of pollution events with a focus on O_3 and CO
  - Utilize tagged O_3 GEOS-Chem model for in-depth analysis of Toronto O_3
- Further integration of measurements and modeling (GEOS-Chem)
- AmmonAQ project in collaboration with LATMOS
  - Analysis of urban air quality focusing on NH_3, using Toronto and Paris as benchmark cities
- Comparison of urban (TAO) and rural (ECCC Egbert) FTIR measurements
  - Egbert is about 80 km north of downtown Toronto (TAO)
  - Egbert FTIR is in good working condition, a new sun tracker has been installed

Egbert FTIR & Sun Tracker

Bruker solar tracker installed here



sunlight

DA8 FTIR→

Egbert FTIR & Sun Tracker

- Egbert can complement TAO data
- As a rural site, it can be a point of comparison for an urban site like TAO
- High-resolution models (GEOS-Chem) can differentiate between the two sites

↑With the housing opened

Acknowledgements

- NSERC CREATE Training Program in Technologies for Exo-Planetary Science (TEPS)

- ABB Bomem, CFCAS, CFI, CRESTech, CSA, ECCC, NSERC, ORDCF, PREA, University of Toronto

- The many interns, students, and postdocs who have made TAO FTIR measurements, including (most recently): Orfeo Colebatch, Ellen Eckert, Lei Liu, Erik Lutsch, and Tyler Wizenberg

Additional Slides: Bootstrap Reanalysis

- Bootstrap resampling is done to find confidence intervals
  - It is an analysis where residuals are randomly redistributed to form a "new" set of data, where another line is refitted
  - This process is repeated (over several hundred to thousands of times)
  - The ensemble of data is analyzed to find the 2-σ confidence interval (2.5% to 97.5% coverage of the data)

(Gardiner et al., 2008)

Additional Slides: Weatherhead Method

- Detection of long-term, linear trends is affected by a number of factors
  - Size of the trend to be detected
  - Time span of the data
  - Magnitude of variability and autocorrelation
- The number of years of data needed to detect a trend strongly depends on (and increases with) the magnitude of variance and the autocorrelation coefficient
- Environmental time series data are often autocorrelated

(Weatherhead et al., 1998)

Additional Slides: More on Biomass Burning

- "As burning occurs, it can release hundreds of years worth of stored carbon dioxide into the atmosphere in a matter of hours"
  - NASA, Earth Observatory (earthobservatory.nasa.gov)
- "Exposure to biomass burning particles is strongly associated with cardiovascular disease, respiratory illness, lung cancer, asthma and low birth weights"
  - Stanford News, 2014 (news.stanford.edu)

Additional Slides: Atmospheric Formic Acid

- Formic acid is one of the most abundant acids in the atmosphere
- Greatly contributes to free precipitation acidity
- Affects aqueous phase chemistry
  - pH-dependent reaction rates
- Naturally produced photochemically, though emission sources (including anthropogenic) also certainly exist

(Millet et al., 2015)

Additional Slides: Retrieval

- A retrieval algorithm called SFIT4 is used to derive vertical profiles and/or columns of trace gases from their absorption and emission spectra:
  - Identify spectral lines of interest
  - Generate a model atmosphere using meteorology data from NCEP and WACCM
  - Use a forward model to simulate a model spectrum
  - Iteratively adjust the *a priori* VMR profile until the model spectrum agrees with the measured spectrum
- SFIT4 is an optimal estimation method (OEM) analysis
  - It uses both measurement data and the *a priori* information
  - It assigns weights to the *a priori* information and the measurement based on each of their uncertainties