Overview of the Long-term Analysis of Toronto-Area Atmospheric Composition

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Scientific Objectives

- Make use of the time series of observations of trace gases measured with an Fourier Transform InfraRed (FTIR) spectrometer situated in downtown Toronto at the University of Toronto Atmospheric Observatory (TAO)
- The scientific objectives discussed in this talk are:
  1. To quantify trends in the time series of trace gas concentrations over Toronto, and for CO, to examine if it is possible to identify what drives the trend
  2. To determine how emissions from biomass burning events affect air quality over Toronto, and whether observations in Toronto be used to quantify emissions
  3. To examine the spatial representativeness of the TAO FTIR NH_3 columns, and temporal variability of NH_3 in Toronto

U of Toronto Atmospheric Observatory (TAO)

Location:
43.66N, 79.40W,
174 m asl

↓ Solar tracker

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

GEOS-Chem

- The GEOS-Chem (geos-chem.org) global 3D chemical transport model (CTM) was used to interpret observational data
  - All GEOS-Chem model runs used in these studies were run in 2° × 2.5° resolution (latitude × longitude), and were driven by MERRA-2 meteorological fields
- Can be run in several modes:
  - Standard mode – full chemistry
    - NH_3 from the model was examined
  - Tagged mode – useful for identifying sources
    - Tagged CO and tagged CH_4 simulations were used

1. Long-term Trends

- Trends of greenhouse gases
  - Quantifying methane (CH_4) and nitrous oxide (N_2O) trends
    - CH_4 and N_2O have a high global warming potential (higher than CO_2)
- Estimating trends of pollutants
  - Ammonia (NH_3); Positive trends observed
  - Carbon monoxide (CO)
  - Tropospheric ozone (O_3); While air quality overall is improving in Toronto, O_3 continues to exhibit exceedances
- The trend was also used to examine the effects of the Lockdown due to COVID-19 on air quality

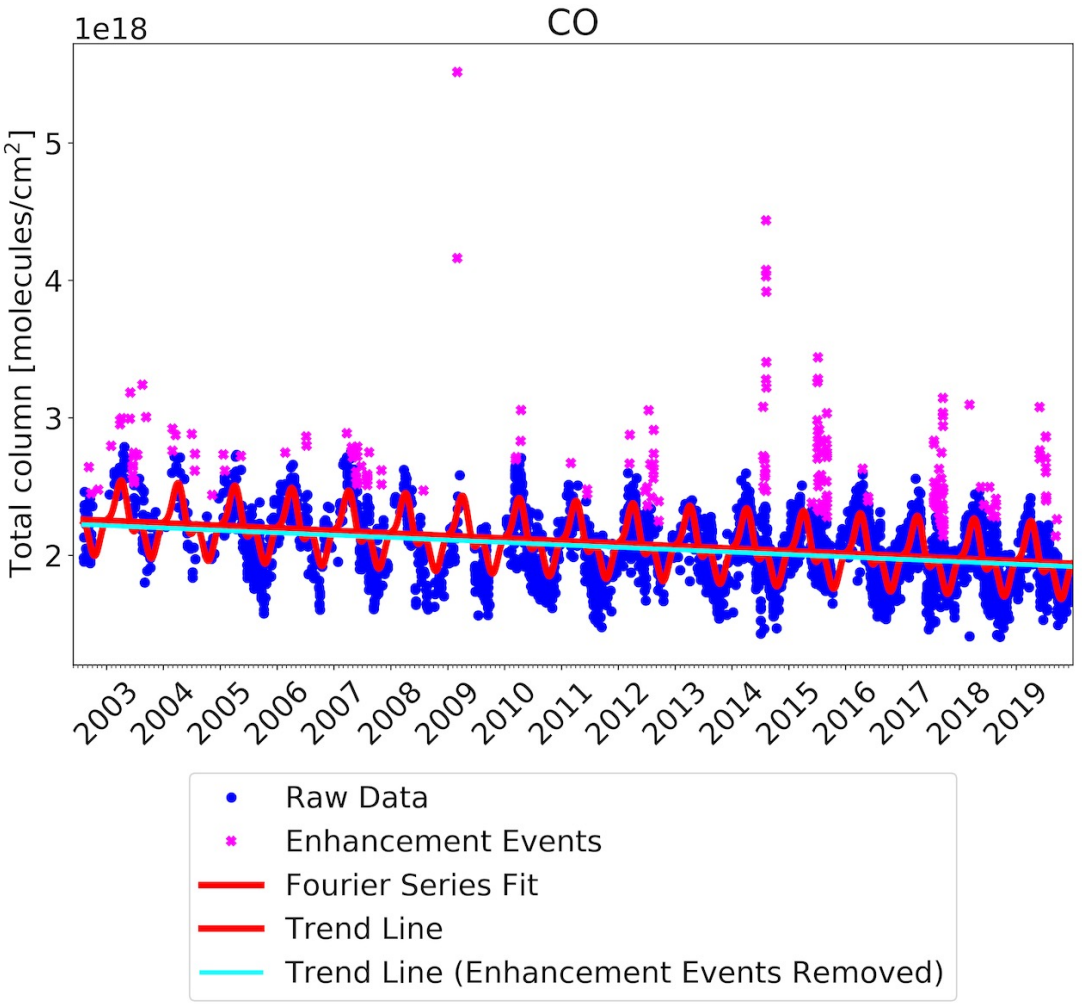
Associated paper: Yamanouchi et al., 2021
Published in ERC (May, 2021)

Trend Analysis

- Trend analysis:
  - Fitting a trended Fourier series
- Enhanced outliers
  - Pollution events can be identified by using residuals of the fit, following the work done by Zellweger et al., 2009
- Bootstrap resampling to find confidence intervals
- For CH_4 and C_2H_6, the trends were separated from 2002-2008 and 2009-2019 (following Franco et al., 2016)
- For O_3, the column was split into tropospheric and stratospheric column
- For CO, a GEOS-Chem tagged run was used to estimate sources of trends
- For CH_4, a GEOS-Chem tagged run was used to estimate major sources

CO

- Negative trend of −0.90 ± 0.07 %/year
- GEOS-Chem tagged CO simulation showed:
  - CH_4 oxidation to be a major source of CO over Toronto, accounting for 30.9 ± 4.4 % of CO
  - North American fossil fuel emissions accounted for 13.9 ± 4.0 %, while Asian fossil fuel sources accounted for 16.8 ± 5.3 %
  - Fossil fuel contributions exhibit seasonality, peaking in winter



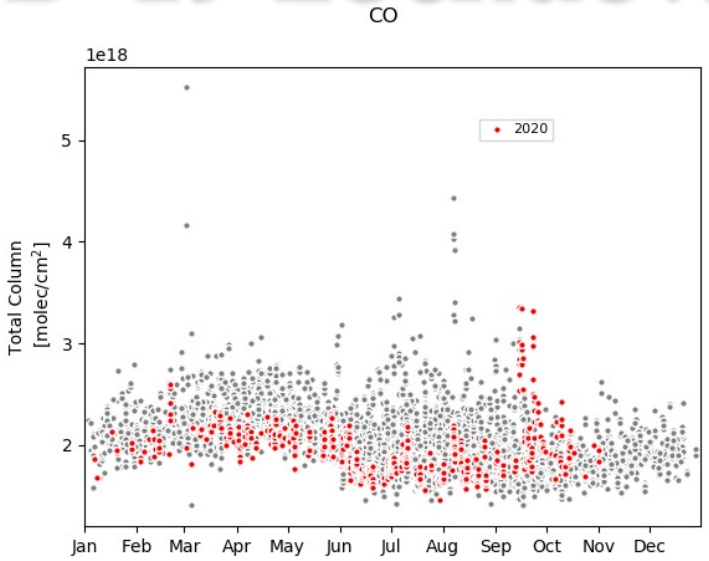
Observed Trends

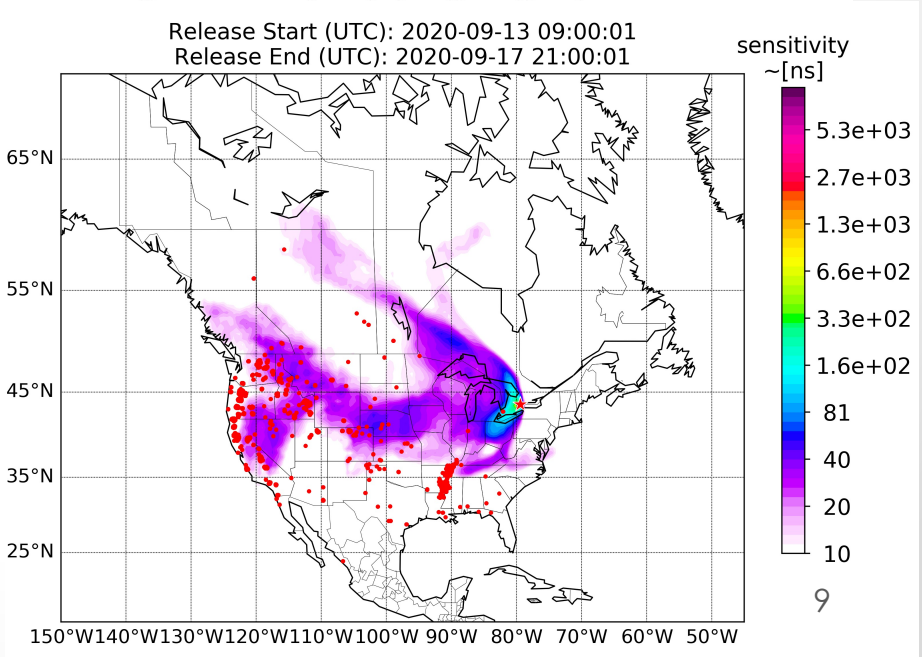
- CH_4
  - 2002 to 2008: 0.26 ± 0.10 %/year
  - 2009-2019: 0.41 ± 0.03 %/year
  - Tagged CH_4 run shows that major sources were wetland emissions, which contributed 31.7 ± 0.5 %
- C_2H_6
  - 2002 to 2008: −0.74 ± 0.73 %/year (decreasing)
  - 2009 to 2019: positive trends of 1.19 ± 0.27
- N_2O
  - 0.28 ± 0.02 %/year
    - This is similar to findings from other FTIRs and an estimate from surface observations given by IPCC (2007)
- O_3
  - Tropospheric O_3 showed a positive trend of 0.28 ± 0.19 %/year
  - Stratospheric O_3 did not exhibit a significant trend
- Other species
  - C_2H_2, HCOOH showed negative trends, while NH_3 was increasing
  - HCl and HF showed negative and positive trends, respectively

Effects of the COVID-19 Lockdown

- Preliminary findings using TAO data:
  - Comparing extrapolated 2020 values using trend data:
    - CO was more than 1σ lower in April
    - Tropospheric O_3 was more than 1σ lower in May and July
  - CO showed a spike in September, 2020
    - Back-trajectory analysis shows this was probably caused by transported wildfire plumes
- The top figure on the right shows 2020 CO total column in red, and 2002-2019 in grey, and the bottom shows back-trajectory analysis to investigate CO spike in October. Red dots indicate fires.
  - Back-trajectory analysis will also be used in the coming slides

Discussed in more detail
in Yamanouchi, 2021 (PhD thesis)





2. 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 (Coogan et al., 2019)
- Quantifying biomass burning emissions and understanding their transport poses a challenge
    - Emissions are sensitive to the type of fuel burnt, and the atmospheric and fire conditions
    - Emitted gases may also undergo chemical transformation in the atmosphere during transport

Associated paper: Yamanouchi et al., 2020
Published in JGR (August, 2020)

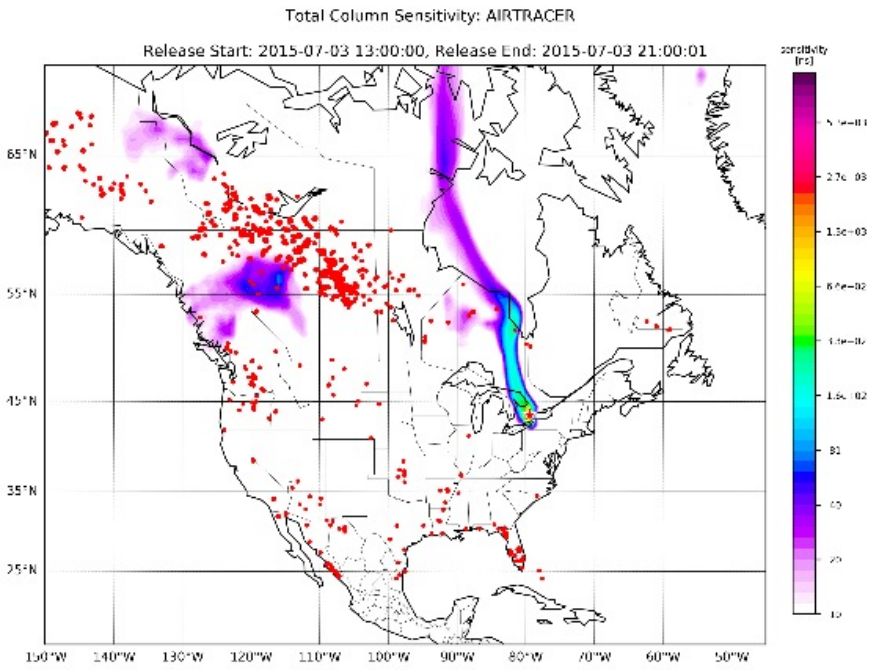
Biomass Burning Analysis

- Enhancements identified in August 2012, July 2015 and September 2017
  - Simultaneous enhancements of CO, HCN, C_2H_6
    - Typical biomass burning species, and often used as tracers
    - For 2015 and 2017, HCOOH and CH_3OH enhancements were also seen
- FLEXPART back-trajectory model used for source attribution
- Emission factors and emission ratios reported
- 2015 event analysis will be presented in detail here

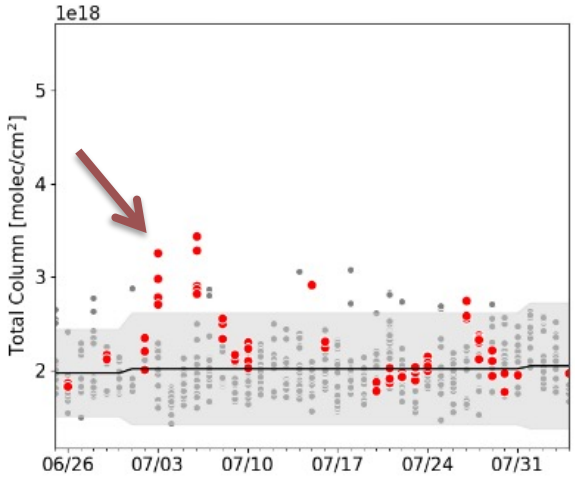
2015 Early July

- Figures below show CO, HCN, and C_2H_6 (2015 in red)
  - Sharp peak in HCN on July 3rd to 6th (indicated by arrows)
  - Gray band indicates 2 standard deviations above and below the monthly mean
- Figure to the right shows the FLEXPART back-trajectory run for July 3
  - The plume likely originated in Alberta and the rest of Western Canada

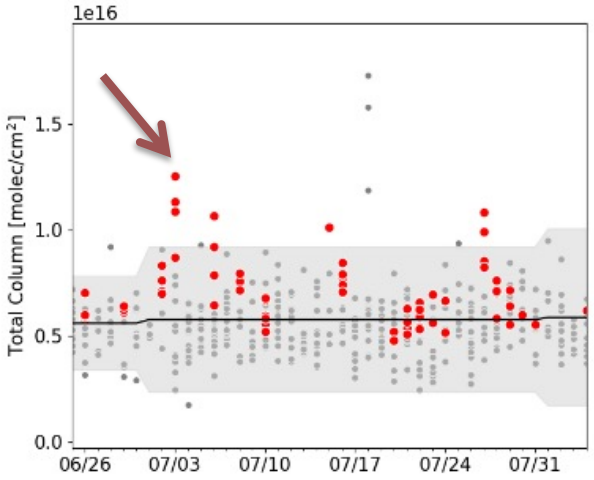
FLEXPART sensitivity plot for 3 July 2015



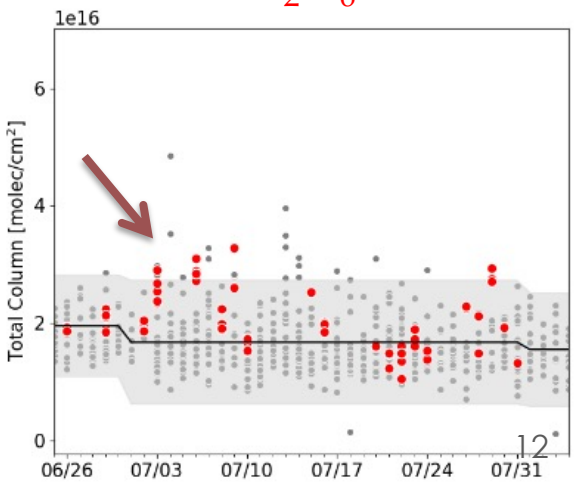
CO



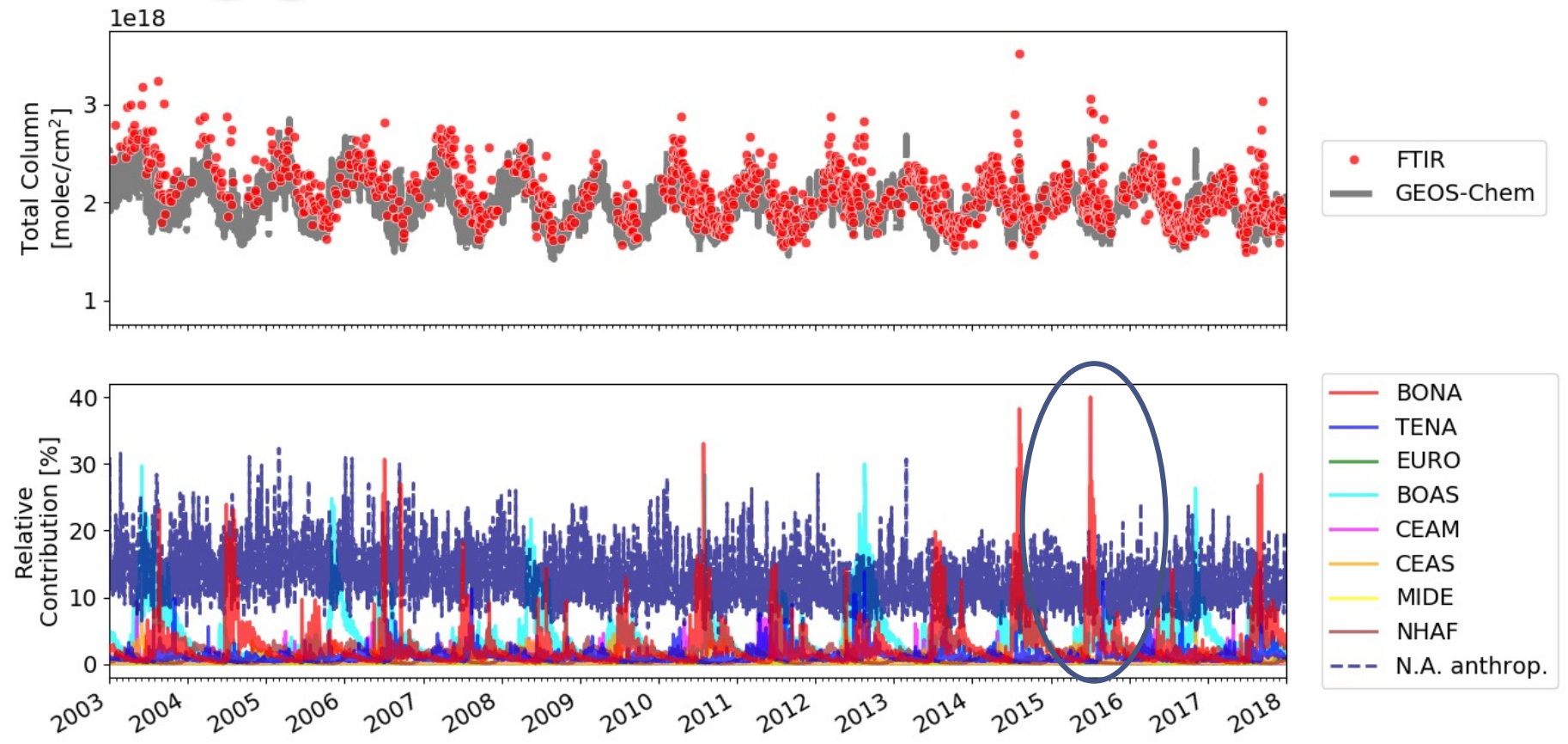
HCN



C_2H_6



Tagged CO Full Timeseries



Note the red spike in 2015

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

Emission Factor Estimates

- Emission factors (EF) were estimated by using literature values (e.g., Akagi et al., 2011) of EF of CO
- Estimates from this study:
  - HCN: 0.32 ± 0.13 to 0.70 ± 0.27 g/kg
  - C_2H_6: 1.17 ± 0.44 to 2.64 ± 0.95 g/kg
  - CH_3OH: 4.17 ± 1.68 to 6.54 ± 2.60 g/kg
  - HCOOH: 5.72 ± 3.04 to 8.51 ± 4.78 g/kg
- The estimates for CH_3OH and HCOOH are likely biased high, as there are many other sources
  - E.g., for the 2015 event, contributions from the wetlands region southwest of Hudson Bay as well as the Pittsburgh/Cleveland area can be seen
- The methodology does not account for chemical production during transport

3. Atmospheric Ammonia

- Ammonia (NH_3) is an important pollutant
  - A source of fine particulate matter in the atmosphere
  - Involved in numerous biochemical exchanges affecting all ecosystems, including acidification and eutrophication of soils and surface waters
- Particulate matter, especially that smaller than 2.5 microns ($PM_{2.5}$), poses serious health hazards
- NH_3 emissions are regulated in some parts of the world (e.g., 1999 Gothenburg Protocol)

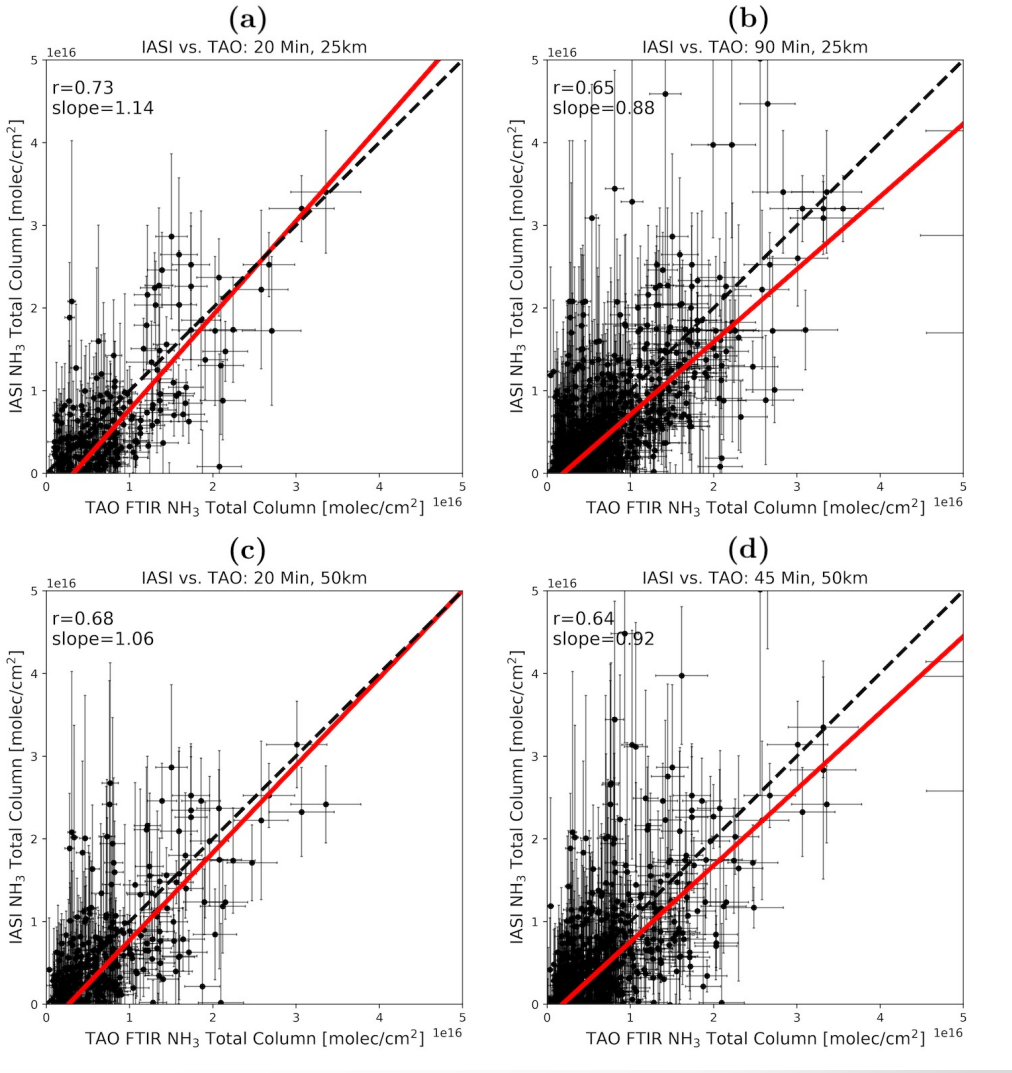
Associated paper: Yamanouchi et al., 2021
Published in AMT (February, 2021)

NH_3 Observations over Toronto

- Analyzed datasets:
  - Total column data from TAO
  - In-situ measurements of NH_3 (ECCC NAPS)
    - 2014-2017 for downtown Toronto
  - IASI satellite NH_3 columns
    - 2008-2018
  - Modeled NH_3
    - GEOS-Chem chemical transport model (v11-01)
      - 2 x 2.5 resolution, 2003-2018
- Datasets were used to quantify NH_3 temporal variability over Toronto
- The multiscale datasets were also compared to assess the representativeness of TAO NH_3 measurements
- GEOS-Chem CTM was also compared against observations

NH_3 Observations over Toronto

- IASI and TAO columns was compared by various coincidence criteria
  - 25 km to 100 km spatial criteria and 20 minutes to 90 minutes temporal criteria
  - Select plots shown
- GEOS-Chem data was compared against both TAO data (point measurement) and IASI (larger regional scale), for assessing model performance on various scales



NH_3 Observations over Toronto

Trends in NH_3 over Toronto observed:

- All show positive trends
- 3.56 ± 0.85, 8.88 ± 5.08, and 8.38 ± 1.54 %/year for TAO, NAPS, and IASI, respectively
- Trends without enhanced outliers were smaller than with; this suggests that enhancements in NH_3 is becoming more frequent and seasonal variability is increasing

The IASI/FTIR comparisons

- Indicates that the TAO FTIR measurements are representative of NH_3 at a city-size scale (~50 km)
  - Similar results found by Tournadre et al. (2020), who found that the FTIR in Paris was capable of providing information about NH_3 variability at a "regional" scale (~ 120 km)
- The highest correlation (R = 0.73) was seen with coincidence criteria of 25 km and 20 minutes

Comparison with GEOS-Chem

- Poor correlation with TAO FTIR for a single grid cell comparison, with R = 0.51 (R^2 = 0.26)
- A more regional scale comparison with IASI (35 N to 53 N, and 93.75 W to 63.75 W) resulted in better correlation R = 0.57 (R^2 = 0.33)

Summary and Discussion

The 18 years of data collected at TAO were analyzed to address three scientific objectives

1. Trends were observed
    - Notable trends: positive trends of CH_4, C_2H_6 (which reversed sign in 2009), N_2O, NH_3, and negative trends of CO
    - Trend used for preliminary assessment of the effects of the COVID-19 Lockdown on air quality in Toronto
2. Three biomass burning events were examined, and emission factors and ratios estimated
    - CH_3OH and HCOOH enhancements also observed, although emissions estimates have biases and uncertainties
3. Observational representativeness of TAO FTIR NH_3 total column estimated to be a city-size scale
    - Similar results found by FTIR in Paris
    - GEOS-Chem CTM at 2 x 2.5 resolution is too coarse for NH_3 comparison with TAO

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Future Work

- Monitoring the atmosphere is a continuous effort
- 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, and a new sun tracker has been installed
- More thoroughly examine the column data from 2020 for the effects on air quality of the lockdown due to the COVID-19 pandemic
- The methodology employed in the biomass burning project can be done on the enhancement event seen in September 2020
- Run GEOS-Chem tagged CO with more specific tags (e.g., CO from North American anthropogenic sources can be divided up into Canada, and Western, Central, and Eastern US)
- Compare nested (higher resolution) GEOS-Chem simulations to TAO NH_3