Tools for independent verification of combustion CO_2

Paul Palmer, Mark Lunt, Mehliyar Sadiq
Samuel Hammer, Cornelia Jäschke, Fabian Maier, Claudius Rosendahl
Stijn Dellaert, Hugo Denier Van Der Gon, Ingrid Super
Gregoire Broquet

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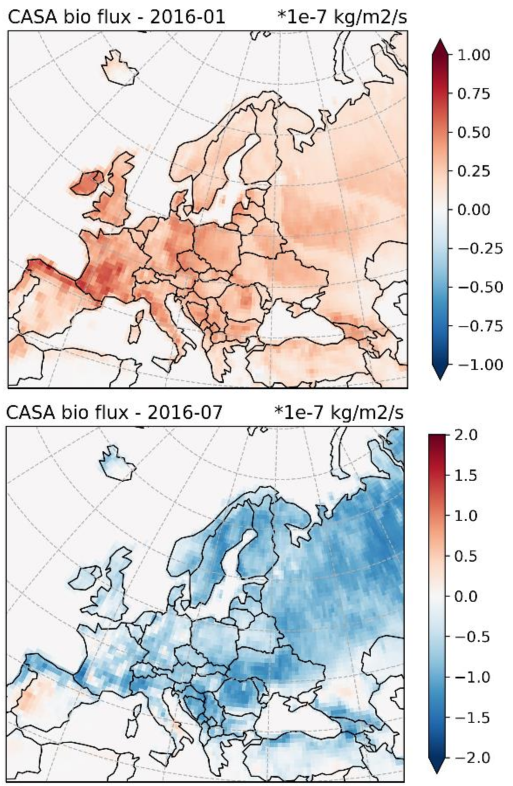
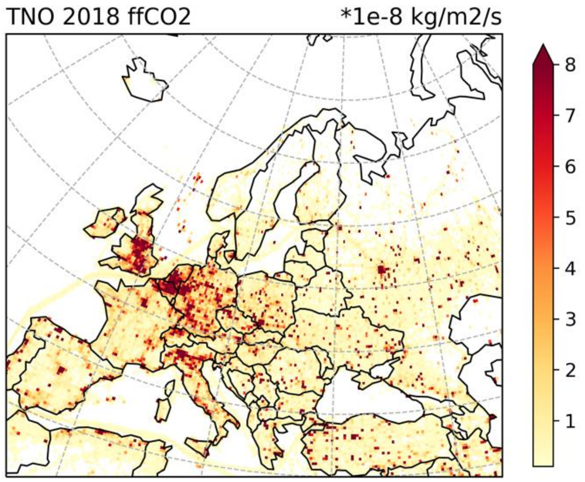
TNO innovation for life
LSCE
VERIFY

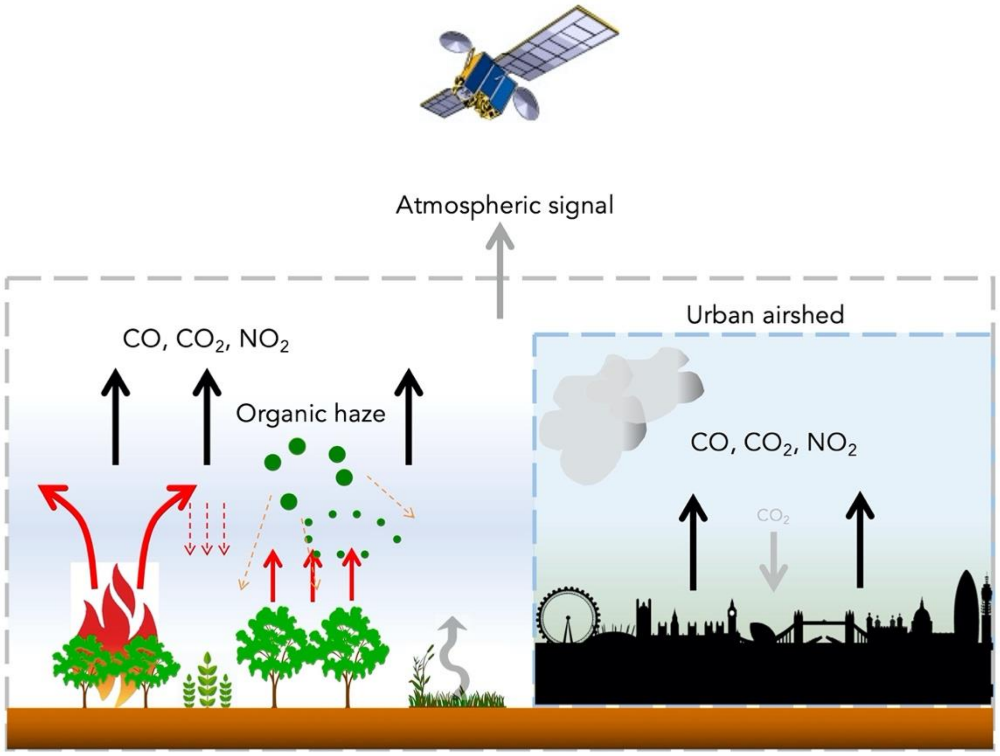
"The crux of the challenge faced by the science and policy communities: effective mitigation of fossil fuel CO_2 emissions within a large-scale, dynamic natural carbon cycle that we do not quantitatively understand."

Bottom-up inventories reveal the mosaic of land use and competing CO_2 fluxes





Top-down estimates rely on an integrated atmospheric signal from the mosaic of emissions

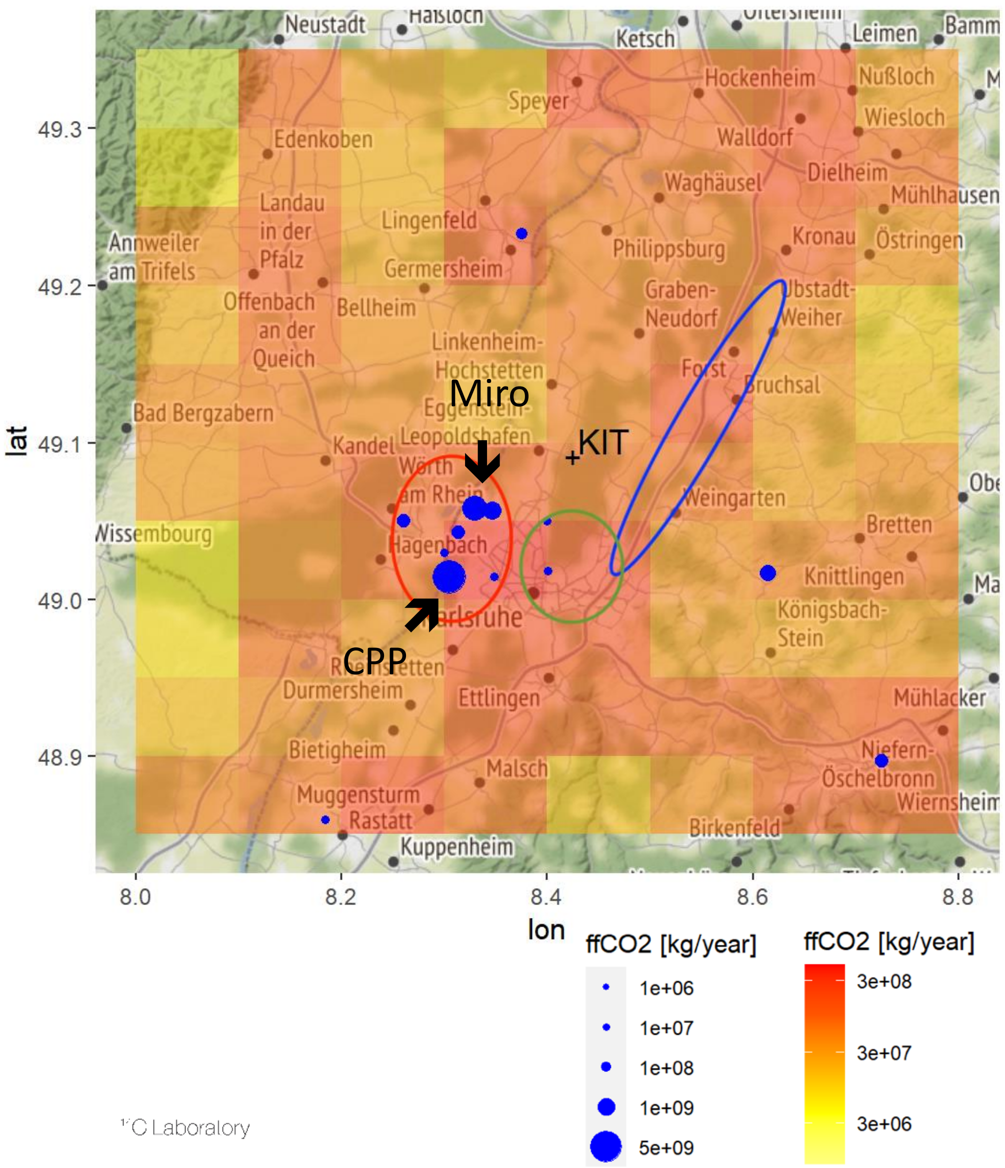


Here, we focus on picking apart the CO_2 combustion signal (fossil fuel and biofuel) using atmospheric gases co-emitted with CO_2

One approach to use directly $^{14}CO2$ to describe $ffCO_2$

Can we use $ffCO_2$:CO:NOx to identify individual sectors?

- ICOS/KIT in-situ measurements of CO_2, CO and NO_x at 200m.
- Flask sampling for ^{14}C-based fossil fuel CO_2 estimates
- Different emission sectors, e.g. from industry, traffic and residential heating
- Strong, point sources around KIT, especially:
- Coal Power Plant Rheinhafen (CPP)
- Miro Refinery



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- Strong…
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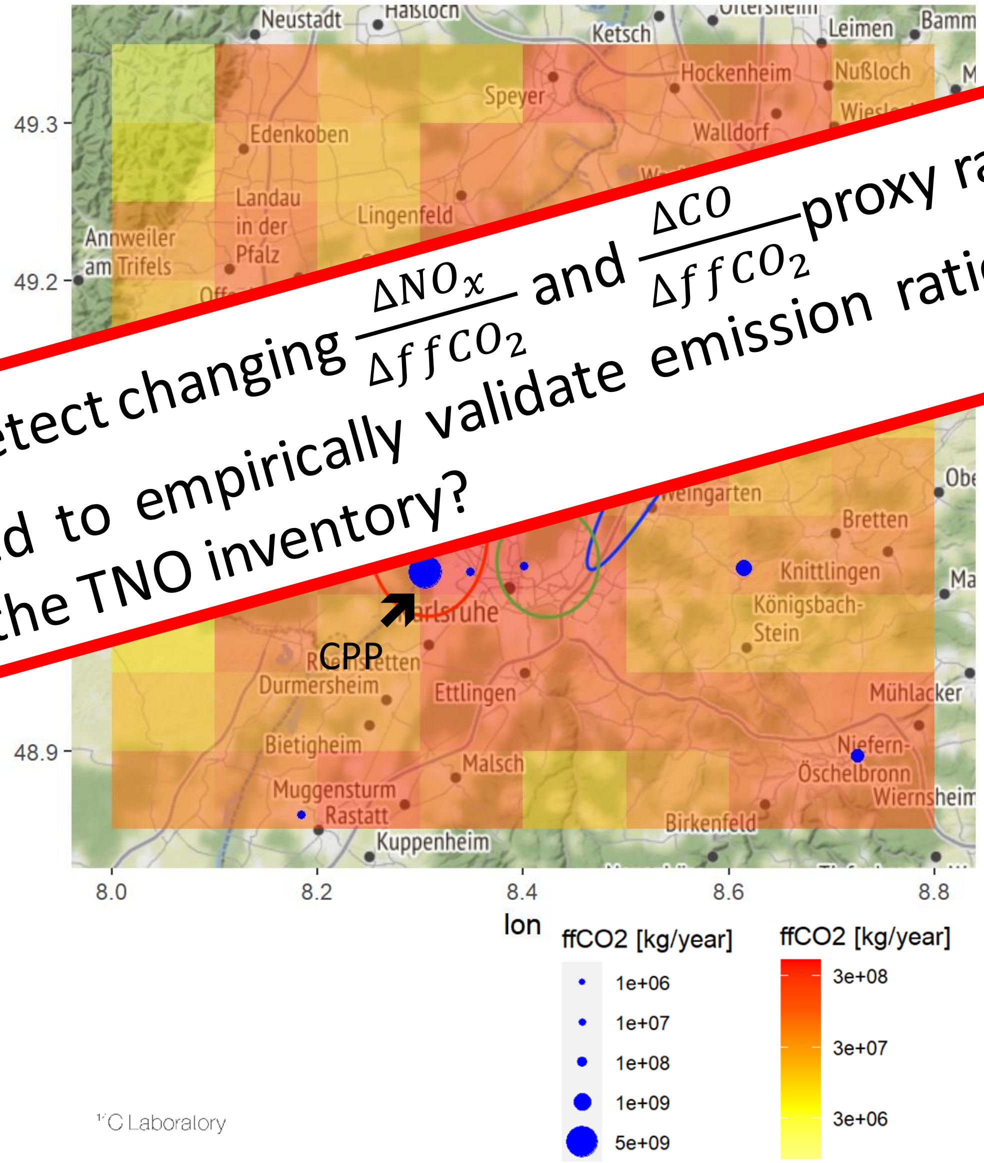
⇒ Can atmospheric observations detect changing $\frac{\Delta NO_x}{\Delta ffCO_2}$ and $\frac{\Delta CO}{\Delta ffCO_2}$ proxy ratios?

⇒ Can these observations be used to empirically validate emission ratios from individual source sectors from the TNO inventory?



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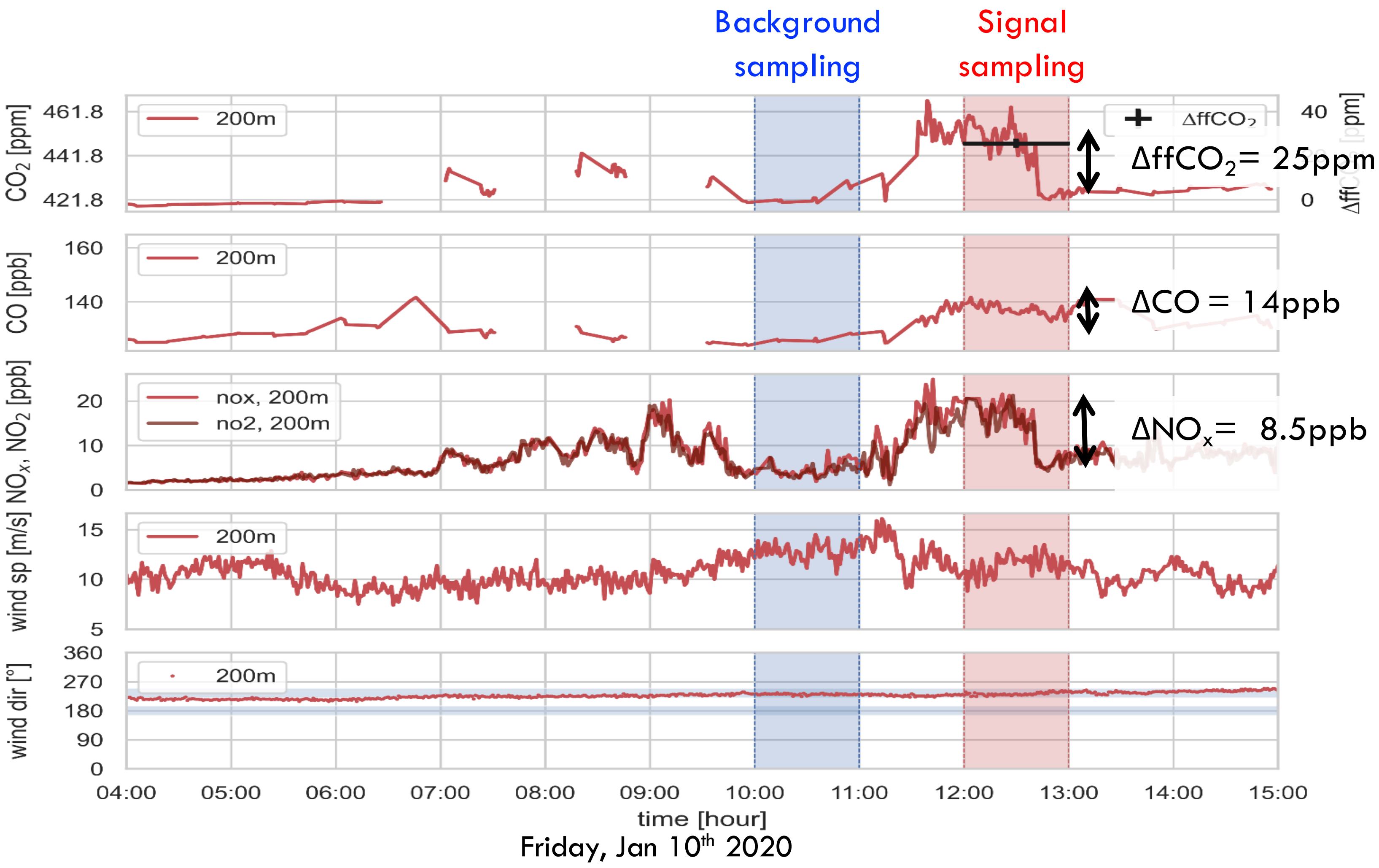
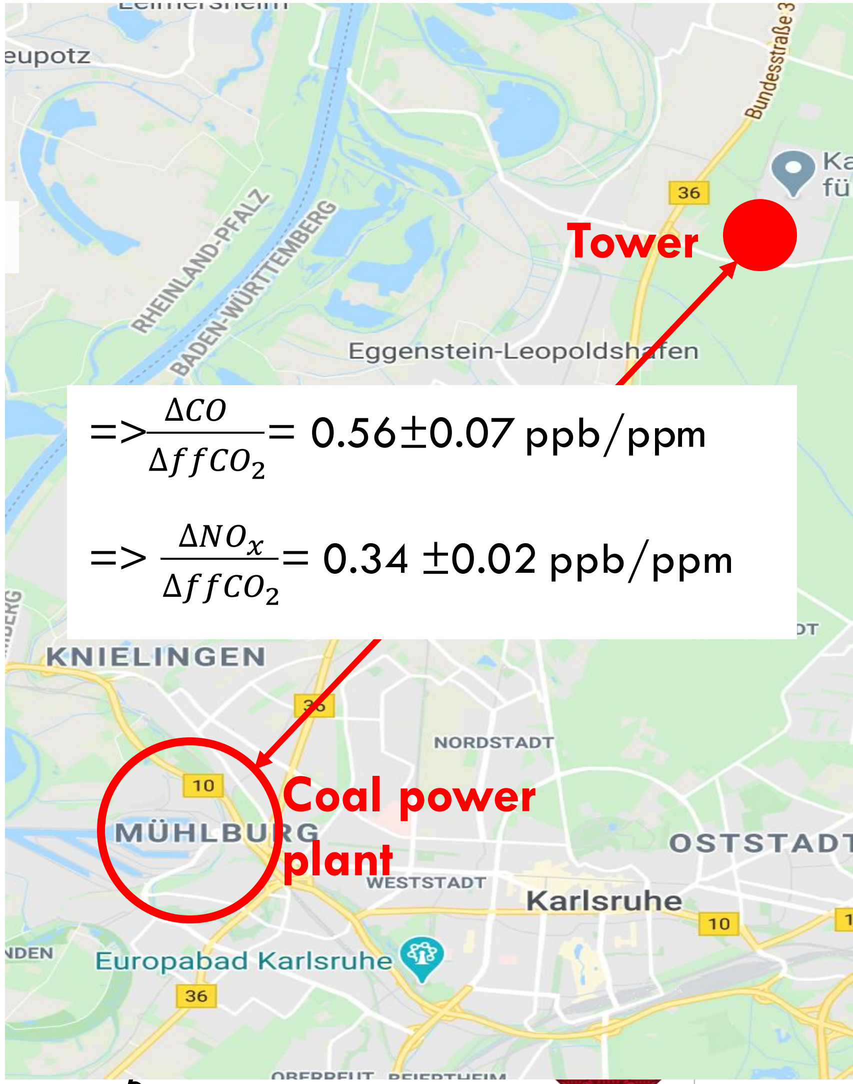
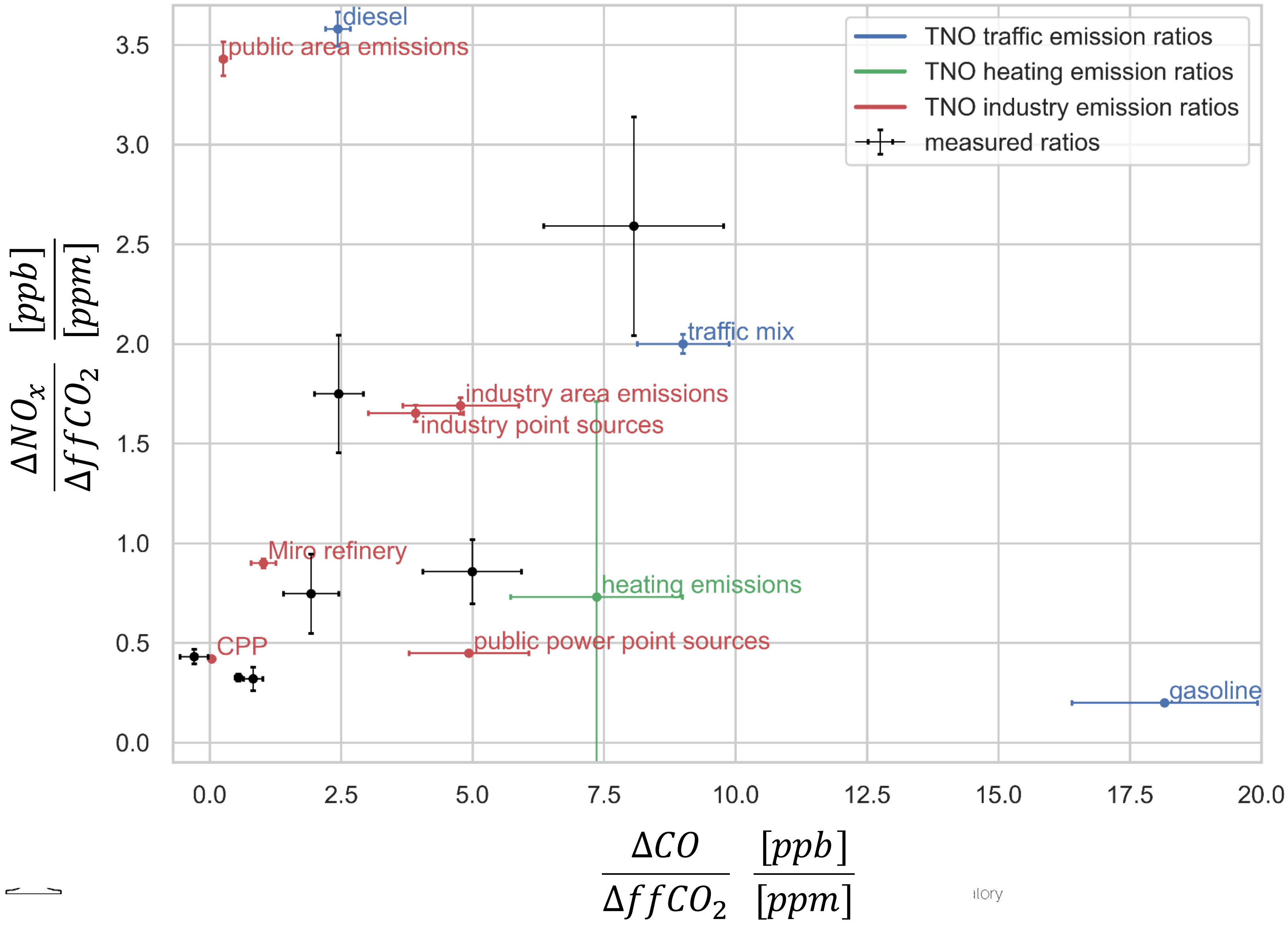
How are atmospheric proxy ratios derived experimentally?

- Example of measurements dominated by CPP:





Comparison of TNO bottom-up and top-down proxy ratios



$$\frac{\Delta NO_x}{\Delta ffCO_2} \text{ vs. } \frac{\Delta CO}{\Delta ffCO_2}$$

- Ratios from TNO **traffic** emissions
- Ratios from emissions of **heating** sector.
- Ratios of different **industry** emissions (Power and industry)
- Ratios from atmopheric data

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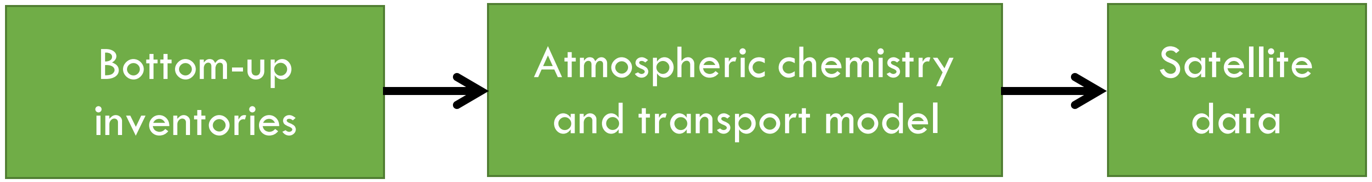
Are bottom-up inventories consistent with satellite observations?



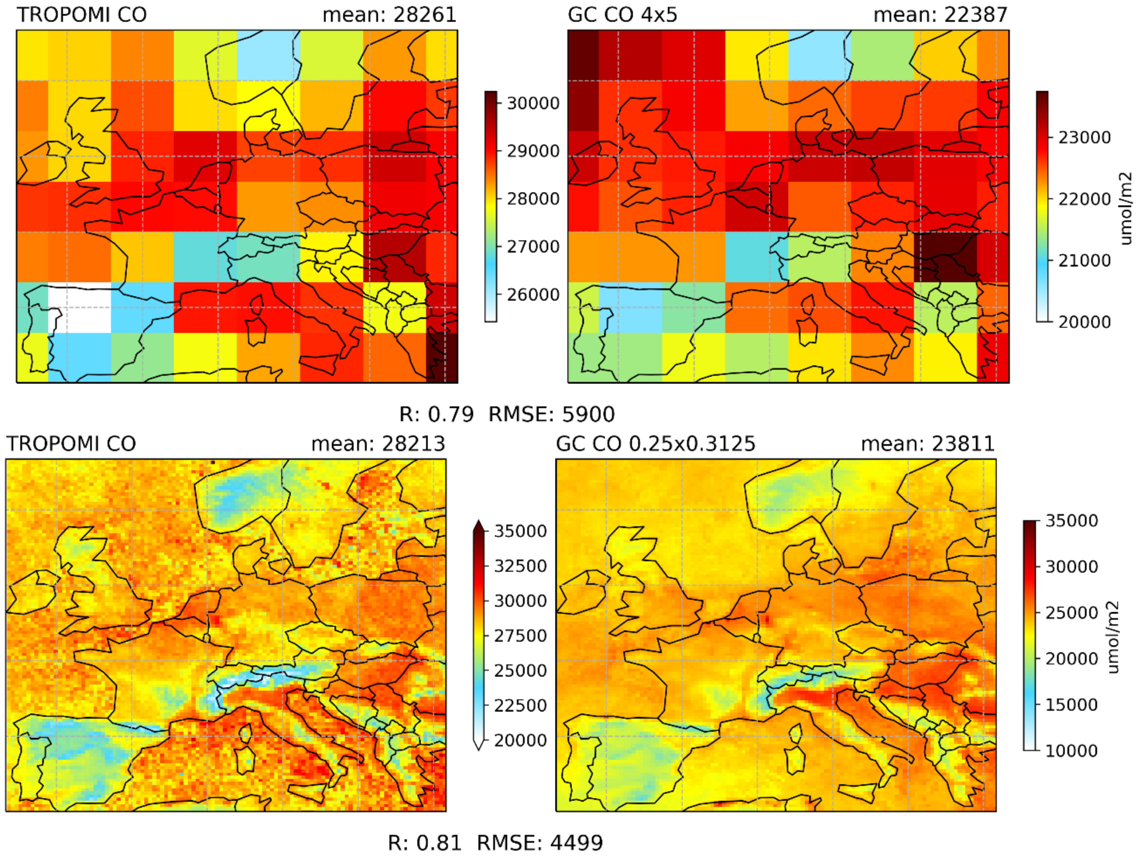
Example: Model and TROPOMI CO columns, July 2018

Global-scale model
Monthly mean
@4°x5°

Nested version of model
Monthly mean
@0.25°x0.3125°

TROPOMI

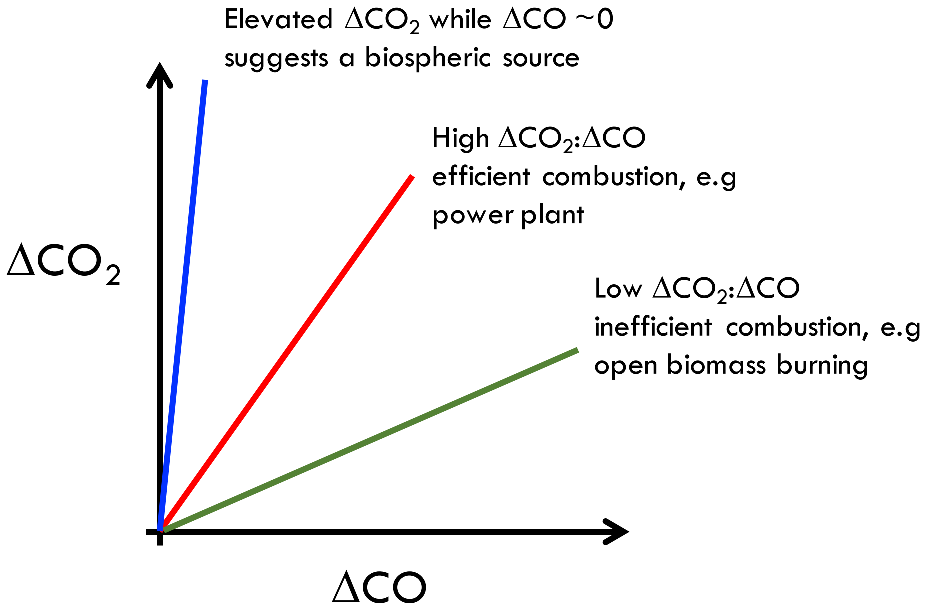
GEOS-Chem (inc TNO emissions)



- Global run underestimates CO columns, but able to capture the spatial patterns observed by TROPOMI.
- Nested model run with TNO inventory corrects underestimation in high CO values, but still underestimate low values.

Using CO_2:CO emission uncertainty correlations to constrain combustion CO_2

Basic idea: use CO to constrain combustion CO_2



CO_2:CO correlations (and their uncertainties) due to the combustion process & atmospheric transport

Here, we focus on the combustion process

Mass of fuel consumed/country/yr

Mass of gas emitted/ (mass of fuel consumed/country/yr)

Emissions = Activity Data x Emission Factor

Common to CO_2 and CO

Anticorrelated between CO and CO_2 via combustion efficiency

$$Emis_i = (AD + \sigma_{AD}) \times (EF + \sigma_{EF})$$

[We use **national-scale values** of AD, σ_{AD}, EF and σ_{EF}]

Monte Carlo approach to estimate error correlations:

- Run thousands of samples to generate emissions uncertainty for each sector for CO and CO_2
- Sum over all emission sectors for CO and CO_2
- Correlate resulting CO_2, CO ensemble

National E_{CO2}:E_{CO} error correlations

Country	E_{CO2}:E_{CO} Pearson correlation coefficient
Belgium	-0.10
France	-0.42
Germany	-0.23
Ireland	-0.62
Italy	-0.42
Lithuania	+0.15
Netherlands	-0.25
Poland	-0.42
Portugal	-0.42
Romania	-0.92
Spain	-0.20
Sweden	-0.22
UK	-0.50

$$Emis_i = (AD + \sigma_{AD}) \times (EF + \sigma_{EF})$$

Strength of correlations depend on the relative weight of σ_{AD} and σ_{EF} .

Limits:

σ_{AD}= 0 then r = -1

σ_{EF}= 0 then r = +1

National scale numbers:
σ_{AD} ~ 1-5%
CO σ_{EF} ~ 20-50%
CO_2 σ_{EF} ~ 1-3%
Balance of uncertainties will be different on smaller scales

Variations in E_{CO2}:E_{CO} error correlations reflect:

- Relative importance of individual sectors to total emissions
- Knowledge about AD and EF and their uncertainties

Closing remarks

- Estimating $ffCO_2$ from atmospheric data is possible
  - Non-trivial methods being developed require careful data analysis
  - Requires closer coordination between inventory, measurement and modelling groups
- What can NGHGI contribute to this science?
  - Spatially and temporally resolved estimates would help greatly, with traceability from emission factors and activity rates and their uncertainties
  - More measurements are always welcome. What, where and how many are open questions.
  - Linking GHGs and air quality gases (e.g. CO, NO_2) represents a promising approach to isolate $ffCO_2$.

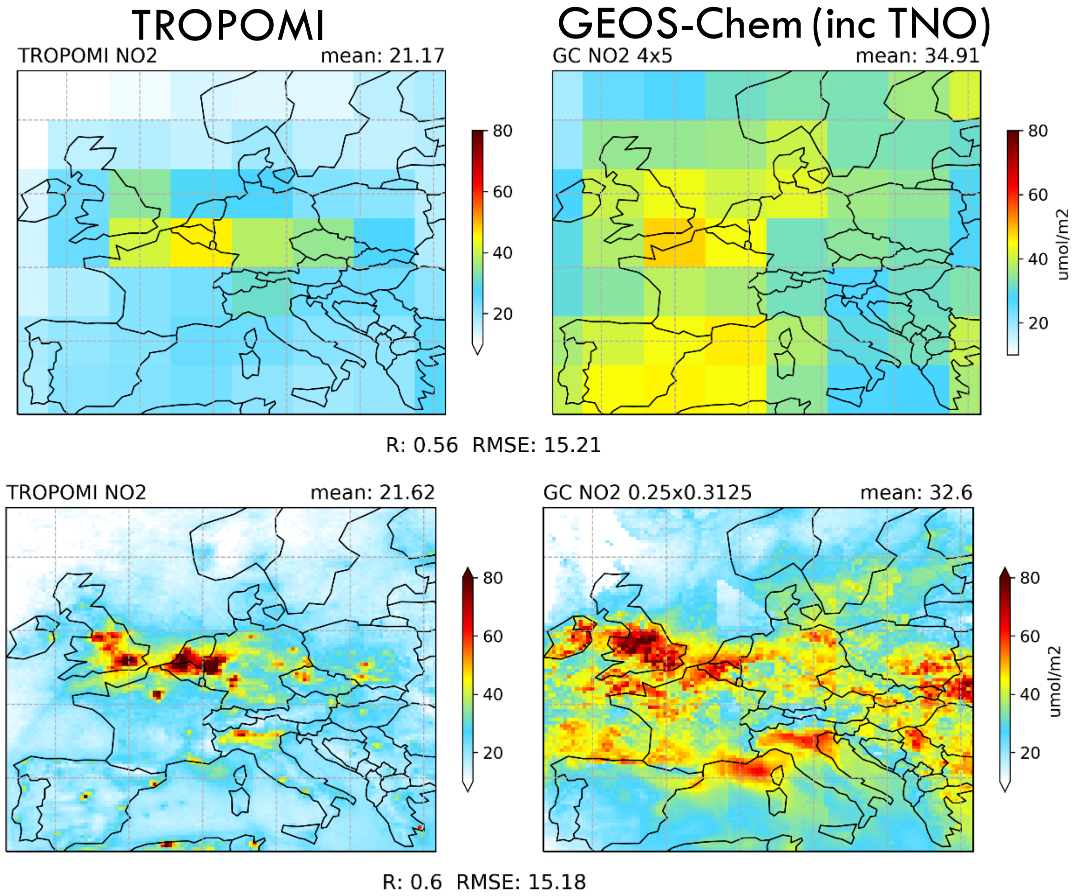
SPARE

Are bottom-up inventories consistent with satellite observations?

Example: Model and TROPOMI NO_2 columns July 2018

Monthly mean @4°x5°

Monthly mean @0.25°x0.3125°



- GEOS-Chem overestimates column NO_2 density over EU, in both global run and nested run.
- Nested run with TNO inventory captures details over UK, France, Belgium and Italy, however, a significant hotspot is simulated over Ukraine, likely due to overestimation in global background.