Atmospheric proxy/ffCO$_2$ ratios - potentials and limitations for regional ffCO$_2$ estimation -

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VERIFY General Assembly meeting, 29.04.2021

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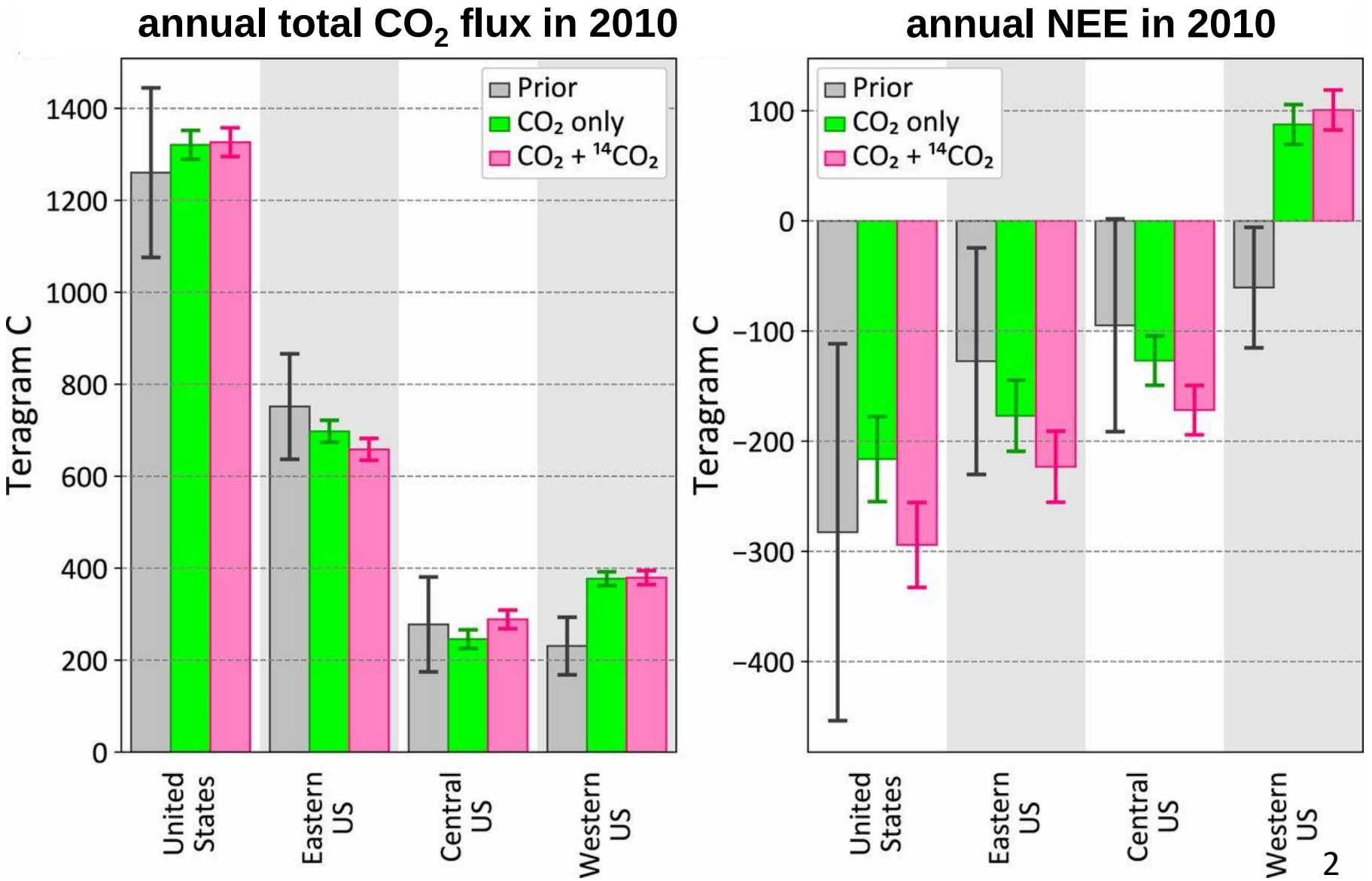
ICOS Integrated Carbon Observation System

Advantages of additional $ffCO_2$ estimates for inverse modelling



Sourish Basu et al.
PNAS 2020;
117:24:13300-13307

PNAS

Observation-based $ffCO_2$ estimation

$^{14}CO_2$

- most direct $ffCO_2$ tracer
- nuclear & other corr. ~10%

- not continuous (yet)
- “only” in situ

Proxies (CO, NO_x,…)

- continuous
- remote sensing

- non-ff sources
- chemically reactive
- process-dependent emission ratios

Aims of proxy/ffCO$_2$ research in VERIFY Task 2.2

1

Determine atmospheric proxy/ffCO$_2$ ratios for major emission sectors

Cross-check inventory emission ratios

2

Diurnal and seasonal variability of atmo. proxy/ffCO$_2$ ratios

Cross-check inventory time profiles

3

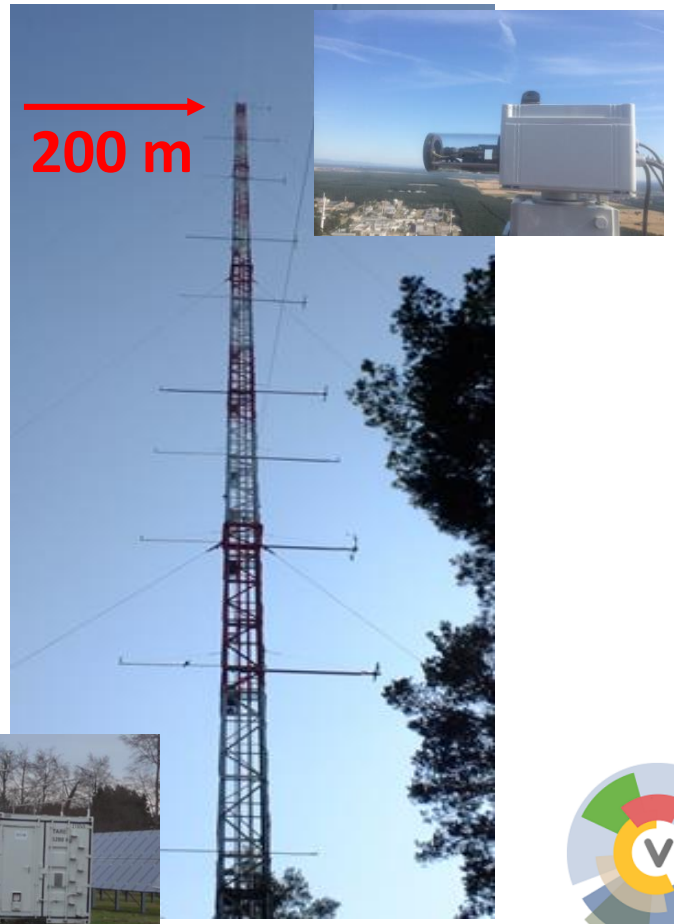
Compare proxy/(ff)CO$_2$ ratios between in-situ and total column

Exploit in situ to total column link

Experimental approaches

ICOS & TCCON station KIT

- In situ and total column comparison
- Temporal variability



In situ: CO_2, CO, NO, NO_2, CH_4, N_2O, ^{222}Rn

Flask: $^{14}CO_2$, $^{13}CO_2$

TC: CO_2, CO, CH_4, NO_2

Mobile laboratories

- Source sector dominated emission ratios

In situ: CO_2, CO, NO, NO_2, CH_4, ^{222}Rn

Flask: $^{14}CO_2$, $^{13}CO_2$

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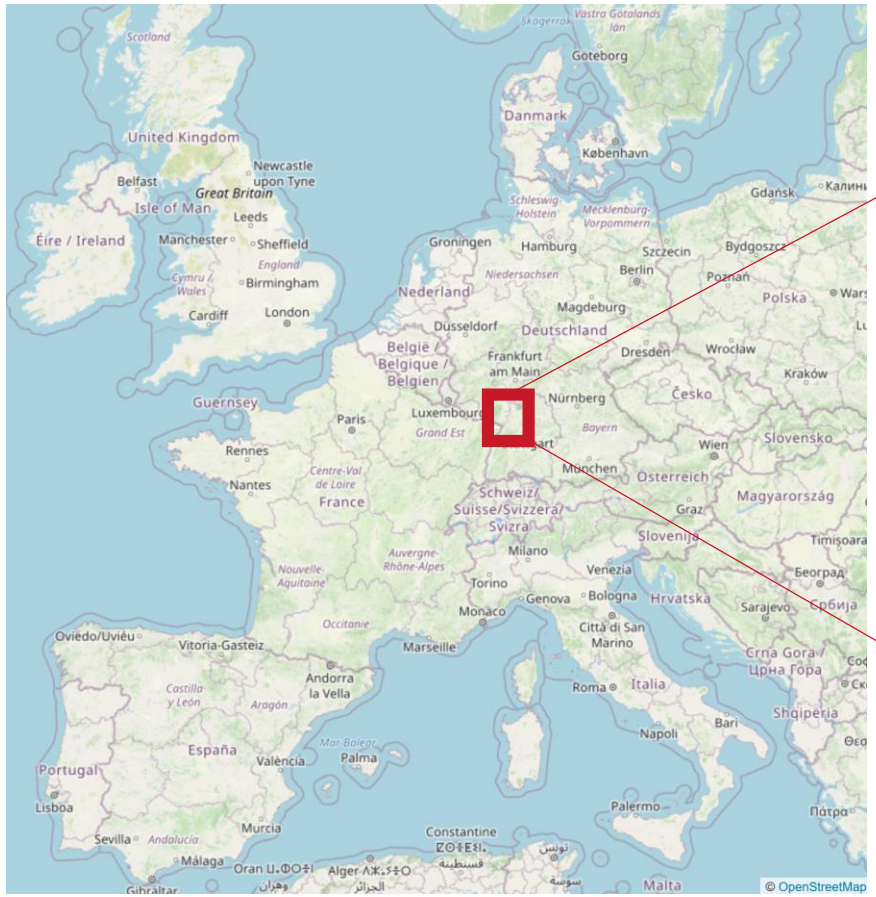
VERIFY case-study area



Map

TNO emission around KIT

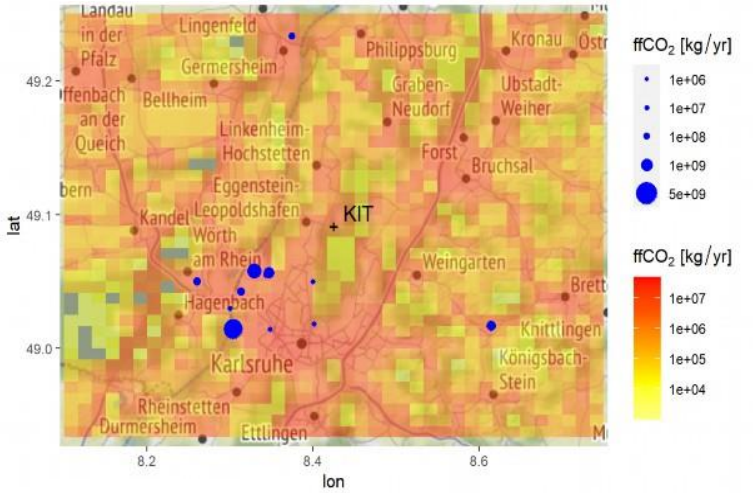
200 m
a. l. g.

KIT Tower

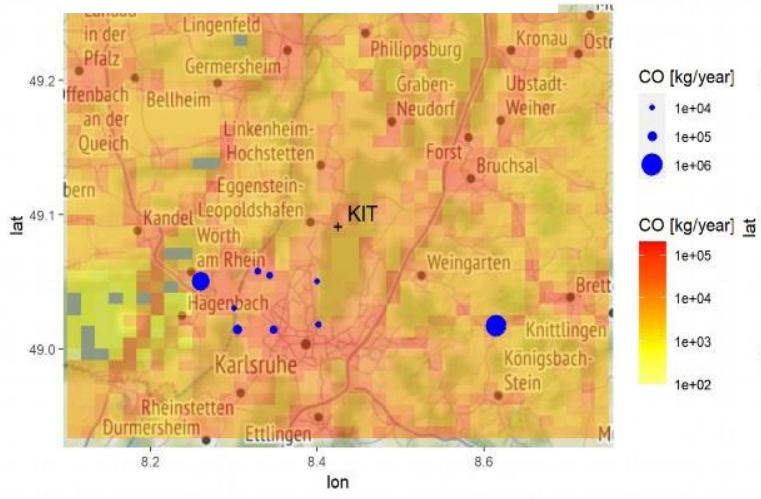
Karlsruher Institut für Technologie

Map

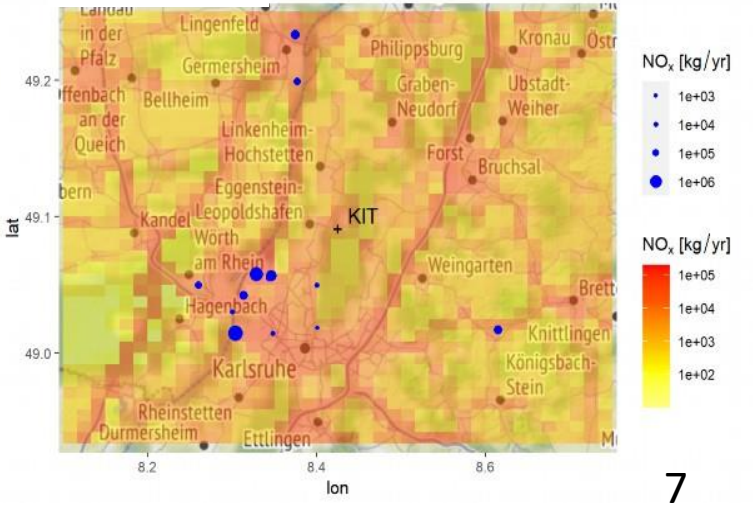
$ffCO_2$



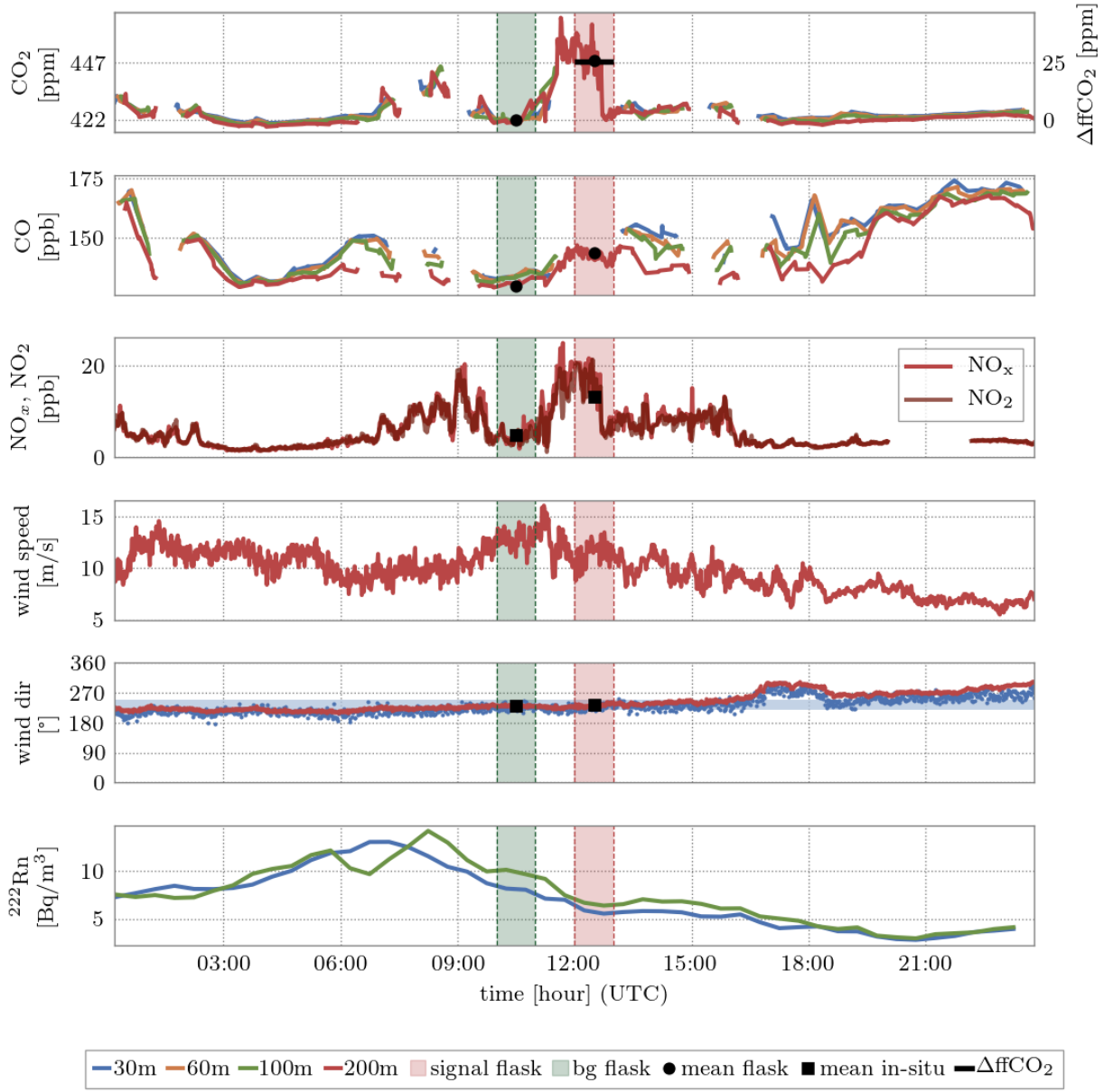
CO (ff and bio)



NO_x



In-situ measurement on January 10, 2020



ΔCO_2 = 26.1±0.1 ppm
$\Delta ffCO_2$= 25.9±1.1 ppm

ΔCO = 14.1±1.4 ppb

ΔNO_x= 8.5±0.1 ppb

$$\frac{\Delta CO}{\Delta ffCO_2} = \mathbf{0.54} \pm 0.06 \frac{ppb}{ppm}$$

$$\frac{\Delta NO_x}{\Delta ffCO_2} = \mathbf{0.33} \pm 0.01 \frac{ppb}{ppm}$$

Aims of proxy/ffCO$_2$ research in VERIFY

1

Determine atmospheric proxy/ffCO$_2$ ratios for major emission sectors

Cross-check inventory emission ratios

2

Diurnal and seasonal variability of atmo. proxy/ffCO$_2$ ratios

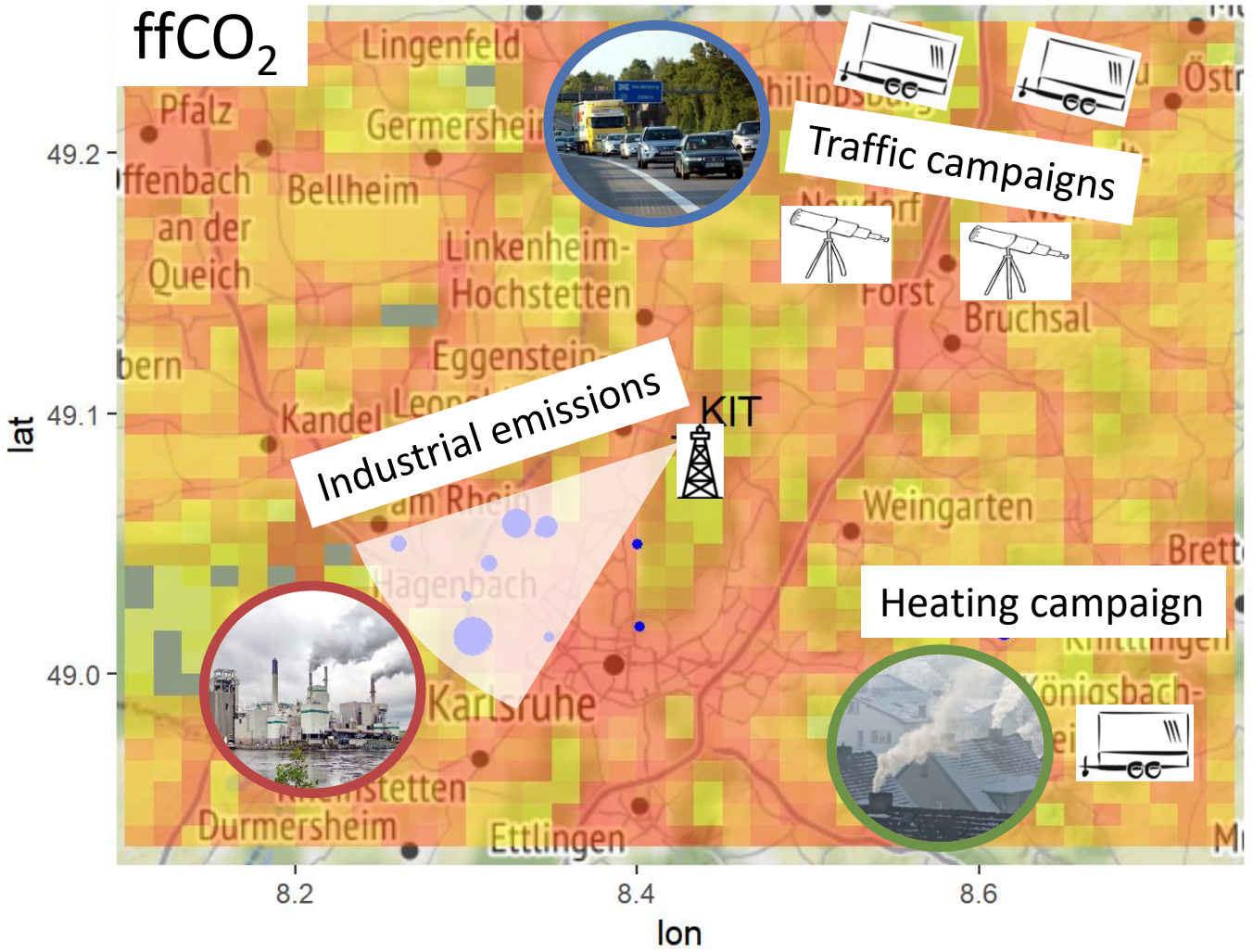
Cross-check inventory time profiles

3

Compare proxy/(ff)CO$_2$ ratios between in-situ and total column

Exploit in situ to TC link

Atmospheric proxy/ffCO$_2$ ratios (sector specific)



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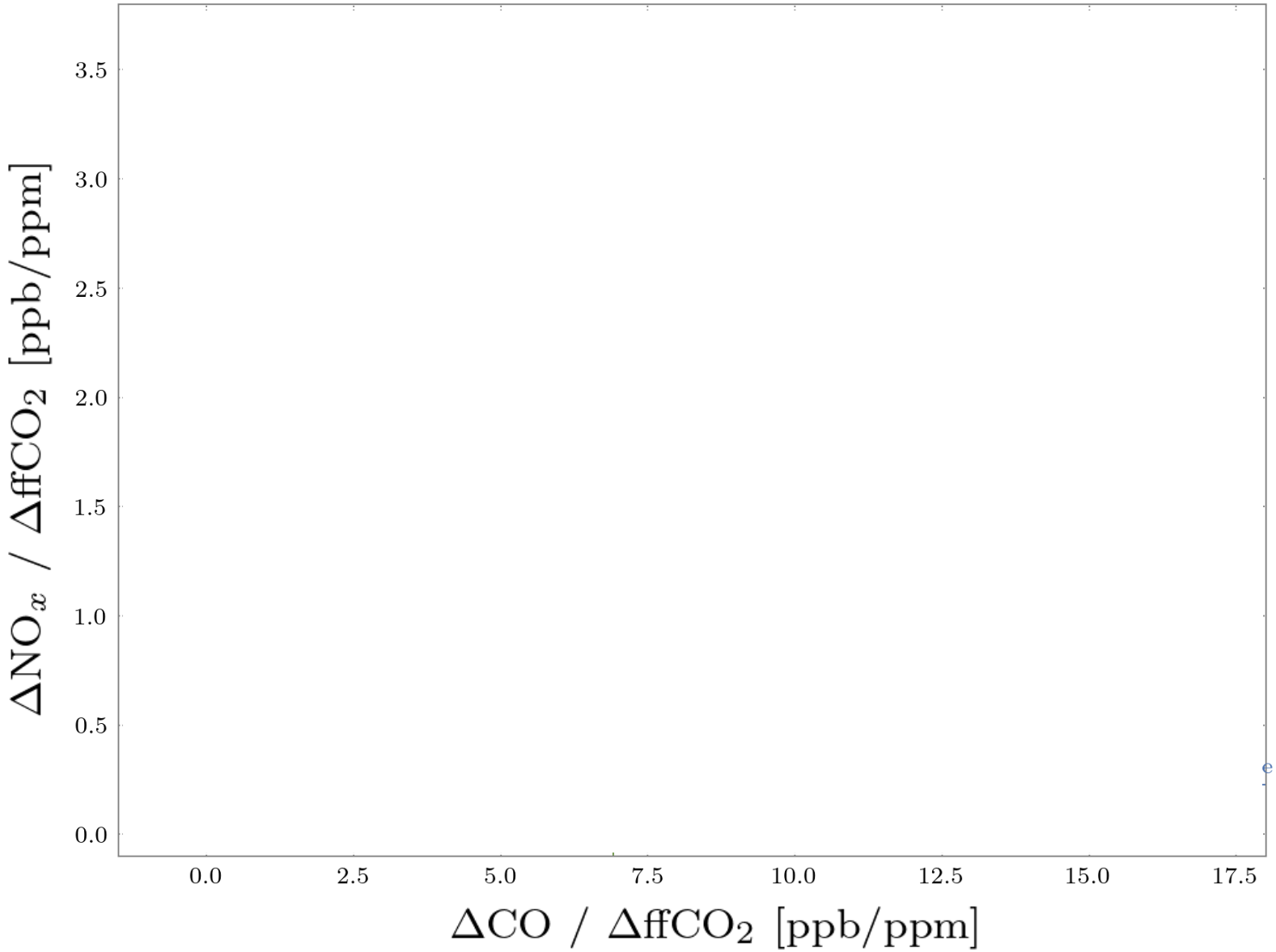
CO and NO_x double-ratio plot



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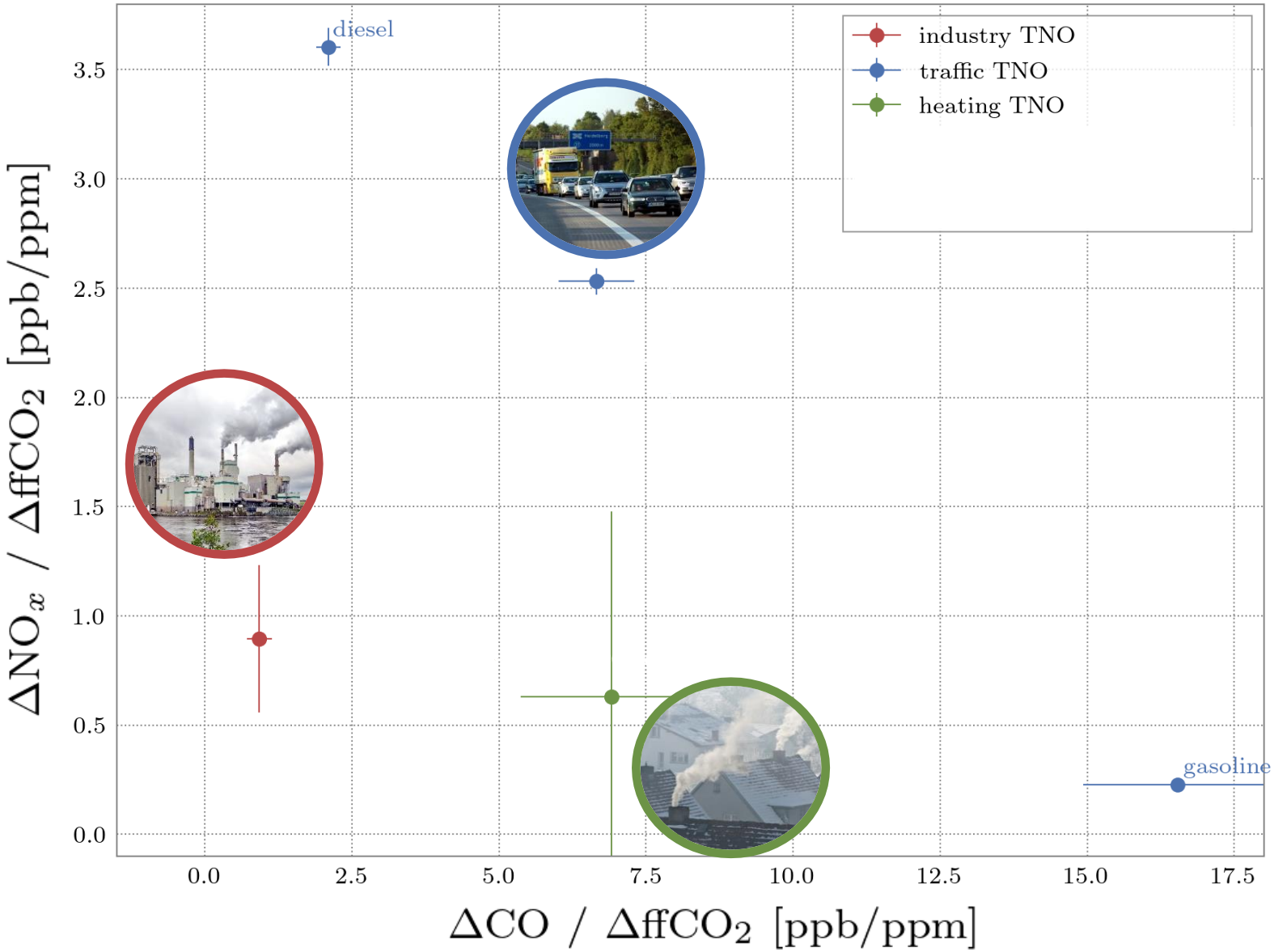
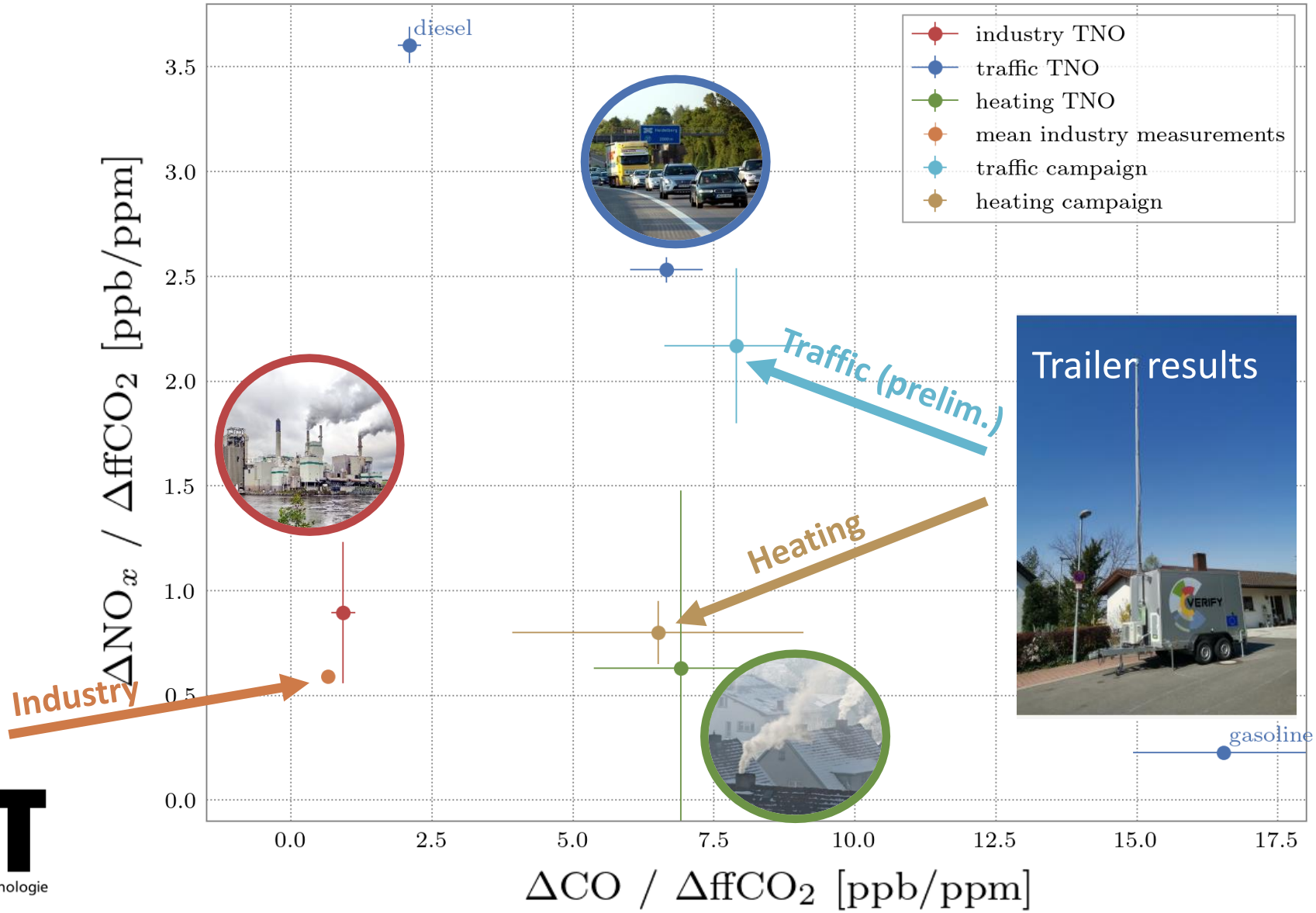


CO and NO_x double-ratio plot



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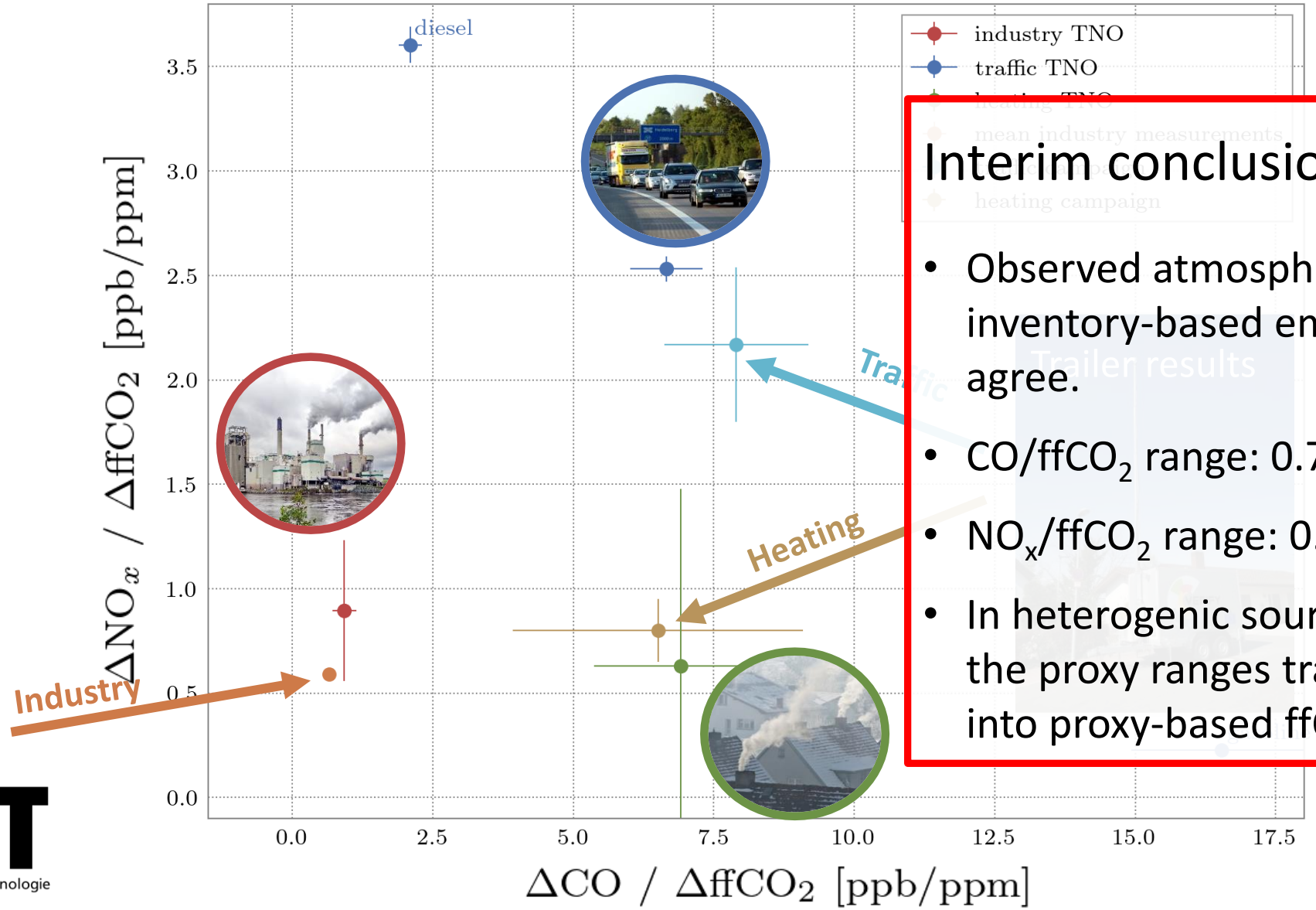
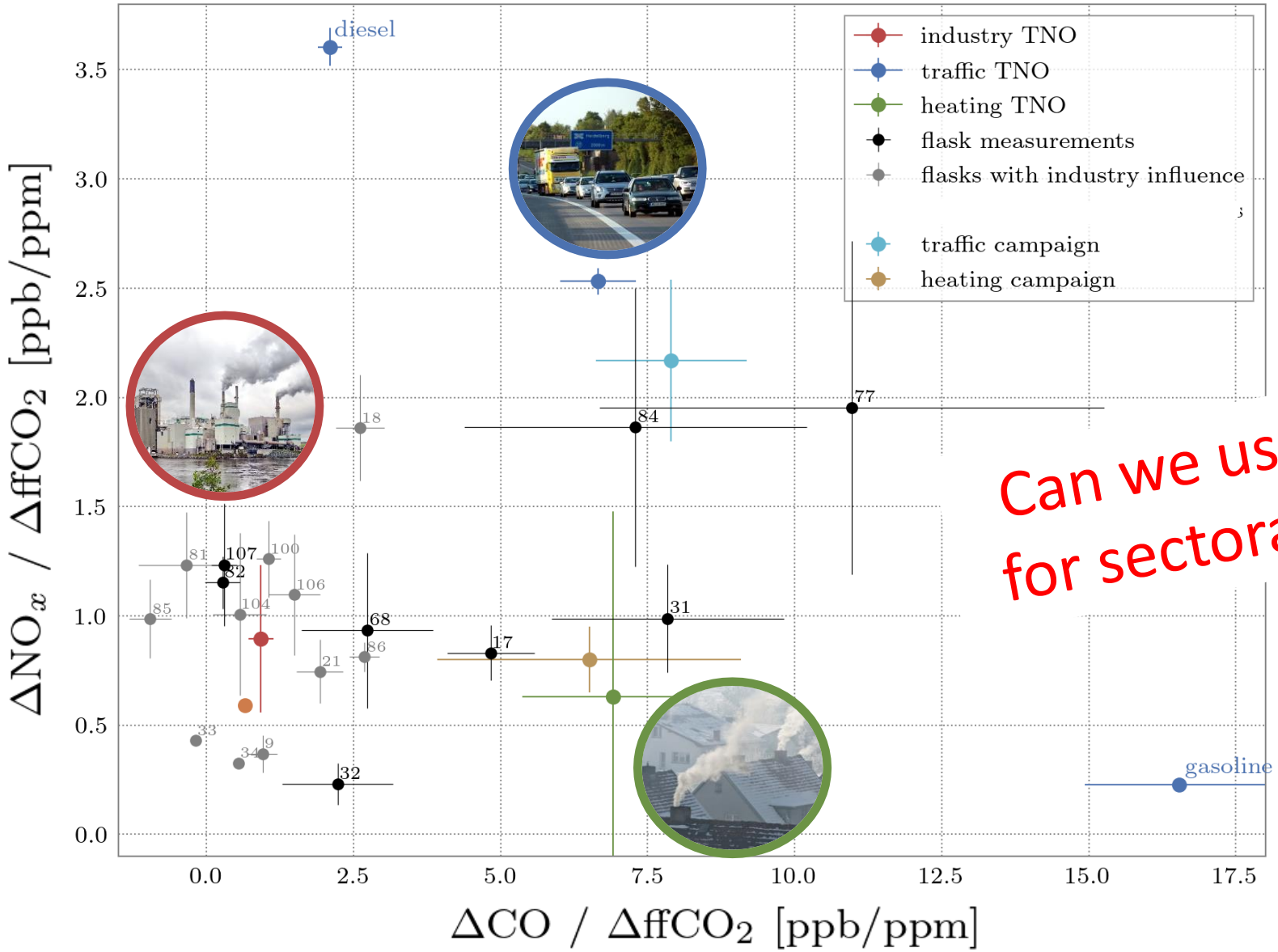
CO and NO_x double-ratio plot



Interim conclusions (1):

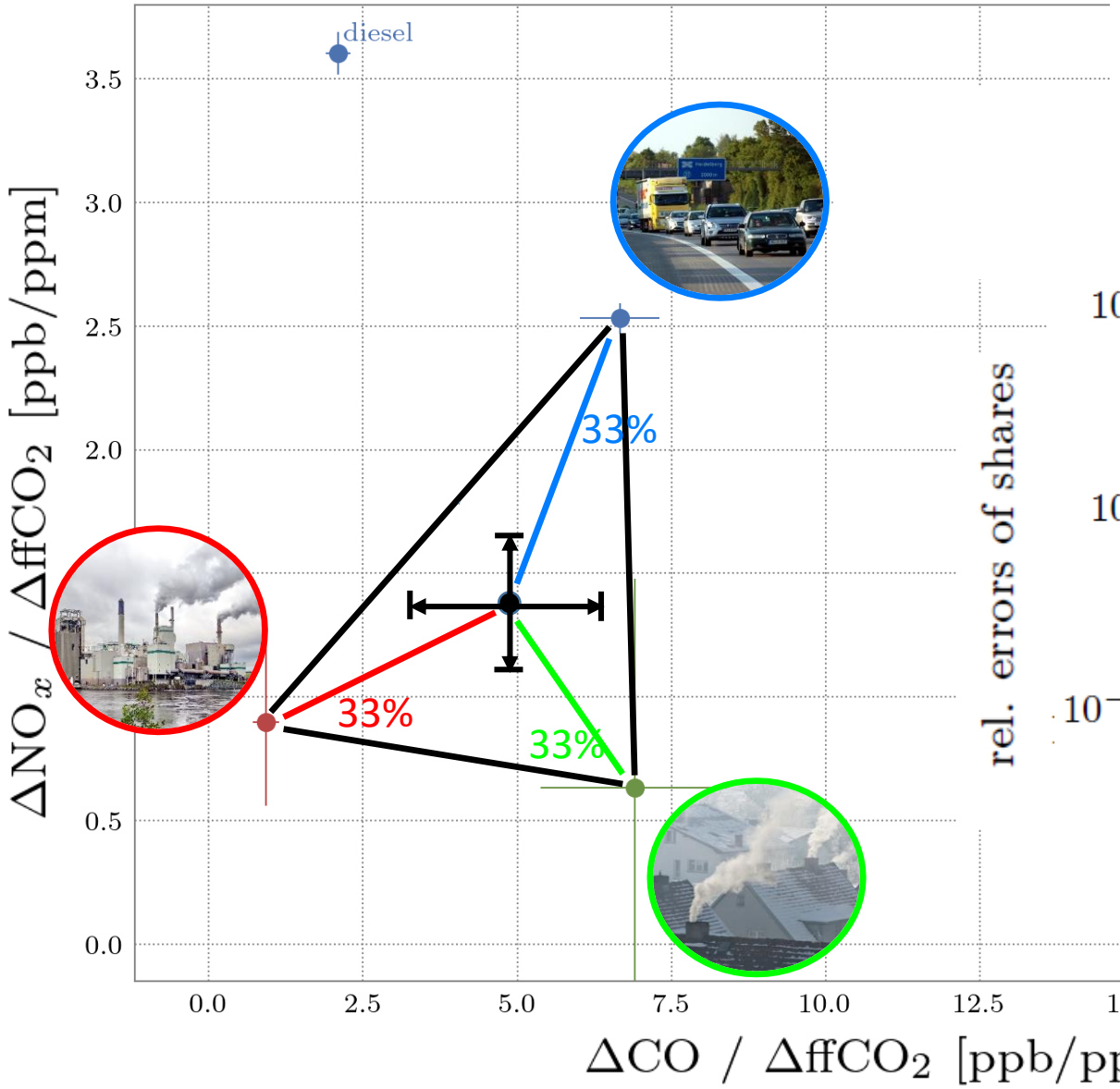
- Observed atmospheric ratios and inventory-based emission ratios do agree.
- $CO/ffCO_2$ range: 0.7 to 8 ppb/ppm
- $NO_x/ffCO_2$ range: 0.6 to 2.3 ppb/ppm
- In heterogenic source landscapes, the proxy ranges translate directly into proxy-based $ffCO_2$ uncertainties.

Turning a disadvantage into an advantage?

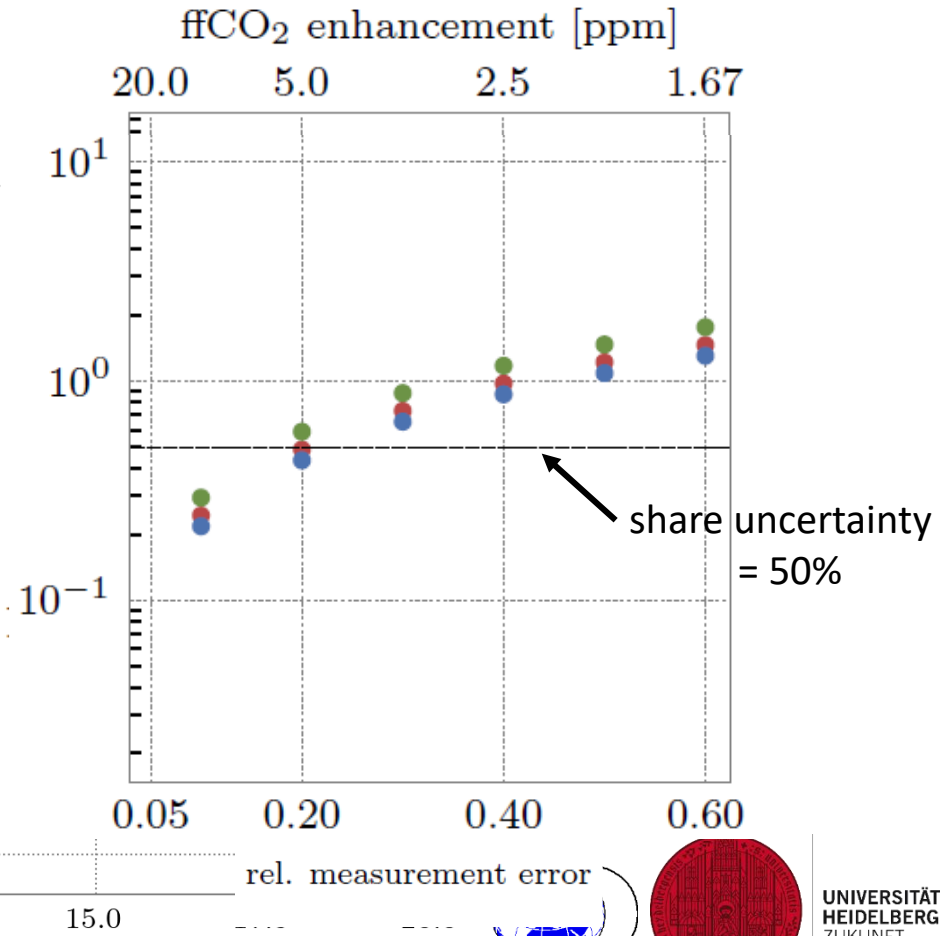


Can we use the proxies for sectoral attribution?

Theoretical potential of source separation?



Separation uncertainty wrt. measurement error?



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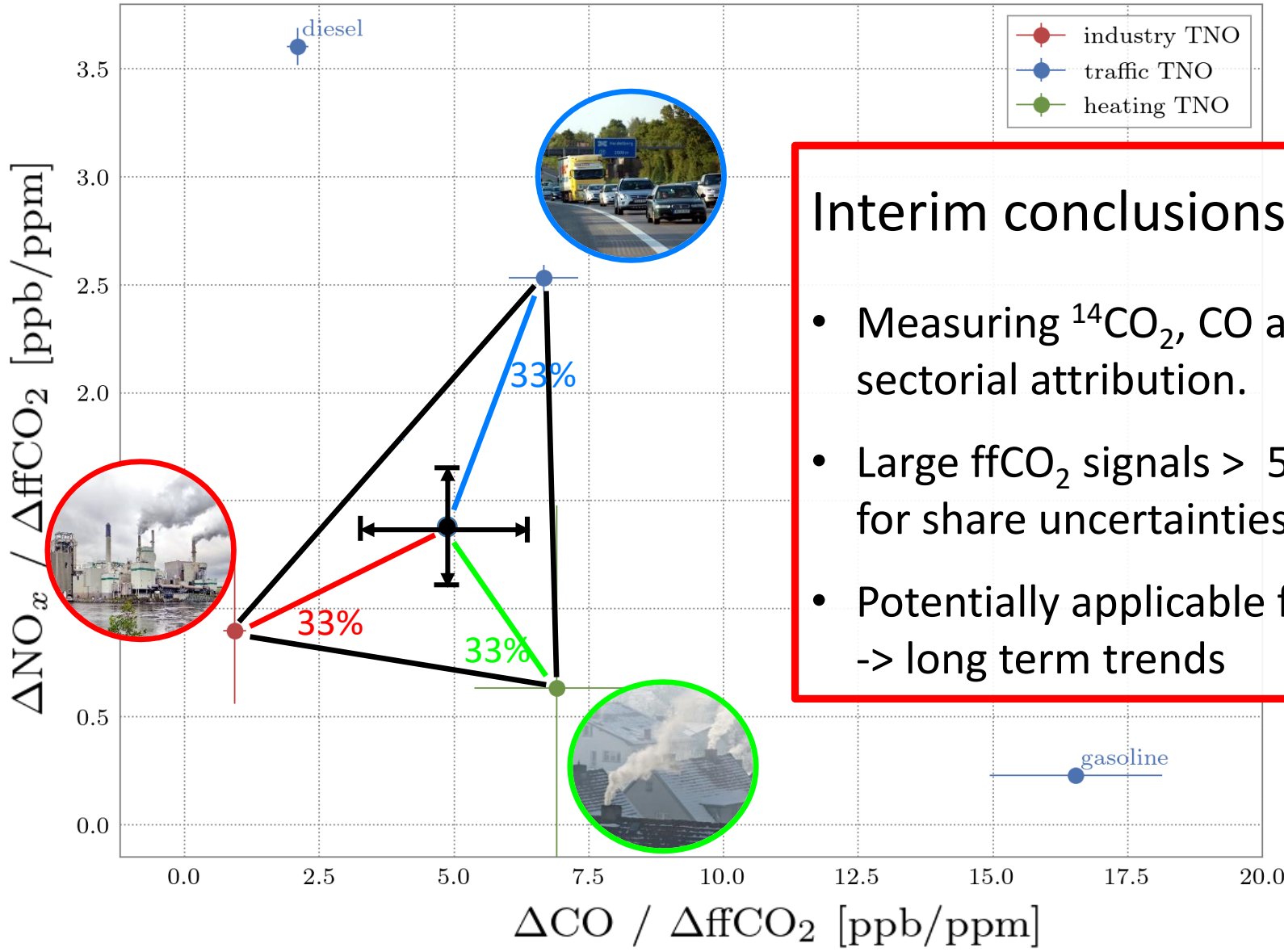
Theoretical potential of source separation?



Interim conclusions (2):

- Measuring $^{14}CO_2$, CO and NO_x allow for sectorial attribution.
- Large $ffCO_2$ signals > 5ppm are needed for share uncertainties below 50%
- Potentially applicable for urban studies -> long term trends

Aims of proxy/ffCO_2 research in VERIFY

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Determine atmospheric proxy/ffCO_2 ratios for major emission sectors

2

Diurnal and seasonal variability of atmo. proxy/ffCO_2 ratios

3

Compare proxy/(ff)CO_2 ratios between in-situ and total column

Cross-check inventory emission ratios

Cross-check inventory time profiles

Exploit in situ to total column link

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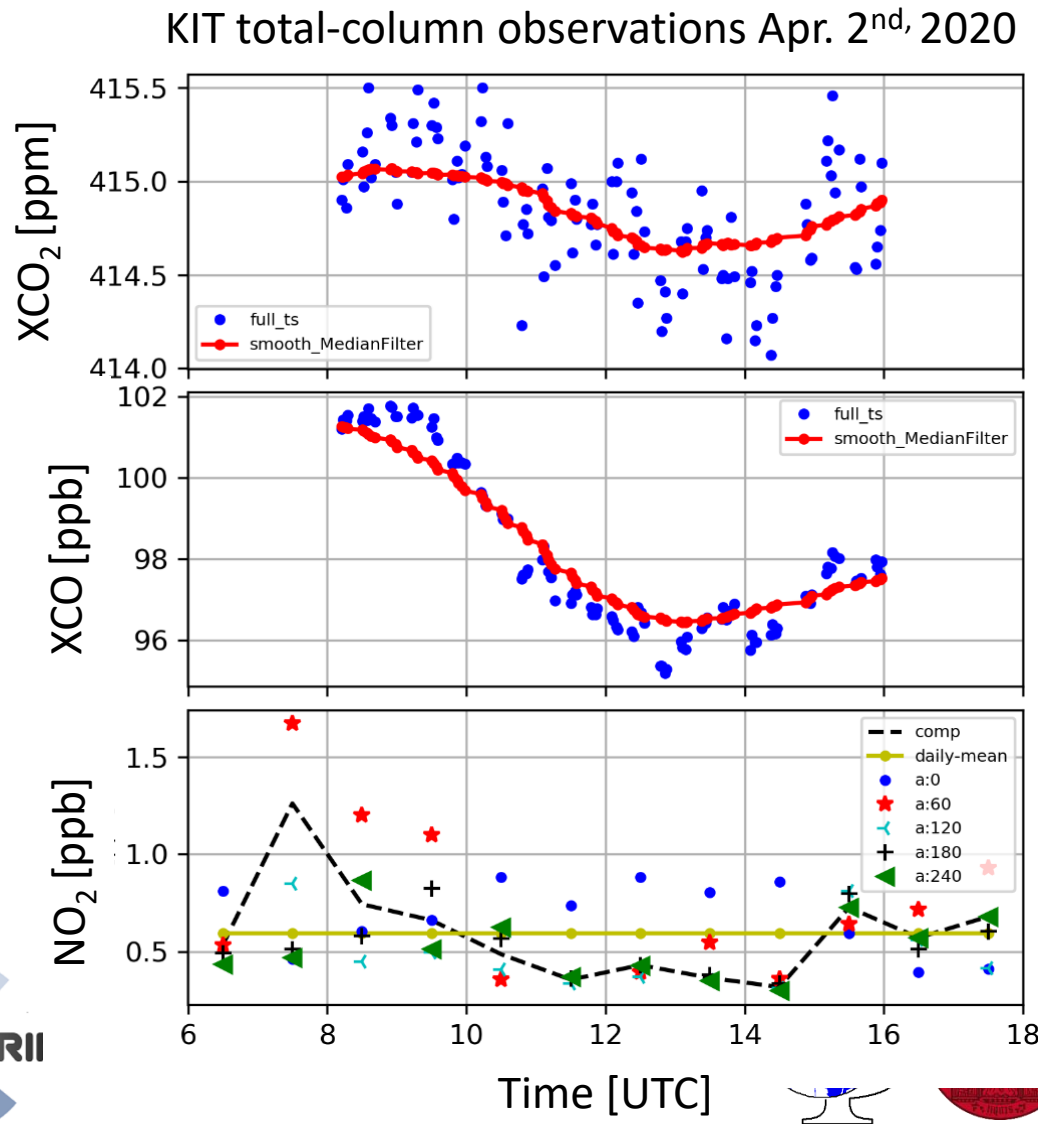
How to determine proxy ratios for total-column measurements?

KIT's working hypothesis:

- Changes in the total column background signal are long-phased.
- Local sources cause short-term changes.



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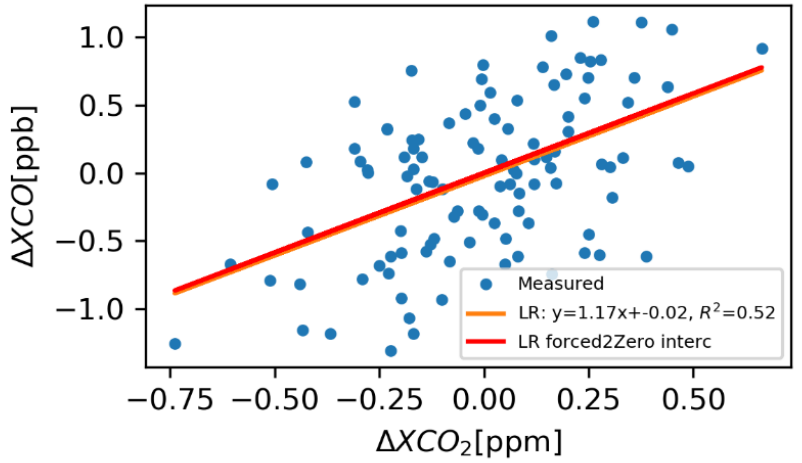
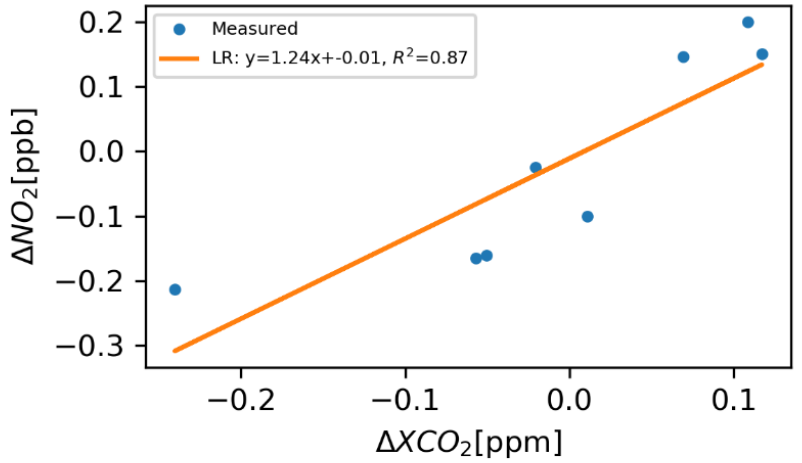
Total-column offset correlations Apr. 2nd, 2020



$$\frac{\Delta XCO}{\Delta XCO_2} = \mathbf{1.2}\,\frac{ppb}{ppm}$$



$$\frac{\Delta NO_2}{\Delta XCO_2} = \mathbf{1.2}\,\frac{ppb}{ppm}$$

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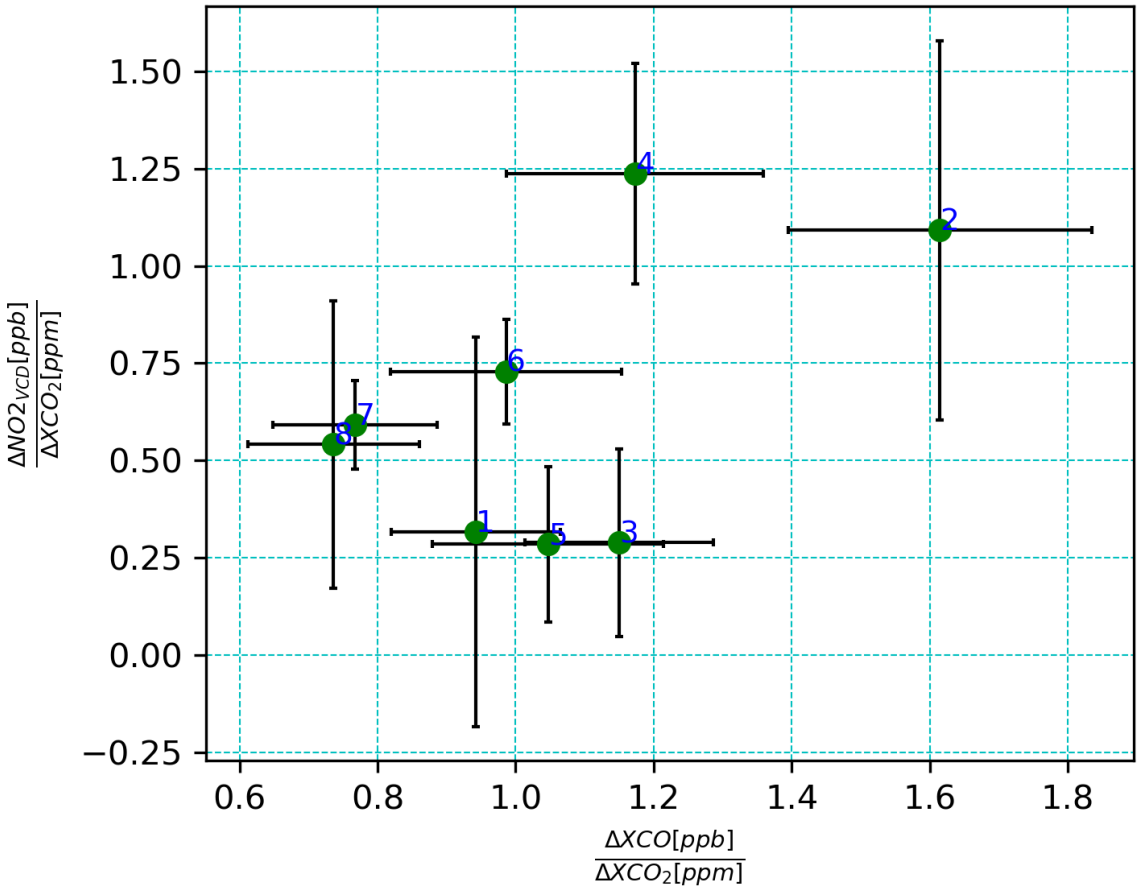
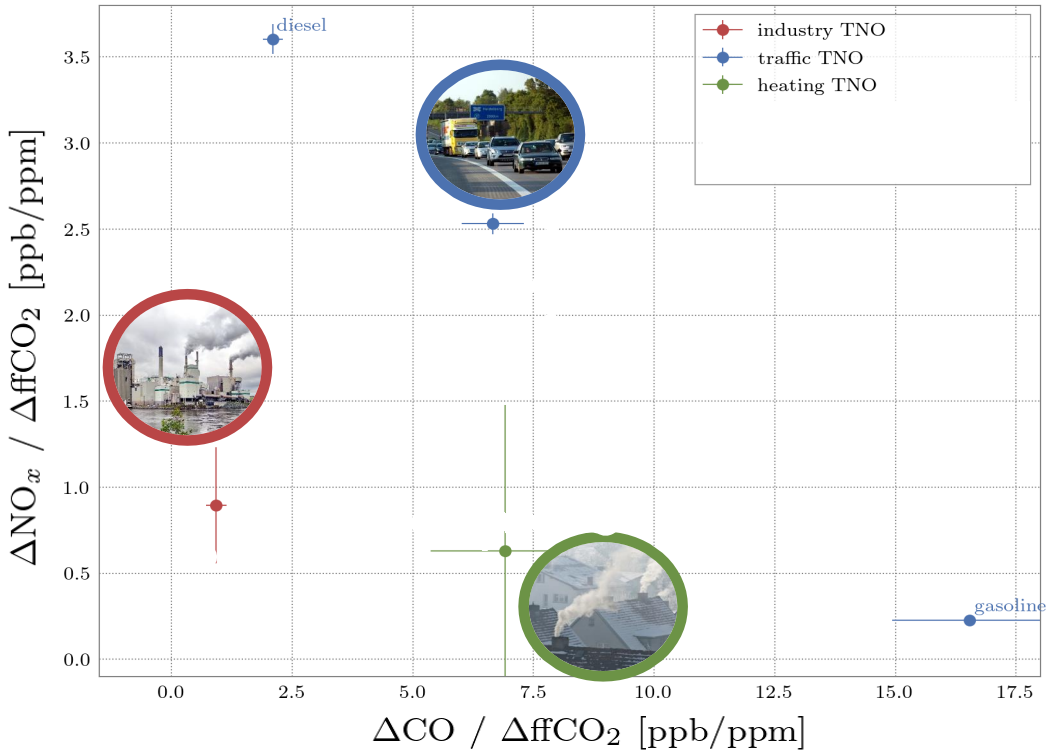
Total-column Xproxy/XCO_2 ratios with $R^2 > 0.5$





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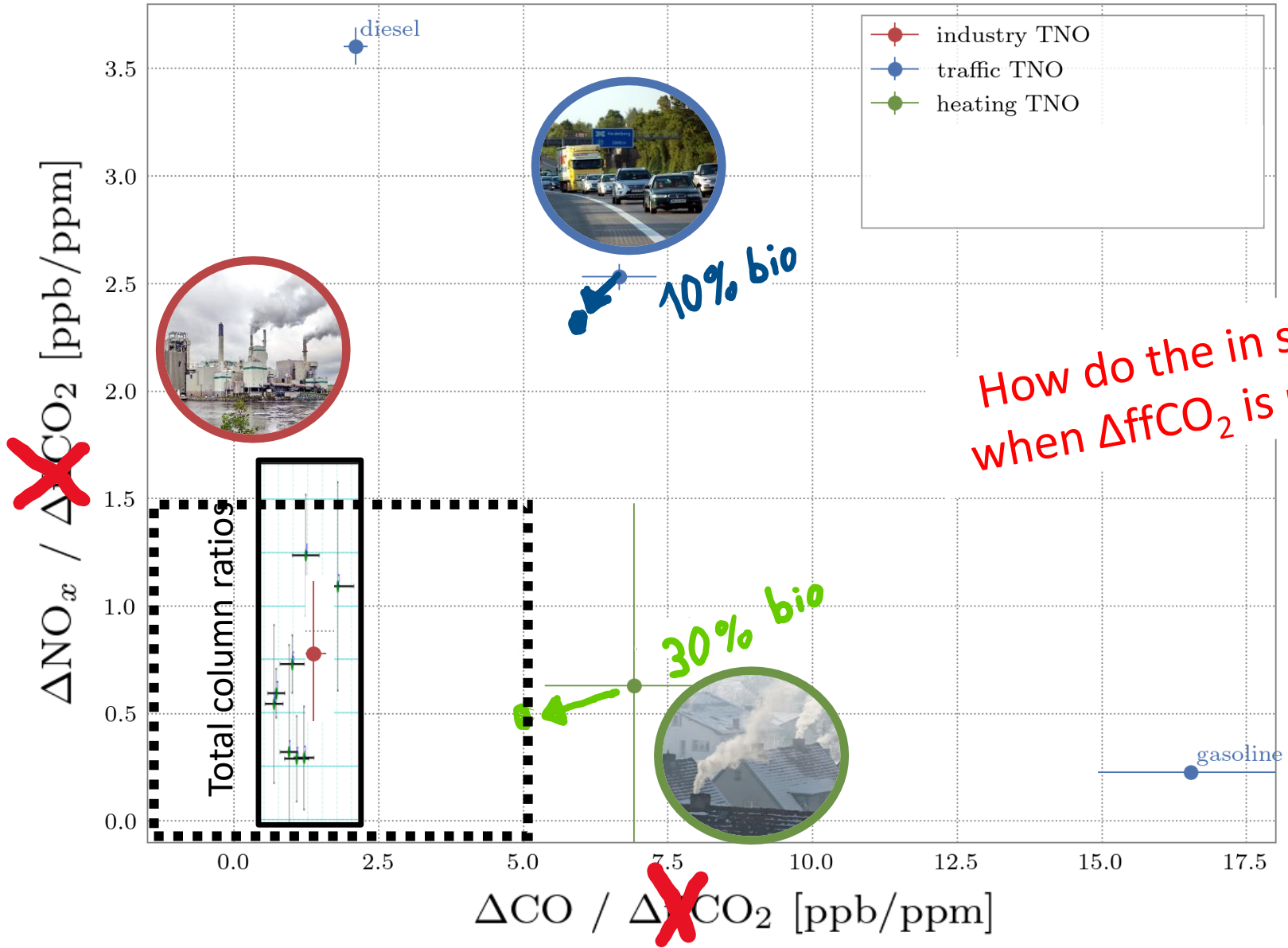
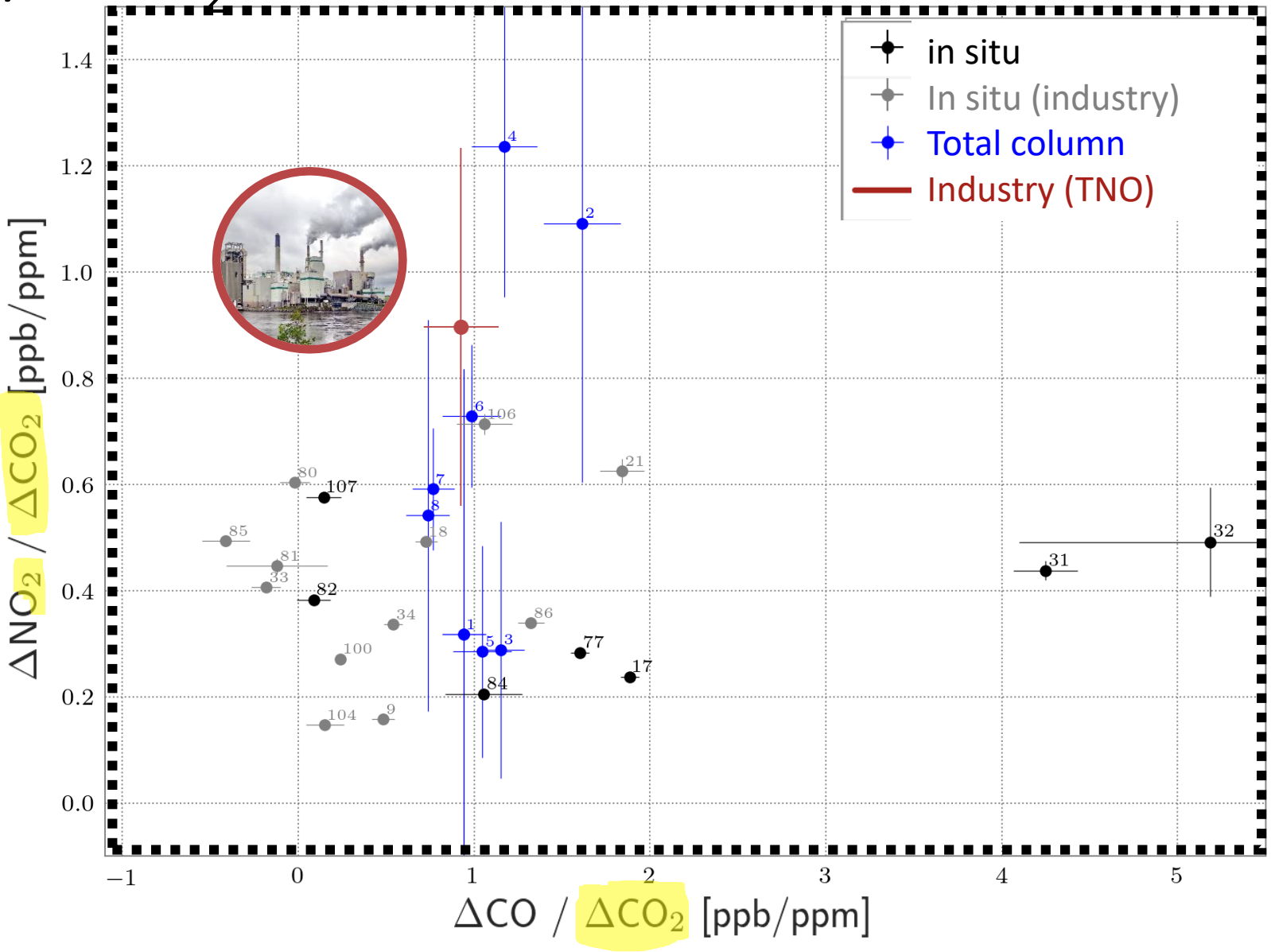
Double-ratio plot – total column ratios

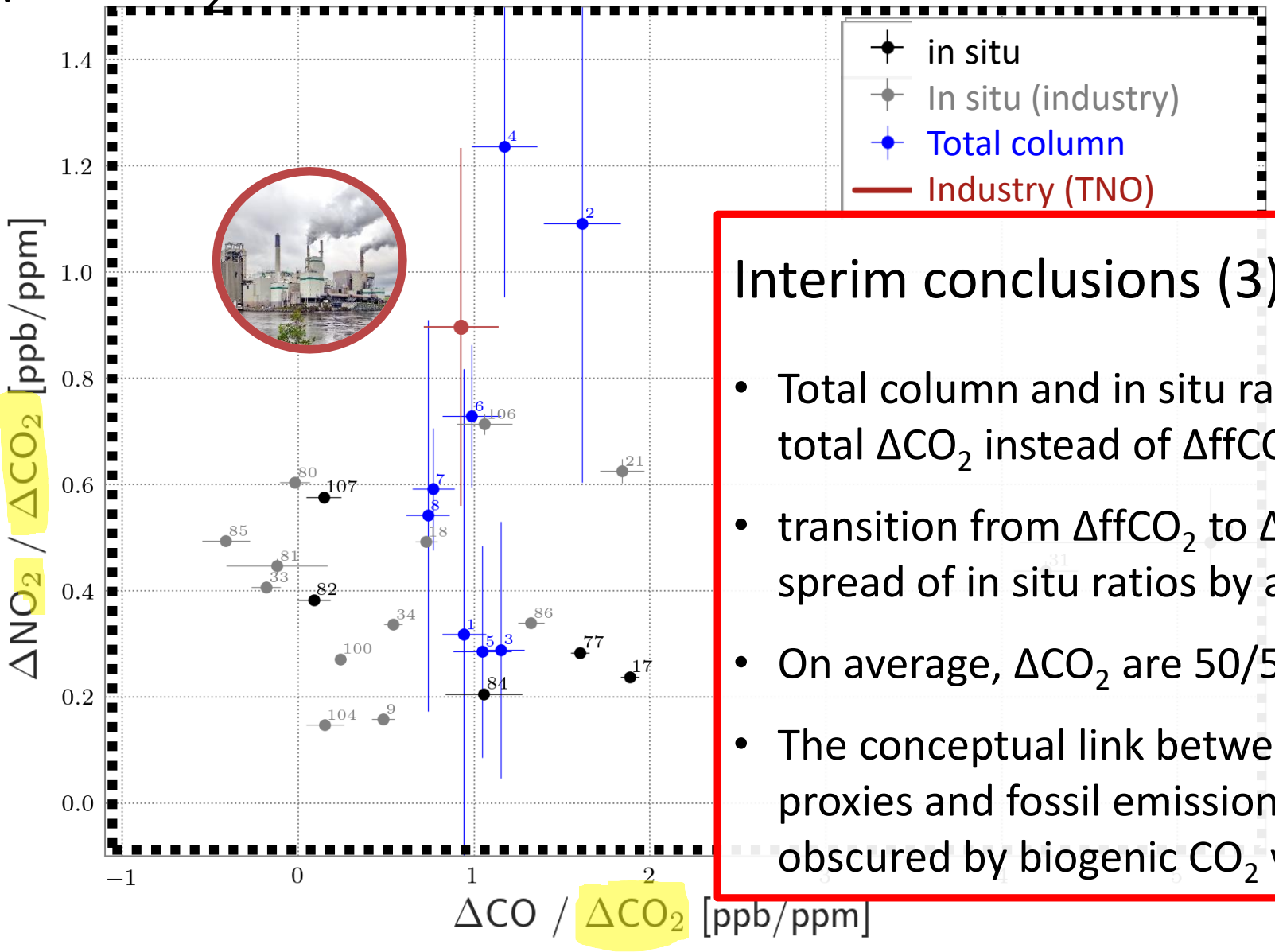


How do the in situ ratios change when $\Delta ffCO_2$ is replaced by ΔCO_2?

Δproxy/ΔCO_2 ratios: in situ and total column



Δproxy/ΔCO_2 ratios: in situ and total column



Interim conclusions (3):

- Total column and in situ ratios coincide if total ΔCO_2 instead of $\Delta ffCO_2$ is used.
- transition from $\Delta ffCO_2$ to ΔCO_2 reduces the spread of in situ ratios by a factor of 2.
- On average, ΔCO_2 are 50/50 bio/fossil
- The conceptual link between co-emitted proxies and fossil emissions is largely obscured by biogenic CO_2 variations.

Summary: Atmospheric proxy/$ffCO_2$ ratios - potentials and limitations for regional $ffCO_2$ estimation -

Potentials

- $^{14}CO_2$, CO and NO_x observations allow for sectoral attribution.
- In heterogeneous emission landscapes, both proxies provide reasonable qualitative $ffCO_2$ estimates.
- The variability of the proxy ratios decreases with increasing distance from the source area.

Limitations

- $ffCO_2$ signals > 5ppm needed
- Quantitative proxy-based fossil fuel estimates are prone to significant uncertainties.
- Problems of air chemistry and non-fossil sources increases.

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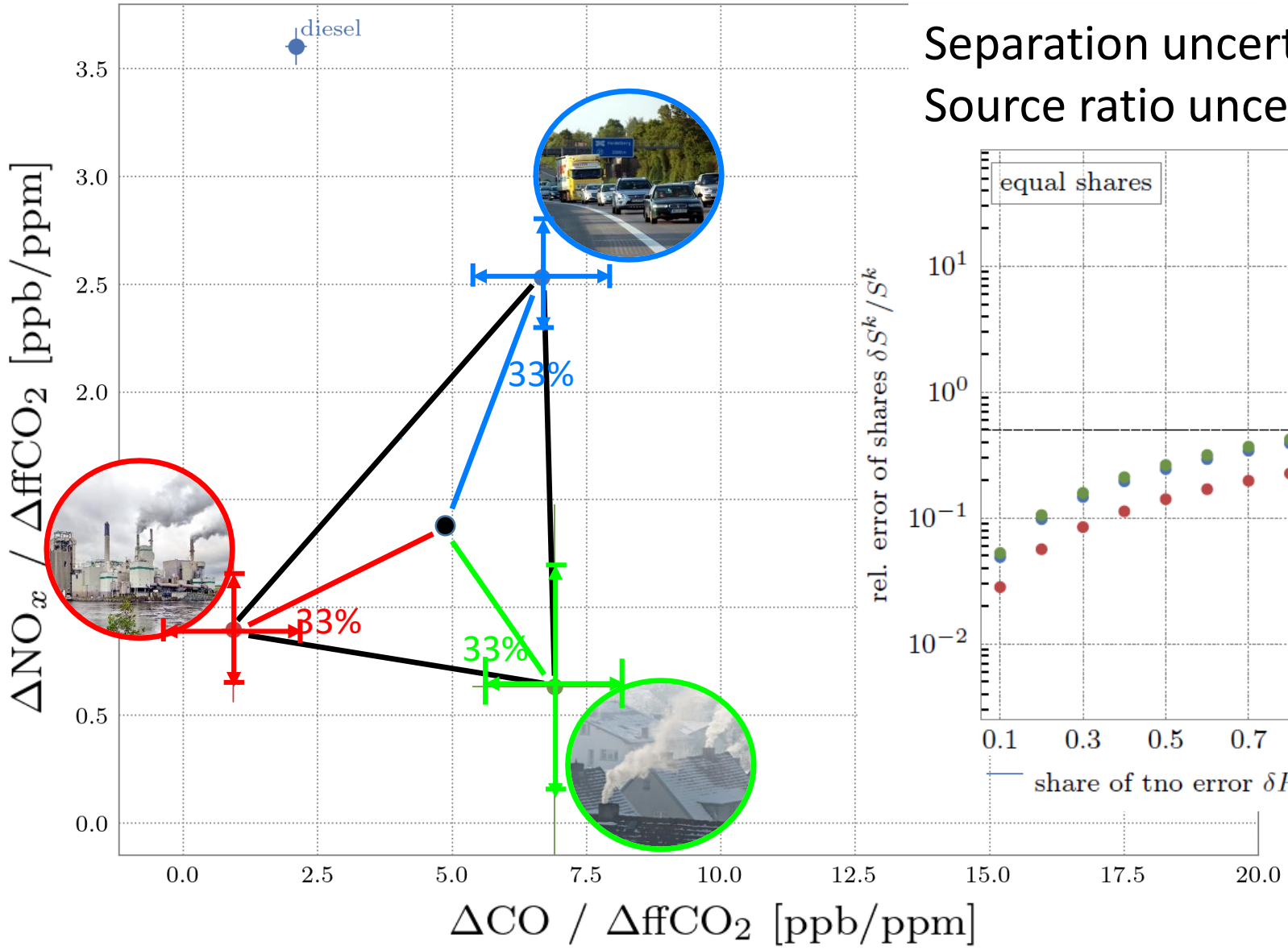
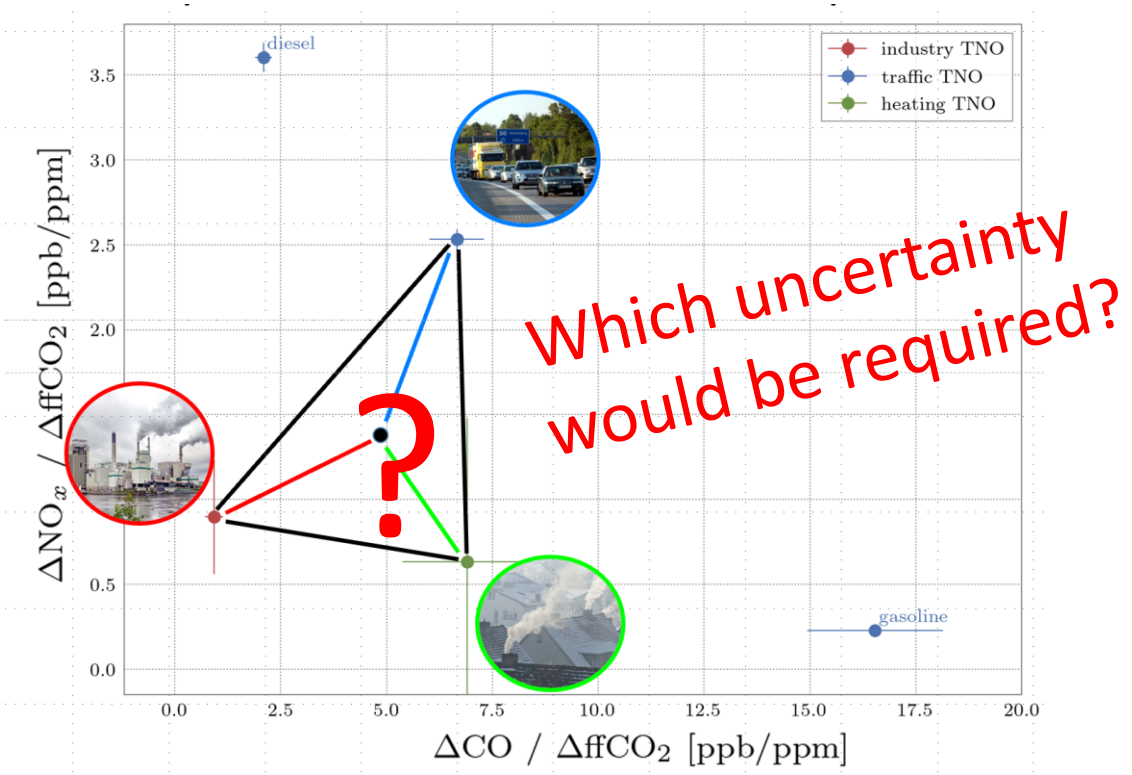
Theoretical potential of source separation?



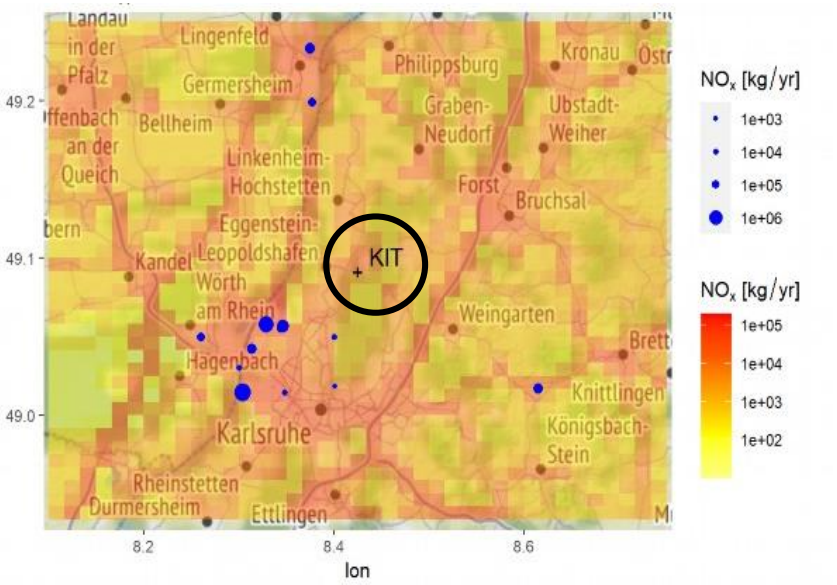
Separation uncertainty wrt.
Source ratio uncertainty?

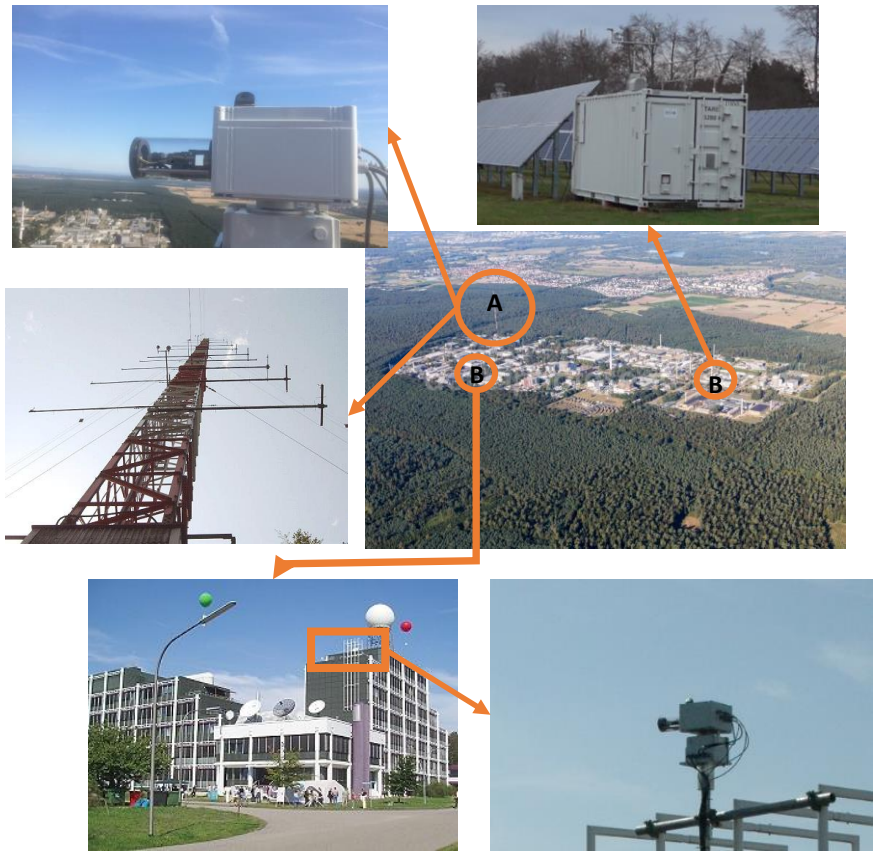
Discussion stimuli

1. Why do the proxy/ffCO$_2$ ratios from the highway campaign agree better with the mean traffic mix?
2. How meaningful is a sectoral attribution with a relative uncertainty of approx. 50%?



TCCON and MAX-DOAS location





MAX-DOAS measurement geometry