# Continuous measurements of the $CH_4$/CO ratio at the remote site of Mt. Cimone and their application for the estimate of regional $CH_4$ emissions.

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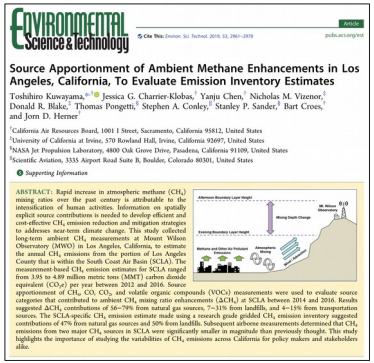
# Motivations


Environmental Science & Technology

Source Apportionment of Ambient Methane Enhancements in Los Angeles, California, To Evaluate Emission Inventory Estimates

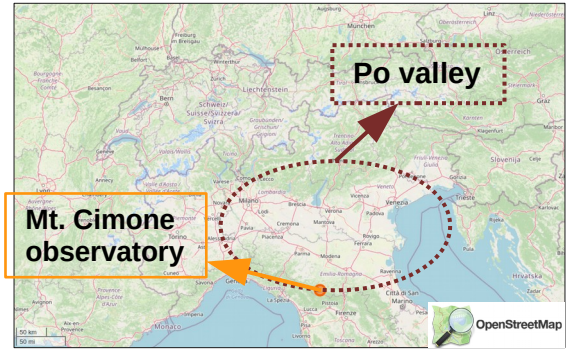
Toshihiro Kuwayama, Jessica G. Charrier-Klobas, Yanju Chen, Nicholas M. Vizenor, Donald R. Blake, Thomas Pongetti, Stephen A. Conley, Stanley P. Sander, Bart Croes, and Jorn D. Herner

[1] Kuwayama, T., et al. (2019). *Source apportionment of ambient methane enhancements in Los Angeles, California, to evaluate emission inventory estimates*. Environmental science & technology, 53(6), 2961-2970.

In-situ measurement of **$CH_4$/CO ratio** at Mt. Wilson observatory were used to evaluate $CH_4$ emissions from Los Angeles city using **inventory-based CO emissions**

$$M_{CH_4}^{estimated}[t] = \left(\frac{CH_4}{CO}\right)^{in-situ} \cdot M_{CO}^{inventory}[t]$$

Evaluated from regressions on hourly $CH_4$ and CO measurements at the observatory

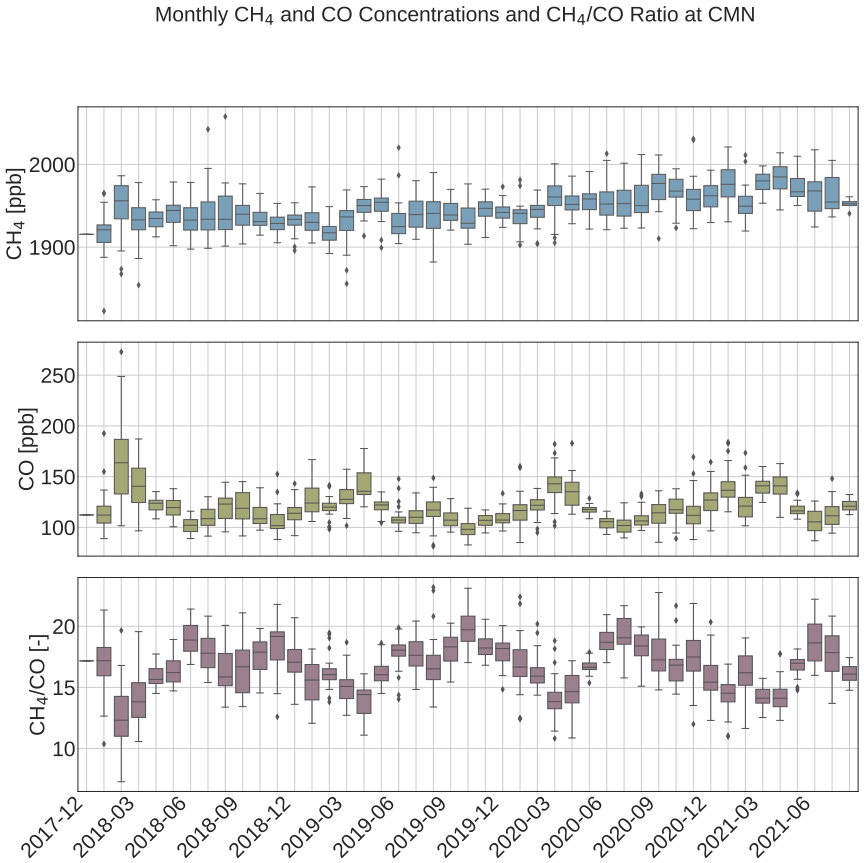



Is it possible to adopt a similar approach to evaluate $CH_4$ emissions occurring over the Northern Italy trough $CH_4$/CO observed at Mt. Cimone?

- More complex emission region: **wider** and **less homogeneous** with respect to Los Angeles
  - Both rural, urban and mountain areas included in the study domain

**A critical assessment of the sensitivity of the considered methodology to different input parameters and settings was carried out.**

# Time series of $CH_4$ and CO at Mt. Cimone



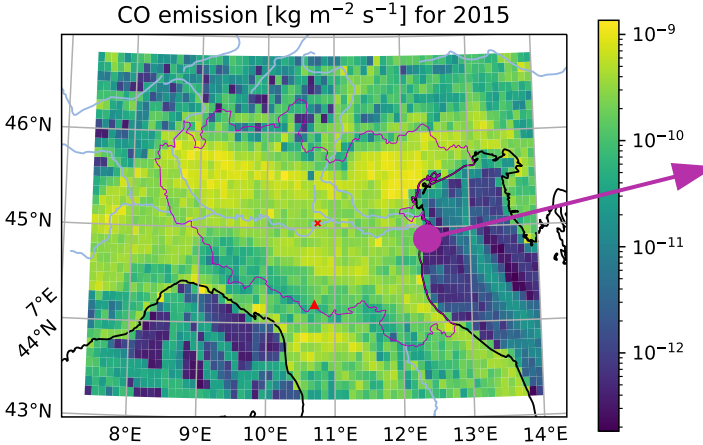

- The Mt. Cimone (CMN) atmospheric site is operated by CNR-ISAC in collaboration with Italian Air Force
- Global Station within WMO-GAW and class-2 atmospheric site in ICOS-RI

- Timeseries in the period Jan 2018 - Dec 2021 were analyzed.
- Measurements were carried out with a CRDS gas analyzer from Picarro following the measurement guidelines defined in ICOS-RI (2020) [2]
- **Hourly averaged** values considered in this work are a mixed time series composed by **(1)** a dataset internally produced by CNR-ISAC from Jan to Apr 2018, **(2)** L2 ICOS Atmosphere Release (May 2018 - Jan 2021), ICOS L1 NRT growing time series (Feb- Dec 2021) [3] [4] . ICOS L1 and L2 data can be accessed by https://data.icos-cp.eu/portal/ .

# Estimation of $CH_4$ and CO emissions from inventories.

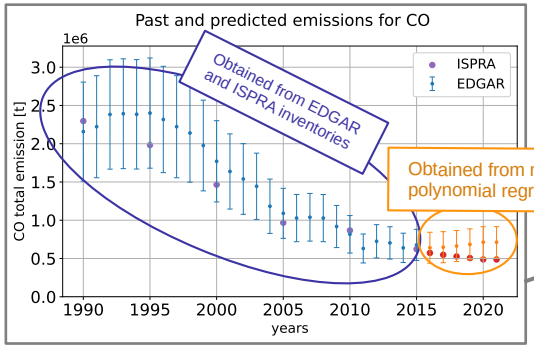
Province-based emission region selection based on **EDGAR** [6,7] (*Emissions Database for Global Atmospheric Research, European Commission Joint Research Center*) and **ISPRA** [8,9] (*Istituto Superiore per la Protezione e la Ricerca Ambientale*) inventories:

EDGAR - spatial resolution: 0.1°x0.1° , time resolution: 1 year
ISPRA - spatial resolution: province-based, time resolution: 5 years




Selected region: greater Po basin domain. emission **footprints** from the **STILT model** [10] were used to define the regional domain which most likely affected CMN

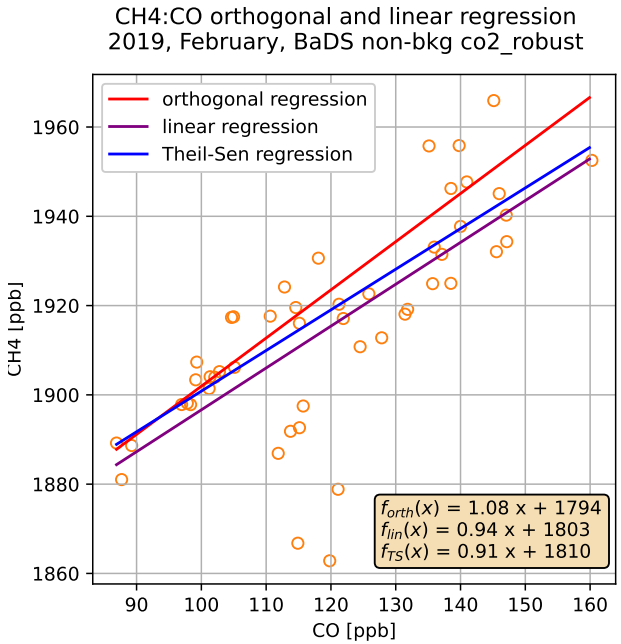
Run on the ICOS carbon portal




- Multiple linear fit to estimate emissions in the years following 2015 (ISPRA, EDGAR CO) or 2018 (EDGAR $CH_4$)
- Ongoing analysis of the last ISPRA release (data up to 2020)

# Parameters and data selection

Different setup were considered with the goal of defining the **sensitivity** of the results by the **input data** and methodology settings:




Sensitivity to the **sub-setting** of in-situ data at CMN:

- All data
- Night-time vs **Day-time**
- Subsetting by **wind direction** (310°-80°; 110°- 180°)
- Application of statistical selection algorithm (BaDSfit [5]) to select **non-background observations**.

Sensitivity to the method used to calculate CH4/CO ratio

- Different time aggregation (monthly, seasonal)
- Different fitting calculation (linear, orthogonal,Theil-Sen)

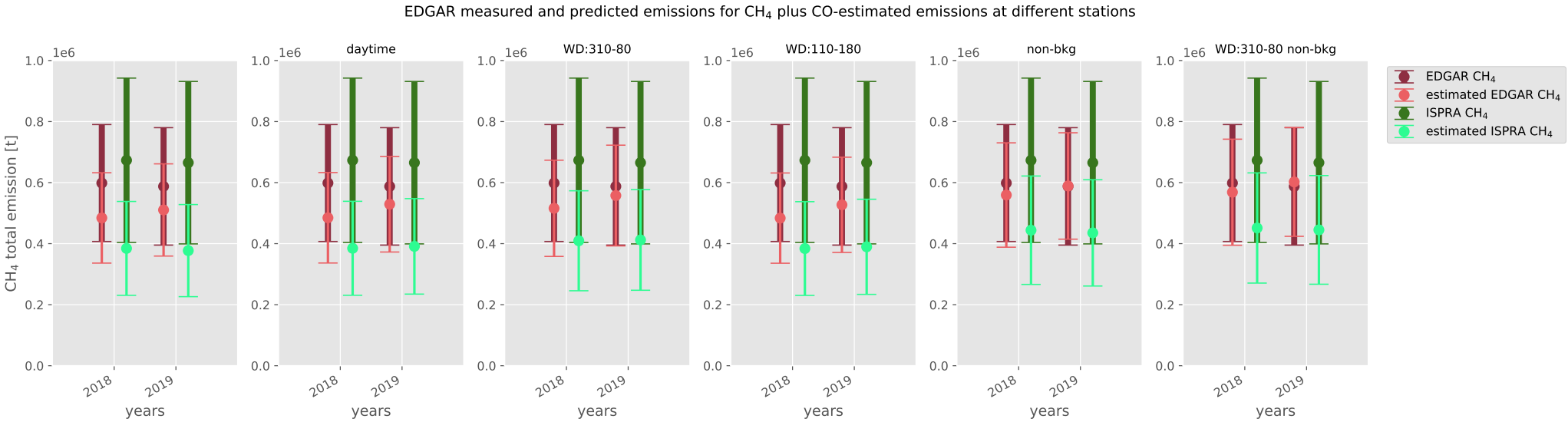
Sensitivity to the source domain boundary selection

- Keeping EDGAR pixels completely inside the source domain
- Keeping EDGAR pixels partially inside the source domain

# $CH_4$ estimated emissions at Mt. Cimone as a function of different setup

Selections over:

- Wind direction (**WD**)
- Daytime
- Emission inventory (**EDGAR**, **ISPRA**)
- **non-bkg** conditions from BaDSfit algorithm




Wind direction (**WD**) selection + **non-bkg** conditions :
Slight improvements with respect to the all data case

Emission inventory (**EDGAR**, **ISPRA**):

- Higher differences
- Differences maximised for the ISPRA case study

# Discussion

**Caveats**:

- To define a limited source emission domain (even if corroborated by STILT) is rather arbitrary
- Absolute mixing ratios of $CH_4$ and CO are used (i.e. assuming that all the $CH_4$/CO variability were related to regional emissions, transport on longer spatial scales neglected)
- Differences between estimated and inventory emissions are larger in the ISPRA with respect to the EDGAR case studies
- Total emissions for 2018-2019 were extrapolated from past emission values

**Next steps:**

- To calculate $CH_4$/CO ratio using **deviations from baseline** (together with the selection of not-background data) could help in **better represent regional emissions**
- Extend the analysis to 2020 ISPRA release (already available) and EDGAR 2022 release (still not available)
- Better evaluation of uncertainties related to the presented method

# Acknowledgements

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# References

[1] Kuwayama, Toshihiro, et al. *"Source apportionment of ambient methane enhancements in Los Angeles, California, to evaluate emission inventory estimates.*" Environmental science & technology 53.6 (2019): 2961-2970.

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