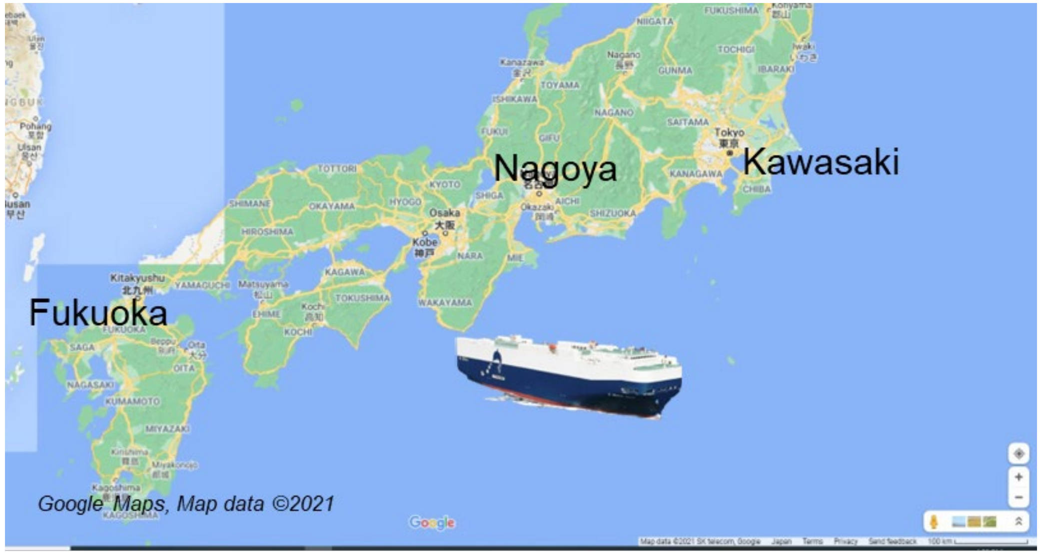
National Institute for Environmental Studies, Japan

UNIVERSITÄT HEIDELBERG ZUKUNFT SEIT 1386

Towards Shipborne Emission Monitoring and Satellite Validation of CO_2, CH_4, CO, and NO_2 Through Simultaneous Columnar and In Situ Observations



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1 Background

Importance

For the global stocktake, we need:

- Improvement of **emission inventories**
- Assessment of the **reduction potential** of anthropogenic GHG and air pollutant emissions

➢ Precise observations with high spatial and temporal coverage

Current situation

The global coverage of in situ observations by public and private networks (ship-, aircraft-, ground-based) and satellite observations is increasing.

New generation satellite missions combine CO_2 and NO_2 observations to better constrain fossil fuel-related emissions and plumes (GOSAT-GW, CO2M).

Problem

Over oceans and coastal regions, reference datasets for carbon cycle studies and satellite validation **remain scarce** (e.g. by TCCON).

TCCON: Total Carbon Column Network (https://tccondata.org/)

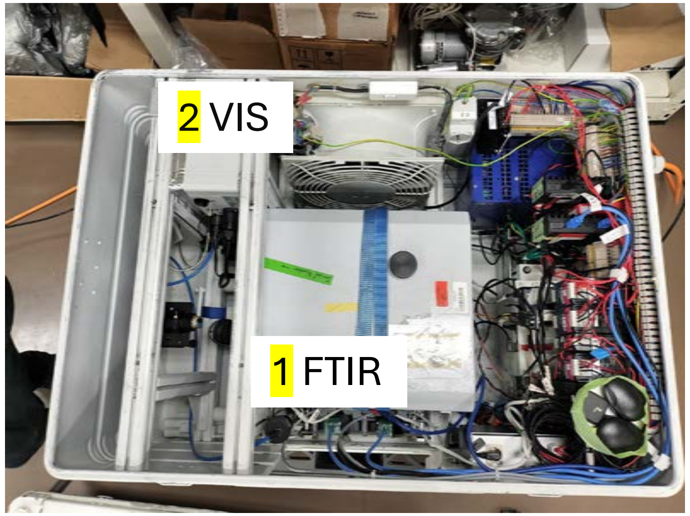
We use **new ship-based remote sensing techniques** as complement to stationary networks for measuring XCO_2, XCH_4, XCO, and VCD NO_2, combined with surface in situ observations.

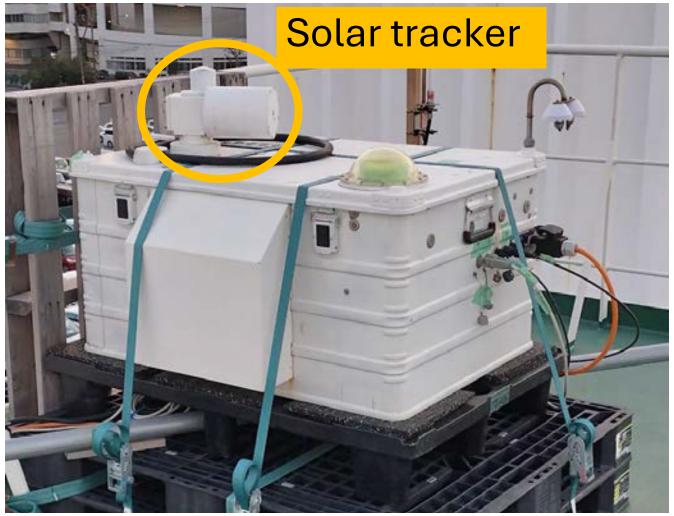
GOSAT-GW: Global Observing SATellite for Greenhouse gases and Water cycle
CO2M: Copernicus Anthropogenic Carbon Dioxide Monitoring

VCD: vertical column density
X[species]: column-averaged dry air mole fraction

a) Semi-automatic FTIR–VIS Spectrometer

(FTIR: Fourier Transform Infrared, VIS: visible spectral range)





1 FTIR spectrometer (Bruker Optics, EM27/SUN) (spectral range 0.91–2.5 μm)
Target: total column (**TC**) & column-averaged dry air mole fractions (**Xgas**) of CO_2, CH_4, CO
(retrieval: *PROFFAST v2.4, KIT, 2024, https://www.imk-asf.kit.edu/english/3225.php*)

2 VIS spectrometer (Ocean Optics, QE-Pro) (spectral range 400.7– 495.9 nm)
Target: vertical column density (**VCD**) of NO_2
(retrieval: *pre-processor (Enders, V. et al., in prep.); QDOAS (Danckaert et al., 2017)*)

TC ≡ VCD

2 Methodology

b) Surface in situ observations

- CAPS NO_2 analyzer (Shoreline Science, CAPS-NO-B-7003C). Target: **NO_2.**
- NO/NO_2/NOx Analyzer (Thermo Scientific, 42iTL). Target: **NO.**
- CRDS analyzer (Picarro, G2401). Target: **CO_2, CH_4, CO.**
- O_3 analyzer (Thermo Scientific, 49i). Target: **O_3.**

CAPS: Cavity Attenuated Phase Shift
CRDS: Cavity Ring-Down Spectroscopy

c) Additional data and filtering

- ERA5 hourly data on pressure levels (*Hersbach et al., 2023; Copernicus Climate Change Service (C3S) Climate Data Store, 2023*)
- Climate TRACE (2023). *Climate TRACE Inventory of Greenhouse Gas Emissions*. Retrieved from https://climatetrace.org.
- Hysplit backward trajectories (*https://www.ready.noaa.gov*)
- Filtering ship exhaust:
  - a) wind direction & speed vs. ship direction & speed (rear funnel)
  - b) abrupt increase and decrease of NO and O_3, respectively.
- 5 min averages

2 Methodology

d) Location

Along major anthropogenic emission sources on Japan's coast between Kanto (Tokyo Metropolitan Area) and Kyushu.

The location was chosen for testing the performance of the semi-automatic FTIR–VIS spectrometer and its application to monitor anthropogenic emissions.

PGN: Pandonia Global Network
TCCON: Total Carbon Column Observing network
COCCON: COllaborative Carbon Column Observing Network

Case study Hiroshima - Kanda

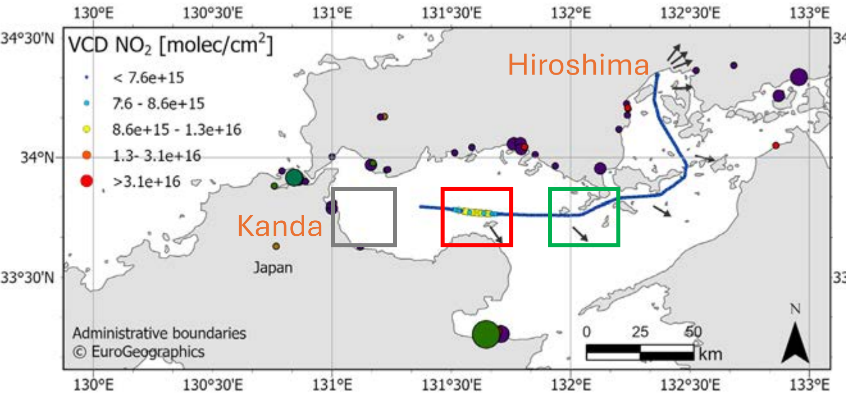
CO_2 emissions
[Mt/Oct 2023]
- <0.10 Mt/Oct
- <0.30 Mt/Oct
- <0.59 Mt/Oct
- <1.01 Mt/Oct
- <1.90 Mt/Oct

- Power plant
- Steel factory
- Oil/gas refinery
- Cement industry

(source: Climate TRACE (2023))

Instrument viewing direction

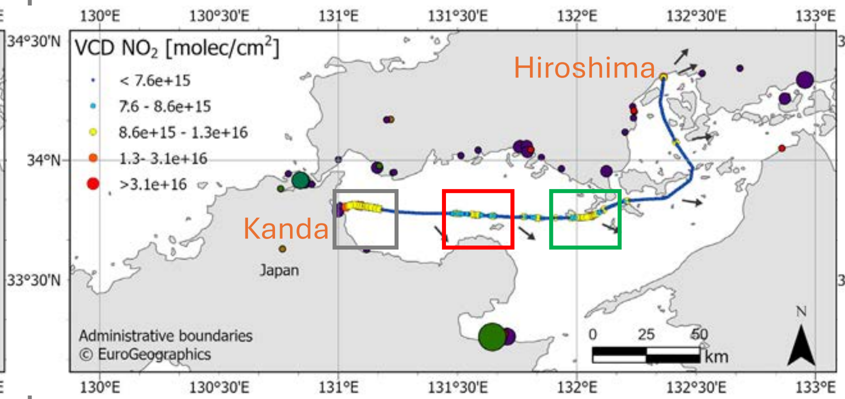
18 October 2023



VCD NO_2 [molec/cm^2]

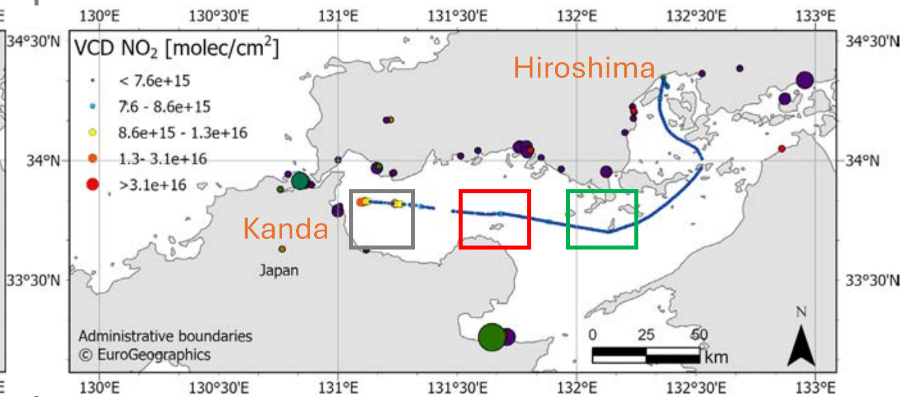
- Low (green box)
- Increased (red box)

25 October 2023



- Increased (green box)
- Increased (red box)

10 April 2024



- Low (green box)
- Low (red box)

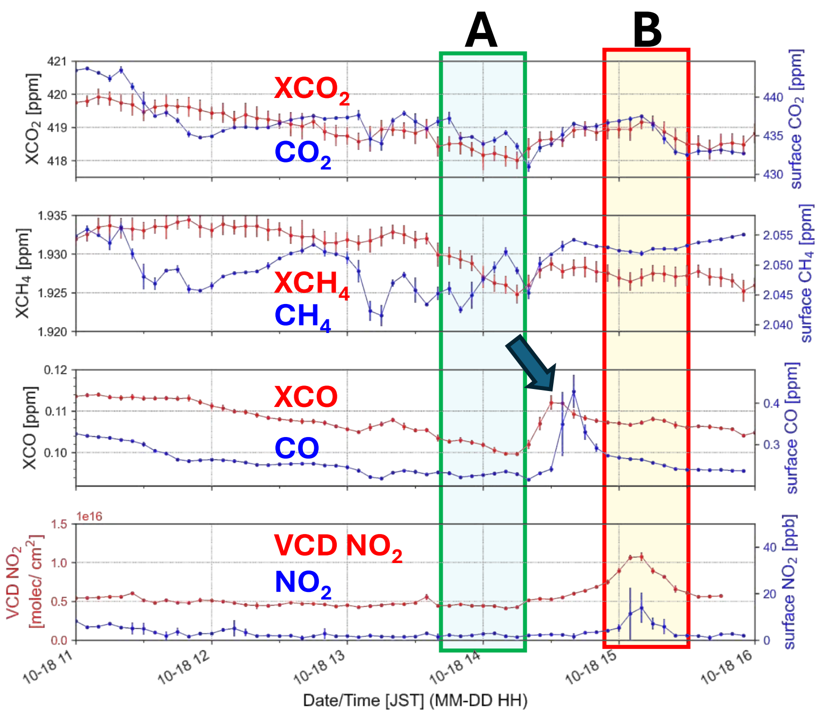
Ship exhaust contaminated (grey box)

Ship's direction: Hiroshima to Kanda

3 Results

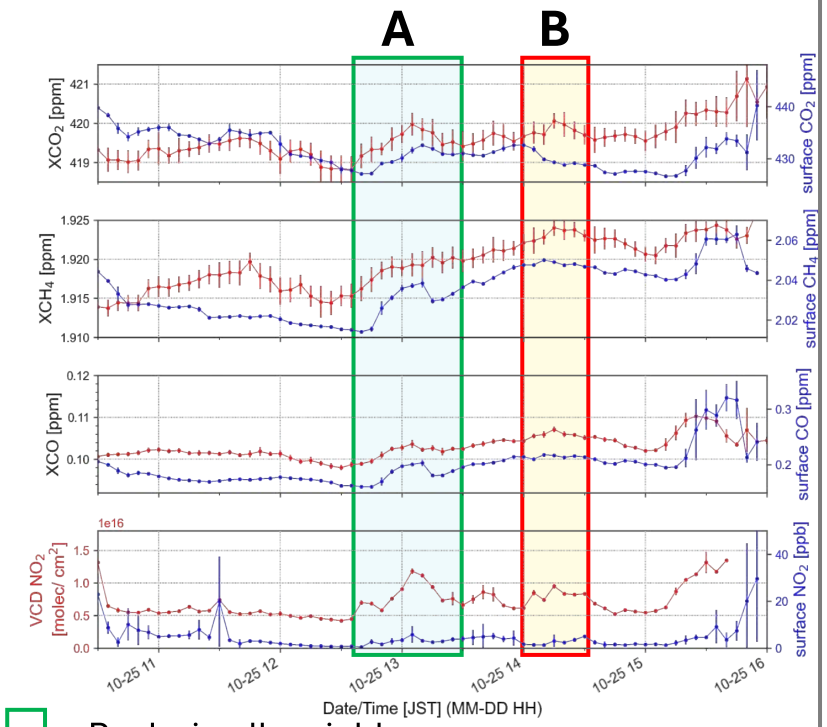
Observations

18 October 2023



- Decreasing trends; lower values than at **B**
- Peaks in CO_2, NO_2; higher values than at **A**
- CO peaks in the column before the surface

25 October 2023



- Peaks in all variables
- Column: Elevated CO_2, NO_2 and CH_4
  Surface: decreasing CO_2 and low NO_2

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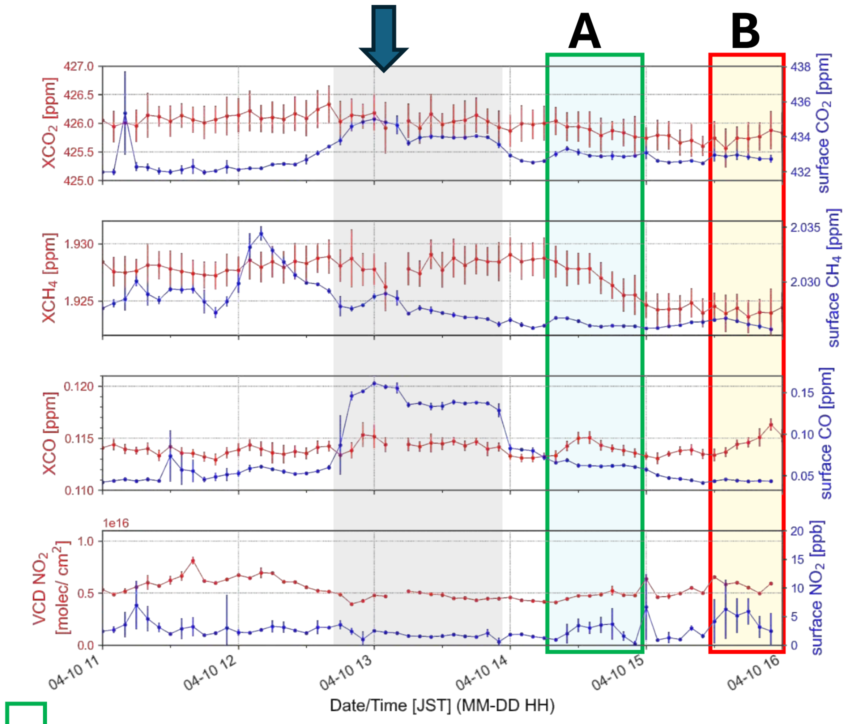


- Small peaks in column CO and surface NO_2
- Small peaks in surface CO_2, CH_4, CO

A: Background conditions
B: Fresh fossil fuel combustion
Lofted, aged CO-rich plume

A: Fresh combustion + CH_4 leaks?
B: Mixed aged industrial emissions

A, B: Lofted CO residual from incomplete combustion/ships?
NO_2: Local fossil fuel combustion
Local incomplete combustion, LNG

3 Results

Observations

6h backward trajectory 500 m (red arrow)

6h backward trajectory 100 m (purple arrow)

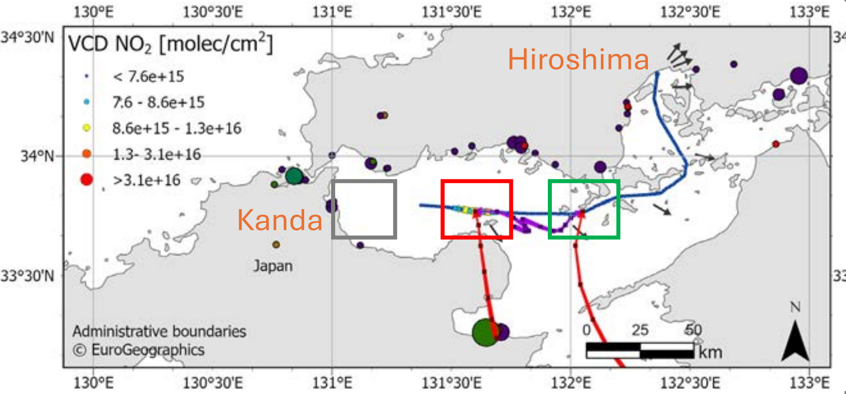
Source: HYSPLIT trajectory model, *https://www.ready.noaa.gov*

- Power plant
- Steel factory
- Oil/gas refinery
- Cement industry

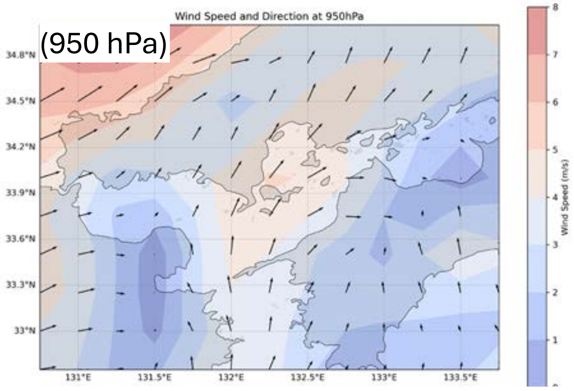
(source: Climate TRACE (2023))

Instrument viewing direction

18 October 2023

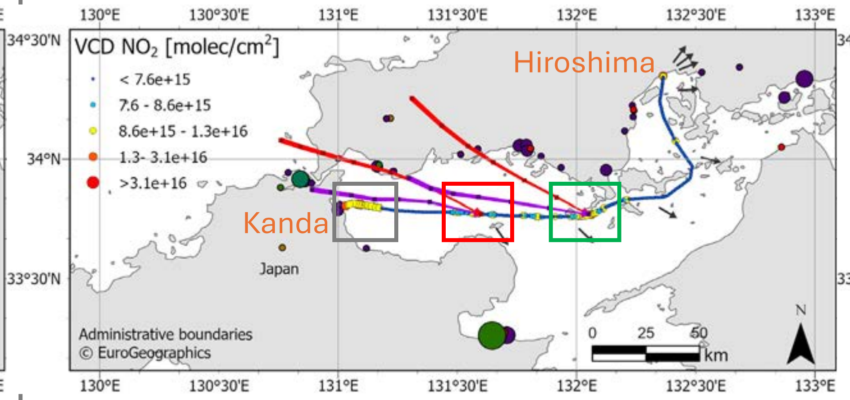


Southerly winds

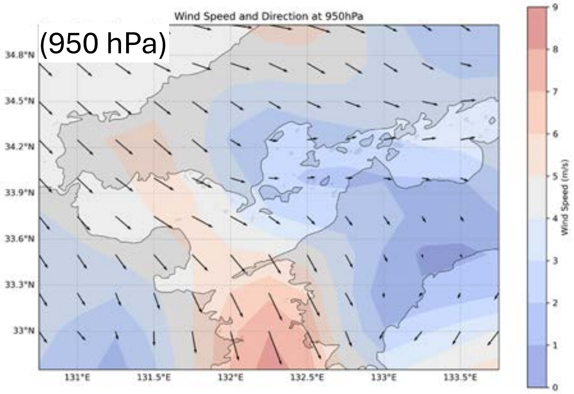


Boundary layer wind direction & speed

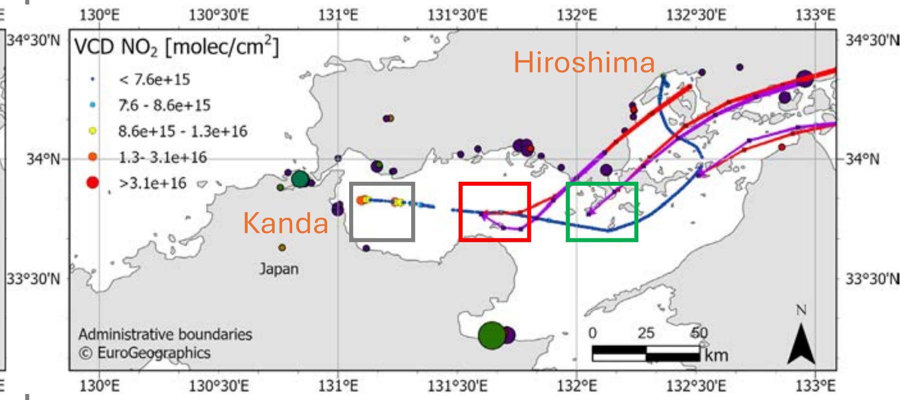
25 October 2023



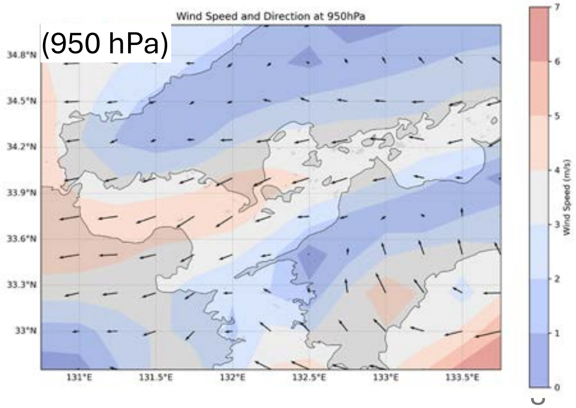
Northwesterly winds



10 April 2024



Easterly/ Northeasterly winds



Source: ERA5, *C3S Climate Data Store, 2023*

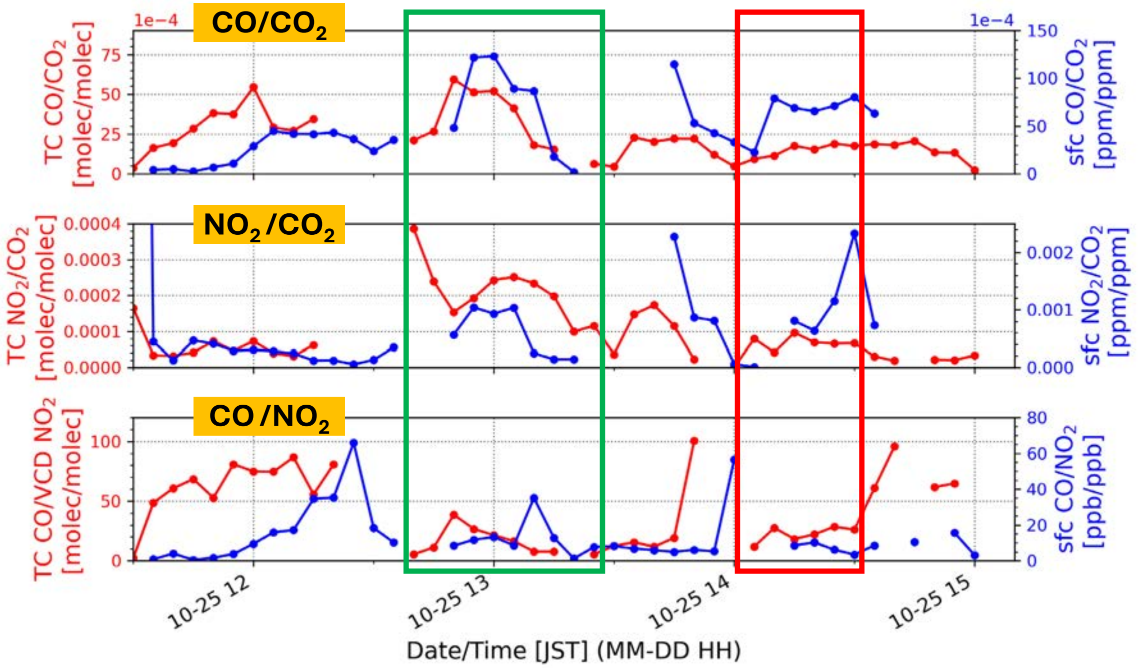
3 Results

Comparison of column-derived and surface in situ ratios

Example 25 October 2023

Background removal: Subtraction of the rolling 5th percentile over a 1-hour centered window (preliminary)

Column derived ratios
Surface in situ derived ratios



A	B
CO/CO_2 = indicator for combustion efficiency	
• Higher ratio • Column ratio is half the surface ratio	• Lower ratio • Column ratio is 1/3 the surface ratio
NO_2/CO_2 = indicator for fossil fuel combustion	
• Higher ratio	• Lower ratio
CO/NO_2 = indicator of combustion type	
Similar ratios	

- Location **A** shows higher CO and NO_2 enhancements relative to CO_2 compared to **B**, suggesting recent, inefficient fossil fuel combustion, while **B** might reflect either more aged emissions or the influence of power plants with emissions controls (NO_X-poor).

  → Emission sources can be power plants (NO_2-rich), mixed with industrial sources and ship exhaust (NO_2 + CO-rich).
- Similar CO/NO_2 ratios at location **A** and **B** suggest similar emission types.

SFC = surface concentration, TC ≡ VCD = total column

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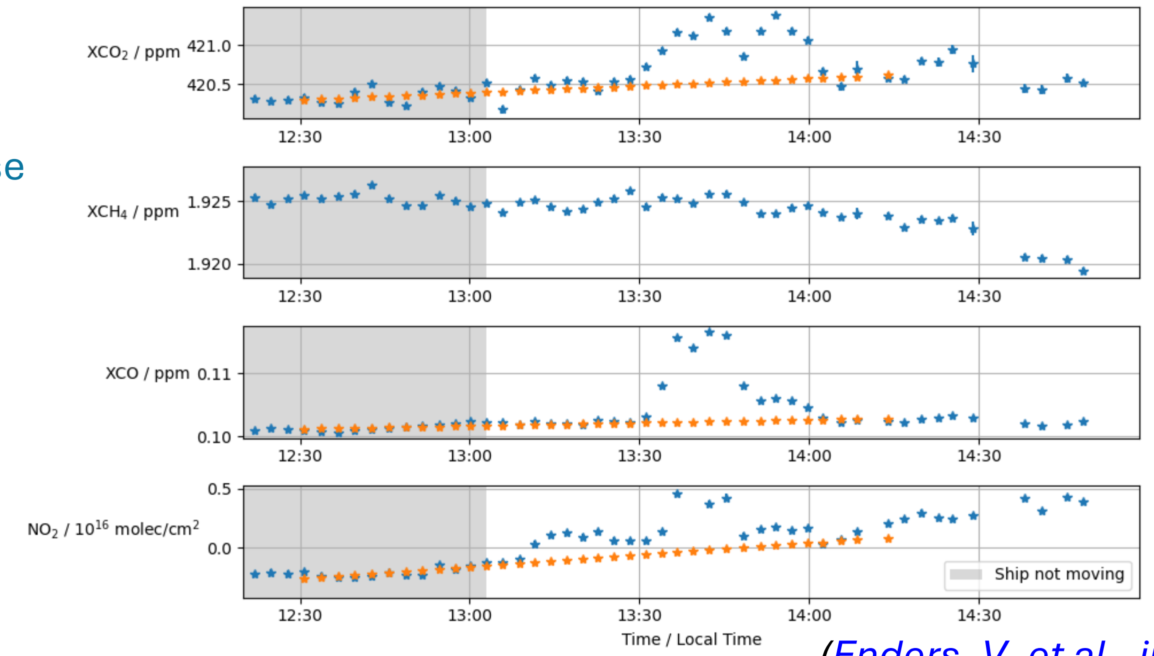
4 Conclusion & Outlook

- Combining observations by the semi-automatic FTIR–VIS Spectrometer with in situ surface observations, we demonstrate in the initial analysis the potential for emission plume detection and its contribution to a better understanding of the vertical distribution of anthropogenic emissions

- **Further analysis includes**
  - A more robust background removal method for estimating enhancement ratios in highly mixed emission regions
  - An assessment of the contribution of the surface emissions to the column observations
  - A more detailed emission source identification, including comparison with high-resolution model results.
  - EM27/SUN retrieval with the upcoming *PROFFAST v2.4.1 release (Feld, L., KIT), which is capable of handling mobile data.*

A detailed description of the instrument setup and a case study comparing emission ratios with inventories is in preparation.

In that case study, distinct plumes allowed the background removal by using least-square line fitting to pre- and post-plume observations (*Luther et al., 2019*).



(Enders, V. et al., in prep.)

Further outlook

- The future goal is to deploy the setup on a ship crossing the Pacific Ocean from North to South for direct validation of columnar satellite observations.
- Ship- and aircraft-based in situ data will complement the observations.