Detecting and assessing trends of CFCs and substitutes from IASI measurements

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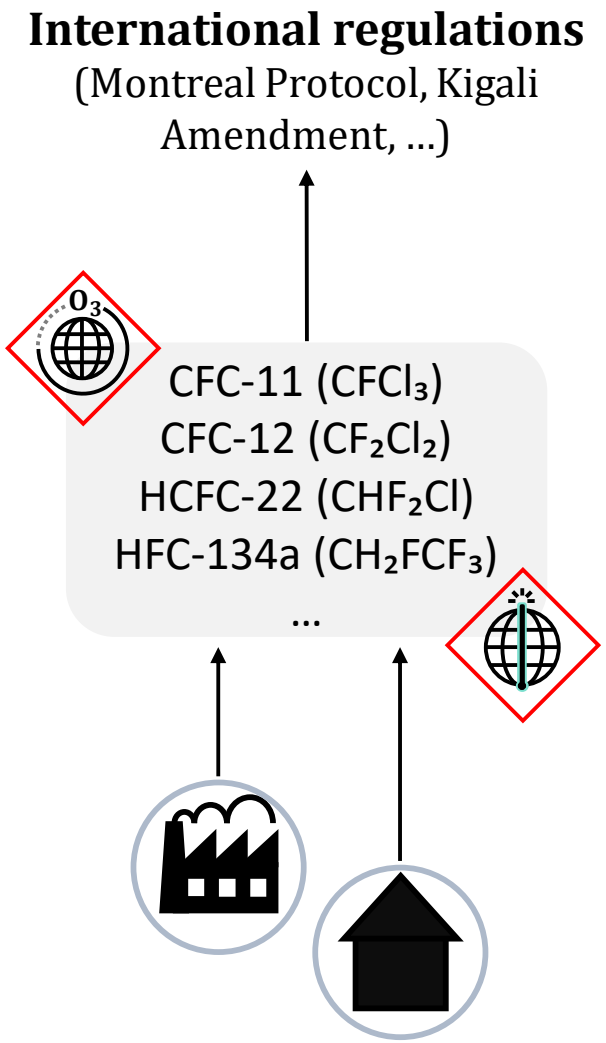
Introduction



The emissions of chlorofluorocarbons (CFCs) and its substitutes have profoundly affected the chemical and radiative equilibrium of the atmosphere. The CFCs play a key role in the **depletion of stratospheric ozone** and are, just as their hydrogenated substitutes, **powerful greenhouse gases**. The emissions of most of these species are presently controlled by **international regulations**. Following the Montreal protocol and its amendments, the concentrations of the CFCs have started to decline in the early-2000. In parallel, the concentrations of the hydrogenated substitutes have increased, and new chlorine species were found at significant concentrations. [WMO, 2018] The exploitation of the measurements from the IASI sounder could play a key role in the monitoring of these species and thereby complement existing surface measurement networks.

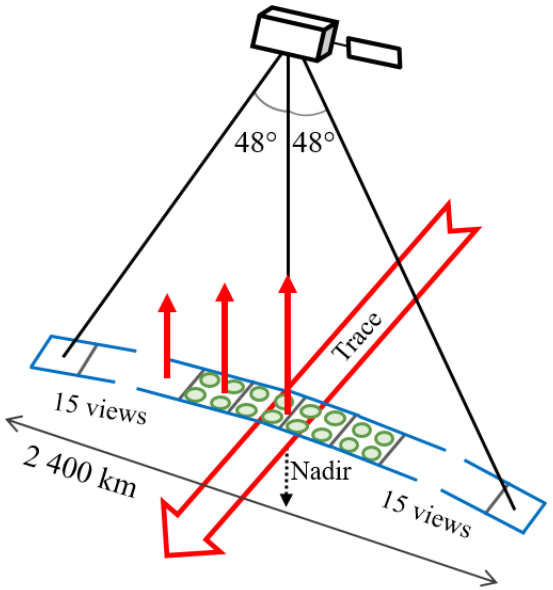
The goal of this study is to **detect the spectral signatures** of halocarbons in **IASI spectra** and to give a first assessment of the **evolution in time** of these species over the **2008 – 2017** period covered by **IASI** on Metop-A.

Infrared Atmospheric Sounding Interferometer

The spatial instrument IASI is an **operational** **hyperspectral** sounder for

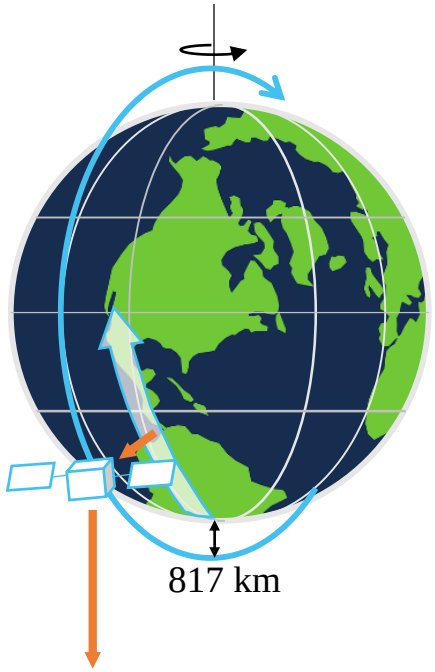
- **meteorology** (priority 1)
- **climate** and **atmospheric chemistry** (priority 2)

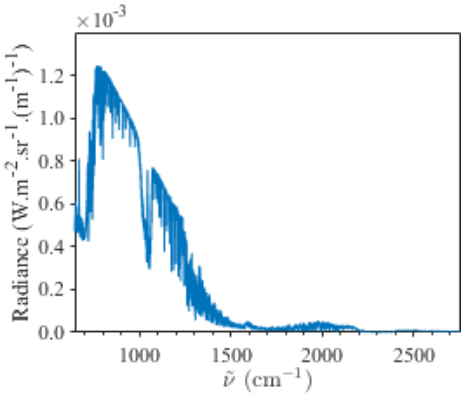
onboard **Metop**, the European meteorological satellites on polar orbit.

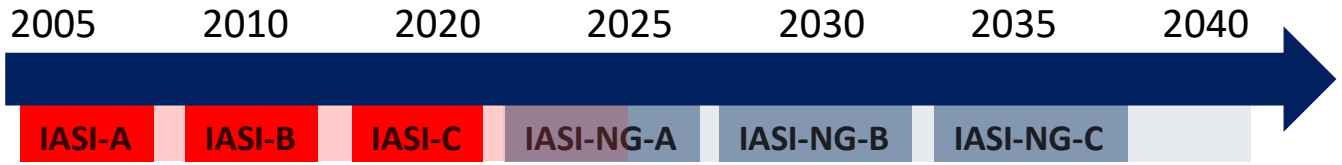


Key points for atmospheric composition

- Near-global coverage twice daily (9:30 and 21:30)
  >1 million observations daily per IASI
- Medium spatial resolution (12 km on-ground at nadir)
- High spectral radiometric performances
- Towards 18 years of global measurements
- Exceptional stability
- IASI-NG next-generation instrument

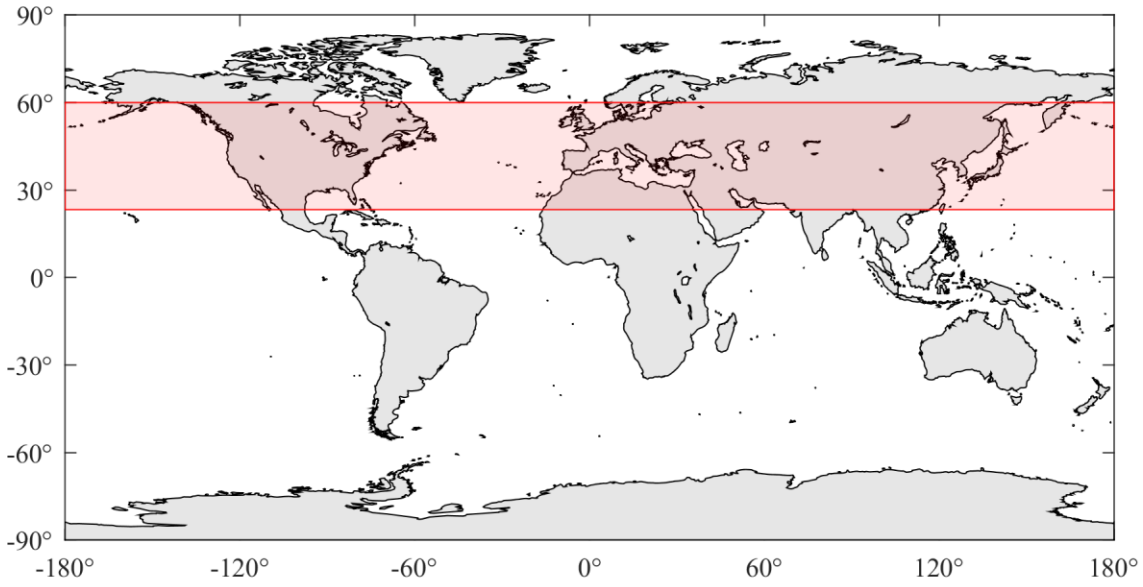






Mean IASI spectra

In this work, we exploit the first decadal record of IASI measurements (**2008–2017**) focusing on the **Northern Middle Latitudes** (23.27°N–66.32°N), examining oceanic (unpolluted) and continental (polluted) regions separately.
The IASI spectra are **monthly averaged** for each year, in order to improve the detection of halocarbons which are weak absorbers.



Whitening transformation

Whitening transformation

It transforms the spectral channels to uncorrelated random variables with unit variance and zero mean.

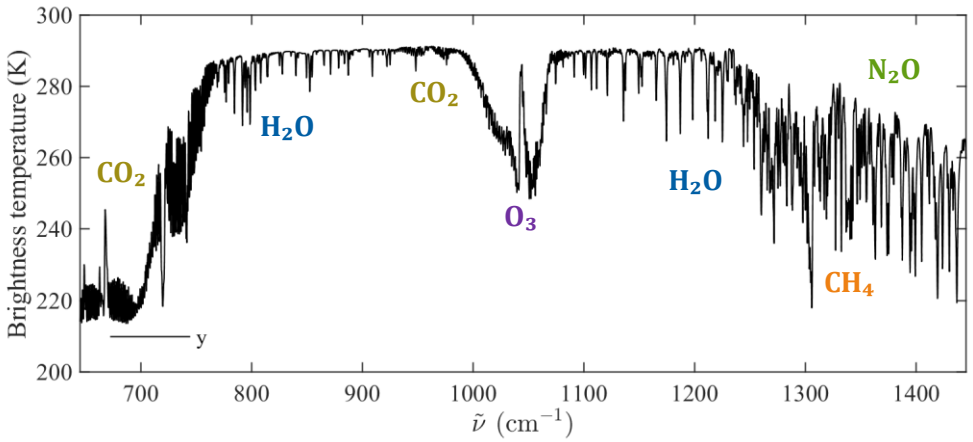
Whitening a spectrum produces another spectrum in which most of the background signal is removed and all spectral aberrations are exposed.

De Longueville, H., Clarisse, L., Whitburn, S., Franco, B., Bauduin, S., Clerbaux, C., et al. (2021). Identification of short and long-lived atmospheric trace gases from IASI space observations. Geophysical Research Letters, 48, e2020GL091742. https://doi.org/10.1029/2020GL091742

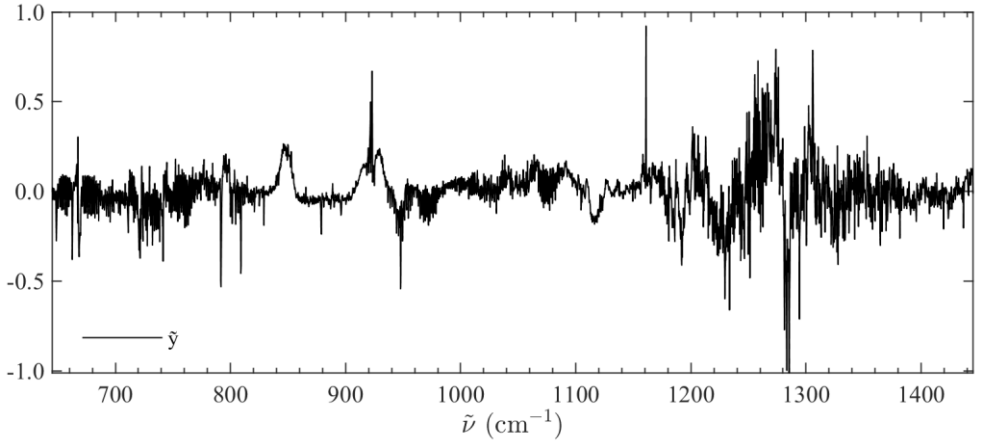
covariance matrix — reference mean spectrum

$$\tilde{y} = S^{-\frac{1}{2}} (y - \bar{y})$$

whitened spectrum — spectrum



whitening →



A positive (negative) whitened signal indicates a decrease (increase) between 2008 and 2017.

Spectral identification

covariance matrix

reference mean spectrum

$$\tilde{y} = S^{-\frac{1}{2}}(y - \bar{y})$$

whitened spectrum

spectrum

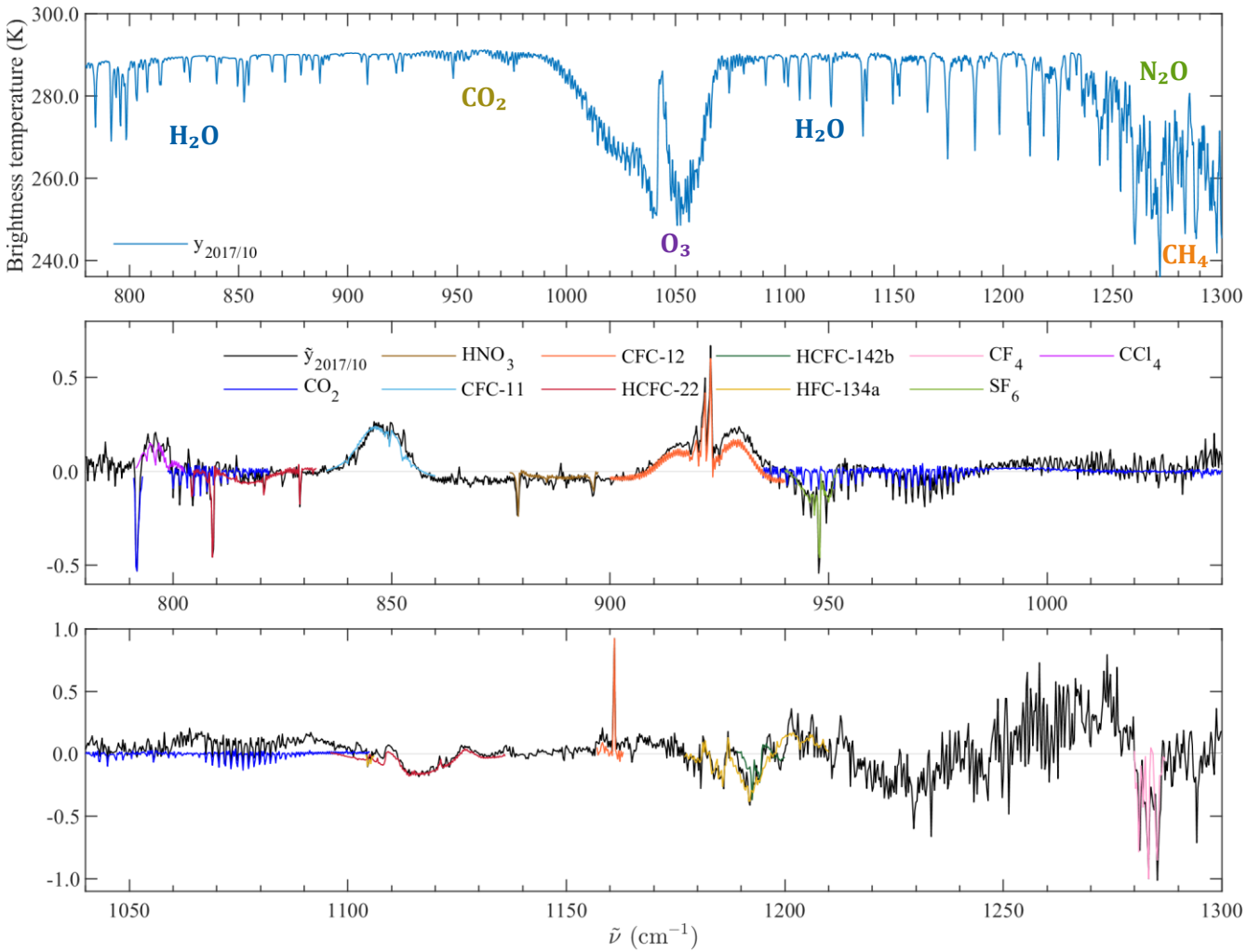
with

- y built from 287 244 clear-sky IASI spectra measured over sea in the Northern Middle Latitudes during October 2017
- $\{\bar{y}, S\}$ calculated from ~7 million clear-sky IASI spectra measured in 2008 over the same area.

Changes in eight halocarbons were identified, these are:

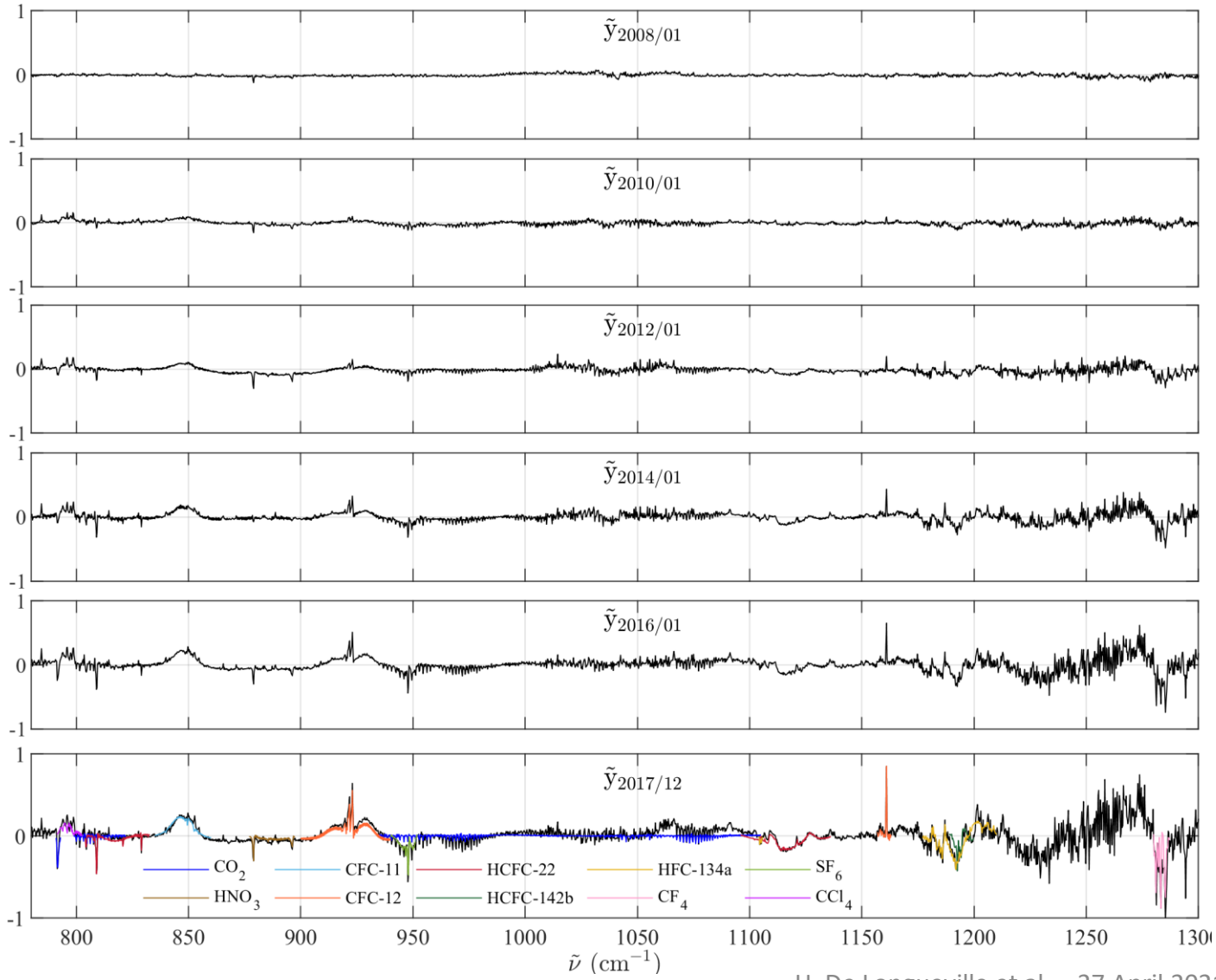
- CFC-11 ($CFCl_3$), CFC-12 (CF_2Cl_2), and carbon tetrachloride (CCl_4) which are detected with a positive value, indicating that their atmospheric abundance was smaller in 2017 than in 2008

- HCFC-22 (CHF_2Cl), HCFC-142b (CH_3CF_2Cl), HFC-134a (CH_2FCF_3), carbon tetrafluoride (CF_4) and sulfur hexafluoride (SF_6) which are detected with a negative value, indicating an increase in their atmospheric concentration between 2008 and 2017



[De Longueville et al., 2021]

Yearly evolution of the atmospheric signal



covariance matrix — reference mean spectrum

$$\tilde{y} = S^{-\frac{1}{2}} (y - \bar{y})$$

whitened spectrum — spectrum

with

- y built from clear-sky IASI spectra measured over sea in the Northern Middle Latitudes for different years/months
- $\{\bar{y}, S\}$ calculated from ~7 million clear-sky IASI spectra measured in 2008 over the same area.

A positive (negative) whitened signal indicates a decrease (increase) over times.

Trends over ten years

Concentration conversion

reference concentration (AGAGE)

$$\hat{x} = x_0 + \left(K^T S^{-1} K\right)^{-1} K^T S^{-1} (y - \bar{y})$$

concentration

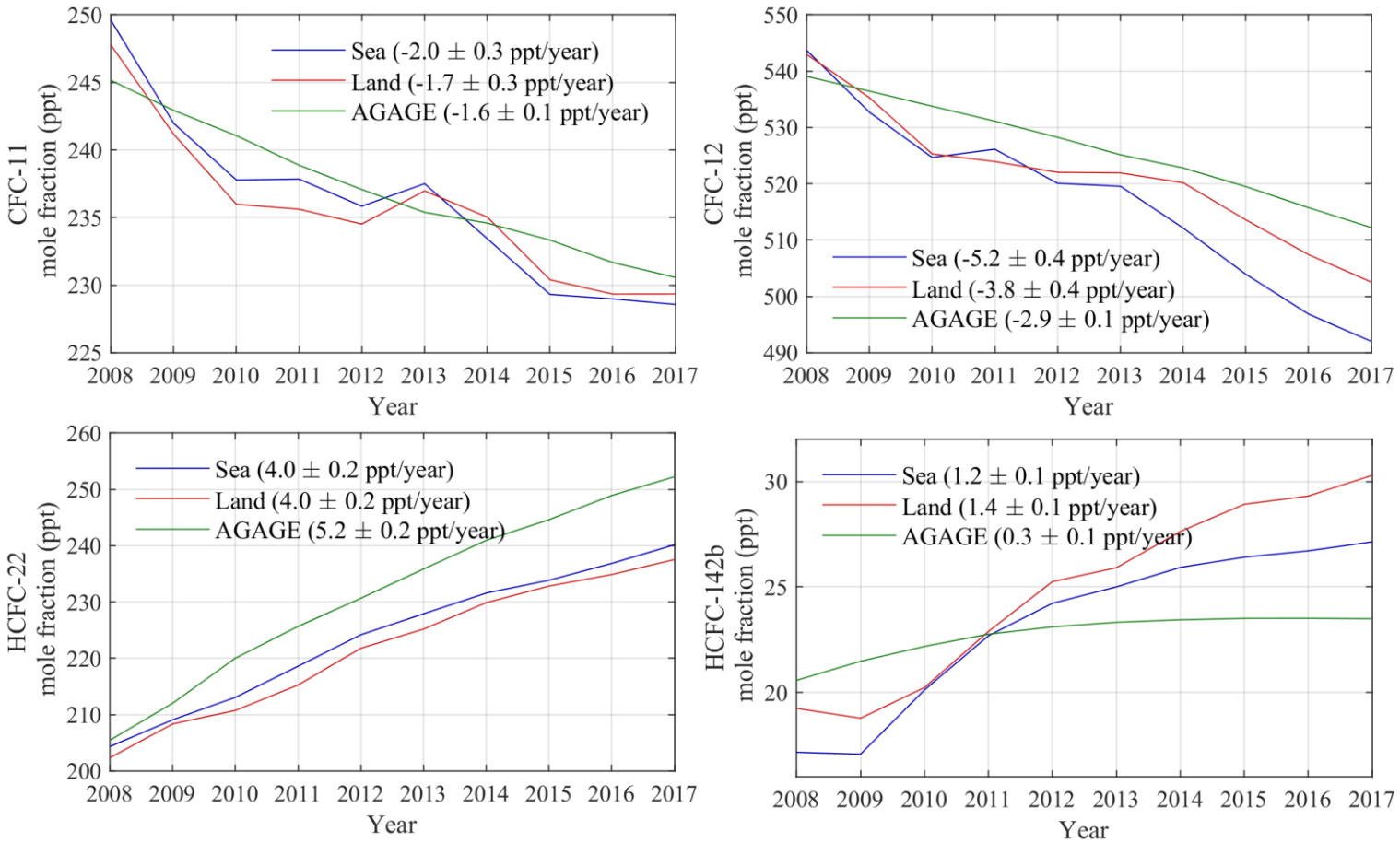
Jacobian

following Walker et al., 2011;

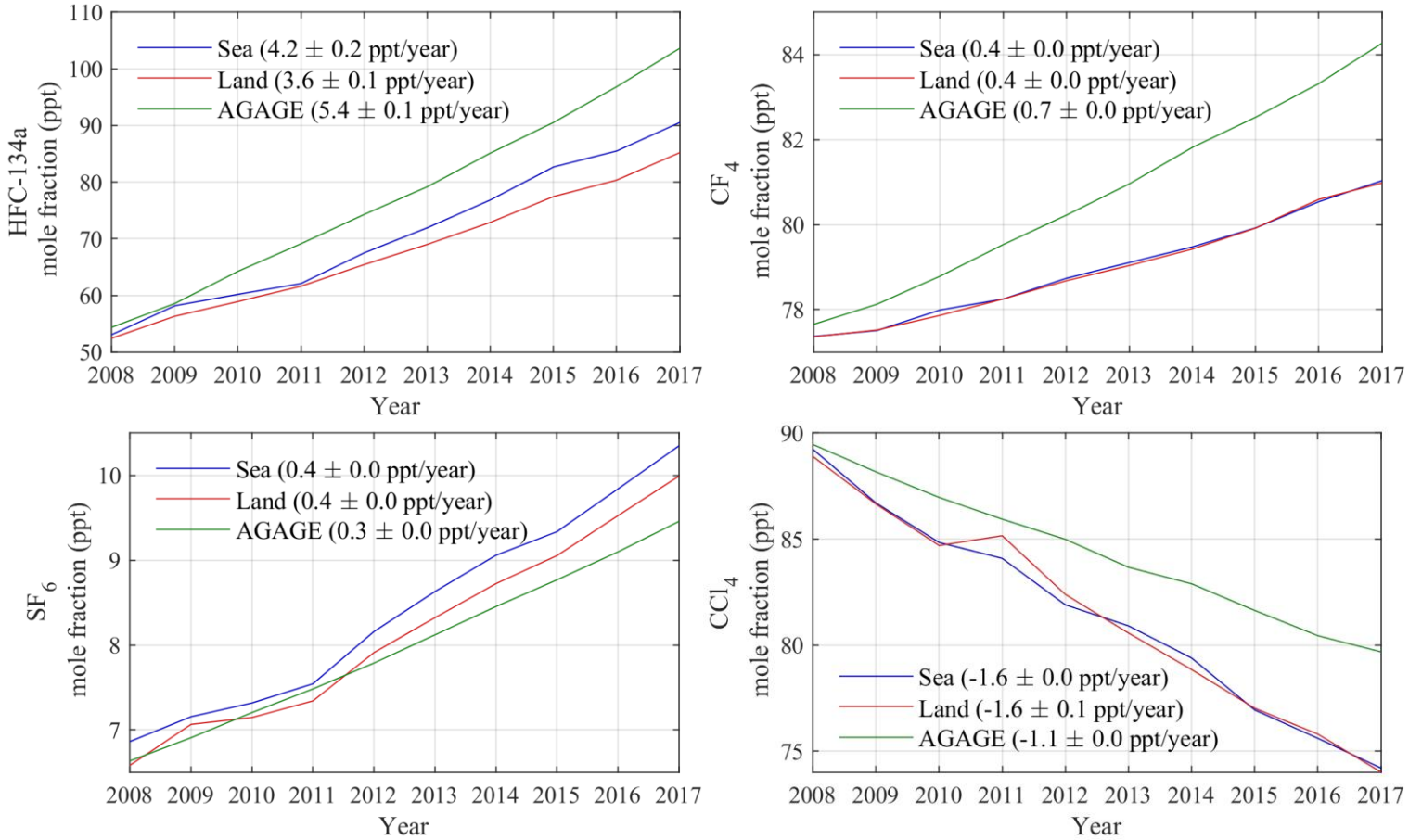
with

- y built from clear-sky IASI spectra measured over sea/land in the Northern Middle Latitudes of each month
- $\{\bar{y}, S\}$ calculated from clear-sky IASI spectra measured in 2008 over the same areas.

The monthly concentrations $\hat{x}$ are averaged over each year and compared to in situ measurements from AGAGE stations.



Trends over ten years



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Conclusion & Perspectives

The application of a whitening transformation to mean IASI spectra has allowed the **unambiguous detection** of **height halocarbons** in **IASI spectra** in the atmospheric window (780-1300 cm^{-1}): CFC-11, CFC-12, HCFC-22, HCFC-142b, HFC-134a, CF_4, SF_6 and CCl_4. The **evolution of their abundance** has been determined for the period **2008–2017** currently covered by IASI; a reasonable agreement with the in situ measurements from AGAGE stations was obtained. While still preliminary, these results are promising and it is anticipated that by refining the methodology it **will be possible** to **monitor precisely**, with IASI and follow-on missions, the **long-term evolution** of these species controlled by international protocols.

IASI → **Global** and **long-term** monitoring of the atmospheric abundance of halocarbons

- → Monitoring of the global evolution of the atmosphere system
- → Verification of the effectiveness of international protocols

References

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• Clerbaux et al., “Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder”, Atmospheric Chemistry and Physics, vol. 9, no. 16, pp. 6041-6054, 2009.

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• Walker et al., “An effective method for the detection of trace species demonstrated using the MetOp Infrared Atmospheric Sounding Interferometer”, Atmospheric Measurement Techniques, vol. 4, no. 8, pp. 1567-1580, 2011.