



# Direct satellite measurements of the radiative forcing of long-lived halogenated gases

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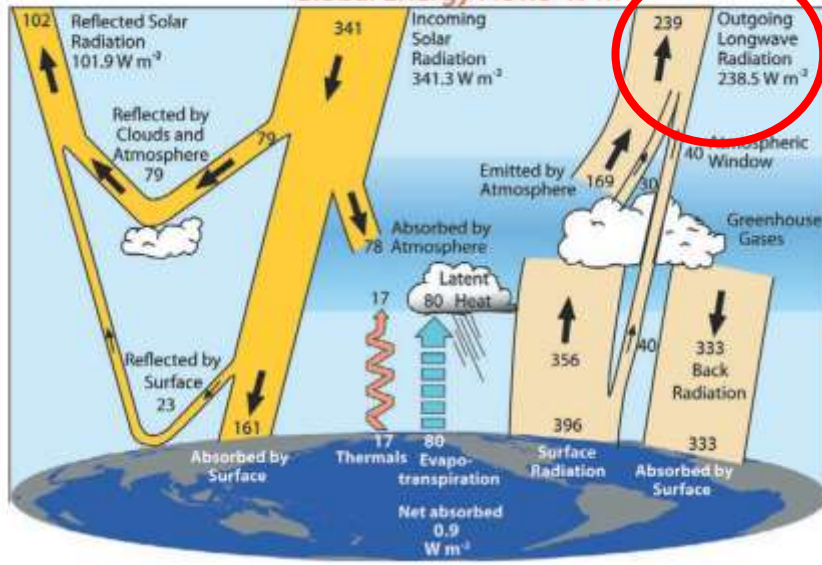
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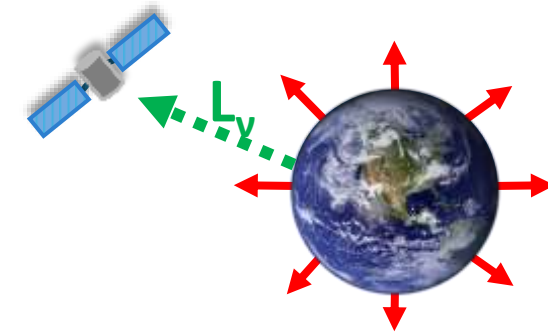
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Global Energy Flows  $\text{W m}^{-2}$ 

# The Earth's Outgoing Longwave Radiation (OLR)

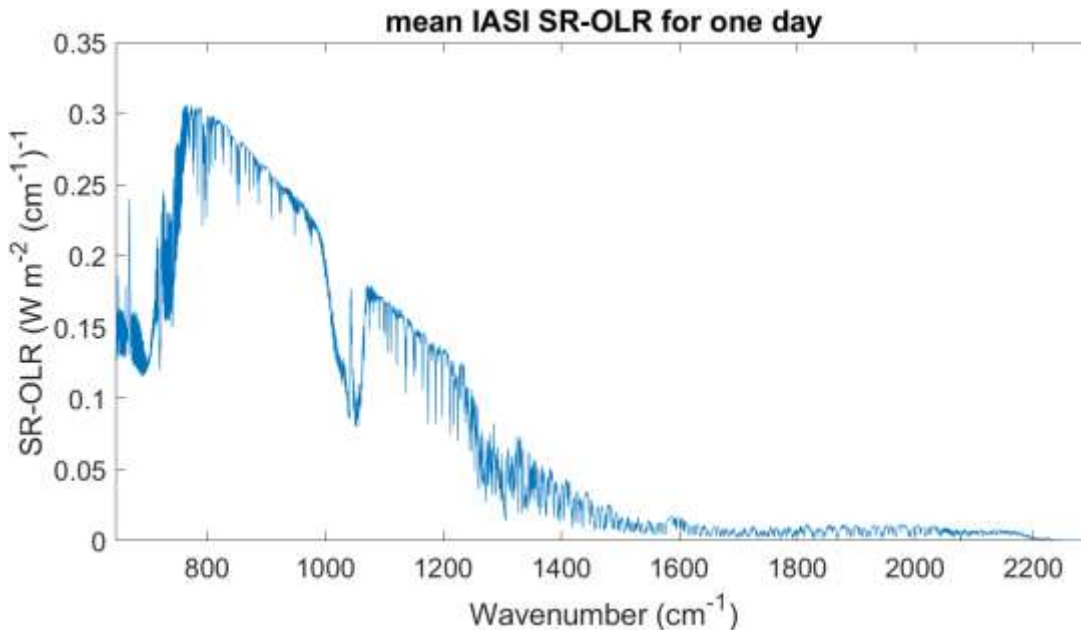
= Total radiation emitted by the Earth-atmosphere system and leaving to space



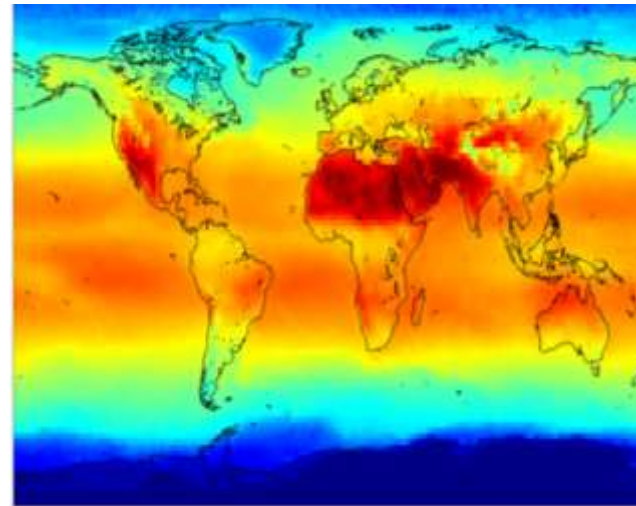
$$F_v = 2\pi \int_0^{\pi/2} L_v(\theta) \sin(\theta) \cos(\theta) d\theta$$

→ A spectrally resolved OLR (SR-OLR) retrieval algorithm from IASI radiances:

$$F_v = \frac{\pi L_v(\theta)}{R_v(\theta)}$$



Example of IASI integrated SR-OLR

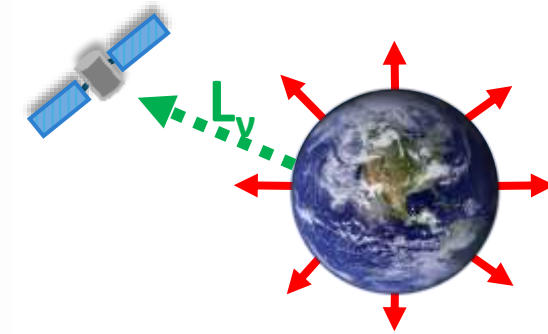


- For clear-sky scenes
- At the spectral sampling of IASI ( $0.25 \text{ cm}^{-1}$ )
- Units: ( $\text{W m}^{-2} (\text{cm}^{-1})^{-1}$ )

**Excellent fundamental climate data record**



# ve Radiation



## Spectrally Resolved Fluxes from IASI Data: Retrieval Algorithm for Clear-Sky Measurements

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### ABSTRACT

Space-based measurements of the outgoing longwave radiation (OLR) are essential for the study of Earth's climate system. While the CERES instrument provides accurate measurements of this quantity, its measurements are not spectrally resolved. Here we present a high-resolution OLR product (sampled at  $0.25 \text{ cm}^{-1}$ ), derived from measurements of the IASI satellite sounder. The applied methodology relies on precalculated angular distribution models (ADMs). These are usually calculated for tens to hundreds of different scene types (characterized by surface and atmosphere parameters). To guarantee accurate results in the range  $645\text{--}2300 \text{ cm}^{-1}$  covered by IASI, we constructed ADMs for over 140 000 scenes. These were selected from one year of CAMS reanalysis data. A dissimilarity-based selection algorithm was applied to choose scenes as different from each other as possible, thereby maximizing the performance on real data, while keeping the number of scenes manageable. A comparison of the IASI OLR integrated over the  $645\text{--}2300 \text{ cm}^{-1}$  range was performed with the longwave broadband OLR products from CERES and the AIRS instrument. The latter are systematically higher due to the contribution of the far infrared to the total IR spectral range, but as expected exhibit generally high spatial correlations with the IASI OLR, except for some areas in the tropical region. We also compared the IASI OLR against the spectrally resolved OLR derived from AIRS. A good agreement was found above  $1200 \text{ cm}^{-1}$  while AIRS OLR appeared to be systematically higher in the atmospheric window region, likely related to differences in overpass time or to the use of a different cloud detection algorithm.

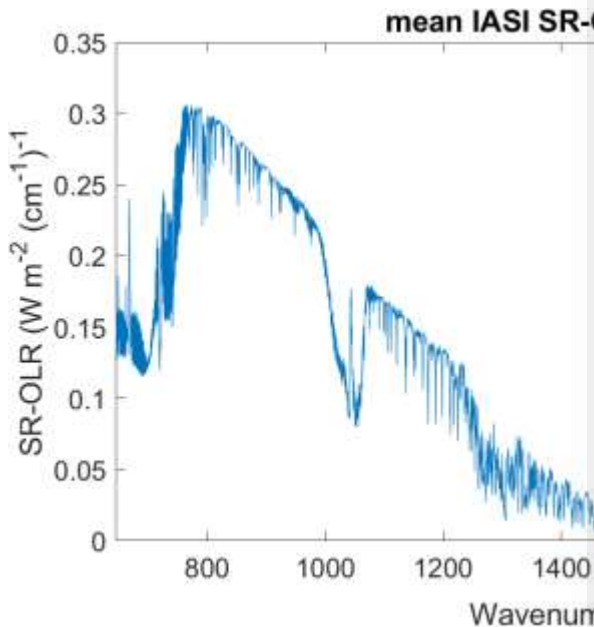
### 1. Introduction

The top-of-the-atmosphere (TOA) thermal flux, also referred to as Earth's outgoing longwave radiation (OLR) ( $\text{W m}^{-2}$ ), represents the total radiation emitted by the Earth-atmosphere system into space. As part of Earth's radiation budget, it reflects how the Earth-atmosphere system balances the incoming solar radiation at the top of the atmosphere and corresponds to about 2/3 of the total outgoing radiation, with the remaining 1/3 being reflected solar radiation (Trenberth et al. 2009). An accurate determination of the OLR is essential to improve our ability to model

Earth's climate system and to monitor climate feedbacks and processes (Brindley et al. 2015). Since the OLR is affected by clouds, aerosols, water vapor ( $\text{H}_2\text{O}$ ), and other greenhouse gases, this requires a good understanding of their impact on Earth's climate. Despite the numerous studies achieved in the last decades (e.g., Soden et al. 2005; Anderson et al. 2010; Y. Huang et al. 2010; Feldman et al. 2011), uncertainties up to a few percent remain, especially for net TOA integrated fluxes (e.g., Loeb et al. 2009; Trenberth et al. 2009; Stephens et al. 2012; Wild et al. 2014).

In principle, infrared sounding satellites are ideal for

→ A spectrally resolved



$$F_v = \frac{\pi L_v(\theta)}{R_v(\theta)}$$

For clear-sky scenes

At the spectral sampling of IASI  
( $0.25 \text{ cm}^{-1}$ )

Units: ( $\text{W m}^{-2} (\text{cm}^{-1})^{-1}$ )

Excellent fundamental  
climate data record

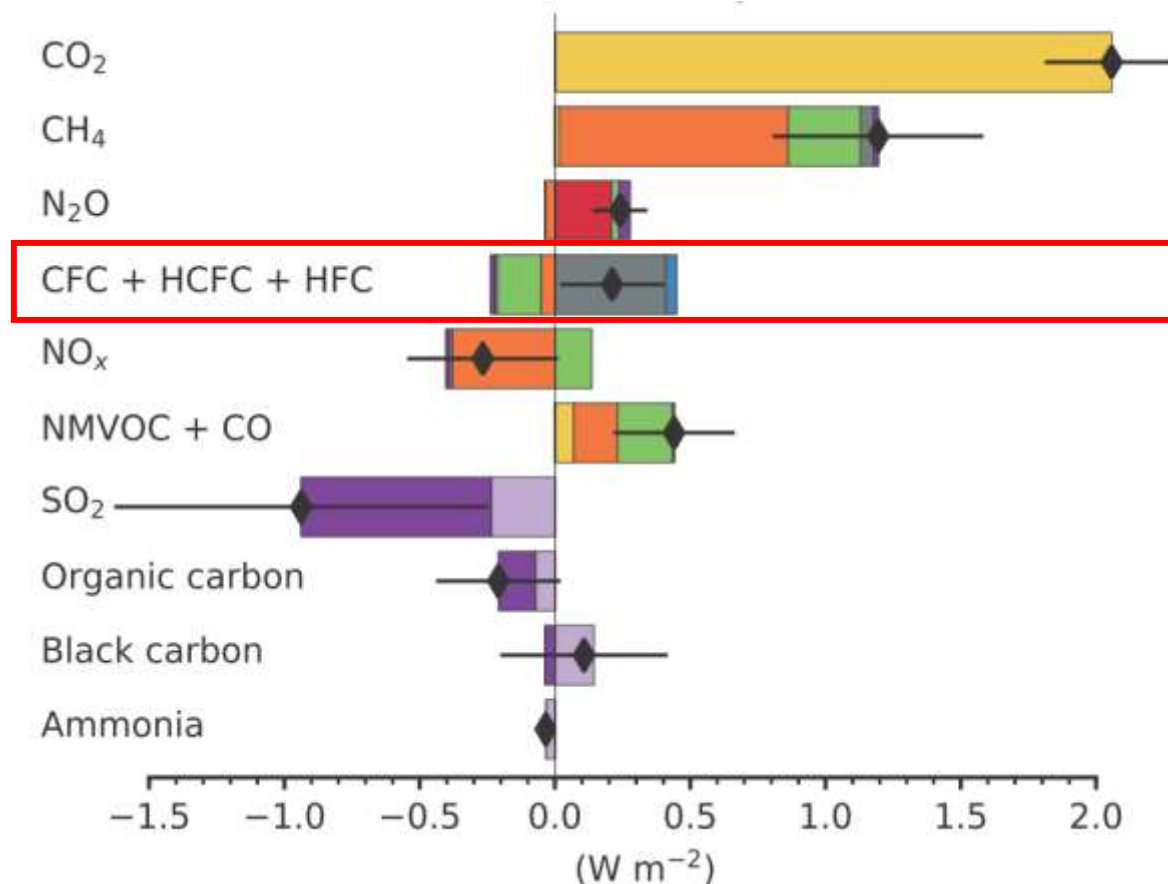
# Exploitation of the SR-OLR dataset:

**Instantaneous Radiative Efficiency (IRE) or forcing (IRF) of halogenated species.**

**IRE ( $\text{W m}^{-2} \text{ppbv}^{-1}$ )** = initial radiative flux imbalance in response to an imposed perturbation of a climate driver (e.g. change in [GHG]).

## Evaluation of the IRE:

- Today, mostly from radiative transfer model calculations for a few idealized atmospheres.
- Alternative approach directly from the changes in the SR-OLR.



IPCC AR6 WG1 (2021) – chapter 6

# IRE of halogenated species

## 1) Starting point:

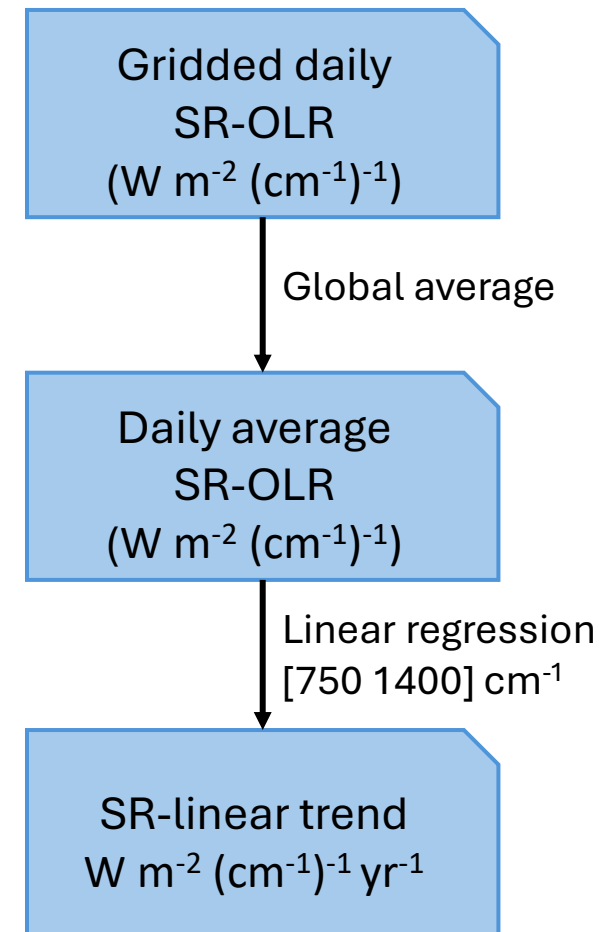
- Clear-sky daily SR-OLR ( $2^\circ \times 2^\circ$ )
- Between 2008 and 2022 (15 years)
- Between 750 and 1400  $\text{cm}^{-1}$  at  $0.25 \text{ cm}^{-1}$  sampling

## 2) Global daily average SR-OLR ( $\text{W m}^{-2} (\text{cm}^{-1})^{-1}$ )

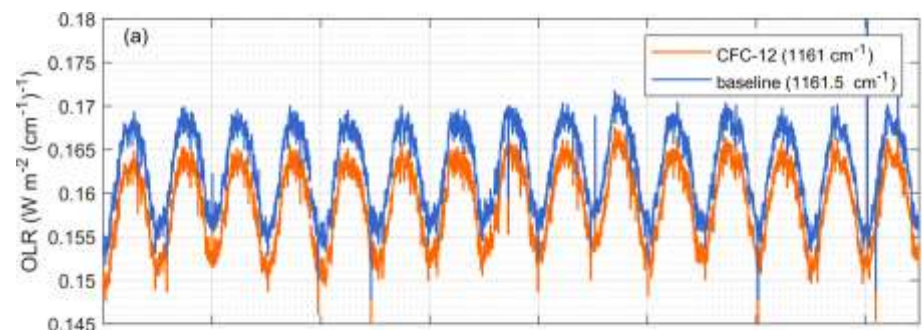
## 3) For each IASI channel: **Linear trend in the SR-OLR** ( $\text{W m}^{-2} (\text{cm}^{-1})^{-1} \text{ year}^{-1}$ )

### → Slope of the linear regression:

- **Surface temperature (baseline trend)**
- **Concentration of absorbing species**
  - ↘ [gas] → ↘ absorption → ↗ OLR (compared to baseline)
  - ↗ [gas] → ↗ absorption → ↘ OLR

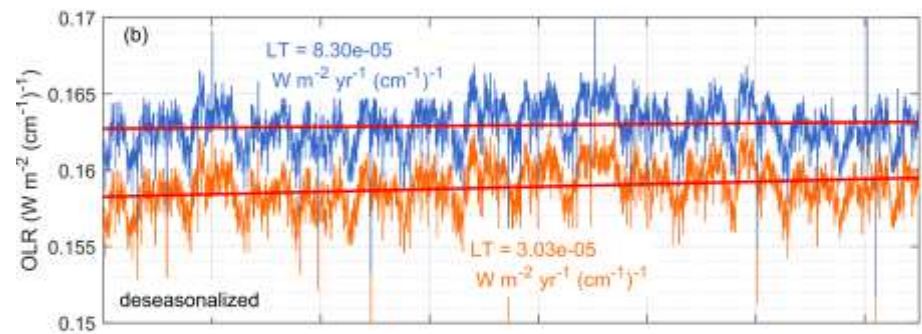


## Example: two IASI channels

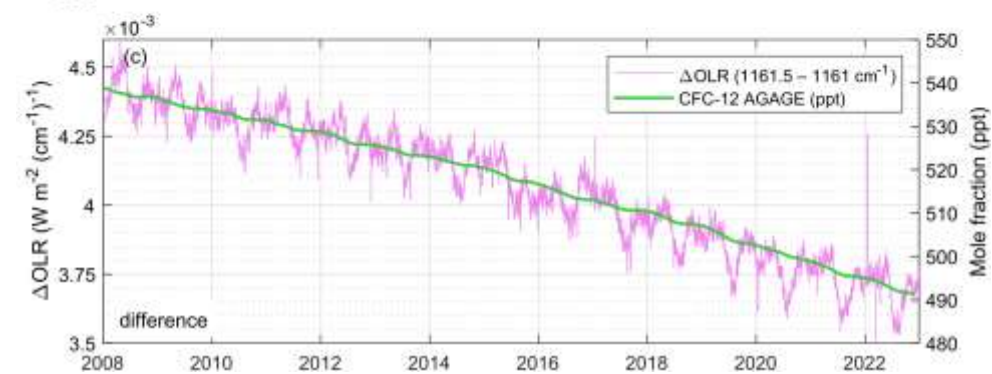


OLR time series

- Baseline channel
- CFC-12 channel



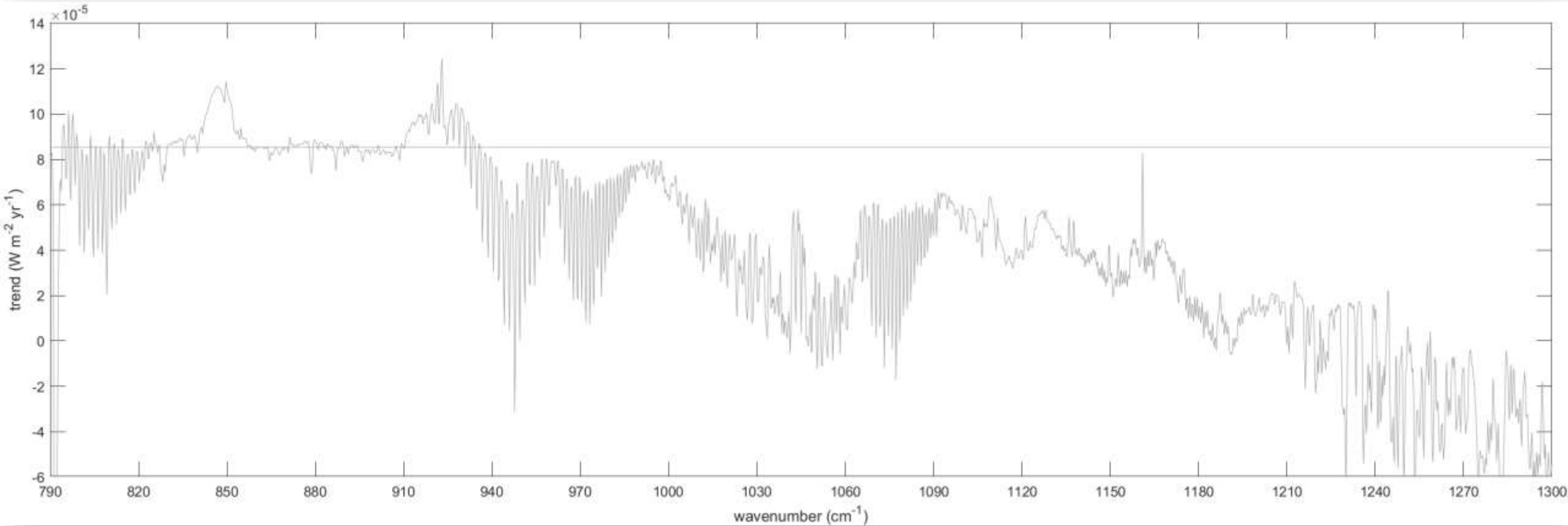
Deseasonalized



Difference

Slope of the linear regression for **each IASI channel**

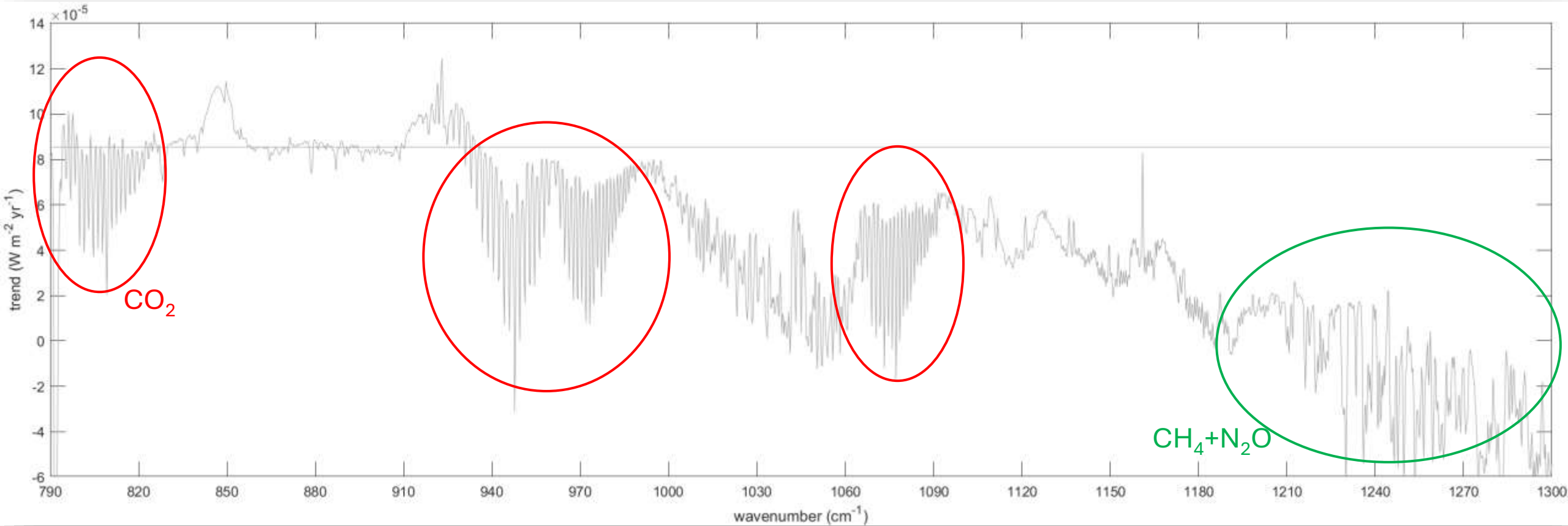
- ... from globally averaged daily SR-OLR
- ... between 2008 and 2022
- ... between 750 and 1400  $\text{cm}^{-1}$





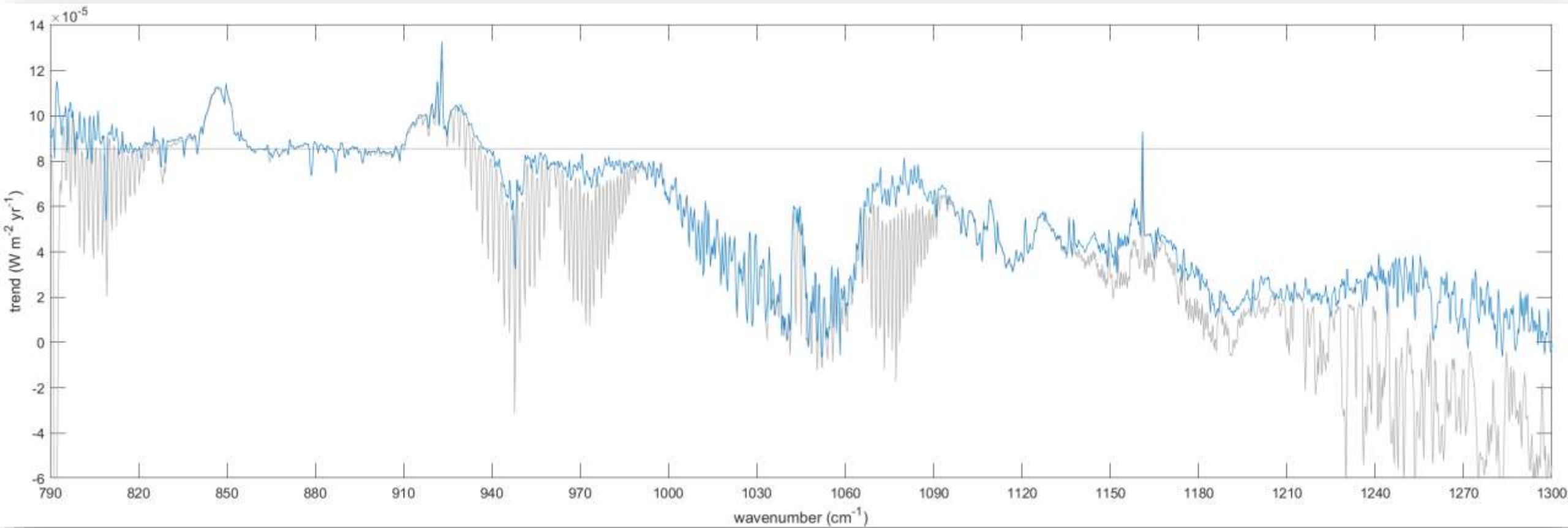
## Slope of the linear regression for each IASI channel

- ... from globally averaged daily SR-OLR
- ... between 2008 and 2022
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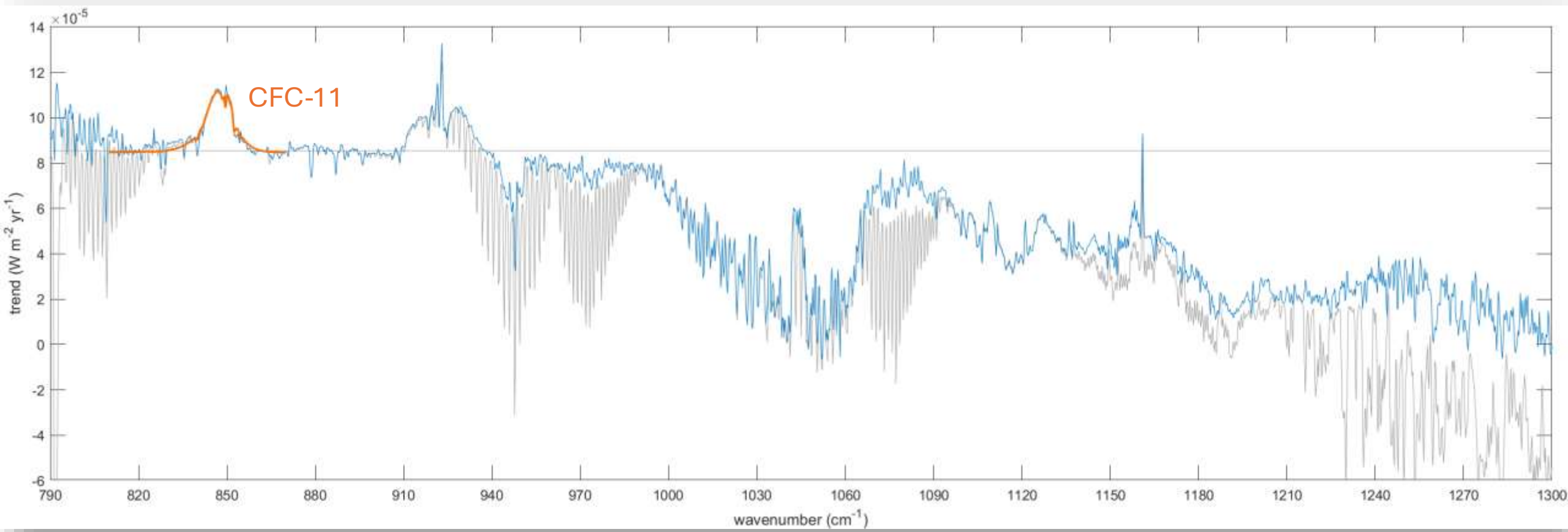




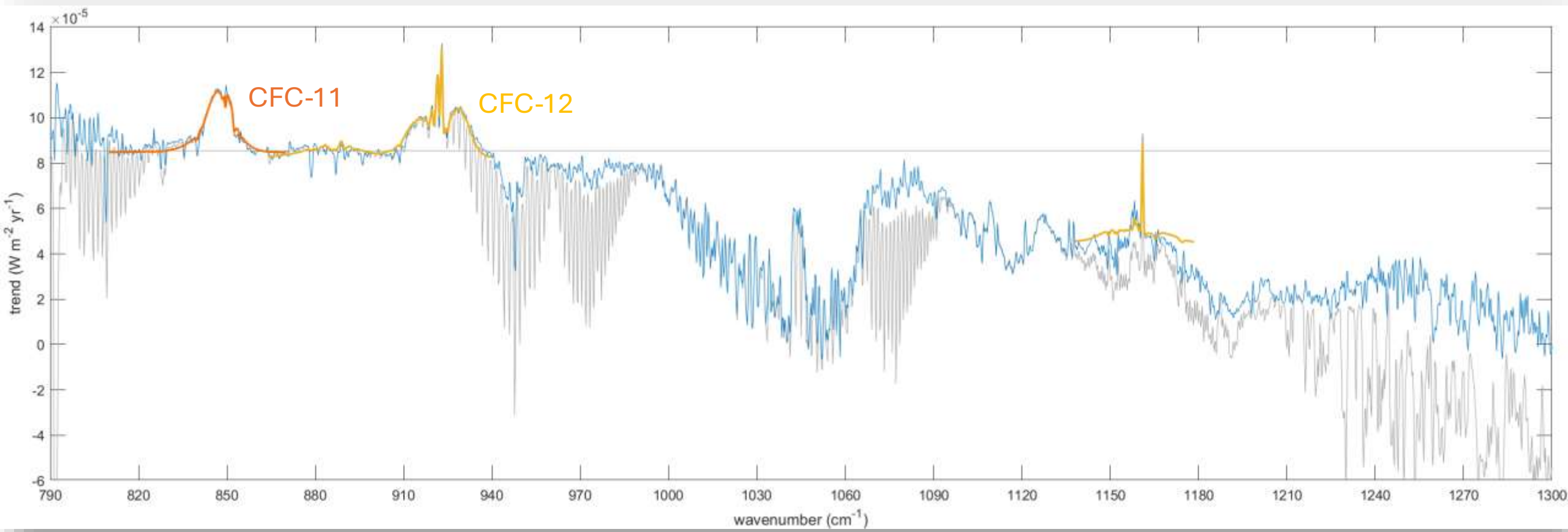
Fit  $\text{CO}_2 + \text{CH}_4 + \text{N}_2\text{O}$  signal and remove their contribution



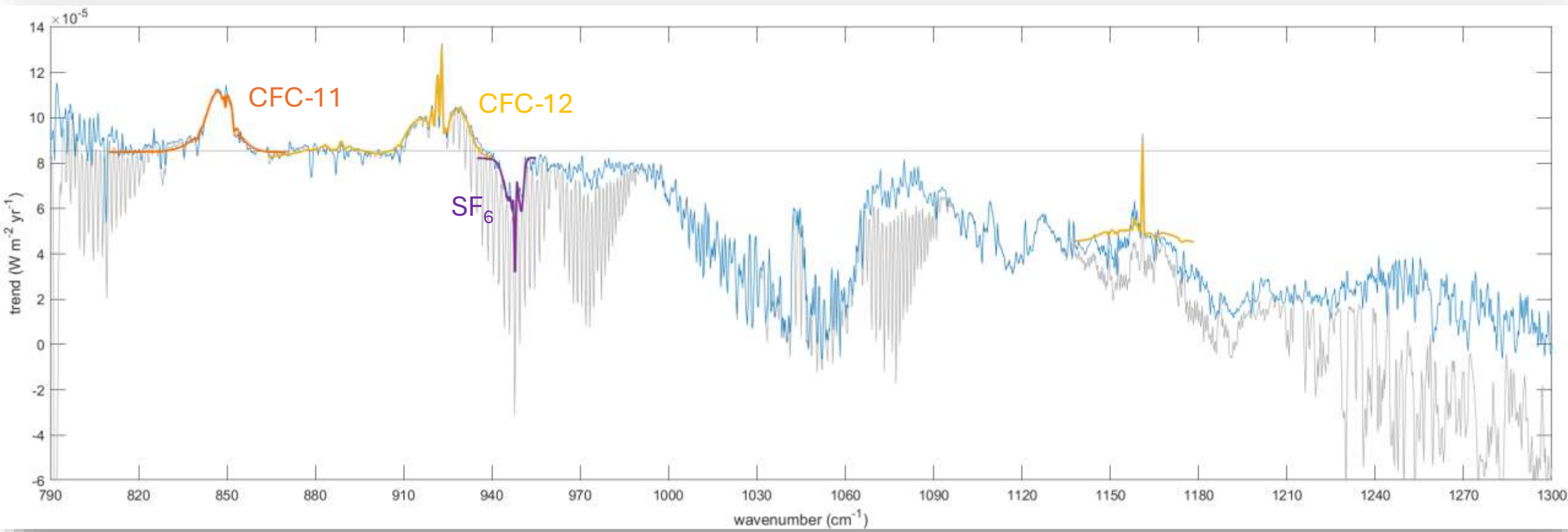
What do we observe ?



What do we observe ?

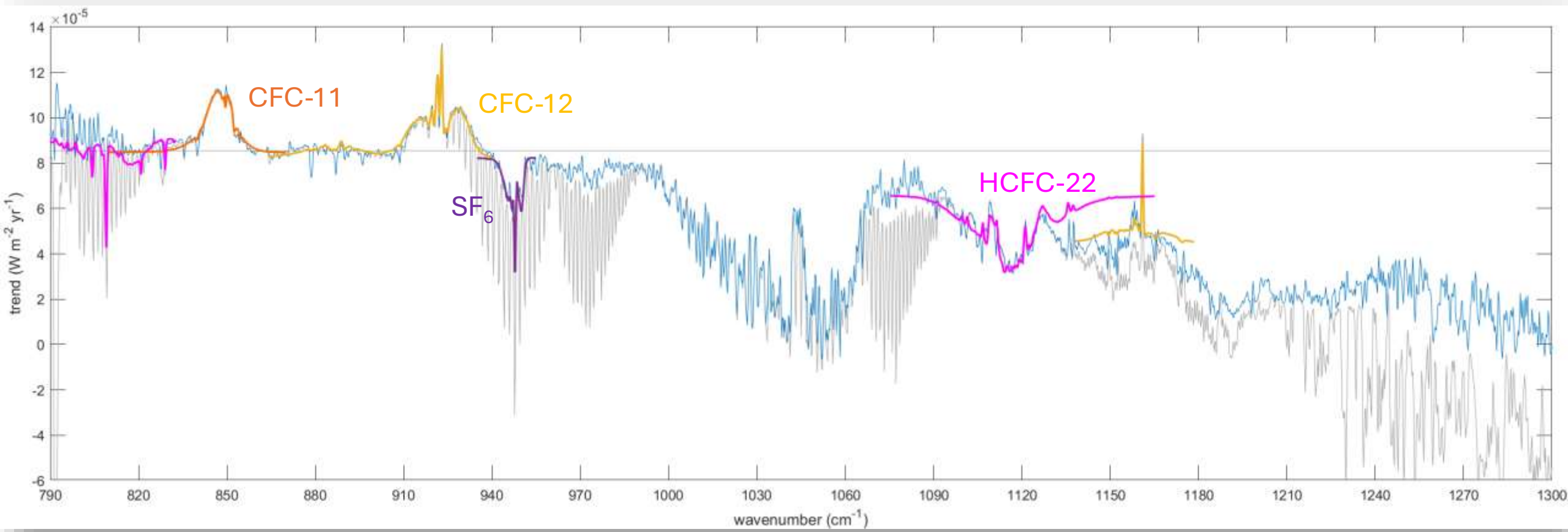


What do we observe ?

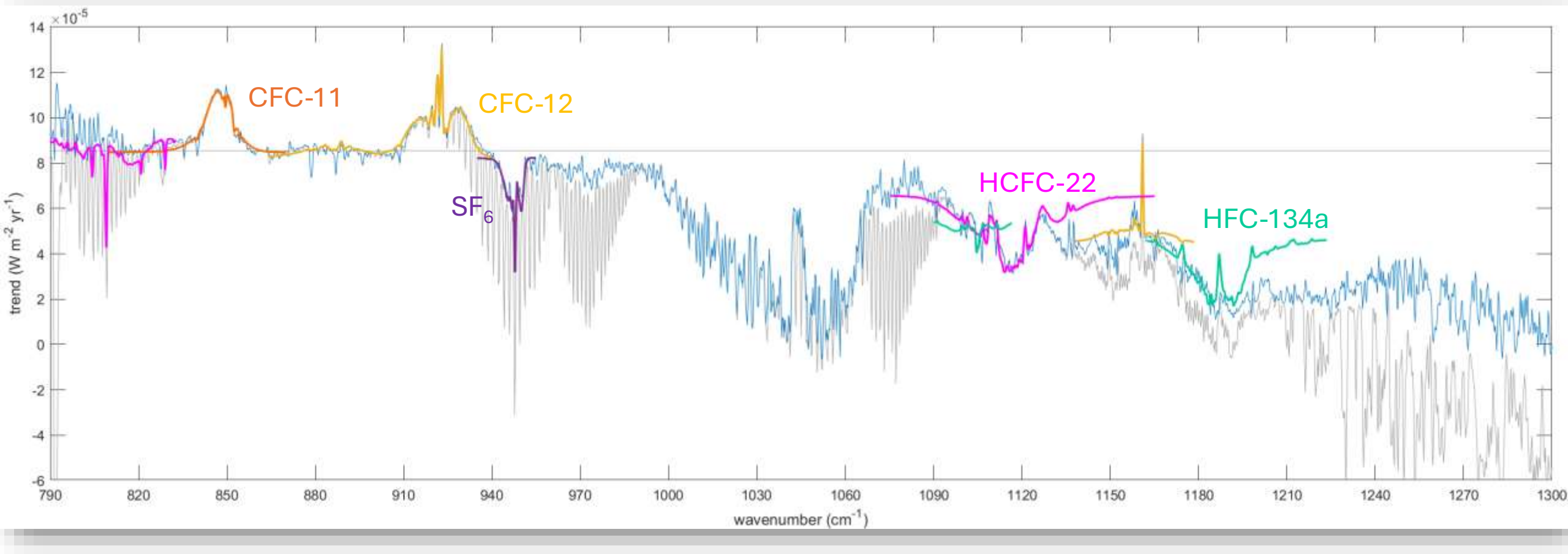




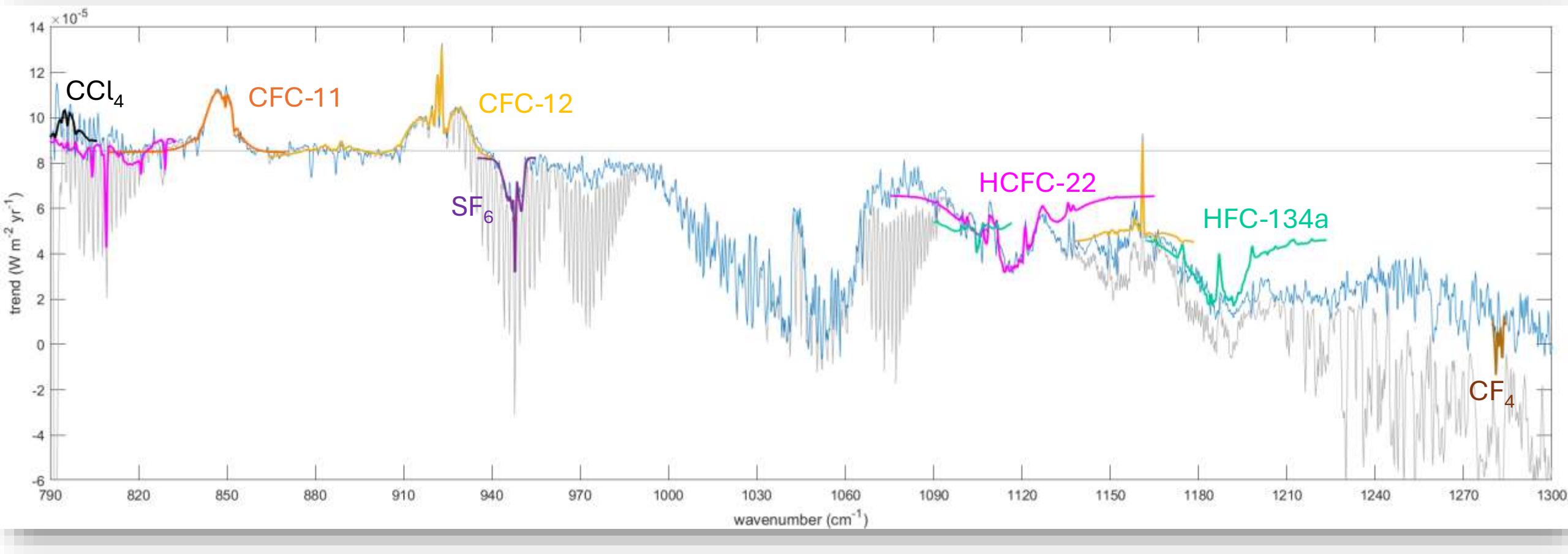
What do we observe ?



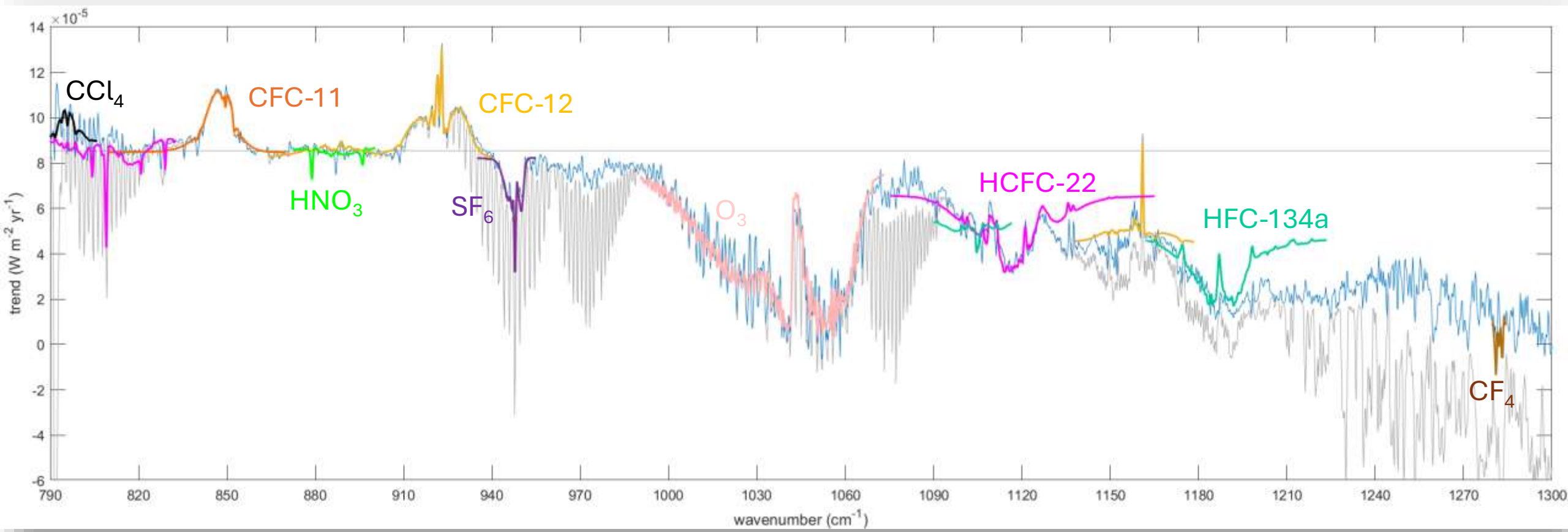
What do we observe ?



What do we observe ?



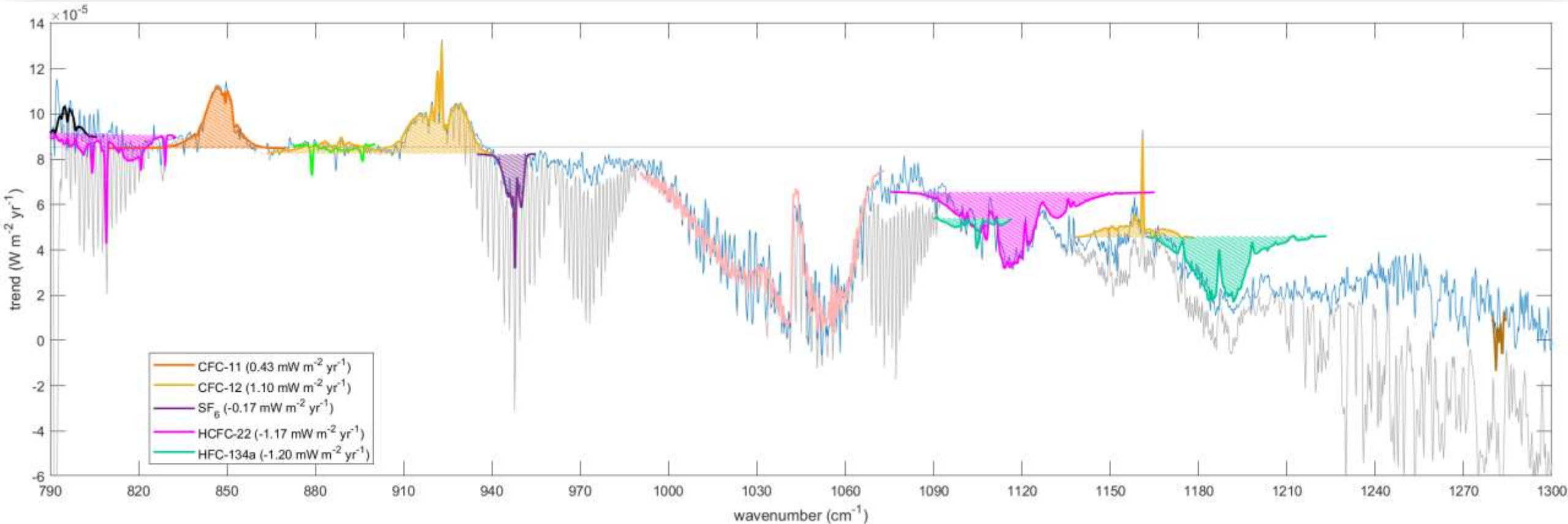
What do we observe ?





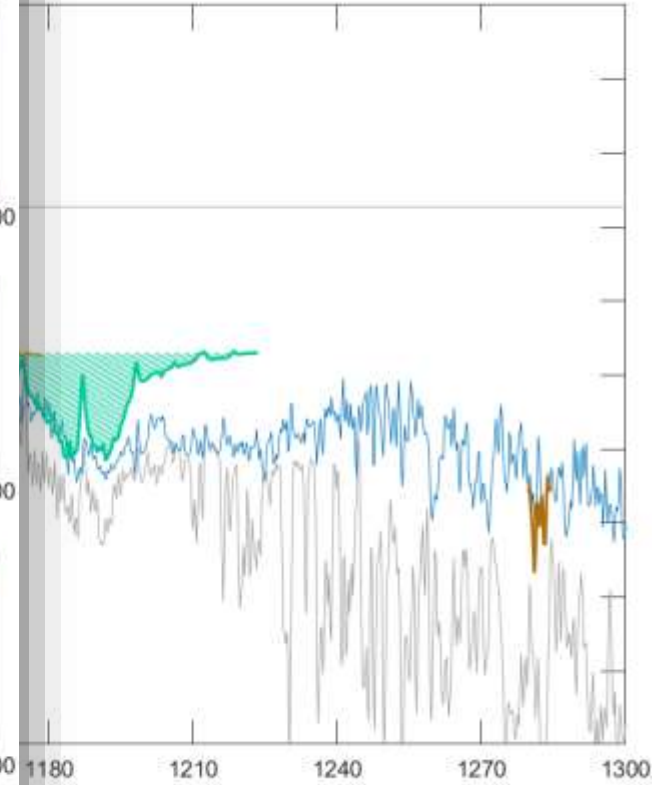
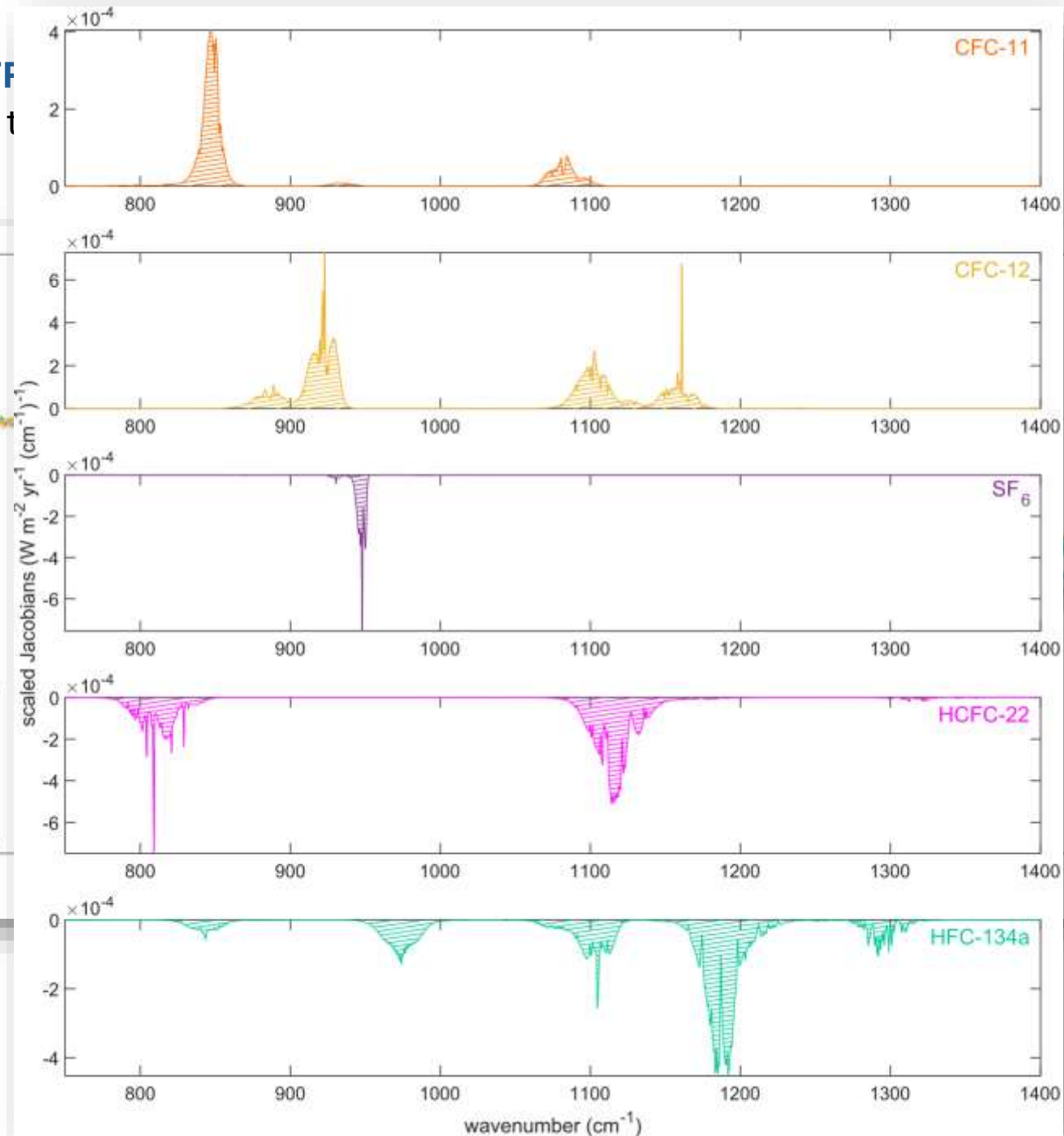
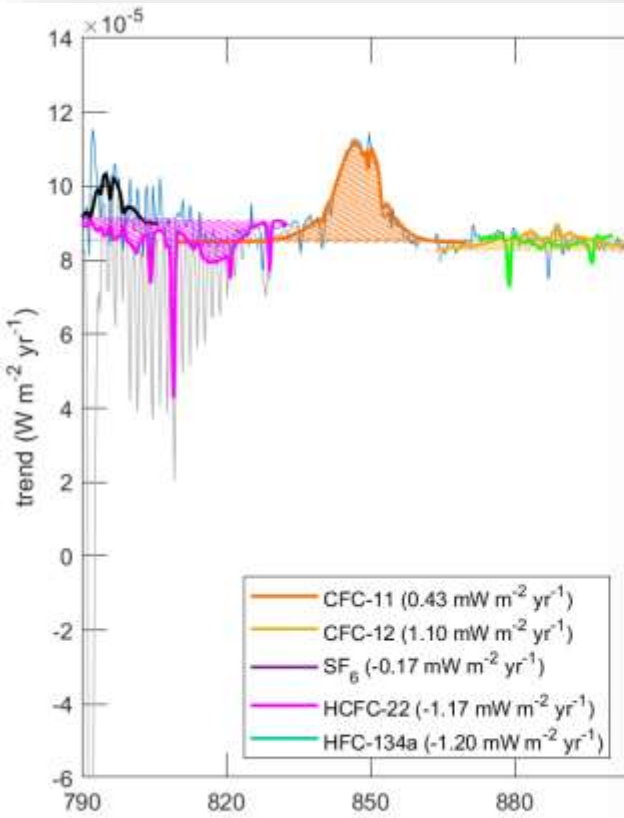
## Forcing rate of change (FRC, $\text{W m}^{-2} \text{yr}^{-1}$ ): CFC-11, CFC-12, $\text{SF}_6$ , HCFC-22 and HFC-134a

→ Fit, scale and integrate the Jacobian

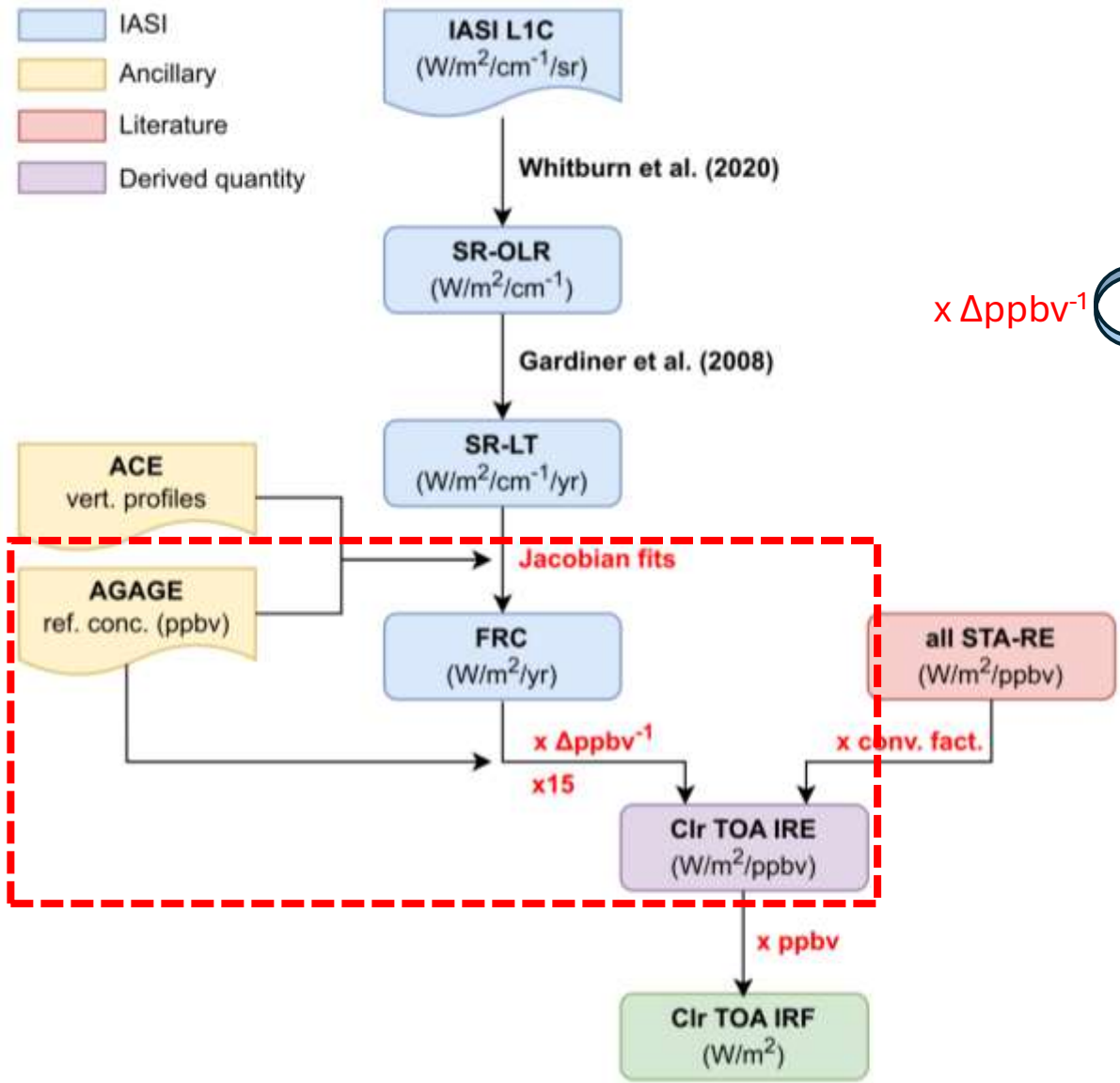


## Forcing rate of change (FR)

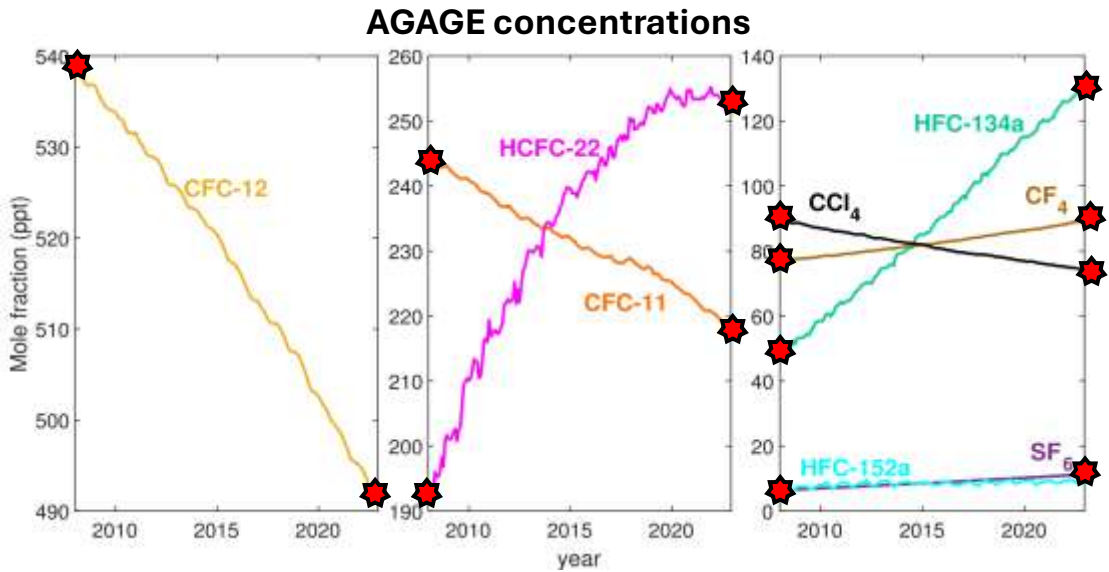
→ Fit, scale and integrate to



# From the FRC (W m<sup>-2</sup> yr<sup>-1</sup>) to the IRE (W m<sup>-2</sup> ppbv<sup>-1</sup>):



	CFC-11	CFC-12	SF <sub>6</sub>	HCFC-22	HFC-134a
AGAGE 2008 (ppt)	243.5	537.5	6.5	196.2	50.4
AGAGE 2022 (ppt)	219.2	493.0	11.1	253.4	128.2
FRC (mW m <sup>-2</sup> yr <sup>-1</sup> )	0.43	1.10	-0.17	-1.17	-1.20
IRE (W m <sup>-2</sup> ppbv <sup>-1</sup> )	0.27	0.37	0.57	0.31	0.23

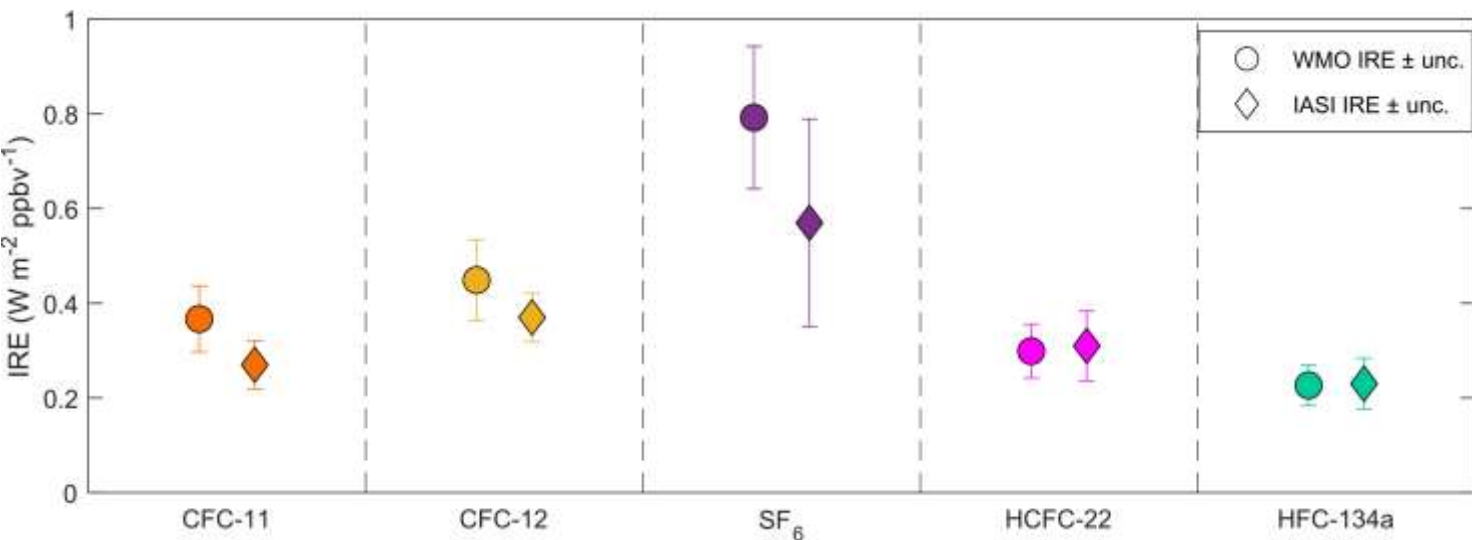


# Uncertainty budget

	CFC-11	CFC-12	SF <sub>6</sub>	HCFC-22	HFC-134a
Fitting range	5%	11%	21%	14%	12%
Jacobian (cross section)	14%	3%	24%	4%	10%
Jacobian (std. atm.)	4%	5%	4%	1%	5%
Jacobian (ref. year)	0%	0%	1%	0%	0%
Long-term T and H <sub>2</sub> O changes	0%	0%	0%	5%	5%
Methodology	11%	6%	21%	18%	16%
<b>Total (RSS)</b>	<b>19%</b>	<b>14%</b>	<b>39%</b>	<b>24%</b>	<b>23%</b>



# Comparison with modeled IRE



Literature:

- All sky
- Adjusted RE

→ Conversion to TOA IRE

	WMO O <sub>3</sub> assess. rep. All STA-RE (W m <sup>-2</sup> ppbv <sup>-1</sup> )	Conversion factor (K. Shine)	WMO O <sub>3</sub> assess. rep. Clr TOA IRE (W m <sup>-2</sup> ppbv <sup>-1</sup> )	IASI Clr TOA IRE (W m <sup>-2</sup> ppbv <sup>-1</sup> )
CFC-11	0.280 ± 0.039	1.31	0.367 ± 0.070	0.27 ± 0.05
CFC-12	0.330 ± 0.0462	1.36	0.422 ± 0.085	0.37 ± 0.05
SF <sub>6</sub>	0.574 ± 0.0804	1.38	0.781 ± 0.151	0.57 ± 0.22
HCFC-22	0.223 ± 0.0312	1.34	0.300 ± 0.057	0.31 ± 0.07
HFC-134a	0.173 ± 0.0242	1.31	0.227 ± 0.043	0.23 ± 0.05

## **Conclusions and perspectives**

- **Alternative approach** for deriving the IRE
- Based on the changes in the SR-OLR
- General good agreement with modeled IREs

### **Advantages:**

- no assumptions on the atmospheric state,
- no need for radiative transfer model calculations

### **Limitations:**

- Absorption in the window region
- Change in concentration

### **Perspectives:**

- IRE of new species with increasing time period of IASI
- IRE of other species outside the window region by fitting simultaneously the interfering species

## **Direct satellite measurements of the radiative forcing of long-lived halogenated gases**

S. Whitburn<sup>1,2,\*</sup>, L. Clarisse<sup>1</sup>, H. De Longueville<sup>1</sup>, P.-F. Coheur<sup>1</sup>, C. Clerbaux<sup>3</sup>, and A. Delcloo<sup>2,4</sup>

<sup>1</sup>Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing (SQUARES), Université libre de Bruxelles (ULB), Brussels, Belgium

<sup>2</sup>Royal Meteorological Institute of Belgium, Brussels, Belgium

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### **Abstract**

While the atmospheric concentrations of ozone-depleting chlorofluorocarbons (CFCs) are gradually declining following regulatory measures, the levels of other halogenated compounds, such as hydrochlorofluorocarbons (HCFCs) and sulfur hexafluoride (SF<sub>6</sub>) continue to rise or are only just starting to stabilize. Most of these halogenated substances are potent greenhouse gases. Their radiative efficiency, which quantifies their impact on the climate, has until now only been estimated indirectly by means of models. Here, we report the clear-sky instantaneous radiative efficiencies (IRE) of CFC-11, CFC-12, SF<sub>6</sub>, HCFC-22 and HFC-134a estimated for the first time directly from experimental data. This is achieved by combining trends observed in 15 years (2008–2022) of spectrally resolved infrared radiance fluxes from the Infrared Atmospheric Sounding Interferometer (IASI) satellite sounder, with concentrations measured from ground and space. Comparisons with literature-reported values point to biases of the order of 1 to 31%. The most significant discrepancies are for CFC-11 and SF<sub>6</sub>, with our estimates being 31% and 28% lower, respectively.