



The Long-Lived greenhouse gas Products Performances (LOLIPOP) CCI+ project

Bianca Maria Dinelli and the LOLIPOP team

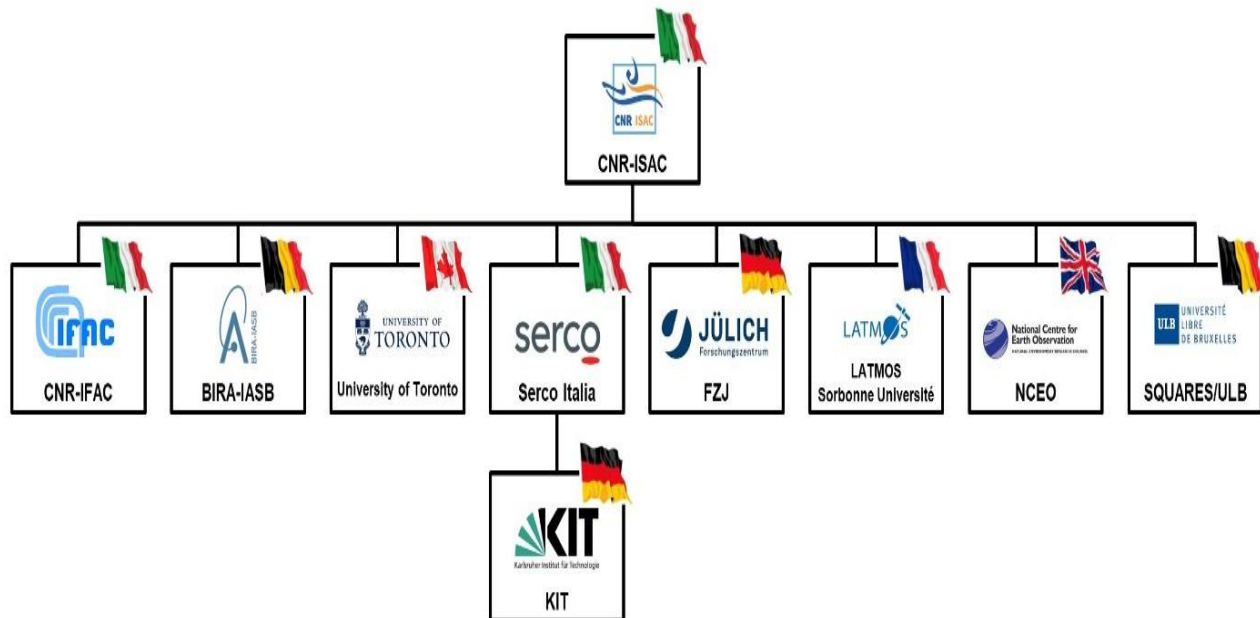
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- For a complete understanding of Earth's climate, it is essential to understand the full basket of atmospheric gases that exhibit a large global warming potential or a strong impact on the ozone layer.
- ESA's Climate Change Initiative is already generating robust satellite-based timeseries for the greenhouse gases water vapour, carbon dioxide (CO₂) and methane (CH₄). However, several **Other Long-Lived GreenHouse Gases (OLLGHGs)** warrant systematic observation, in particular, nitrous oxide (N₂O) and halogenated carbon compounds (CFCs, HFCs, HCFCs, PFCs) are also considered by GCOS as Essential Climate Variables (ECVs, GCOS-defined Essential Climate Variables).
- These gases have **long atmospheric lifetimes**, exhibit **significant global warming potentials** and provide a major contribution to the radiative forcing uncertainty estimates. Nitrous oxide and chlorine-containing OLLGHGs are also the main source of the **anthropogenic ozone depletion** and are regulated internationally under the 1989 UN Montreal Protocol.
- **Several satellite instruments provide information on the atmospheric abundance and distribution of the OLLGHGs** (Envisat MIPAS, Scisat ACE-FTS, MetOp IASI, and Aura HIRDLS). They can provide a valuable multi-mission resource for monitoring and understanding the role OLLGHGs in the atmosphere over the last two decades providing complementary information to the ground-based in-situ monitoring networks.
- To foster the full exploitation of these satellite data, in November 2023 ESA has started the **Long Lived greenhouse gases PrOducts Performance (LOLIPOP)** project.

- The goal of the project is to determine if the actual set of satellite measurements is good enough to be used in climate science and services so that the construction of a harmonized and consistent dataset of satellite measurements can go ahead.
- If this is not the case, the final goal is to suggest actions to be taken to either improve the quality of satellite measurements of the OLLGHGs (through new retrieval techniques applied to existing satellite missions) or to develop dedicated satellite missions for their monitoring.

- Perform an **analysis of the state of the art** of the satellite measurements of the OLLGHGs.
- Perform an **analysis of the users' needs**, to which the OLLGHGs project is intended to respond.
- Perform a **thorough quantitative and qualitative assessment of satellite measurements of the OLLGHGs**, with the aim of providing users with guidance on which data sets would be best for their particular needs and application.
- **Investigate the benefit of the use of satellite observations** in several end user applications through three case studies;
- **Disseminate the project results** to the user community and foster the discussions between data users and data producers;
- **Organise a user workshop** to collect feedback from the users on the work performed in the project;
- **Provide suggestions for future work** on the OLLGHGs, prioritising the actions to be taken, building on the outcome of the whole project.



Scientific and Project Coordination

• **CNR-ISAC:** B. M. Dinelli and E. Castelli **with the support of Serco Italia** : M. Cardaci, G. Brizzi and M. Valeri

Retrieval team

• **CNR-IFAC:** P. Raspollini, M. Gai

• **LATMOS-SU:** C. Clerbaux

• **SQUARES-ULB:** P. Coheur, L. Clarisse and S. Whitburn

• **U. Toronto:** K. Walker

• **KIT:** G. Stiller

Validation team

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U. Toronto: K. Walker

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• **NCEO:** M. Chipperfield, and J. Harrison

• **CNR-ISAC:** F. Fabiano

ESA Technical Officers:

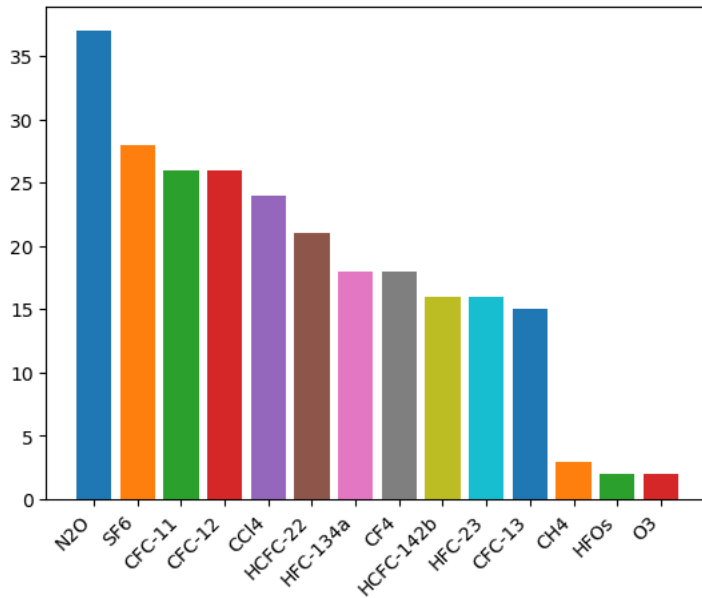
Simon Pinnock

Claire McIntosh

- A review of the user needs has been performed through both a literature review and a questionnaire distributed among the possible users
- The literature review highlighted that
 - **there are three main groups of applications** that will benefit from and improved OLLGHGs dataset:
Climate model applications, Chemistry-climate models, Emission inversion
 - There is the need to monitor the OLLGHGs concentrations for climate change mitigation
- In climate models the OLLGHGs are used as aggregate with a vertical distribution that is prescribed and tuned according to sparse surface measurements
- The distributed questionnaire revealed that the user requirements do change according to the application and the examined gas

From the survey: the two communities: climate/chemistry modelling and chemistry/emissions/transport studies have different needs

Priority list



Requirements related to applications

- 1) Horizontal resolution:
 - low (few degs lat/lon) for climate applications and general chemistry modeling;
 - very high (0.1 deg) for emission/transport studies
- 2) Vertical resolution/range: < 3 km (~ 1 km).
 - Should at least include the troposphere and stratosphere, the higher the better.
- 3) Similar Accuracy requirements: 10% (2-5%). Stricter requirement for N₂O (up to ~0.1 ppb).

An inventory of the available datasets from limb and nadir satellite measurements has been performed for **11 OLLGHGs**: N₂O, CFC-11, CFC-12, CFC-113, CF₄, HCFC-22, HCFC-142b, HFC-23, HFC-134a, SF₆, CCl₄.

- The reported datasets are well consolidated and most of them are available online. Almost all datasets have been validated or inter-compared with other measurements.
- In some cases, different datasets are produced exploiting the measurements of the same instrument (i.e., for MIPAS there are MIPAS-ESA and MIPAS-IMK).
- Several sparse measurements before 2002 were found. For some of them only Level 3 data (daily averages) are available, for other (the oldest ones), only Level 1 data are available.

The document is available on-line in the LOLIPOP web site

<https://climate.esa.int/en/projects/long-lived-greenhouse-gas-products-performances-lolipop/>

Inventory of satellite products – the document



It is an interactive document

For each instrument there is:

A Summary Table

Contains information on

- the type of product,
- the observation geometry
- the temporal and spatial coverage
- the characterization of the product
- the data format.

| | | | | | | | | | | | | | |
|-------------|-----|---|-----|------|----|------|-----|------|--|------|-----|--------------------|--|
| lolipop cci | | ESA Climate Change Initiative "Plus" (CCI+) | | | | | | | | | | Page 16 | |
| | | OLLGHGs INVENTORY (URD) | | | | | | | | | | [D1.1] LOLIPOP_URD | |
| | | | | | | | | | | | | Version 1.0 | |
| | | | | | | | | | | | | 15/03/2024 | |
| N2O | CFC | | | HCFC | | | HFC | | | CCl4 | SF6 | | |
| | 11 | 12 | 113 | CF4 | 22 | 142b | 23 | 134a | | | | | |

2.4 MIPAS-ESA/ENVISAT

| MIPAS-ESA N ₂ O products | |
|-------------------------------------|---|
| Product type | VMR profile |
| Level 2 processor | Optimised Retrieval Model |
| Data version | L2 V8.22 |
| Reference | https://doi.org/10.5194/amt-14-7975-2021 ; https://earth.esa.int/eogateway/documents/20142/37627/README_V8_issue_1_0_20201221.pdf |
| Geometry | Limb |
| Temporal coverage | 2002-2012 |
| Spatial coverage | Global |
| Horizontal resolution | It depends on the meas. modes, around 400-500 km for NOM |
| Vertical resolution | About 4 km up to 30 km, slow degradation with altitude above |
| Useful vertical range | 6-60 km |
| Spectroscopic database | Spectroscopic Database: HITRAN_mipas_p4.45 is based on HITRAN08 (Rothman et al., 2009), but spectroscopic parameters for the molecules O ₂ , SO ₂ , OCS, CH ₃ Cl, C ₂ H ₂ and C ₂ H ₆ are taken from HITRAN 2012 (Rothman et al., 2012). |
| Spectral range | Microwindows in 1140.725-1291.95 cm ⁻¹ |
| Product characterization | Random error (and CM), systematic error, AK |
| Data Format | NetCDF |
| Contact | Piera Raspollini |
| Data download | https://hm-atmosds.eo.esa.int/oads/access/collection/EnvisatMIPASL2PS |
| Recommendation | - |

THE INSTRUMENT

Short description in section [INSTRUMENTS](#) → [MIPAS short description](#)

DATA OVERVIEW

The MIPAS level2-v8 database, along with the values of tangent pressures, temperatures, and VMR profiles of all the retrieved molecules, includes also some important products that can be used as diagnostic tools to characterise the quality of the reported results. Among them, the averaging kernels, the covariance matrices that map the random measurement noise onto the solution, and a few quality flags. All the products are stored in NetCDF files.

The L2 V8.22 dataset is described in: <https://doi.org/10.5194/amt-14-7975-2021>. The algorithm used for the reprocessing is described in: <https://doi.org/10.5194/amt-15-1871-2022>.

| | | | | | | | | | | | | | |
|-------------|-----|---|-----|------|----|------|-----|------|--|------|-----|--------------------|--|
| lolipop cci | | ESA Climate Change Initiative "Plus" (CCI+) | | | | | | | | | | Page 17 | |
| | | OLLGHGs INVENTORY (URD) | | | | | | | | | | [D1.1] LOLIPOP_URD | |
| | | | | | | | | | | | | Version 1.0 | |
| | | | | | | | | | | | | 15/03/2024 | |
| N2O | CFC | | | HCFC | | | HFC | | | CCl4 | SF6 | | |
| | 11 | 12 | 113 | CF4 | 22 | 142b | 23 | 134a | | | | | |

CONCLUSIONS AND VALIDATION

Results of the validation with MIPAS balloon in Wetzel et al., 2022: <https://doi.org/10.5194/amt-15-6669-2022>.

Results of the validation vs ACE-FTS and ground-based measurements reported in the readme file: https://earth.esa.int/eogateway/documents/20142/37627/README_V8_issue_1_0_20201221.pdf.

The comparison results show a globally (without the Antarctic) and vertically consistent MIPAS N₂O V8 bias of about 5 % positive and a similar spread, meaning that median differences are at the edge of being significant. The V8 (and V7) N₂O bias is slightly reduced with respect to the V5 and V6 bias results in the full resolution period, yet at the cost of a small bias increase in the optimised resolution period. Note however that the smoothed difference profile shape does not seem to be in agreement with the MIPAS balloon comparisons (also at Kiruna). The large comparison uncertainties moreover make it difficult to detect seasonal dependences or trends. Positive bias for N₂O (10 %–20 %) below 35 km (within combined systematic errors); especially N₂O pronounced for N₂O in the lowermost stratosphere around 15 km. Somewhat larger positive deviations also in the tropics around 30 km.

MIPAS exhibits a significant positive bias of about 5% with respect to the ground-based FTIR measurements. In the comparison to the balloon observation, this positive bias is even more pronounced reaching values typically between 10 and 20%. This holds for both MIPAS observation periods (FR and OR mode) and different geographical regions.

FILTERING AND DATA QUALITY

The quality of the retrieved profiles is determined on the basis of four criteria, two providing information on the successful convergence of the retrieval iterations, one on the capability of the retrieval to reproduce the measurements, and one on the presence of outliers in the retrieval error.

To provide an easy way to remove unreliable data, a final post-quality flag, summarising the outcome of the four quality criteria, is reported in the output files.

Take all profiles with `post_quality_flag=0`.

DATA AVAILABILITY

The data are available after registration at <https://doi.org/10.5270/EN1-c8hgqx4> (European Space Agency, 2021). Their utilisation is subject to ESA's Earth Observation Terms and Conditions.

The information has been divided into two types of files: a standard one and an extended one. The standard files, one for each orbit and retrieved species, contain the information commonly required by the data users. Its filetype label is "2PS", and it is compliant with the Climate and Forecast convention (CF-1.6, Eaton et al., 2011) and with the Attribute Convention for Data Discovery (ACDD-1.3, ESIP, 2015). Extended files, identified by the filetype label "2PE", are also provided for each species and each orbit. They are "thought" for diagnostics and for advanced users, who need complete information about the retrieval process. This includes the full state vector (retrieved profiles, atmospheric continuum, and instrumental offset), along with the full CM and AKM, and additional information about the retrieval.

Other information

Information on

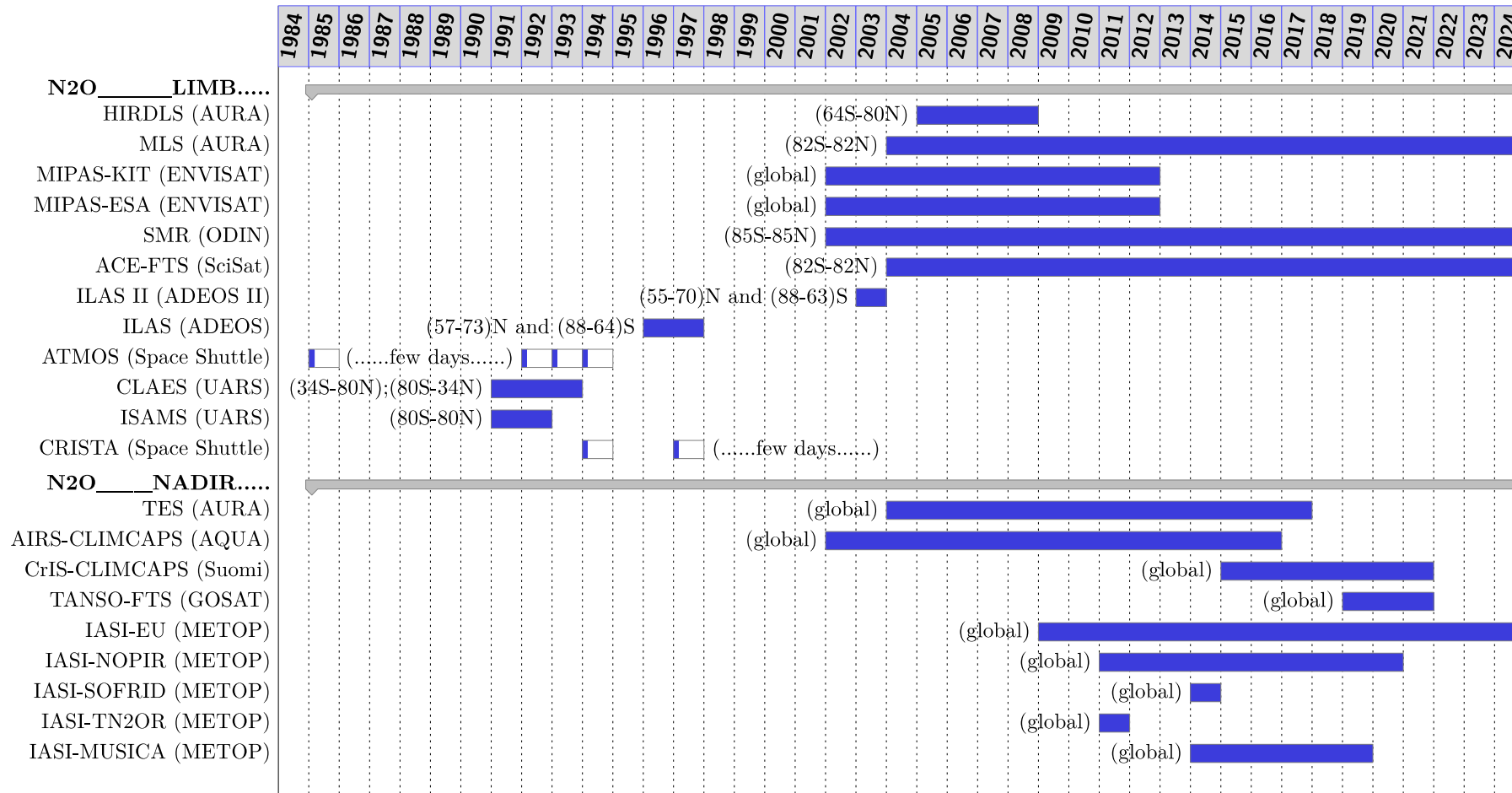
- the instrument
- The dataset validation
- The quality of the data
- Availability of the data

The document is available on-line in the LOLIPOP web site

<https://climate.esa.int/en/projects/long-lived-greenhouse-gas-products-performances-lolipop/>



Inventory of satellite products: tables – N₂O

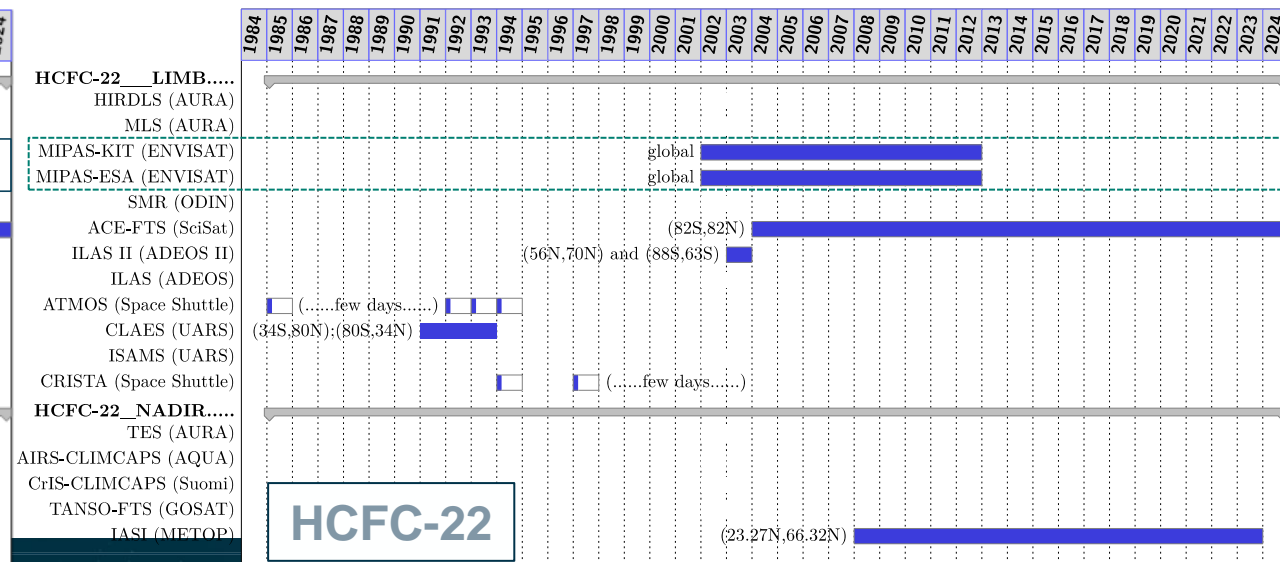
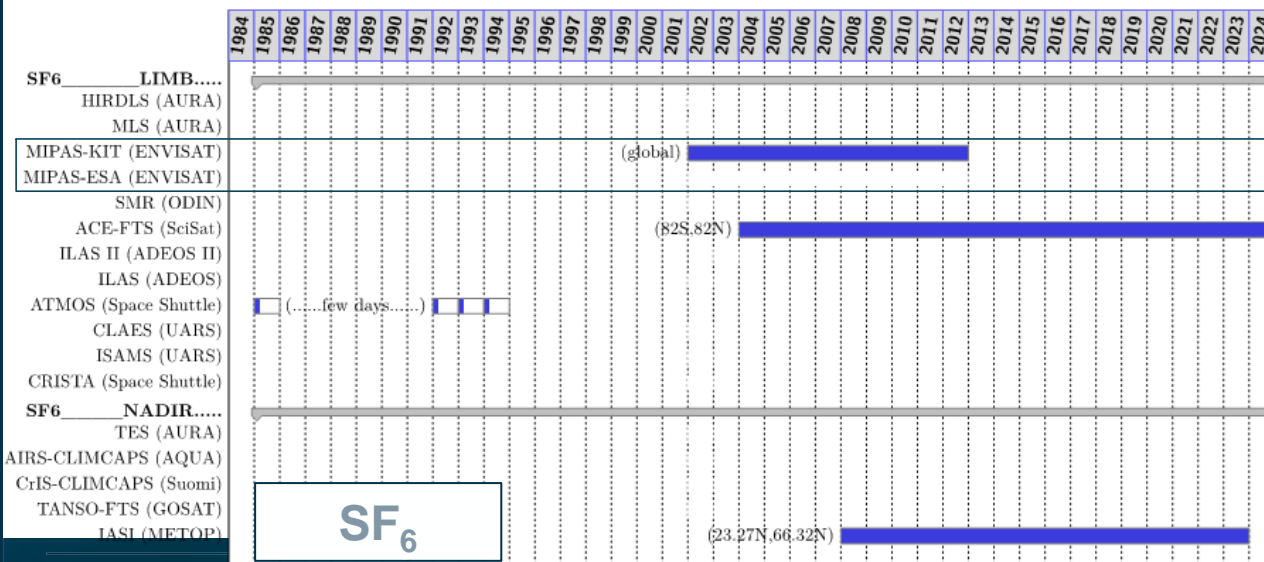
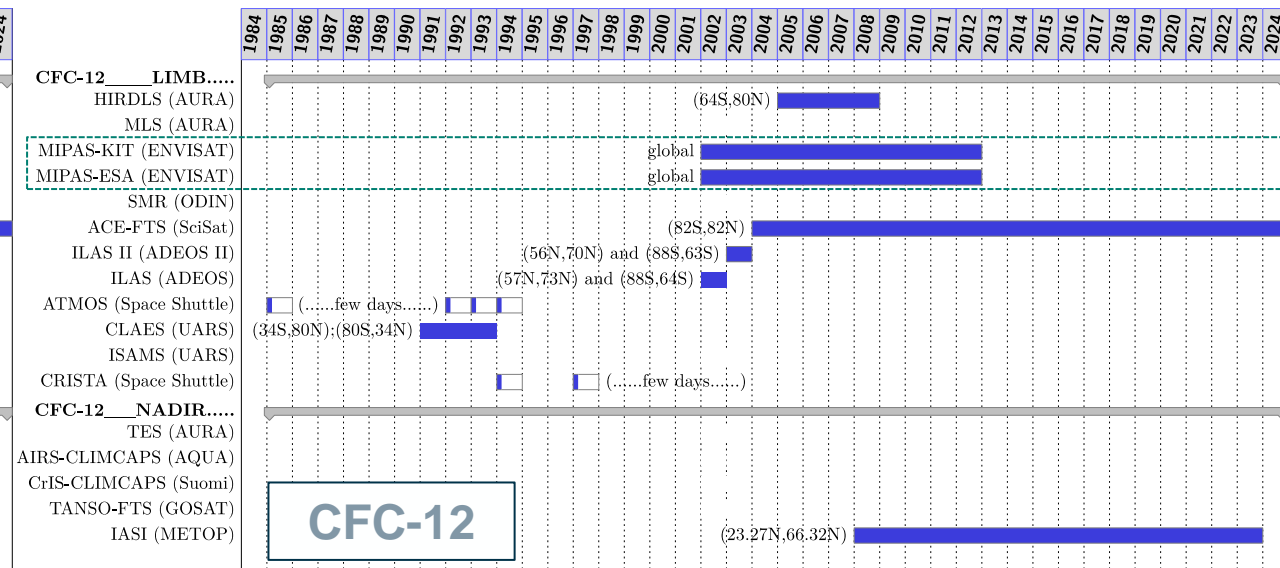
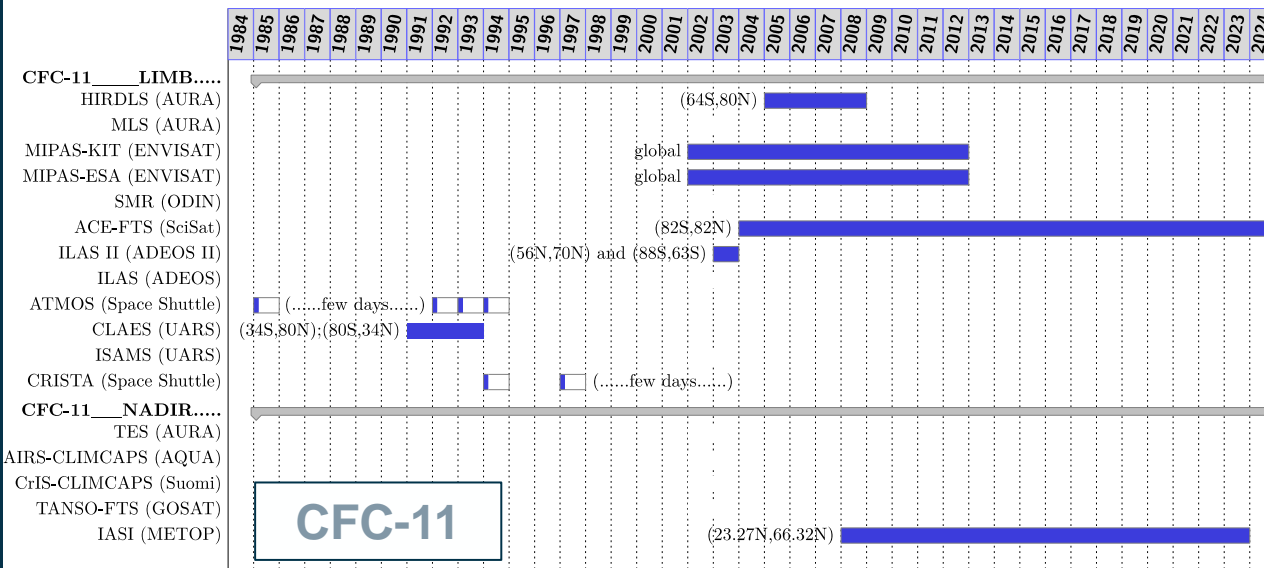


VMR profiles for limb observations

Total/partial columns or VMR profiles for nadir observations



Inventory of satellite products

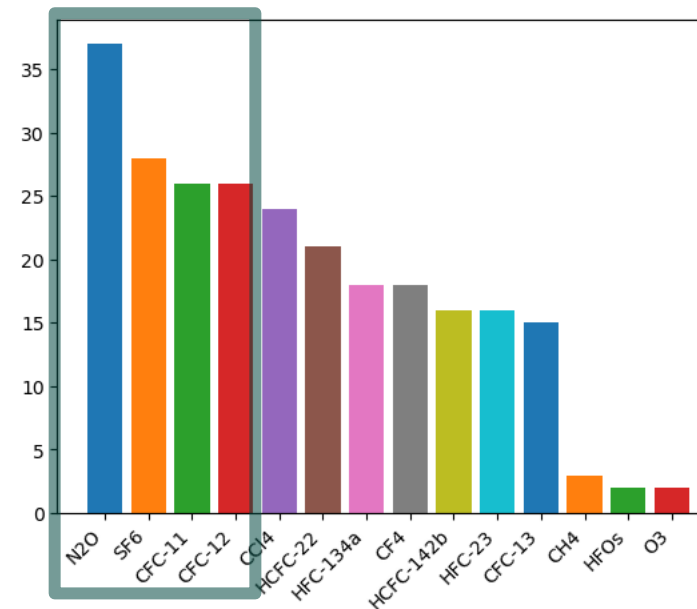


Selection of the products to harmonize and validate



- Based on the outcomes of the literature review, users' needs and satellite products inventory, a selection of the data to be included in the homogenization and validation exercise has been performed.
 - a priority list of the species of interest has been compiled
 - Higher priority has been given to products with an elevated level of maturity.
- As a result, the species N_2O , SF_6 , CFC-11 and CFC-12, retrieved from both limb and nadir measurements, have been selected for the harmonization and validation exercise
- Main focus on the data measured after 2002

Priority list from questionnaire



The products to be harmonised and validated have been divided into two sets: nadir and limb.

- Harmonization will be done converting all the data into a common format
- Validation will use an “as uniform as possible” approach. Collocation criteria:
 - Nadir :100 km/500 km within 2 hours
 - Limb : 500km within 6 hours
- Metric: average bias, scatter (standard deviation on the biases), correlation, long-term stability and seasonality
- Reference data
 - Nadir : Ground based stations and networks NDACC – TCCON – AGAGE
 - Limb : Balloon borne measurements: Solar Occultation (MkIV Balloon FTIR spectrometer) – limb emission (FTIR spectrometers MIPAS-B and GLORIA-B) – In-situ (HIPPO)
- For limb observations only – intercomparison among satellite datasets

Satellite dataset available: NADIR and LIMB

NADIR

N₂O:

TES, AIRS-CLIMCAPS, CrIS-CLIMCAPS, TANSO-FTS FOCAL, IASI_EUMETSAT, IASI_NOPIR, IASI_SOFRID, IASI_TN2OR, IASI_MUSICA

CFCs:

IASI_ULB

Large variability in data content and vocabulary

N₂O

HIRDLS (Aura): 2005-2008
MLS (Aura): 2004-present
MIPAS (ENVISAT) 2002-2012 (ESA and IMK-IAA)
SMR (Odin): 2002-present
ACE-FTS (SCISAT): 2004-present

LIMB

Halogenated compounds

HIRDLS: **CFC-11, CFC-12**
MIPAS: **CFC-11, CFC-12, CFC-113***, **HCFC-22, CCl₄, SF₆***, **CF₄** (*IMK-IAA only)
ACE-FTS: **CFC-11, CFC-12, CFC-113, HCFC-22, HCFC-141b, HCFC-142b, HFC-23, HFC-134a, CCl₄, SF₆, CF₄**

Individual formats in the LOLIPOP cluster



Unified Data format

Directory structure

File format

Vocabulary and attributes

| name | lat-lon | Profile/Total Column | AVK |
|----------|---------|----------------------|-----|
| TES | Y | P + TC | Y |
| CLIMCAPS | Y | P | N |
| FOCAL | Y | TC | N |
| EUMETSAT | Y | TC | N |
| NOPIR | Y | P + TC | Y |
| SOFRID | Y | P + TC | Y |
| TN2OR | Y | P | Y |
| MUSICA | Y | P | Y |
| ULB | N* | TC | N |

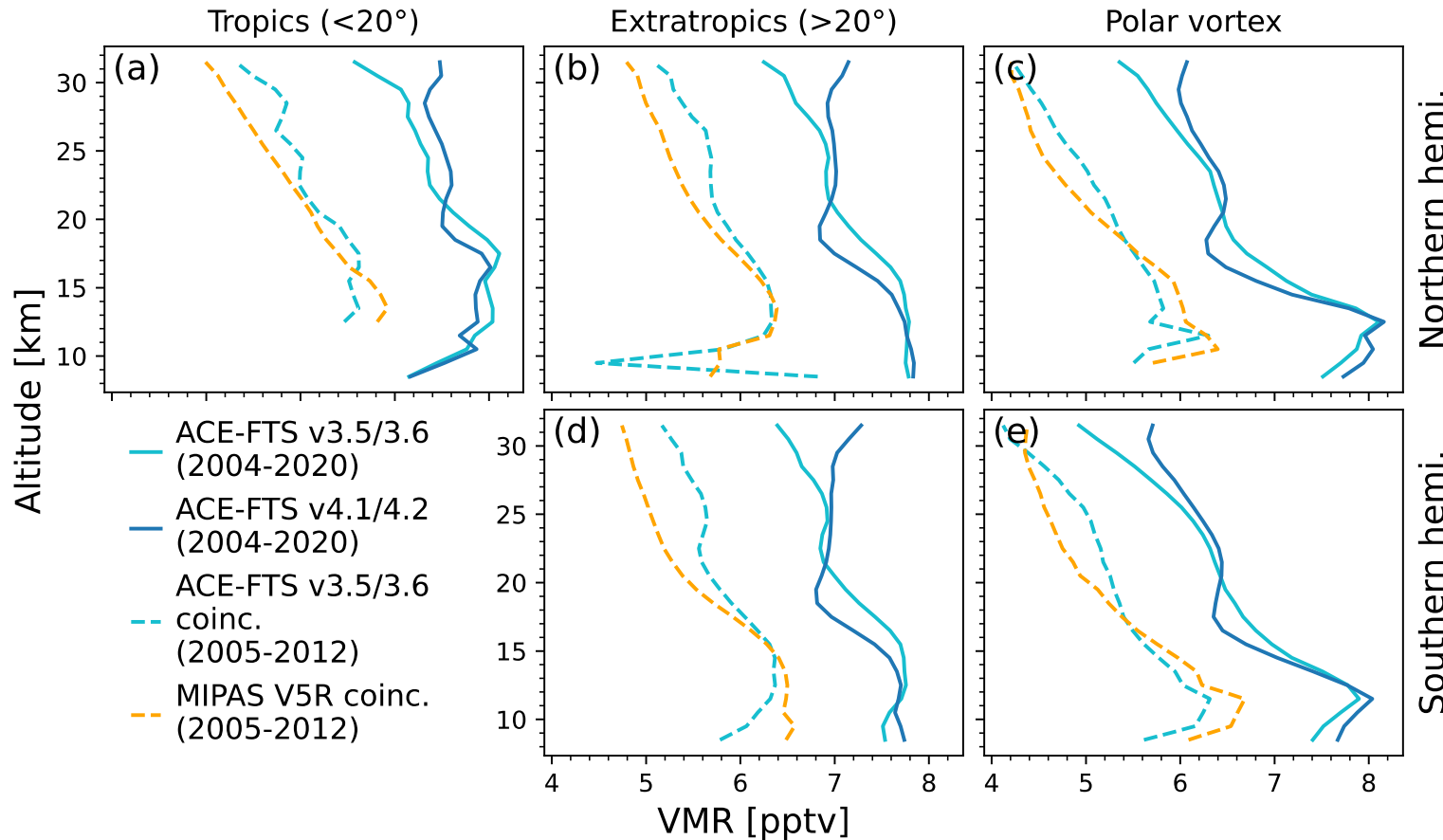
* Average over single large domain

- Convert the actual content so that the quantity of interest matches with that of the reference data
- Harmonize and Convert file format so it abides by CCI standards

Validation of LIMB data – some examples



EXAMPLE: ACE-FTS/MIPAS SF₆ COMPARISONS



Compare two versions of ACE-FTS with each other and with coincident (dashed lines) MIPAS profiles

- MIPAS-ACE within 12 h / 1000 km and on same side of polar vortex edge.
- ACE-FTS (2004-2020) compare same occultations.

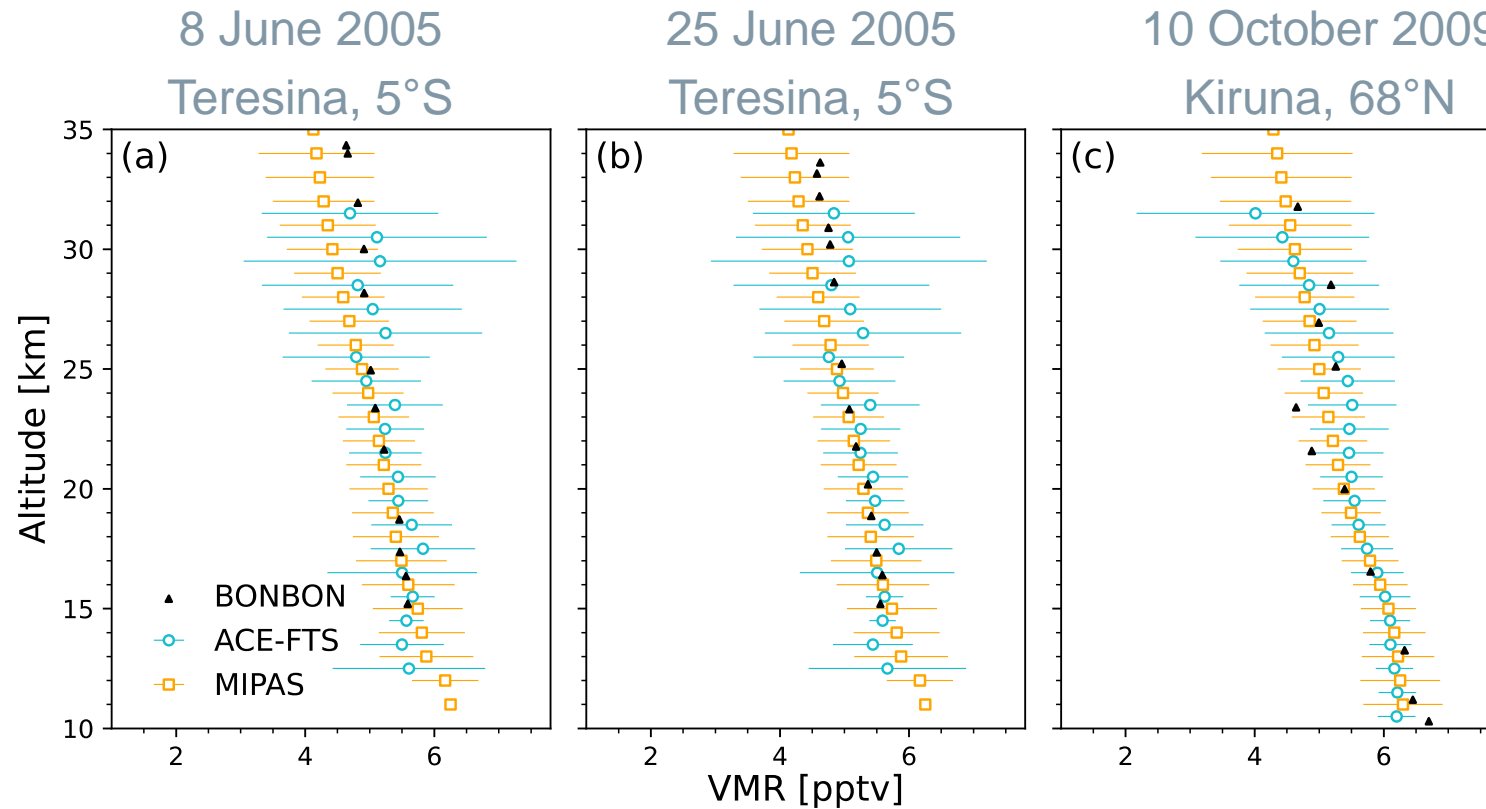
Polar vortex defined as all measurements with an absolute scaled potential vorticity greater than $1.6E-4 \text{ s}^{-1}$.

Increase in SF₆ over time can be seen in ~1 pptv difference between 2005-2012 and 2004-2020 comparisons.

Validation of LIMB data – some examples



EXAMPLE: ACE-FTS/MIPAS SF6 COMPARISONS WITH IN SITU BALLOON MEASUREMENTS



Compare ACE-FTS and MIPAS profiles from within 90 days and 5° latitude of the BONBON measurements.

Error bars represent one standard deviation of satellite profiles.

The main target of the user case studies is to demonstrate the potential benefits of an improved dataset of OLLGHGs satellite observations for the users in the climate and atmospheric modelling research community.

Three user case studies have been selected:

1. Sensitivity of historical climate model simulations to the OLLGHG climatology
2. Study of the radiative forcing of OLLGHG **Not yet started**
3. Monitoring of stratospheric chlorine levels and their impacts on ozone recovery

Case Study 1: Sensitivity of historical climate model simulations to the OLLGHG climatology

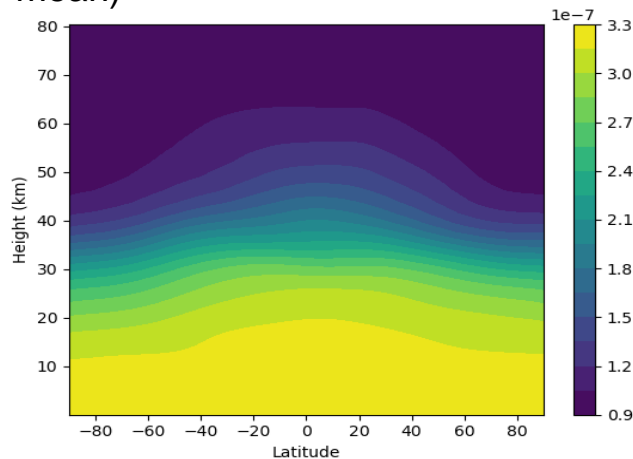


What is the impact of an updated latitude-height climatology of minor GHGs in climate models?

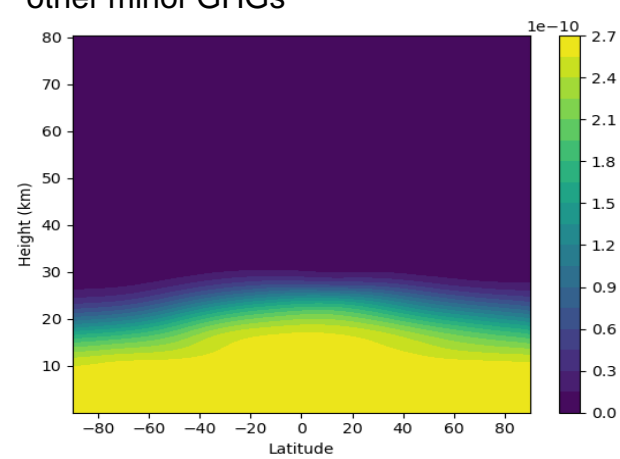
Objectives:

- Evaluate the **sensitivity of the simulated climate** to changes in the distribution of minor GHGs
- **Implement an updated climatology** of GHGs in the EC-Earth climate model

N₂O climatology in EC-Earth3 (annual mean)



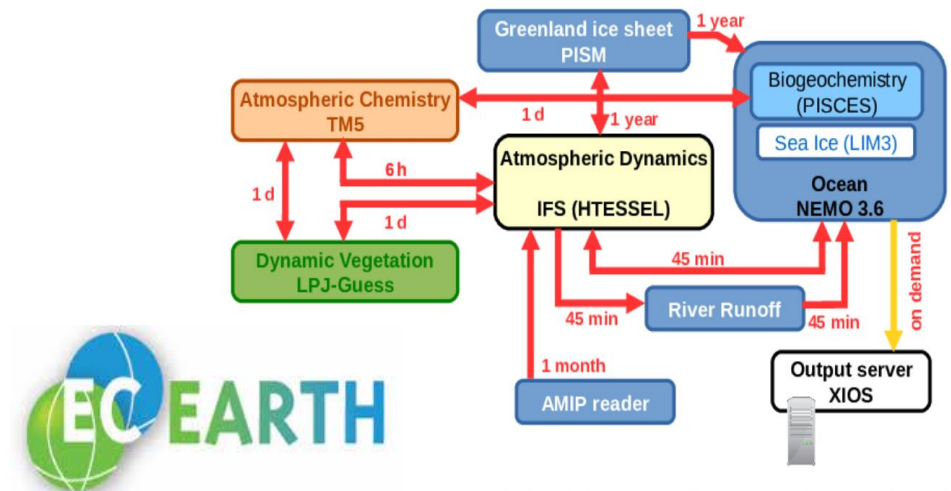
Reference CFC-11 climatology in EC-Earth3 (annual mean), used also for CFC-12 and other minor GHGs



EC-Earth3 (4) is an Atmosphere-Ocean Coupled General Circulation Model, which includes:

- atmospheric model IFS cy37 (cy48);
- ocean model NEMO v3.6 (v4);
- Sea-ice model LIM2 (SI3).

EC-Earth participated in the last climate model intercomparison project **CMIP6**



Case study 2: Study of the Radiative Forcing of OLLGHGs

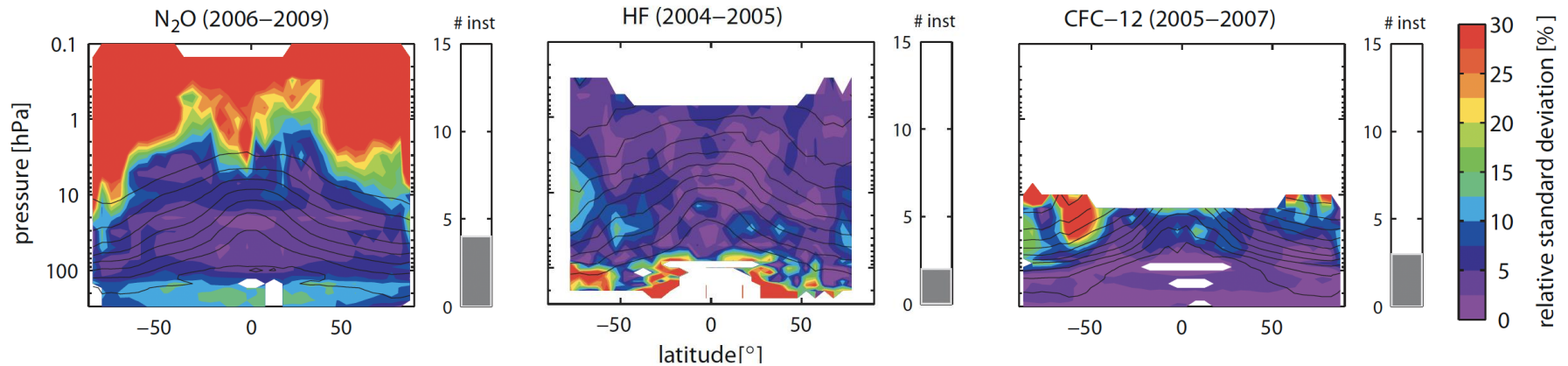
Radiative forcing (RF) refers to the amount of energy imbalance in the Earth's atmosphere caused by changes in the concentration of greenhouse gases, aerosols, and solar radiation.

Not yet started

Objectives:

- Estimate the RF of long-lived GHGs using an off-line radiative transfer model, SOCRATES
- Quantify uncertainties in RF due to measurement uncertainties, uncertainties in long-term trends, and the use of vertically resolved versus spatially uniform distributions of long-lived GHGs.

Figure:
Uncertainties in key long-lived GHGs as derived from available satellite limb sounders.



SPARC Report No.8, SPARC Data Initiative (Eds.Hegglin & Tegtmeier) 2017

SOCRATES = Suite Of Community Radiative Transfer codes based on Edwards and Slingo radiative transfer model (Edwards and Slingo, 1996)

Case study 3: Monitoring of stratospheric chlorine levels and their impact on ozone recovery



National Centre for
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NATURAL ENVIRONMENT RESEARCH COUNCIL



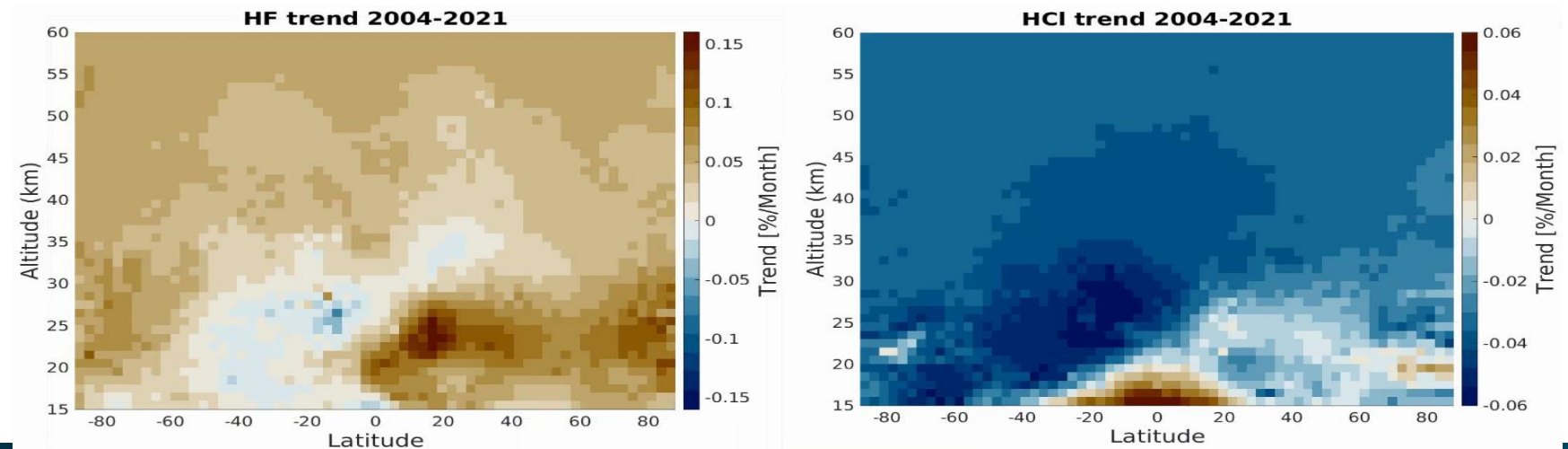
How have stratospheric chlorine levels changed over recent decades and how have these affected ozone recovery?

The team will use a combination of direct observations (mainly **ACE-FTS**), atmospheric modelling (**TOMCAT**, www.see.leeds.ac.uk/tomcat) and machine-learning-based data assimilation (**ML-TOMCAT**, Dhomse et al., 2021).

Objectives:

- Compare ACE-FTS, TOMCAT, ML-TOMCAT chlorine datasets in order to investigate differences and better understand biases in the models.
- Calculate trends in chlorine OLLGHGs (source gases), reservoir, and total Cl_y in order to determine implications for stratospheric ozone.
- Assess how much stratospheric O_3 has changed (increased/recovered) due to the decreasing halogen source species.

A first look at trends from ML-TOMCAT



The LOLIPOP project is in its first year of operations

The first objective (provide the state of the art of OLLGHGs satellite measurements and of the user requirements) has been met and the documentation is available on the project web site

The harmonization and validation of the OLLGHGs datasets (for both Nadir looking and Limb looking instruments) just started

Two of the three case studies are ongoing, one will start in November

A user workshop will be organised next year

We hope to succeed in demonstrating that the examined OLLGHGs data are mature and can join the other CCI datasets

LOLIPOP web site

<https://climate.esa.int/en/projects/long-lived-greenhouse-gas-products-performances-lolipop/>



THANK YOU