LOng-LIved greenhouse gas PrOducts Performances (LOLIPOP)

Product Validation Plan (PVP)

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1 Introduction

1.1 Scope of the document

This document is the Product Validation Plan (PVP) version 1.1 (v1.1), which is deliverable 2.1 of the ESA CCI LOLIPOP project, which outlines the general validation strategy used within this project. Note that the definitive validation strategy will be outlined in the Product Validation and Intercomparison Report (PVIR D2.3).

The LOLIPOP (Long-lived greenhouse gas products performances) project is looking into, and assessing the quality of, currently available long-lived greenhouse gas satellite measurements excluding carbon dioxide and methane (which fall under ESA's GHG CCI project). In essence, this concerns measurements (limb and nadir) of N_2O and chlorine- and fluorine-containing species. Information gathered in this project will further the R&D needed to generate new Other Long-Lived GreenHouse Gas (OLLGHG) Essential Climate Variable (ECV) satellite-derived data products.

The products to be validated were determined within WP1300, for nadir and limb configured satellite instruments.

Note that ideally, ECV products shall meet GCOS (Global Climate Observing System) data product requirements, which are typically denoted into 3 tiers (Threshold, Breakthrough and Goal). However up until its most recent installment (**/GCOS-245/** in 2022), N_2O and Halogenated species were not featured in the requirements. With this latest update, only N_2O was added. These requirements are geared towards profile measurements alone (threshold vertical resolution at 3 km, threshold horizontal resolution of 1000 km) and were written in light of establishing future missions. Therefore, these may be inadequate to assess N_2O products from current or legacy missions. Other listed threshold quality parameters for atmospheric N_2O mole fractions are a temporal resolution of 168 hours, a 2-sigma measurement uncertainty of 0.3 ppb, and a stability of 0.2 ppb per decade. Instead, we will use the quality indicators as brought forward by LOLIPOP's own data user survey [RD-1].

1.2 Reference Document

[RD-1] D1.1_LOLIPOP_URD version 1.1

1.3 Acronyms and Abbreviations

Abbreviation	**Meaning**
ACE-FTS	Atmospheric Chemistry Experiment - Fourier Transform Spectrometer
AGAGE	Advanced Global Atmospheric Gases Experiment
CCI	Climate Change Initiative

CFC	ChloroFluoroCarbon
ECV	Essential Climate Variable
ESA	European Space Agency
FoM	Figure of Merit
FTIR	Fourier Transform InfraRed
GCOS	Global Climate Observing System
GLORIA-B	Gimballed Limb Observer for Radiance Imaging of the Atmosphere – Balloon
HCFC	HydroChloroFluoroCarbon
HIPPO	HIAPER Pole-to-Pole Observations
LOLIPOP	Long Lived greenhouse gas PrOducts Performances
MIPAS-B	Michelson Interferometer for Passive Atmospheric Sounding - Balloon
NDACC	Network for the Detection of Atmospheric Composition Change
PVIR	Product Validation and Intercomparison Report
PVP	Product Validation Plan
TCCON	Total Carbon Column Observing Network

2 General Nadir product validation strategy

For the nadir data products, two types of comparisons will be undertaken.

- Intercomparisons will be made between nadir satellite measurements when at least two different satellite systems provide retrievals for a given gas. These measurements should overlap sufficiently in time and space to ensure that a meaningful comparison can be made.
- Validation comparisons will be made between nadir satellite measurements and difference reference data sources as outlined below.

No comparisons between nadir and limb satellite measurements are currently foreseen. If new nadir data products for CFCs, HCFCs, or HFCs are retrieved in this project, this may be added as appropriate.

2.1 Reference data

2.1.1 NDACC

The Network for the Detection of Atmospheric Composition Change (NDACC) is an international network of atmospheric composition sounding instruments (https://ndacc.larc.nasa.gov/). For this work package, we are particularly interested in the ground-based solar-absorption remote sensing FTIR measurements as they sample virtually the same atmospheric column as the nadir satellites (although potentially with differing vertical sensitivities). For its retrievals, NDACC used an optimal estimation approach **/Rodgers 2000/** using either the SFIT **/Pougatchev 1995/** or PROFITT **/Hase 2006/** retrieval algorithms. Standard target species are HCHO, C_2H_6, HCN, CO, HCl, HF, N_2O, CH_4, HNO_3, $ClONO_2$, and O_3. For these species a harmonized retrieval approach has been agreed upon by all partners. However, partners are free to retrieve (and do so) other species such as CFC-11, CFC-12 and HCFC-22 (i.e. **/Zeng 2020/**, **/Pardo Cantos 2022/**, **/Zhou 2016, 2024/**), but it should be noted that harmonization between different station might be less strict, nor are all stations within the network involved (see Table 1 for a list of NDACC stations and the species of interest they retrieve). More detailed information on NDACC can be found in **/De Mazière et al. 2018/**.

Figure 1: Map of all FTIR NDACC sites.

Station	Lat	Lon	Alt (m)	CFC-11	CFC-12	SF6	HCFC-22
Eureka	80.06	−86.42	610				X
Ny Ålesund	78.92	11.92	24				X
Thule	76.52	−68.77	225				X
Harestua	60.22	10.75	59				X
St. Petersburg	59.88	29.83	20	X	X		X
Bremen	53.10	8.85	3				X

Jungfraujoch	46.55	7.98	3580	X	X	X	X
Toronto	43.60	79.36	17				X
Rikubetsu	43.46	143.77	380				X
Boulder	40.04	−105.24	1612				X
Tsukuba	36.05	140.12	3				X
Hefei	31.91	117.19	29	X	X		
Izaña	28.3	−16.48	2370				X
Réunion St.Denis	−20.90	55.49	85	X	X	X	X
Réunion Maido	−21.08	55.38	2155	X	X	X	X
Wollongong	−34.41	150.88	31				X
Lauder	−45.04	169.68	370	X		X	X
Arrival Heights	−77.83	166.66	200				X

Table 1: NDACC data availability for the validation of priority halogenated LOLIPOP species (CFC-11, CFC-12, and SF_6, with an option for HCFC-22).

2.1.2 TCCON

The Total Carbon Column Observing Network (**/Wunch et al. 2011/**) is a network of ground-based FTIR instruments that provide highly accurate and precise column-averaged dry-air mole fractions of atmospheric components, including CO_2, CH_4, N_2O, HF, CO, H_2O and HDO.

A key difference with respect to NDACC is that TCCON uses a profile scaling retrieval approach instead of optimal estimation. It also uses the simultaneously retrieved O_2 column, to derive the dry-air mole fractions. The network as a whole has undergone extensive validation using aircraft in situ overpass measurements and, as a result, a small correction is applied to the data to align with international measurement standards. For N_2O an airmass independent correction factor of 0.9821 ± 0.0098 is applied, next to small airmass dependent correction factors applied on a per retrieval window basis. This implies a total network uncertainty of ~1%. More information can be found on the TCCON wiki (https://tccon-wiki.caltech.edu/Main/DataDescriptionGGG2020).

In a study by **/Zhou et al. 2019/** comparing XN_2O measurements at combined TCCON/NDACC sites, it was found that the mean differences (NDACC–TCCON) at these sites ranged between −3.32 and 1.37 ppb (−1.1 %–0.5 %) with standard deviations between 1.69 and 5.01 ppb (0.5 %–1.6 %), which are within the uncertainties of the two datasets. The NDACC N2O retrieval has good sensitivity throughout the troposphere and stratosphere, while the TCCON retrieval underestimates a deviation from the a priori in the troposphere and overestimates it in the stratosphere.

We also need to take into account that both NDACC and TCCON rely on direct sun measurements. Thus clear-sky conditions are a must and for high latitude sites no data is available during local winter.

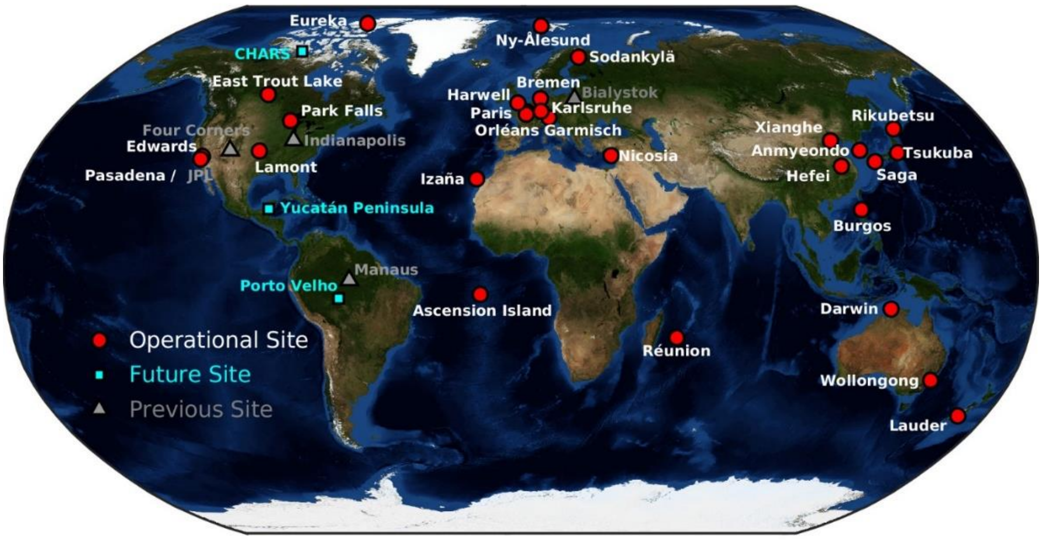

Figure 2: Map of all FTIR TCCON sites.

2.1.3 In Situ

While ground-based remote sensing data are ideal counterparts to nadir sounding satellite products, for CFCs particularly, the obtained reference dataset may be very limited. Therefore, ground-based or mobile platform (balloon, aircraft, etc.) born instruments could be of great use, particularly when assessing long term trends. Here, we'll look specifically at the ALE/GAGE/AGAGE missions started in 1978, which are based on gas chromatography and mass spectrometry, and which determined the atmospheric concentrations of halocarbons at various locations **/Prinn et al. 2000/**. The 3 phases (The Atmospheric Lifetime Experiment ALE 1978-1986, The Global Atmospheric Gases Experiment GAGE 1981-1996 and The Advanced Global Gases Experiment AGAGE 1993-present) are primarily driven by changes in the instrumentation, which allowed the monitoring of additional gases at ever-greater frequency. Currently, the network measures CFC-11, CFC-12, CFC-113, $CHCl_3$, CCl_4, CH_3CCl_3, CH_4 and N_2O) at all stations (see Figure 3 for the geographical distribution) and many more at selected sites (see https://agage.mit.edu/instruments for more information).

Figure 3: Map of all AGAGE stations.

2.2 Strategy

The general principles of each validation revolve around finding adequate collocation criteria between satellite and reference data that strike a balance between obtaining a statistically significant dataset and limiting the risk of introducing additional biases due to spatial and temporal differences between the reference and satellite measurement. We will strive to make these colocation parameters as uniform as possible for the different algorithms involved, but in reality, this may not be practically possible when comparing a high and low density dataset. As a starting point, we will test 2 collocation criteria: pairing satellite and remote sensing data that are taken within 2 hours of one another and are located no further than either 500 km or 100km apart. Typically, satellite products contain information regarding their vertical sensitivity, either in the form of a vertical sensitivity profile or a (column) averaging kernel combined with an a priori profile. If no such information is available, valuable information can still be obtained in comparisons with reference data, albeit more restricted. The exact formulation on how to deal with these inherent differences depends in part on how the sensitivity information is presented to the users. In all cases, we will refer to the respective user guides, and we will implement them accordingly. However, we can formulate validation principles.

One principle is that when the retrieved satellite data contains a priori information, we will take this into account in our comparisons. This we do by changing the a priori in one of the datasets by its counterpart. Since we want to look at the performance of various competing algorithm products, we will use the reference data a priori profiles as the common a priori.

As an example (the exact formulation may depend on how the a priori information is provided), to limit the impact of the a priori, we adjust the satellite dry air mole fraction using the FTS a priori as in

$$\hat{c}_{S,adj} = \hat{c}_S + \sum_l pw_l\,(1 - A_l)(x^l_{F,a} - x^l_{S,a})$$

where, $\hat{c}_S$ represents the originally retrieved satellite column-averaged dry air mole fraction, l is the index of the vertical layer, A_l the corresponding column averaging kernel of the satellite algorithm, $\mathbf{x}_{S,a}$ and $\mathbf{x}_{F,a}$ are the satellite and FTS a priori dry air mole fraction profiles, respectively. pw_l is the pressure weight associated with level or layer l.

Likewise, to address the differences in vertical sensitivity we apply either the satellite averaging kernel or vertical sensitivity onto the FTS data to obtain a hypothetical satellite retrieved value if the FTS profile were to correspond with the true state of the atmosphere.

Important to note here is that unlike NDACC, which directly yields retrieved profiles ($x_{F,r}$), TCCON provides total column dry air mole fractions only. So, we will apply this vertical sensitivity correction onto the scaled TCCON a priori, where the scaling factor takes into account the actual retrieval (which is based on the scaling of an a priori profile) as well as the post retrieval corrections to bring TCCON in line with in situ measurements. Thus, the scaled TCCON profile ($x_{F,r}$) corresponds to

$$x_{F,r} = x_{F,a} \times \hat{c}_{F,r} / \hat{c}_{F,a}$$

where $x_{F,a}$ is the TCCON a priori profile. $\hat{c}_{F,r}$ and $\hat{c}_{F,a}$ are the TCCON retrieved and a priori column-averaged dry air mole fractions.

The adjusted FTS dry air mole fraction then corresponds to

$$\hat{c}_{F,adj} = \sum_l pw_l\,(x^l_{F,a} + (x^l_{F,r} - x^l_{F,a})A_l)$$

where, pw_l again represents the pressure weight associated with the level or vertical layer with index l and A_l the corresponding column averaging kernel of the satellite algorithm. $\mathbf{x}_{F,a}$ and $\mathbf{x}_{F,r}$ are the FTS a priori and scaled dry air mole fraction profiles, respectively. Again, the exact formulation may depend on how the sensitivity information is presented to the user.

This approach should minimize the inherent differences between satellite and ground-based remote sensing retrievals, regardless of the algorithm and target species involved. If partial column information is available, we will also look at these layers.

2.3 Quality Parameters

Quality indicators, or so-called Figures of Merit (FoM), that will be derived from the intercomparisons at each individual reference site are the bias, scatter and long-term stability. Using multiple reference sites, we can look at the overall bias, scatter and stability as well as the variability of these parameters over the networks. We will also assess the user declared uncertainties.

When 2 (or more) algorithms retrieve the same satellite product (which is the case for IASI N_2O), we will determine the statistical significance of their FoM differences by calculating the probability that the obtained sample variances all stem from the same population. When both products also cover the same timeframe and spatial coverage, we will also generate identical resolution gridded monthly or seasonal (depending on data density) maps for direct comparison.

Optionally (if time permits and if the vertical sensitivity differences are significant enough to merit such an exercise) we will use 3D model fields as an intermediate to take into account the respective differences in vertical sensitivity. In this case, instead of a direct comparison we evaluate the respective biases towards the model.

Also note that the above-mentioned methodology and FoM parameter determination will be primarily applied to our N_2O algorithm analysis. Currently the NADIR halogenated retrieval products are restricted to IASI. This particular product (**/De Longueville, 2021**) does not retrieve absolute concentrations. Instead, it tracks the evolution of CFC concentrations with respect to a reference year (2008) over the Northern mid latitudes (23.27°N–66.32°N). In other words, the primary focus of our validation lies in checking its ability to capture the long-term evolution of these compounds. In this respect, using the AGAGE network data is certainly feasible despite obvious differences in sampled atmosphere. In the same vain, the sometimes very limited FTIR reference dataset (Table 1, but even more limited when taking the IASI products latitudinal range into account) should not be an impediment either.

3 General Limb product validation strategy

For the limb data products, two types of comparisons will be undertaken.

- Intercomparisons will be made between vertically resolved satellite measurements when at least two different satellite systems provide retrievals for a given gas. These measurements should overlap sufficiently in time and space to ensure that a meaningful comparison can be made.
- Validation comparisons will be made between vertically resolved satellite measurements and difference reference data sources as outlined below.

No comparisons between limb and nadir satellite measurements are currently foreseen. If new nadir data products for CFCs, HCFCs, or HFCs are retrieved in this project, this may be added as appropriate.

3.1 Reference data

3.1.1 Balloon-borne Fourier transform infrared (FTIR) Spectrometers

Deploying FTIR spectrometers on balloon platforms enables the measurement of atmospheric profiles from a near-space vantage point. For this work package, we will focus on measurements from three balloon-borne FTIR instruments: MkIV FTIR **/Toon, 1991/**, MIPAS-B (e.g., **/von Clarmann et al., 1993/**), and GLORIA (**/Friedl-Vallon et al., 2014/**; **/Riese et al., 2014/**; **/Johansson et al., 2022/**). The MkIV measures using the solar occultation technique (as used by ACE-FTS) and MIPAS-B and GLORIA are limb emission instruments like MIPAS. All provide a profiling view of the atmosphere similar to the limb sounders to be validated in this work package.

Details on the flights by these three FTIR spectrometers since 2000 are given in the tables below.

Table 2 provides details for the GLORIA and MIPAS instruments and Table 3provided details for the MkIV instrument. For these instruments, the locations of the launch sites are as follows: Kiruna/Esrange, Sweden; Aire sur l'Adour, France; Teresina, Brazil; Timmins, Canada; and Ft. Sumner, New Mexico, USA. The suite of gases that can be retrieved from the balloon-borne FTIR measurements is given in Table 4. Note, not all species may be available from each flight depending on conditions and development of processor.

Date	Instrument	Launch Site	Reference
2001-01-11	MIPAS-B2	Kiruna/Esrange	**/Hoepfner et al. 2002/**
2002-02-12	MIPAS-B2	Kiruna/Esrange	https://stratocat.com.ar/fichas-e/2002/KRN-20020212.htm
2002-09-24	MIPAS-B2	Aire sur l'Adour	**/Wetzel et al. 2022/**
2002-12-07	MIPAS-B2	Kiruna/Esrange	**/Oelhaf et al. 2003/**
2003-03-20	MIPAS-B2	Kiruna/Esrange	**/Wetzel et al. 2022/**
2003-07-03	MIPAS-B2	Kiruna/Esrange	**/Wetzel et al. 2022/**
2005-06-14	MIPAS-B2	Teresina	**/Wetzel et al. 2022/**
2008-06-06	MIPAS-B2	Teresina	**/Wetzel et al. 2022/**
2009-03-11	MIPAS-B2	Kiruna/Esrange	**/Wetzel et al. 2022/**
2010-01-24	MIPAS-B2	Kiruna/Esrange	**/Wetzel et al. 2022/**
2011-03-31	MIPAS-B2	Kiruna/Esrange	**/Wetzel et al. 2022/**
2014-09-07	MIPAS-B2	Timmins	**/Wetzel et al. 2017/**
2021-08-21	GLORIA-B	Kiruna/Esrange	https://stratocat.com.ar/fichas-e/2021/KRN-20210821.htm
2022-08-23	GLORIA-B++	Timmins	https://stratocat.com.ar/fichas-e/2022/TMS-20220823.htm

Table 2: Details of balloon flights with the MIPAS-B and GLORIA FTIR instruments.

Date	Tangent Latitude (N) (at 20 km)	Tangent Longitude (E) (at 20 km)	Minimum Altitude (km)	Balloon Altitude (km)	Launch Site	Event
2000-03-15	67.8	34.2	11	29	Kiruna/Esrange	Sunrise
2002-12-16	64.4	31.2	12	31	Kiruna/Esrange	Sunrise
2003-04-01	68.3	35.2	11	32	Kiruna/Esrange	Sunrise
2003-09-19	34.3	-113.3	7	36	Ft. Sumner	Sunset
2004-09-23	33.8	-109.2	11	38	Ft. Sumner	Sunset
2005-09-20	35.2	-114.1	11	39	Ft. Sumner	Sunset
2005-09-21	34	-110.3	13	29	Ft. Sumner	Sunrise
2007-02-07	67.9	21	-	34	Kiruna/Esrange	Ascent#
2007-02-22	67.9	21.1	25	34	Kiruna/Esrange	Ascent#
2007-09-22	35.2	-114.1	10	38	Ft. Sumner	Sunset
2007-09-23	34	-110.3	13	38	Ft. Sumner	Sunrise
2011-09-23	34.5	-108.8	6	39	Ft. Sumner	Sunset
2011-09-24	35.7	-96.3	14	40	Ft. Sumner	Sunrise
2014-09-13	36.2	-112.5	7	39	Ft. Sumner	Sunset
2014-09-14	35.6	-103.5	8	40	Ft. Sumner	Sunrise
2016-09-27	36	-110.5	11	39	Ft. Sumner	Sunset
2019-10-07	33.4	-109.7	7	38	Ft. Sumner	Sunset
2021-09-25	33.5	-107.5	8	39	Ft. Sumner	Sunset
2021-09-26	33	-97.8	10	40	Ft. Sumner	Sunrise

Table 3: Details of balloon flights with the MkIV FTIR instrument. # indicates that the balloon burst upon reaching float altitude. All details from https://mark4sun.jpl.nasa.gov/m4data.html.

Balloon FTIR spectrometer	Species retrieved
MIPAS-B	**N_2O**, **CFC-11**, **CFC-12**, **SF_6**, CFC-113, HCFC-22, CCl_4, CF_4
GLORIA	**N_2O**, **CFC-11**, **CFC-12**, **SF_6**, CFC-113, HCFC-22, CCl_4, CF_4
MkIV	**N_2O**, **CFC-11**, **CFC-12**, **SF_6**, CFC-113, HCFC-22, CCl_4, CF_4, HCFC-142b, HFC-23

Table 4: Typical gases retrieved from the balloon-borne FTIRs. The species indicated in bold are those four prioritized in this project.

3.1.2 In situ aircraft measurements

Measurements from the HIPPO project **/Wofsy 2011/** will be investigated as possible additional sources of in situ reference data for the limb product validation. Lack of temporal and spatial overlap in the airborne and satellite measurements may preclude the inclusion of all of these independent datasets. Therefore, this work will be done on a best effort basis.

3.2 Strategy

The general principles for the validation comparisons aim to find adequate collocation criteria between satellite and reference data to balance the competing needs of obtaining a statistically significant dataset and limiting the risk of introducing biases. We will strive to make these collocation parameters as uniform as possible as to aid the interpretation of data sets, but in reality, this may not be practically possible when comparing a high and low density dataset. As a starting point, we will use within 500 km and 6 hours, based on previous validation efforts. Using zonal mean comparisons for lower density satellite measurements will also be investigated (e.g., /**Kolonjari 2024**/).

Since the vertical sampling and resolutions of the limb profile measurements and balloon FTIR observations are quite similar (2-5 km, typically), these comparisons will initially use the native profiles without any account taken of the averaging kernels. The need to apply the averaging kernels will be evaluated as time permits in the comparisons.

If sufficient spatial and temporal overlaps are found between the satellite measurements and the HIPPO aircraft observations, a comparison strategy based on that of **/Martinez-Alonso 2014/** will be utilized for the HIPPO in situ profiles. This will employ “looser” criteria than the balloon-borne profile comparisons described above.

3.3 Quality Parameters

Quality indicators that will be derived from the comparisons with the reference balloon-borne measurements are the bias and scatter. If sufficient flights and coincidences are available for individual satellite datasets, the long-term stability will also be examined. The combination of multiple reference flights can be used to look at the overall bias and scatter as well as the variability of these parameters.

Similar quality indicators will be derived for the intercomparisons between satellite data products. Profile-to-profile comparisons will be made by identifying pairs of coincident profiles using a strategy similar to that for the reference balloon-borne measurements.

The focus for these evaluations will be on N_2O, CFC-11, CFC-12, and SF_6 as agreed following the completion of WP1300. All species selected have at least two satellite systems making observations of these gases (and in some cases, multiple retrieval processors). Also, the two balloon-borne FTIRs and the HIPPO aircraft provide profile measurements of these species.

From the User’s Needs survey, N_2O, SF_6, CFC-11, and CFC-12 were the top four requested gases of interest – those of highest priority. The next set identified (in requested order) were CCl_4, HCFC-22, HFC-134a, CF_4, HCFC-142b, HFC-23, and CFC-113. Of these, HCFC-22,

CCl_4, and CF_4 it will be possible to make both intercomparison and validation assessments because there are satellite and reference measurements available. For CFC-113, HCFC-142b, and HCF-23, the focus will be on limited validation assessments as only one satellite data set is available for comparison with reference measurements.

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