GLYoxal Retrievals from TROPOMI (GLYRETRO)

Sentinel-5p + Innovation - Theme 1: CHOCHO

Scientific Roadmap

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authors : Christophe Lerot, Leonardo Alvarado, Andreas Richter, Trissevgeni Stavrakou, Michel Van Roozendael

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Applicable Documents

[AD01] Sentinel-5p Innovation (S5p+I) - Statement of Work - EOP-SD-SOW-2018-049, Issue 2, 20/08/2018.

[AD02] Sentinel-5p+Innovation: Theme 1; Glyoxal Retrievals from TROPOMI (GLYRETRO): Requirement Baseline Document, BIRA-IASB, issue 2.0, S5p+I_CHOCHO_BIRA_RB, 01/11/2019.

[AD03] Sentinel-5p+Innovation: Theme 1; Glyoxal Retrievals from TROPOMI (GLYRETRO): ATBD, BIRA-IASB, issue 3.1, S5p+I_CHOCHO_BIRA_ATBD, 22/12/2021.

[AD04] Sentinel-5p+Innovation: Theme 1; Glyoxal Retrievals from TROPOMI (GLYRETRO): Product Validation Report, IUP-Bremen, issue 2.1, S5p+I_CHOCHO_BIRA_VR, 22/12/2021.

[AD05] Sentinel-5p+Innovation: Theme 1; Glyoxal Retrievals from TROPOMI (GLYRETRO): Product User Manual, BIRA-IASB, issue 1.1, S5p+I_CHOCHO_BIRA_PUM, 22/12/2021.

Reference Documents

[RD01] Sentinel-4 L2 Processor Component Development–Project Management Plan, DLR, S4-L2-DLR-PMP-1004, issue 2.1, 31/05/2017.

[RD02] Sentinel-5 Level-2 Prototype Processor Development Requirements Specification, ESA, S5-RS-ESA-GR-0131, issue 1.7, 29/06/2018.

[RD03] S5L2PP: Record of agreements from negotiation; S5L2PP proposal consortium; ST-ESA-S5L2PP-NOT-003; Issue 1.1, 02/09/2016.

[RD04] S5L2PP Glyoxal ATBD, BIRA-IASB, BIRA-ESA-S5L2PP-ATBD-004, issue: 4.1, 24/11/2021.

1 Introduction

The purpose of this document is to assess the maturity of the TROPOMI glyoxal tropospheric column product developed within the S5p+Innovation program, to identify its current limitations and to define possible further developments to improve the product. Based on this, current scientific and operational requirements are discussed and possibly reviewed. This document also describes what would be the specifics and needs for an operational production of the TROPOMI glyoxal data.

2 Product maturity assessment

Scientific requirements are useful to support the discussion on the product maturity. They have been defined in the context of the preparation of the operational L2 processors of the future Sentinel-4 and -5 instruments [RD01-03] and are given in Table 1. While one single total uncertainty requirement is defined for Sentinel-4, two separate values are defined for the random and systematic components of the uncertainty in Sentinel-5. They are further discussed in the requirement baseline document [AD02].

Table 1 : Uncertainty Requirements on glyoxal column retrievals defined for the Sentinel-4 and -5 missions.

	Uncertainty (Threshold)	Conditions
Sentinel-4	7 x 10^{14} molec.cm^{-2} or 50% (least stringent)	SZA < 60° VZA < 60° cloud fraction < 20% VCD > 5 x 10^{14} molec.cm^{-2}
Sentinel-5	Random error: < 1.5 x 10^{15} molec.cm^{-2} Systematic error: < 2.5 x 10^{14} molec.cm^{-2} or 50% (least stringent)	SZA < 70° VZA < 70°

The TROPOMI glyoxal product developed within this study [AD03] relies on a strong heritage. Indeed, first glyoxal tropospheric column retrievals using a DOAS methodology have been demonstrated more than 15 years ago using SCIAMACHY spectra (Wittrock et al., 2006). Since then, the approach has been successfully applied by different teams on different instruments (e.g. Alvarado et al., 2014; Chan Miller et al., 2014; Lerot et al., 2010; Vrekoussis et al., 2010) and all those studies contributed to the development of this TROPOMI product. The DOAS approach itself is not to be demonstrated any longer since applied for decades in numerous applications from ground, air and space instruments.

The main glyoxal DOAS settings used for the spectral fits are also relatively well established. They vary marginally from a study to another. Sensitivity tests and observing system simulation experiments have shown that the spectral region encompassing the two strongest glyoxal absorption bands is optimal. Nevertheless, small changes in the DOAS settings may substantially affect the retrieved glyoxal columns owing to its low optical depth as discussed in the next section. The method to compute air mass factors is also quite common; the different algorithms mostly differ from each other in the used input data and in the way cloud/aerosols are considered. There is therefore a consensus on the fact that glyoxal columns can reliably be retrieved using the DOAS method.

The algorithm developed for TROPOMI within this study has also been applied to other satellite instruments, i.e. OMI, GOME-2A and GOME-2B. The different data sets obtained have been intercompared and a very satisfactory consistency is found with mean differences of less than 20%. In addition, a significant effort has been done in validating the satellite products with a series of MAX-DOAS measurements. It has been found that the seasonal variability observed by the satellite and ground-based instruments agree well with high correlation coefficients. Observed biases are generally within the defined requirements, at the exception of specific conditions. Please refer to the verification report [AD04] or Lerot et al., (2021) for more details. Those elements give confidence into the maturity of the developed product. Table 2 gives estimates of the maturity level of the product for typical conditions. Those estimates are based on the validation results and experience built during the product development. Causes for lower maturity in specific conditions will be discussed in the next section.

The data production has greatly benefited from the large experience of the consortium in the development of UV-Visible DOAS satellite products and of its implication in the TROPOMI operational activities. This has been essential to facilitate the treatment of the L1 data, to establish the in-house processing chains and to generate a TROPOMI glyoxal product with a format very similar to that of the operational products. At the end of this study, a 4-years TROPOMI glyoxal data set is available and easily ingestible by any user familiar with the TROPOMI operational products (See Product User Manual [AD05]). Access to the data can be granted via the project website, which facilitates their exploitation.

Table 2 : Estimates of the maturity level of the TROPOMI glyoxal product for typical conditions.

	Maturity level Low	Maturity level Medium	Maturity level High	Comment
Tropics			x	
Anthropogenic conditions			x	Medium if contamination by aerosols
Low sun elevation (mid/high latitude during wintertime)		x		Low bias identified in validation
Fire events		x		High signal; AMF uncertainties larger
Oceans		x		High sensitivity to the DOAS settings

There is an increasing interest from the user community to exploit glyoxal data, of which the potential to provide information on non-methane volatile organic compounds emissions (NMVOC) (e.g. Cao et al., 2018; Fu et al., 2008; Stavrakou et al., 2009) and on the production of secondary organic aerosols (e.g. Hallquist et al., 2009; Knote et al., 2014) is more and more recognized. Since the product developed within the study has been made publicly available, access to the database has been requested by an important number of users. Although currently in development, we can expect from their respective applications interesting results on the mid-term in a number of different domains:

- Evaluation of emission inventories in China and at the global scale.
- Evaluation of glyoxal output from different CTMs, including the CAMS system.
- Inverse modelling of NMVOC emissions using CTMs.
- Study of particular events, such as COVID-19 lockdown or extreme fire events.
- Comparisons with independent measurements from ground-based or airborne systems, validation of other CHOCHO satellite retrievals, including from the GEMS instrument.

3 Scientific limitations and area for improvements

Despite its good maturity, a number of limitations are known to affect the product. Some are fundamental and related to the glyoxal characteristics while others might be reduced with further future developments. We discuss below the main limitations associated to each algorithmic step as well as the possible improvements.

3.1 DOAS fits

The glyoxal optical depth is very low, typically one order of magnitude smaller than that of other spectral signatures in the same spectral region (e.g. NO_2, O2-O2, Ring, etc). The first consequence of this weak absorption is that the product is prone-to-noise. Individual glyoxal measurements are generally meaningless since the level of noise is larger than the usual atmospheric glyoxal columns. This level of noise can be easily reduced by averaging together several observations at cost of the spatial or time resolution. The huge amount of TROPOMI data allows nevertheless maintaining a high level of spatial-temporal details compared to past satellite missions. The second consequence of the weak glyoxal optical depth is that its retrieval is very sensitive to any not-perfectly-fitted spectral structure originating from calibration limitations, inaccurate cross-sections or defects in the forward model. This effect is more difficult to mitigate as cross-correlations with other spectral signatures generally cause systematic biases. Depending on the origin of the problematic spectral signature, local systematic biases can therefore be introduced in the product. The background correction procedure (see ATBD [AD03]) aims at reducing such possible biases in the product but it efficiently removes only row or latitudinal-dependent biases. Local biases introduced by specific conditions (e.g. due to an enhanced signal of any other trace gas or the presence of clouds/aerosols) will remain in the product. This limitation is very difficult to circumvent as we depend mainly on the quality of the available cross-sections and on the L1 data calibration. Only the forward model can possibly be improved in case of identified issue. For example, this has been the case during the study to mitigate biases introduced in case of scene brightness heterogeneity or of very strong NO2 absorption.

3.2 Air Mass Factors

The accuracy of the air mass factors calculations significantly depends on the input data. For this glyoxal product, the most critical inputs are the surface albedo and the a priori glyoxal vertical profile shape.

For the surface albedo, we still rely on the OMI Lambertian-equivalent reflectivity (LER) database (Kleipool et al., 2008). It has the advantage to be compatible with the S5p overpass time but has a too coarse spatial resolution for TROPOMI and neglects the viewing zenith angle dependence of the reflectances. It would be of great benefit to use in future the directionally dependent LER TROPOMI database that has been developed as part of another project of the ESA S5p+Innovation program. The current version is based on TROPOMI L1 data v1.0.0 of which the radiometric calibration is imperfect, resulting in surface albedo biased high in the UV-Visible region. A L1 data reprocessing is planned in the coming months with an improved calibration and the next version of the TROPOMI albedo database is expected to be more accurate.

A priori glyoxal profile shapes over lands are provided by the BIRA-IASB chemical-transport model MAGRITTE running at 1°x1° resolution. Those have been modelled for the year 2018 using corresponding emission inventories. Using one single year of profiles is reasonable for emissions characterized by a regular and recurrent cycle such as biogenic or anthropogenic emissions. However, this scheme leads to larger profile-related uncertainties for irregular events such as fire events. In addition, uncertainties would be larger in case of long-term emission changes. The TROPOMI time series being still relatively short, this source of uncertainties is likely limited. To reduce as much as possible those uncertainties, it would be beneficial to reprocess backward the AMFs based on profiles calculated with emission inventories provided specifically for every year. It has to be noted that those optimized emission inventories are provided with a certain delay, preventing their use for forward processing. Over oceans, the models do not reproduce the glyoxal columns observed from space both in terms of spatial distribution and of magnitude. For this reason, we use one profile that has been measured a few years ago by an airborne MAXDOAS during a campaign over the Pacific Ocean instead of the CTM output. Using this single profile for all oceans and all seasons is certainly a large source of error. To date, the interpretation of the glyoxal signal over oceans remains under debate and production mechanisms remain unclear in those regions. Any progress made in future on this aspect would allow reducing AMF uncertainties.

For computing the air mass factors, we use a very simple scheme assuming a clear atmosphere without any cloud or aerosols. Quality assurance values are low in case of cloud contamination in order to filter out easily such scenes. Other tropospheric products use relaxed cloud filtering in combination with a cloud correction in the AMF computation. This allows the exploitation of a larger number of observations. However, this is not applicable for glyoxal since an artificial increase of the slant columns is observed over bright scenes (e.g. because of cloud coverage). This increase originates from spectral cross-correlation with other absorbers as described in the previous section. This fundamental limitation will remain unless future developments in the spectral fit allow mitigating the cross-correlations. Explicit corrections for aerosols require availability of input aerosol properties and is only feasible for specific case studies rather than at the global scale.

3.3 Background correction

The background correction aims at reducing as much as possible the presence of biases in the product by using the retrievals in a remote reference sector (i.e. the Pacific Ocean). Nevertheless, it also suffers from uncertainties, mostly related to our current limited knowledge of the glyoxal concentrations in this reference sector. The current approach assumes a homogeneous distribution of the glyoxal concentration with a fixed reference column of 1×10^{14} molec.cm^{-2}, based on the work by Sinreich et al. (2010). The choice of this value and of the spatial distribution assumption has a significant impact on the retrieved glyoxal columns at the global scale.

Provided the different existing product uncertainties associated to the oceanic glyoxal fields, it would be beneficial to have more independent glyoxal measurements in remote oceans to improve our knowledge on (1) its spatial and vertical distribution and (2) the magnitude of its vertical columns.

Table 3 summarizes the main scientific limitations discussed above for the different steps of the algorithm and the related possible solutions or future developments that might improve the product. In the next section, we will discuss the technical aspects and current limitations to make the production of the TROPOMI glyoxal retrievals operational.

Table 3 : Main limitations in the different algorithmic steps and possible solutions to reduce the associated uncertainties.

	Limitations	Key Factors/Solutions
Spectral fits	Sensitive to spectral interferences (fundamental limitation)	Improvement of absorption cross-sections Accuracy of L1 calibration
AMF	Albedo database too coarse and neglects anisotropy	Use S5p DLER climatology V2 when available

	Profile shape database based one single year Oceanic AMFs based one single profile Simple clear-sky model	Simulate profiles for several years and reprocess AMF Needs for independent oceanic measurements Use of a more sophisticated model requires mitigation of spectral interferences in the DOAS fit. Explicit aerosol correction for specific studies.
Background correction	Magnitude and spatial distribution of reference glyoxal columns uncertain	Needs for independent oceanic measurements

4 Specific needs for operational activities.

4.1 Algorithm structure and interdependencies

Figure 1 presents the flow diagram of the glyoxal retrieval algorithm. It is conceptually close to other DOAS products already running in the TROPOMI operational environment (e.g. the formaldehyde product), which would greatly facilitate its operationalization. The sketch includes all different steps necessary to the CHOCHO retrieval, i.e. (1) the slant column density derived from the measured reflectance spectrum, (2) the AMF computation for the conversion of the slant column into a vertical column and (3) the background normalization required in the case of weak absorbers such as glyoxal. The interdependencies with static (purple boxes) and dynamic (light orange boxes) input data and other L2 products, such as clouds and aerosols are also represented. The DOAS fit also uses as reference mean radiance spectra, which needs to be preprocessed. Similarly, the background correction relies on a database of slant columns retrieved in a reference sector, which needs to be regularly updated, ideally on a daily basis.

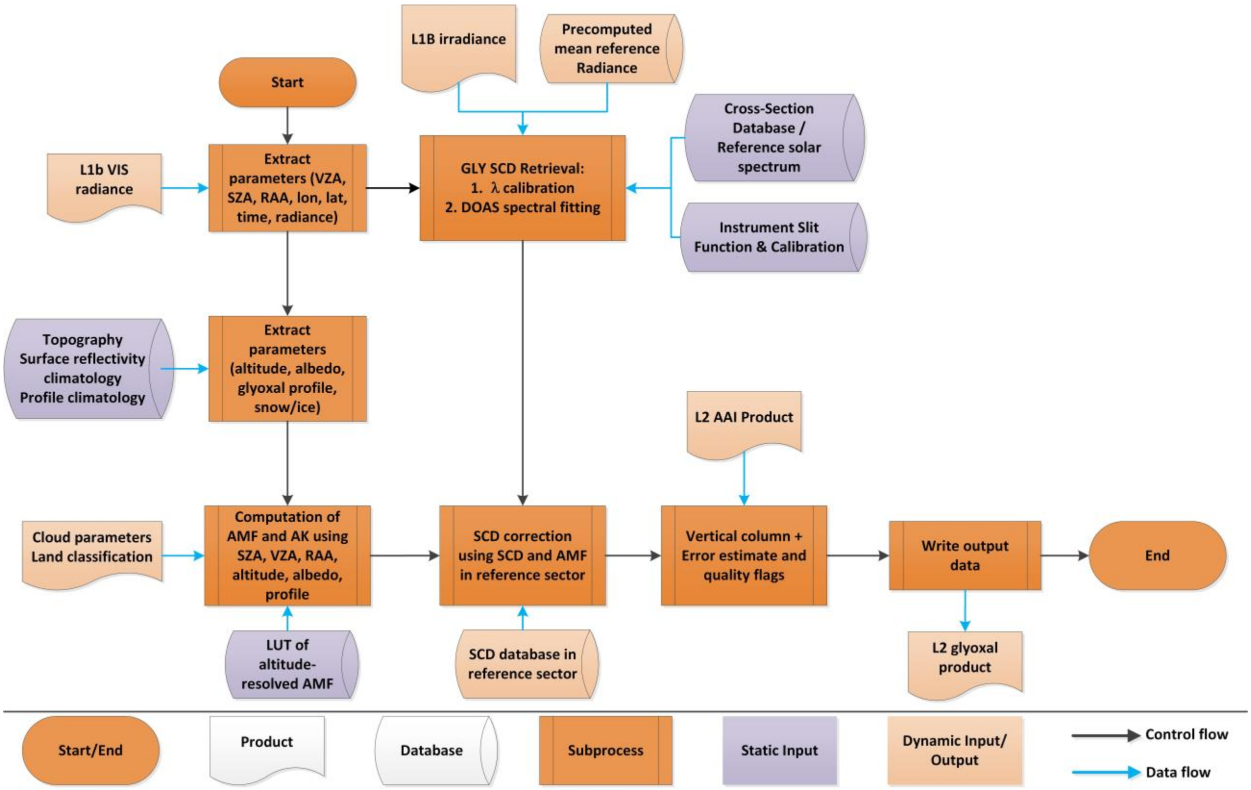

Figure 1 – General flowchart of the glyoxal DOAS retrieval algorithm.

In a scientific context, there is a large flexibility with the input data handling as there is no time requirement for the processing. In an operational environment, the constraints are much more important and all input/output data need to be orchestrated in order to guarantee the retrieval success, while meeting much more stringent time constraints, e.g. a near-real time delivery. A few aspects would need to be adapted for a forward operational processing:

- *Handling of the reference radiance and SCD databases:* The DOAS fits use mean radiances as reference. Those are obtained by averaging many spectra falling into a reference sector located in the Pacific Ocean. In the scientific processing, we use mean radiances computed based on spectra recorded in the Pacific Ocean the same day as the data under DOAS treatment. Having such a time proximity is ideal to reduce efficiently the impact of any distortion present in the spectra. On the other hand, if there is no data available for a given day in the reference sector, no retrieval will be performed in the scientific processing. The background correction aims at reducing possible systematic biases in the final vertical columns and the applied corrections rely on slant columns derived also in a reference Pacific sector the same day as the retrievals of interest. Time proximity of the reference data is also important for this scheme. In a forward operational processing, the two approaches would need to be modified to rely only on past recent measurements, ideally from the previous day. In case of missing data in the reference sector, a mechanism using older data needs to be developed in order to avoid as much as possible data gaps in the operational production. Such mechanisms have been implemented for the HCHO and SO2 TROPOMI operational products and operationalization of the glyoxal algorithm may benefit from this. This has also been comprehensively addressed in the context of the preparation of the Sentinel-5 L2 prototype processors [RD04].

- *Source for auxiliary data:* Glyoxal retrievals are performed based on the spectral data of the band 4 of the instrument, which is also the baseline for NO_2 retrievals. Those two products share many common input data: the cloud fraction retrieved in the same spectral region, topography, surface classification and snow/ice flag. In the scientific glyoxal algorithm, we extract directly from the TROPOMI offline NO_2 product the needed data. This is very convenient for several reasons: (1) The number of dependencies with auxiliary data sources is limited; (2) the input is directly available for every TROPOMI ground pixel, without having to apply any tessellation algorithm or interpolation method. Depending on the offline NO2 product presents the disadvantage that there is a delay of about one week before the glyoxal vertical columns can be processed. In an operational context, this would need to be changed in order to avoid this link. Input data should be extracted directly from their original source so that the glyoxal product can be processed in parallel to the NO_2 product and not sequentially any longer.

- *A priori profiles*: The scientific algorithm currently relies on one year of glyoxal profile shapes modelled with BIRA-IASB CTM MAGRITTE. Using such a fixed database is straightforward to use, including in an operational environment. As mentioned above, the variability of the emissions and meteorology is however not well captured with such an approach. The current operational DOAS products rely on forecasts of profiles with the TM5 model for the near-real time processing and on the reanalysis for the offline processing. This approach certainly considers better the variability of the atmospheric conditions. Unfortunately, the number of models offering glyoxal output is limited and, to our knowledge, none with such a forecast capability. The increasing interest in modelling glyoxal might offer new perspectives in future, which would allow to improve this aspect and to enhance the consistency with other operational products.

4.2 Computational performance and output format.

The structure of the algorithm is very similar to that of the operational HCHO and SO_2 products. The different modules are executed sequentially, i.e. the DOAS fit, the AMF computation and the application of the background correction procedure. Using the current BIRA-IASB computing infrastructure, the process of one TROPOMI orbit requires less than 40 minutes using one single core. No major issue from the computational performance point of view is anticipated for a possible operationalization of the glyoxal product.

The current output format of the L2 glyoxal files produced by this scientific algorithm follows closely the current conventions of the operational products, in terms of filename and content structure as well as variable name nomenclature and units. Provided information is also very similar and includes:

- glyoxal vertical column, slant column and air mass factor
- measurement time and geolocation (center and corner), taken from the L1b product
- retrieval diagnostics including error estimates, averaging kernel and a priori information.
- quality flags.
- input parameters

Following the operational products, all variables provided in the L2 glyoxal files are organized in the structure sketched as in Figure 2. Detailed information can be found in the Product User Manual [AD05]. A possible operationalization would require consolidation of the metadata, of the quality assurance values in order to enhance the consistency with current operational products. Flags should also be further developed in order to trace better any event occurring during the processing and leading to any lower quality/missing data.

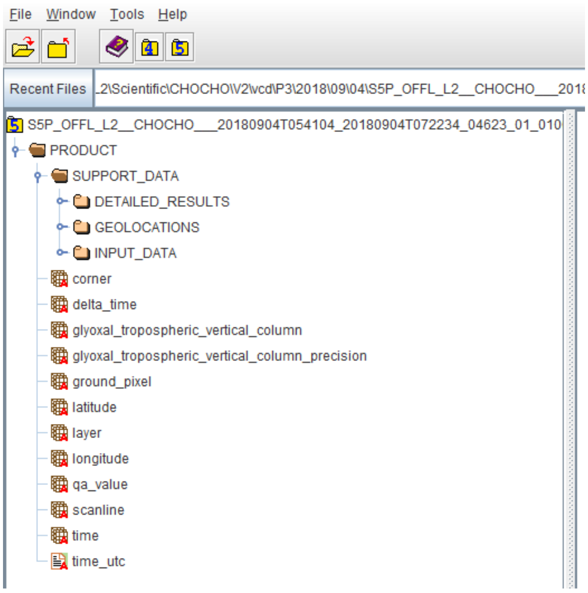

Figure 2 : L2 glyoxal file structure

5 Conclusions and perspectives

Overall, the TROPOMI glyoxal retrieval algorithm developed within the S5p+I framework is reasonably mature. Technically speaking, it benefited from the significant involvement of the consortium into the development of other operational products of which the retrieval principles are similar. It allowed establishing robust scientific processing chains providing as output L2 files compatible with the format and content of the operational products. An actual operationalization of the algorithm would be achievable, after adaptation of a few elements of the algorithm, mostly related to the handling of input data and the consolidation and tracing of the processing chain.

From the scientific point of view, this scientific product could rely on the large experience of the consortium in the retrieval of glyoxal columns for many years. Requirements have only been defined for glyoxal in the framework of the preparation of the future Sentinel-4 and -5 missions. Intercomparison and validation activities carried out during the study showed that the product is of good quality and that the requirements are generally met. Those requirements appear to be realistic and more stringent values would be challenging to meet. Provided the high level of noise in glyoxal products, it is important to specify different requirements for the random and systematic error components. The weak glyoxal optical depth leads to some fundamental limitations difficult to circumvent such as the level of noise or interferences with other spectral signatures. Nevertheless, the product could be further improved in case of future reprocessing activities. In particular, the surface albedo and a priori profile databases needed for air mass factor computation should be updated and possibly harmonized with other sensors to consolidate the analyses of the glyoxal variability and of possible long-term changes.

A significant contribution to the product uncertainty is due to the limited knowledge of the glyoxal concentrations over oceans. This remaining issue impacts the product not only over oceans via the air mass factor calculations but also at the global scale since the background correction relies on a reference sector taken over the Pacific ocean. The origin of the current large discrepancy between available remote glyoxal measurements, in situ data and models remains unclear. No production mechanism allows reproducing the distribution of the glyoxal columns observed from space. It cannot be excluded either that spectral interferences cause part of this signal. In order to make progress on this pending issue, having more independent oceanic measurements providing additional information on the glyoxal concentrations and its spatial distribution would be beneficial.

The potential of glyoxal measurements to derive information on NMVOC emissions is more and more recognized by the user community. This scientific TROPOMI glyoxal product, which is available vie the project website and has been advertised via conferences and publications, has been requested by a number of external teams for developing their own applications. Different type of studies are covered such as comparisons with independent measurements, optimization of emission inventories, evaluation of model output, etc. We can reasonably anticipate that our product will be of valuable interest for many other future studies.

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