Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observations

FRM$_4$DOAS

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Lead author:

F. Hendrick (BIRA-IASB)

Contributing authors:

C. Fayt, M. Friedrich, and M. Van Roozendael (BIRA-IASB)

S. Beirle, S. Dörner, and T. Wagner (MPIC)

U. Frieß and L. Tirpitz (IUP-Heidelberg)

A. Richter, T. Bösch, and L. Alvarado (IUP-Bremen)

A. Piters (KNMI)

K. Kreher (BK Scientific)

M. Navarro, O. Puentedura, C. Prados Román, and M. Yela (INTA)

A. Bais and D. Karagki (AUTH)

R. Querel (NIWA)

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Abbreviations and acronyms

AMF	Air Mass Factor
AOD	Aerosol Optical Depth
ATBD	Algorithm Theoretical Baseline Document
AUTH	Aristole University of Thessaloniki
BIRA/BIRA-IASB	Royal Belgian Institute for Space Aeronomy
BOREAS	Bremen Optimal estimation REtrieval for Aerosols and trace gaseS
CAMS	Copernicus Atmospheric Monitoring Service
CESAR	Cabauw Experimental Site for Atmospheric Research
CCN	Contract Change Notice
CINDI	Cabauw Intercomparison of Nitrogen Dioxide Measuring Instruments
CINDI-2	Cabauw Intercomparison of Nitrogen Dioxide Measuring Instruments 2
CPS	Central Processing System
DHF	Data Handling Facility
DLR	Deutsches Zentrum für Luft- und Raumfahrt
DOAS	Differential Optical Absorption Spectroscopy
DOFS	Degree of Freedom for Signal
DOI	Digital Object identifier
dSCD/DSCD	Differential Slant Column Density
ESA	European Space Agency
ESRIN	ESA European centre of excellence for exploitation of Earth observation missions
EUMETSAT	European Organisation for the Exploitation of Meteorological Satellites
EVDC	ESA Atmospheric Validation Data Centre
FOV	Field of View
FTP	File Transfer Protocol
FRM4DOAS	Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observations
GEOMS	Generic Earth Observation Metadata Standard
GPS	Global Positioning System
GOME	Global Ozone Monitoring Instrument
HDF	Hierarchical Data Format
HPC	High Performance Computer
IAP-CAS	Institute of Atmospheric Physics-Chinese Academy of Sciences
INTA	Instituto Nacional de Tecnica Aeroespacial
IUP-UB	Institute of Environmental Physics, University of Bremen
KNMI	Royal Netherlands Meteorological Institute
LP-DOAS	Long-path DOAS
MAPA	Mainz Profile Algorithm
MAX-DOAS	Multi-Axis Differential Optical Absorption Spectroscopy
MMF	Mexican MAX-DOAS Fit
MPIC	Max Planck Institute for Chemistry
NDACC	Network for the Detection of Atmospheric Composition Change
netCDF	network Common Data Form
NIDFORVAL	S5P NItrogen Dioxide and FORmaldehyde VALidation
NRT	Near Real Time
OEM	Optimal Estimation Method
OMI	Ozone Monitoring Instrument
PGN	Pandonia Global Network
PI	Principal Investigator
PRD	Pearl River Delta

QA/QC	Quality Assurance/Quality Control
RAA	Relative Azimuth Angle
R&D	Research and Development
RIVM	Rijksinstituut voor Volksgezondheid en Milieu
RMS	Root Mean Square
RTM	Radiative Transfer Model
S-4	Sentinel-4
S-5	Sentinel-5
S-5P	Sentinel-5 Precursor
SAF	Satellite Application Facility
SAOZ	Système d'Analyse par Observations Zénithales
SCD	Slant Column Density
SNR	Signal-to-noise ratio
SZA	Solar Zenith Angle
TEMIS	Tropospheric Emission Monitoring Internet Service
TRL	Technical Readiness Level
UHEID	University of Heidelberg
UV	Ultra-Violet
VDAF	Sentinel-5P Validation Data Analysis Facility
VCD	Vertical Column Density
VIS/vis	Visible
VZA	Viewing Zenith Angle
WBS	Work Breakdown Structure
WFDOAS	Weighting Function DOAS
WP	Work Package
XML	Extensible Markup Language
YAML	Ain't Markup Language

Project deliverables table

This table contains the project deliverables that are used as reference in the present document.

Deliverable number	Deliverable name	Link
D2	Network Scientific Requirements Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D2_Network_Scientific_Requirements_Document_20171116_final.pdf
D3	MAX-DOAS Instruments Review Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D3_MAXDOAS_Instruments_Review_v0.4_20171127.pdf
D4	MAX-DOAS Calibration and Operations Best Practices Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D4_MAXDOAS_Best_Practices_Document_20180110_v1_0.pdf
D5	MAX-DOAS Algorithm Round-Robin Definition and Results Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D5_MAXDOAS_Algorithm_Round_Robin_Document_20180130_final.pdf
D6	MAX-DOAS Algorithm ATBD	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D6_MAXDOAS_Algorithm_ATBD_v02_20180130.pdf

D7	MAX-DOAS Network Processing System Technical Requirements Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D7_MAXDOAS_Network_Processing_System_Technical_Requirements_20180130.pdf
D10	Processing System Validation report	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D10_system_validation_190618.pdf
D11	Intercomparison Campaign Requirements Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D11_Campaign_Requirements_Document_20161021.pdf
D12	Intercomparison Campaign Technical Requirements Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D12_Technical_Requirements_Document_20161021_final.pdf
D13	Intercomparison Campaign Planning Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D13_Campaign_Planning_Document_20161021_final.pdf
D14	Intercomparison Campaign Data Protocol	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D14_Campaign_Data_Protocol_20161021_final.pdf
D16	Consistency of MAX-DOAS aerosol and trace gas profiling during CINDI-2	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_D16_v2.1_Submission.pdf
D17	NDACC MAXDOAS DOI policy and procedure document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_CCN02_D17_NDACC_MAXDOAS_DOI_and_procedure_document_v2_0_20200615.pdf
D18	NDACC MAX-DOAS instrument certification procedures document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_CCN02_D18_NDACC_MAXDOAS_Certification_Procedures_document_v2.0_20200617.pdf
D20	MAX-DOAS Network Operational Processing System Architecture Design Document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_CCN02_D20_MAXDOAS_Network_Operational_Processing_System_Architecture_Design_Document__v2.0_20200611.pdf
D21	Operational Processing System Validation and Test Report document	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_CCN02_D21_Operational_Processing_System_and_Test_Report_document_v1.0_20200623.pdf
D26	NDACC MAX-DOAS Service Performance Assessment Report	https://frm4doas.aeronomie.be/ProjectDir/Deliverables/FRM4DOAS_CCN03_D26_NDACC_MAXDOAS_Service_performance_assessment_report_v2.0_20210415.pdf

Project summary

The first central NRT (24h latency) processing system for MAX-DOAS instruments has been developed in the framework of the FRM_4DOAS project. The system, which includes state-of-the-art retrieval algorithms for tropospheric and stratospheric NO_2 vertical profiles, tropospheric HCHO vertical profiles, and total O_3 columns, has been successfully demonstrated on daily submitted radiance spectra from 14 stations operated by the project partners and on measurements taken during the CINDI-2 intercomparison campaign held in Cabauw (The Netherlands) in September 2016.

One of the most important tasks during the first half of the project main phase was the selection of the baseline tropospheric profiling algorithms to be implemented in the processing system. This selection has been done through a dedicated Round-Robin intercomparison exercice involving the retrieval algorithms available within the FRM_4DOAS team, together with the underlying RTMs. It has consisted in simulating slant column densities (SCDs) of HCHO, NO_2 and O_4 for a set of atmospheric scenarios and viewing geometries using partners' RTM in view of their evaluation. After this first step, a reference dataset of differential slant columns (dSCDs) of HCHO, NO_2 and O_4, calculated using the median of the ensemble of simulated SCDs, has been compiled and distributed among the FRM_4DOAS participants. A quantitative quality assessment of the profiling algorithms has then been performed by comparing the profiles retrieved using this reference dataset with the initial ('true') aerosol and trace gas profiles. From this evaluation which also included technical aspects like the computing time, MPIC-MAPA and BIRA-MMF yielded on overall the most accurate results of the parameterized and OEM-based algorithms, respectively. Therefore they were selected as community algorithms to be implemented in the FRM_4DOAS centralized processing system.

Also important for the harmonisation of the MAX-DOAS data sets was the establishment of best practices for instrument calibration and operation. It was based on the individual experience of the project partners and on the outcomes of the past MAX-DOAS intercomparison campaigns, in particular CINDI-2. The most relevant guidelines and recommendations have been gathered in a quick and concise check-list which has been made available to the DOAS Community through the FRM_4DOAS web site. The MAX-DOAS Community has been also solicited by the FRM_4DOAS consortium to fill in a questionnaire for assessing the Technical Readiness Level of their current instruments and data processing. 21 groups (including the 6 FRM_4DOAS partners) representing a total of 61 (MAX-)DOAS instruments operated worldwide expressed their interest and willingness to provide their radiance spectra to the FRM_4DOAS centralised processing system and to be involved in future community efforts for improving (MAX-)DOAS standards. The evaluation of the received questionnaires showed that most instruments meet the technical specifications for being included in FRM_4DOAS but efforts should be put on instrument calibration and data process automation. It is also important to note that almost all of them expressed difficulty for financially supporting their (MAX-)DOAS measurements in a long-term perspective.

The second half of the main phase of the project has been mostly dedicated to the development of the central processing system code. Based on the technical requirements that were established for such a processing system by the FRM_4DOAS consortium, it has been decided to implement an event-driven architecture which uses a succession of asynchronous modules (e.g. QDOAS or stratospheric NO_2 profiling modules) triggered by the presence of queues which contain a continuously updated list of input files to process, the execution of a module being controlled by an independent entity called wrapper. It should be noted that significant effort has been put on the modularity of the code and associated routines, so that all modules can work independently and new tools like e.g. a new version of a profiling algorithm can be easily plugged in.

The processing system has been tested first on daily submitted radiance spectra files from 9 of the 14 selected partners' stations. Based on this daily processing, a technical verification has been performed, in particular the successful and timely creation of log, history, and output files corresponding to the different steps has been thoroughly checked. In a second step, CINDI-2 spectra files from a selection of instruments have been ingested and the retrieval results have been used for the validation of the processing system through comparison to data available from the campaign. Overall, the FRM_4DOAS MAPA and MMF profiling algorithms worked well on the CINDI-2 data sets but also on the University of Bremen profiles retrievals at three stations used as additional source of validation data.

As part of project CCN02 and CCN03, the NDACC MAX-DOAS Service has been developed based on this prototype central processing system. The main technical development steps included the optimisation of the prototype processor and its conversion into an automated central facility, an assessment of the products maturity through new extended verification and validation exercises, the evaluation of the central processor performance in terms of computing time and data storage, the set-up of the NDACC and EVDC databases for accepting FRM_4DOAS files, and the development of a level-2 data download web page for spectra file submitters accessible from the project website. In addition to the technical aspects, significant effort was also put in the creation of the administrative framework for future service upscaling, like the assignment of DOIs to the different data sets, the set-up of a user data policy based on the Creative Commons licensing approach, and the drafting of a new NDACC affiliation and certification procedure protocol for MAX-DOAS instruments.

The NDACC MAX-DOAS Service has been launched in November 2020, with the start of daily submission of GEOMS HDF files of tropospheric NO_2 and total O_3 products for 14 partners' stations to the NDACC rapid delivery database, with mirroring on EVDC. The other FRM_4DOAS products (tropospheric HCHO and stratospheric NO_2 vertical profiles) have been assessed as not mature enough for public release. They will be further consolidated as part of the ESA FRM_4DOAS-2.0 R&D and Copernicus operational follow-up projects.

Introduction

This document is the final report of the ESA project FRM4DOAS (Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observation; ESA Contract n° 4000118181/16/I-EF). The aim of this project was to further harmonize MAX-DOAS systems and data sets, through the

- Specification of best practices for instrument operation and calibration
- Demonstration of a centralised NRT (near-real-time/6-24h latency) processing system for MAXDOAS instruments operated within the international Network for the Detection of Atmospheric Composition Change (NDACC)
- Establishment of links with other UV-Visible instrument networks, e.g. PGN (Pandonia Global Network; see https://www.pandonia-global-network.org/)
- Support to the CINDI-2 campaign

Although for this project, the target species were tropospheric and stratospheric NO_2 vertical profiles, total O_3 columns, and tropospheric HCHO profiles and the central processing system was tested on a limited number of stations (see Table 1 below) and on CINDI-2 intercomparison campaign data, one of the major objectives was to collect and create the necessary information, guidelines and infrastructure which can be the basis for a more comprehensive network including many more MAX-DOAS instruments and covering a larger number of MAX-DOAS products. Such reference network will play a crucial role in the validation of current and future atmospheric composition satellite missions, in particular the ESA Copernicus Sentinel missions S-5P, S-4, and S-5.

Table 1: MAX-DOAS sites integrated in the demonstration central processing system.

Location	Lat (°N)	Long (°E)	Alt (m) a.s.l	Owner
Ny-Alesund, Norway	79	12	20	IUP-UB
Harestua	60	11	600	BIRA-IASB
Bremen, Germany	53	9	46	IUP-UB
De Bilt, The Netherlands	52	5	0	KNMI
Uccle, Belgium	51	4	95	BIRA
Mainz, Germany	50	8	50	MPIC
Heidelberg, Germany	49	8	115	UHEID
Thessaloniki	41	23	80	AUTH
Xianghe, China	40	116	178	BIRA/ IAP-CAS
Athens, Greece	38	24	532	IUP-UB
Izana, Spain	28	-16	2367	INTA
La Réunion Maïdo, France	-21	55	2158	BIRA-IASB
Lauder, New-Zealand	-45	170	370	NIWA
Neumayer	-71	-8	50	UHEID

The project's Work Breakdown Structure (WBS) has been organised around the main technical tasks of the project as illustrated in Figure 1.

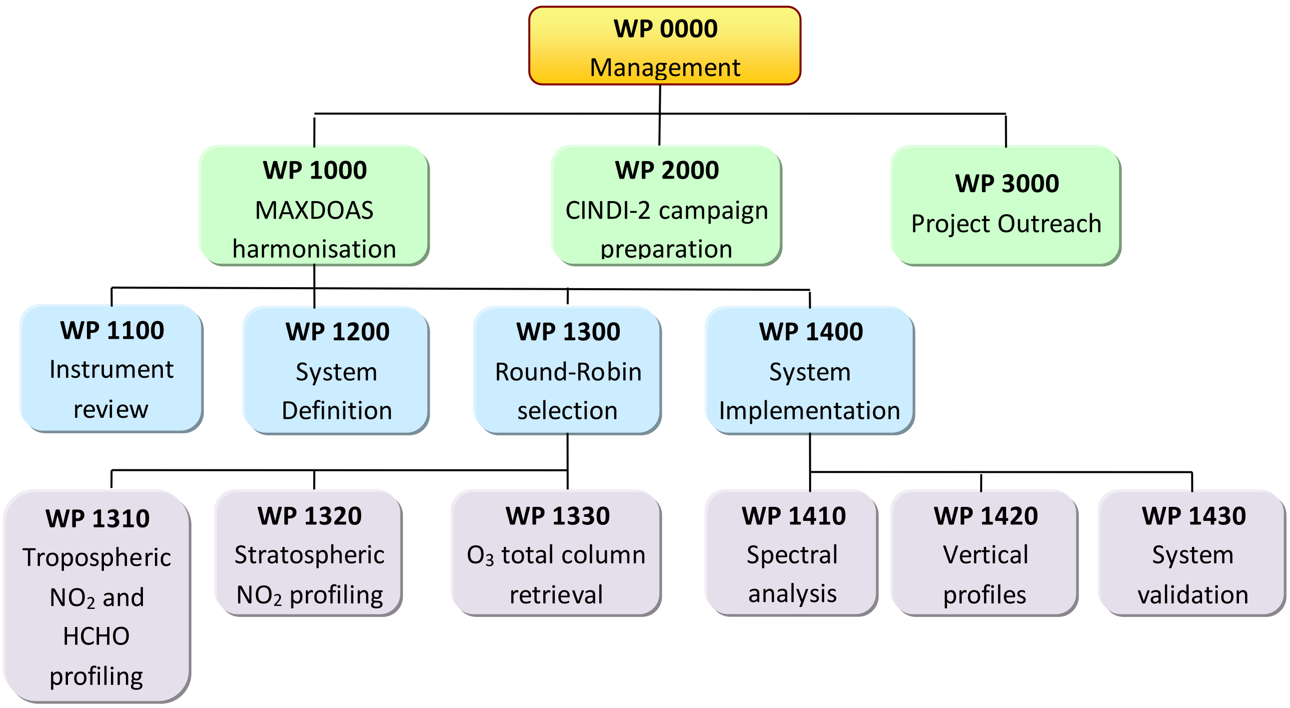


Figure 1: Work Breakdown Structure (WBS).

As part of WP1100, the starting point of the project has consisted in a review of the status of MAX-DOAS instruments and networks, including technological design, scientific quality, reliability, calibration status, documentation, data processing and data availability, peer-reviewed publications. This also included the definition of the technical requirements for an operational MAX-DOAS processing system addressing all necessary issues, i.e. input/output, processing performance, QA/QC, data formatting, etc.

In a second step, a Round-Robin intercomparison exercise of profiling algorithms involving all key actors in MAX-DOAS research in Europe were organised as part of WP 1300. This activity focused on tropospheric NO_2 and HCHO vertical profiling (WP 1310), stratospheric NO_2 vertical profiling (WP 1320), and O_3 total column retrievals (WP 1330). The output of this WP was the selection of baseline algorithms ready for implementation in the processing system.

Based on the output of WP1100 and 1300, the processing system architecture has been defined as part of WP 1200.

The main step of the project has been the development of the centralised processing system code within WP 1400. This code includes modules for DOAS spectral analysis (WP 1410), trace gas vertical profiling (WP 1420), and processing system validation (WP 1430). The final step in WP 1400 was to evaluate the performance of the system and to test it on a selection of partners' stations (see Table 1) and CINDI-2 measurement data sets (WP 1430).

During the first year of the project, some resources were dedicated in WP 2000 to the preparation and planning of the CINDI-2 campaign that was held in September 2016 in Cabauw (The Netherlands).

Preparation tasks included the writing of planning documents and scientific coordination with the local organiser of the campaign (KNMI).

Finally, WP 3000 was dedicated to project outreach.

The main phase of the project ended on 20-21 November 2018 with a final meeting organized in ESRIN. It was then followed by two successive Contract Change Notices (CCN), CCN02 (08/2019-05/2020) and CCN03 (10/2020-03/2021). Based on the prototype version of the FRM_4DOAS Central Processing System developed in WP1400, CCN02 was aiming at the creation of the framework and the development of the operational code needed for a kick-off of the NDACC MAX-DOAS Service. This activity was carried out in the perspective of a service upscaling (more stations and products) expected in the future operational phase of FRM_4DOAS. The main objective of CCN03 was to launch and maintain the NDACC MAX-DOAS Service in a demonstration mode. Demonstration mode meant that GEOMS output files are catalogued on the NDACC and EVDC data handling facilities without any operational commitment with respect to validation servers like CAMS or VDAF. During the CCN03 period, the NDACC MAX-DOAS Service has delivered only the data products that were assessed as mature during CCN02, i.e. lower tropospheric NO_2 vertical profiles and total O_3 columns.

The present final report gives an overview of the main achievements of the FRM_4DOAS phase I project (see Section A) and its successive CCNs (see Section B). Project outreach and conclusions and perspectives are given in Sections C and D, respectively. The project deliverables used as references in this document are listed in the Project deliverables table at the beginning of this report and are publicly available on the FRM_4DOAS web site at https://frm4doas.aeronomie.be/index.php/documents.

A. Main achievements of the FRM4DOAS project's phase I (July 2016-November 2018)

1. MAX-DOAS network and instruments review

The review of the current status of MAX-DOAS instruments and networks is a crucial step for establishing the instrument and network scientific requirements for satellite validation and the instrument calibration and operation best practices. The results of this review are extensively described in two deliverables, 'MAXDOAS Instruments Review Document' (D3) and 'MAXDOAS Network Scientific Requirements Document' (D2). In the present report, we will give only the main outcomes of these review activities.

1.1 MAX-DOAS instrument and network scientific requirements

1.1.1 Instrument requirements

Instrument requirements for MAX-DOAS instruments to be used in satellite validation networks are driven by the requirements on the quantities measured by the satellite, both in terms of the relevance of these quantities and in terms of the necessary precision and accuracy. In many cases, a combined measurement of all relevant quantities is useful as this makes best use of a given infrastructure. Exceptions are monitors for volcanic emissions or other measurements in remote regions such as the Arctic having limited access to shelter, power, and data bandwidth where simple instruments provide advantages.

Some aspects to be taken into account when evaluating instrument requirements are:

- Does the wavelength coverage allow measurement of all species of interest?
- Does the signal to noise ratio (SNR) allow measurement uncertainties low enough to provide meaningful validation for satellite observations?
- Does the instrument perform measurements at different elevation angles in order to separate tropospheric and stratospheric column amounts?
- Are pointing accuracy and FOV good enough to enable profile inversion?
- Can measurements be performed at different azimuth angles to evaluate horizontal gradients?

An overview on qualitative requirements for some instrumental parameters is given in Table 2. These values are based on experience and should not be seen as quantitative threshold requirements.

Table 2: MAX-DOAS Instrument requirements for satellite validation measurements.

	NO_2	HCHO	Glyoxal	SO_2	O_3 UV	O_3 Visible*
Wavelength coverage [nm]	400 – 500	340 – 360	400 – 500	305 – 330	320 – 350	450 - 550
Spectral resolution	< 1 nm	< 0.6nm	< 1 nm	< 0.5 nm	< 0.8 nm	< 1nm
SNR	3000 - 4000	4000	4000	3000	3000	3000 - 4000
Elevation angles trop. Columns	30°, 90°					
Elevation angles profiles	1°, 3°, 5°, 10°, 30°, 90°					
Elevation accuracy profiles	<= 0.2°					
FOV	<= 1.5°					
Solar zenith angles to be covered in twilight geometry	75° – 94°					

* Total column from zenith twilight observations

The accuracy requirements for air quality satellite measurements and how does the ground-based MAX-DOAS instrument accuracy compare to those requirements is also an important aspect for establishing MAX-DOAS instrument requirements. Table 3 presents an overview of the accuracy requirements for air quality satellite measurements as determined in the framework of the Copernicus Sentinel 5 Precursor mission (see https://sentinels.copernicus.eu/documents/247904/2506504/S5P-Level-1b-L2-numbered-validation-requirements.pdf) and the typical accuracy achievable using ground-based DOAS and MAX-DOAS instruments, as extracted from the recent peer-reviewed literature. It can be seen that that for the FRM_4DOAS target species (NO_2, HCHO and total O_3), the accuracy of DOAS/MAX-DOAS measurements currently meets the requirements for satellite measurements. An harmonization effort like FRM_4DOAS, through a centralized processing using harmonised settings and the specification of instrument operation best practices could therefore further decrease the accuracy threshold values of MAX-DOAS measurements listed in Table 3.

Table 3. Accuracy requirements for satellite measurements and corresponding accuracy estimates for DOAS and MAX-DOAS measurements (based on the literature).

Species	Data Product	Accuracy requirements for satellite measurements	Theme	Accuracy of DOAS/MAXDOAS measurements
Ozone	Total column	3.5-5%	A3/B1	5% (Hendrick et al., 2011)
NO2	Stratospheric column	<10%	A3	<10% (Hendrick et al., 2004)
	Tropospheric column	25-50%	B1/B3	15% (Hendrick et al., 2014; Vlemmix et al., 2011)
HCHO	Tropospheric column	40-80%	B1/B3	20% (Franco et al., 2015; Vigouroux et al., 2009).
SO2	Enhanced stratospheric column	30-50%	A3	--
	Tropospheric column	30-50%	B1/B3	25% (Wang et al., 2014)
Glyoxal	Tropospheric column	1.2e14 molec/cm^2 or 60%	B1	30% (Sinreich et al., 2010; Mahajan et al., 2014).

A3 – Ozone layer assessment
B1 – Air quality protocol monitoring
B3 – Air quality assessment

The other instrument requirements which have been identified in the project are the instrument operation requirements like e.g. measurement time and frequency which need to be adapted to satellite overpasses, the regular monitoring of instrument performance, the data evaluation, format, and delivery. Further details on those requirements can be found in the deliverable D2.

1.1.2 Network requirements

The geographical layout of an optimal MAX-DOAS validation network is determined by the spatial variability and distribution of the parameters of interest. Species with long atmospheric lifetime, such as CO_2, need fewer stations than reactive gases such as NO_2. Thorough validation requires a spatial distribution of the measurements covering:

- Both hemispheres
- All relevant latitudes from the tropics to polar regions
- Background regions and sites where high concentrations are expected
- Regions having different conditions with respect to parameters which can potentially affect measurement quality such as albedo, cloud cover, aerosol loading, topography

Which regions to target for appropriate sampling of the range of measurement values depends on the trace gas or measurement quantity of interest. Relevant regions have been identified in FRM$_4$DOAS and an overview of those is shown in Table 4.

Table 4: Regions of interest for satellite validation measurements.

Species	Background	Hot spot
NO_2	• Rural areas on all continents • Oceanic areas	• Industrial areas on all continents • Shipping regions • Biomass burning areas in Europe, South America, Africa, Asia • Soil emission areas (Savannah, agricultural areas) • Lightning regions, if possible separated from other sources
Formaldehyde and Glyoxal	• Sparsely vegetated area • Oceanic regions • Desert regions	• Biogenic emission regions (boreal forests, rain forests) • Biomass burning areas in Europe, America, Africa, Asia • Industrial hotspots (Po valley, PRD, Houston area, ..) • Continental outflow areas
SO_2	• Rural areas • Oceanic regions	• Volcanic emission regions (degassing + explosive) • Industrial hot spots (ore mining, power plants, shipping regions) • Oil and gas production regions
Ozone	• Rural regions • Oceanic regions • Polar regions (ozone depletion events)	• Biomass burning regions • Continental outflow regions • Industrial regions

Ideally, the measurement locations should be chosen to provide data representative for an area of the size of the satellite pixel or larger. Since the satellite pixel size has decreased in recent years (e.g. 3.5 x 5.5km² of the TROPOMI instrument), finding stations representative for a full satellite pixel but also finding satellite pixels measuring a similar quantity as the ground-based observations have been made easier. However, it should be noted that for small satellite pixels, the spatial averaging by MAX-DOAS observations (5 – 20 km) may become a limiting factor.

The considerations listed above apply mainly for an operational validation network. It has to be complemented by validation campaigns providing data from additional instruments and also moving platforms in order to better characterize spatial gradients or for example the latitudinal distribution. For MAX-DOAS measurements in particular, the nature of the network (a heterogeneous network of different instruments funded and operated by various institutes and universities) limits the applicability of the criteria listed in Table 4 as only part of the instruments are deployed with satellite validation being the main application target.

1.2 MAX-DOAS instruments and networks review

A large number of UV-Vis DOAS (Differential Optical Absorption Spectroscopy) instruments are operated worldwide for the regular monitoring of NO_2, O_3, HCHO, and several other species. While the overall measurement principle of DOAS is the same for all instruments, there are many different approaches to instrument design and operation. These differences are driven by the various scientific applications for which the instruments are developed (stratospheric research, air pollution monitoring, monitoring of volcanic emissions, power plant emission monitoring, process studies, …), but also by cost and ease of deployment which are important factors for the establishment of networks. As many of the instruments were designed and built by individual research groups, there is a wide diversity of instruments (e.g. 'research grade' versus commercial spectrometers) in operation today, and comparability of measurement results is an issue that needs to be addressed for integration of data from all these instruments into a centralised processing system.

There are different ways of categorizing DOAS instruments, possible options being viewing modes, target quantities, size and quality of spectrometers and detectors used, or their participation in networks. In FRM_4DOAS, a mixed approach is taken, separating the instruments by their viewing options (MAX-DOAS, zenith-sky and direct-sun) but also by existing or developing networks (MAX-DOAS, PGN, SAOZ). It is based on information collected as part of the activities of the NDACC (Network for the Detection of Atmospheric Composition Change) UV-vis Working Group (see http://ndacc-uvvis-wg.aeronomie.be/) and the EUMETSAT Atmospheric Composition (AC) SAF, and the NIDFORVAL S-5P validation project coordinated by BIRA-IASB.

The geographical distribution of operational SAOZ/Zenith-sky DOAS instruments as on January 2018, as well as MAX-DOAS and Pandora systems, is provided in Figure 2. The corresponding list of instrument can be found in deliverable D3.

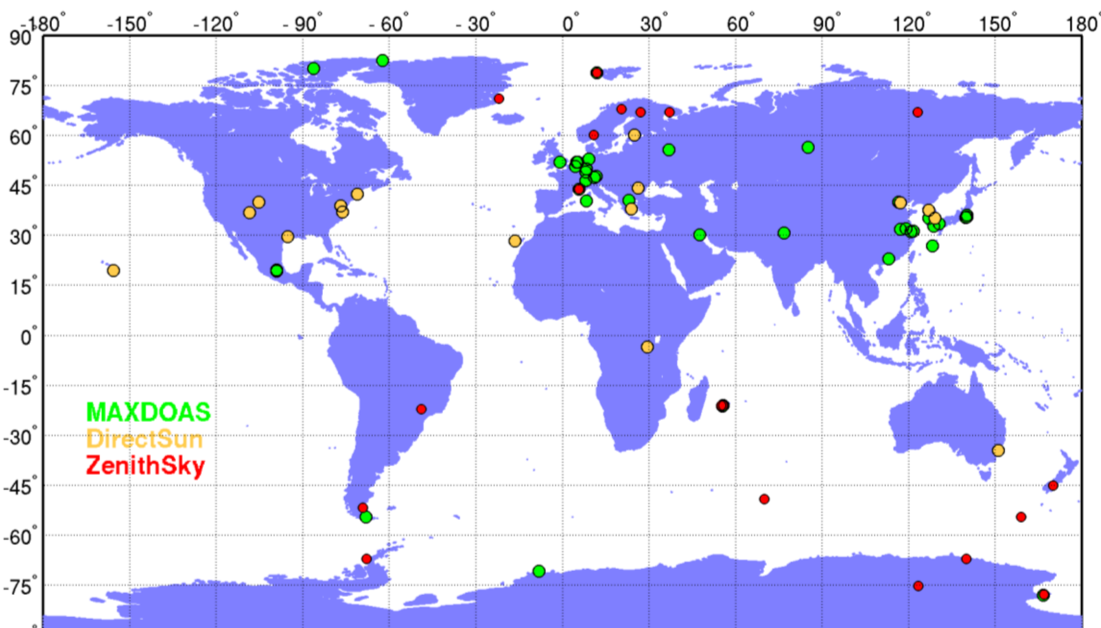


Figure 2: Geographical distribution of MAX-DOAS, PGN (direct sun), and SAOZ/Zenith-sky DOAS monitoring sites in operation globally as on January 2018.

1.3 FRM_4DOAS demonstration stations

1.3.1 Technical Readiness Level evaluation

A crucial step when considering an instrument for inclusion in the FRM_4DOAS processing system is the evaluation of its Technical Readiness Level (TRL). This is important to a) understand the type and status of the instrument, b) evaluate the usefulness of the instrument for the network and c) provide feedback to the instrument owners with respect to possible development needs. Within FRM_4DOAS, it has been decided to collect information (instrument technical characteristics and calibration/operation procedures) for the TRL evaluation through a dedicated questionnaire which is available on the FRM_4DOAS website at http://frm4doas.aeronomie.be/index.php/frm4doas-questionnaire. In addition to instrument type, location, corresponding principle investigator, and institution contact details, it includes questions about:

- Instrument details (spectral resolution and coverage, fields of view, pointing abilities and speed, type of spectrometer and detector, cooling of detector, temperature stabilisation of instrument, …)
- Calibration and characterisation of instrument (slit function, straylight, FOV, pointing accuracy, …)
- Instrument operation procedure (automatic operation and calibration, QA/QC tests, operation documentation,…)
- Procedures for data transfer from the station to the institution
- Procedures for spectra calibration
- Questions about the willingness to be part of the FRM_4DOAS processing system and possible constraints
- List of publications and projects where corresponding data are used

The technical specifications collected for the CINDI-2 campaign (see deliverable D13) have been also used as additional source of information in the evaluation. It is important to point out that depending on the intended application of a DOAS instrument, very different approaches can be taken for the set-up and operation of the instrument. Also, again depending on the field of application, different levels and means of instrument characterisation are needed. Here, the focus is on the use of data from DOAS instruments for a centralised processing in view of satellite data validation. Therefore, the technical readiness for the FRM_4DOAS processing should not be confused with a quality judgement, let alone an assessment of the scientific quality of measurements performed with the instrument.

The TRL evaluation of the FRM_4DOAS stations can be found in deliverable D3. It shows that the relevant criteria for instrument specifications, characterization and calibration, and operation are met to a large extent by all FRM_4DOAS demonstration instruments.

1.3.2 Potential network extension

In a second step, the potential for extending the FRM_4DOAS centralised processing to additional stations has been evaluated by circulating the questionnaire to the whole DOAS Community via e-mail (an official questionnaire release e-mail was sent by ESA on 04/05/2017) but also through dedicated presentations at major conferences/workshops (e.g. EGU2017, 8th International DOAS Workshop in Yokohama). So far, 21 groups (including the 6 FRM_4DOAS partners) representing a total of 61 (MAX-

)DOAS instruments operated worldwide expressed their interest in being part of the FRM$_4$DOAS processing and provided their self-assessment to BIRA-IASB. The geographical distribution of potential FRM$_4$DOAS stations is presented in Figure 3; the corresponding list of stations can be found in deliverable D3.

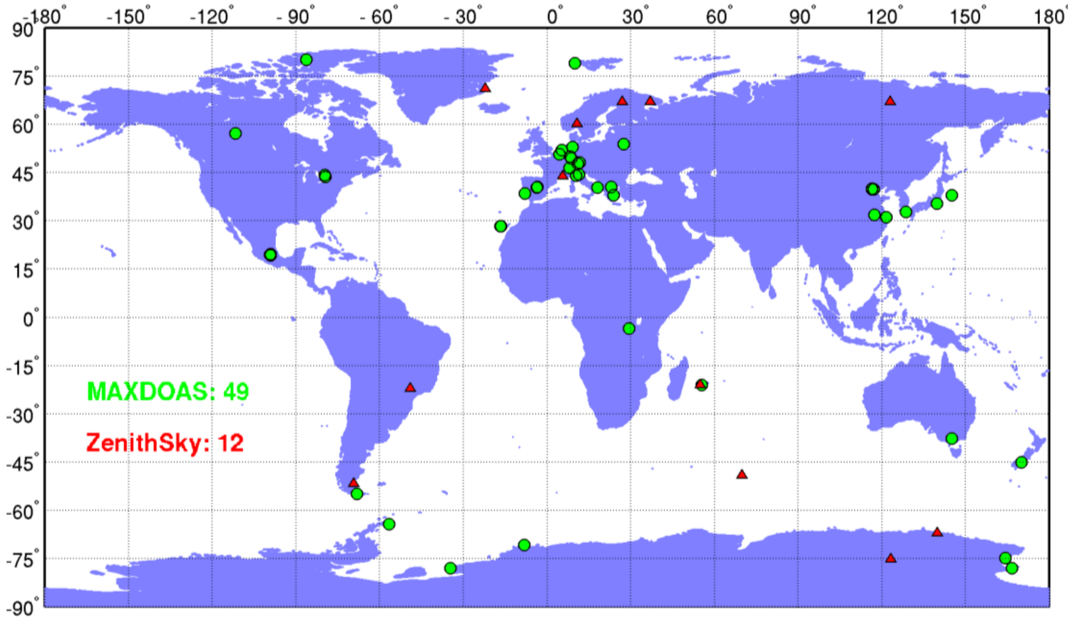


Figure 3: Potential stations that could be included in the FRM$_4$DOAS processing system according to the replies received to the questionnaire (status as on January 2018).

All groups expressed their willingness to provide their radiance spectra to the FRM$_4$DOAS centralised processing system and to be involved in future community efforts for improving (MAX-)DOAS standards. The evaluation of the received questionnaires shows that most instruments meet the technical specifications for being included in FRM$_4$DOAS but efforts should be put on (1) the characterization and calibration of the instruments, and (2) the automation of the processes producing calibrated radiance spectra ready for DOAS analysis. In that sense, calibration activities carried out during the CINDI-2 campaign should contribute to improve point (1) since most of those groups have participated in CINDI-2. Finally, it is also important to note that almost all of them expressed difficulty for financially supporting their (MAX-)DOAS measurements in a long-term perspective.

2. CINDI-2 campaign

In parallel to the development of the processing system which was the main task in FRM$_4$DOAS, some resources were also dedicated to the preparation and planning of the CINDI-2 campaign which took place in September 2016 at the CESAR site in Cabauw (The Netherlands). This campaign was a follow-up of the successful CINDI campaign (Piters et al., 2012) and focused on the Intercalibration of a large number of DOAS and MAX-DOAS systems in view of their contribution to the validation of air quality satellite sensors (in particular the Sentinels 4, 5 and 5P). Preparation tasks included the scientific coordination with KNMI which was the local organiser of the campaign, and the writing of the following planning documents:

- Intercomparison Campaign Requirements Document (D11)
- Intercomparison Campaign Technical Requirements Document (D12)
- Intercomparison Campaign Planning Document (D13)
- Intercomparison Campaign Data Protocol (D14)

CINDI-2 MAX-DOAS and ancillary measurement data sets have been used for the Round-Robin exercise and the validation of the FRM_4DOAS centralised processing System, as shown in Sections 4.2.2.7 and 5.5.2. CINDI-2 activities also strongly contributed to the TRL and harmonisation level evaluations of the current MAX-DOAS network and to the definition of the MAX-DOAS best practices, especially in terms of calibration procedures (see Kreher et al., 2020).

The off-line centralised QDOAS processing of CINDI-2 spectra performed by BIRA-IASB in the framework of the semi-blind intercomparison study allowed to optimise the DOAS analysis settings to be implemented in FRM_4DOAS. Moreover, it also showed that a centralised processing can improve the overall agreement between the different groups as illustrated in Figure 4.

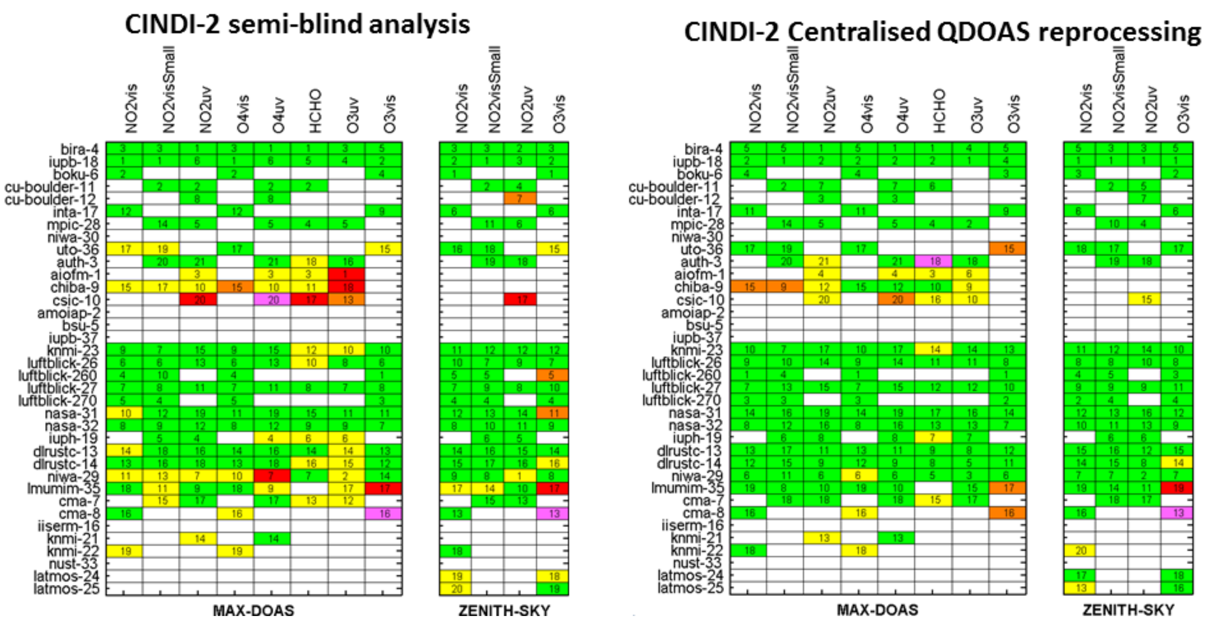


Figure 4: Assessment matrix based on the semi-blind analysis (left plot) and on the centralised QDOAS reprocessing (right plot) for all 36 CINDI-2 MAX-DOAS instruments. Green indicates that all three selected assessment criteria have been fulfilled, yellow means that one criterion is not satisfied, orange means two are not, red means all 3 criteria have not been met and pink indicates that this data set has at least one extreme outlier. In each data product category, groups are sorted by increasing values of the median DOAS fit RMS. A white cell indicates that no data is available.

3. MAX-DOAS Best Practices Definition

Guidelines and recommendations for best practice approaches to calibration and operation of MAX-DOAS instruments were collected based on partners' experience and outcomes from the previous (MAX-)DOAS intercomparison campaigns, especially CINDI-2. These best practices are extensively described in deliverable D4 ('MAX-DOAS Calibration and Operations Best Practices'). From this document, which is expected to remain a living document with corrections, extensions and other changes implemented by the MAX-DOAS community in the coming years, a quick and concise check-list with the most important requirements has been created and is described below. This check-list is available at https://frm4doas.aeronomie.be/ProjectDir/Guidelines/ FRM4DOAS_Operation_Calibration_Guidelines _v1_1_20180316.pdf) and is summarised below.

3.1 Instrument guidelines

- Wavelength resolution better than 0.8 nm (UV) and 1.5 nm (visible)
- Sampling better than 5 (i.e. <0.16 nm for UV and <0.3 nm for visible)
- FOV of 1.5° or better in vertical direction
- GPS signal / time server for proper time information
- Quartz fibre / polarisation scrambler for removal of polarisation features
- Optical low pass filter for UV instruments for straylight removal
- Wavelength coverage should follow NDACC / CINDI-2 recommendations as far as possible. At least one O_4 band covered well.

3.2 Operation guidelines

- SZA range up to 85° for MAX-DOAS and up to 94° for zenith-sky observations
- Zenith-sky observations at least every 30 minutes during MAX-DOAS observations
- At least one zenith-sky measurement per degree SZA at twilight
- At least 1°, 2°, 3°, 5°, 10°, 30° elevation in scan
- Off-axis viewing directions away from the sun (if possible)
- Dark signal daily (or continuously if possible with instrument, see below for details)
- Slit function on a regular basis (daily but at least once per year, see below for details)
- Horizon scan on a regular basis (daily but at least once per week, see below for details)

3.3 Data processing guidelines

- Apply dark signal correction
- Apply wavelength calibration using line lamp or Fraunhofer atlas
- Apply non-linearity correction if necessary
- Average spectra to reach SNR of at least 3000 (vis) or 4000 (UV)
- Use prescribed format for spectra (see http://frm4doas.aeronomie.be/ProjectDir/Guidelines/L1_format_20180308_v1.pdf for details)

3.4 Instrument calibration guidelines

3.4.1 Before first operation

- Straylight if possible
  - Measurements of white light source with and without cut-off filters

- Nonlinearity
  - Simple white light source or very clear day zenith observations
  - Measurements at different exposure times
  - Ratio of dark signal (and smear) corrected intensities as function of larger intensity
- Polarisation dependency
  - Simple white light source with polariser mounted in turntable
  - Measurements at different polariser positions
  - Ratio to (arbitrary) reference measurement
- Slit function including T-dependence if not T stabilised
  - Line lamp with many emission lines Or line lamp at different grating positions
  - If necessary: Variation of instrument temperature over expected T-range
- FOV
  - Simple white light source (with diffuser and aperture if needed)
  - Sufficient distance to light source (> 3 m)
  - Vertical and horizontal scan in at least FOV / 10 ° steps
- Pointing accuracy
  - Strong light source at know position and altitude (if possible)
  - Horizon scan (in different azimuths if possible)
  - Solar scan at different solar elevation and azimuth angles (if possible)

3.4.2 During operation

- Dark signal daily (or continuously if possible with instrument)
  - Minimum 2 measurements (very short and very long exposure time)
  - Or all used exposure times
  - Average at least over 20 measurements
- Slit function on a regular basis (daily but at least once per year)
- Horizon scan on a regular basis (daily but at least once per week)
  - Short integration time measurements from at least –3° … + 3° in 0.1° steps towards unobstructed horizon
- If possible: regular solar scans
  - At different SZA and VZA angles,
  - both in the vertical and horizontal
  - To derive pointing accuracy and FOV

4. FRM_4DOAS Round-Robin exercise

The objective of the MAX-DOAS algorithm Round-Robin within WP1300 was to review the strengths and weaknesses of the MAX-DOAS profile retrieval algorithms available in the scientific community in order to jointly define a community algorithm that has been implemented as baseline in the centralized processing system. In addition to HCHO and NO_2 profiles, the ability of the retrievals to reconstruct the aerosol profile needed to be assessed since aerosol profiles retrieved from MAX-DOAS O_4 measurements represent an important intermediate data product that serves as input for the trace gas profile inversion.

The definition of the community algorithm was based on intercomparison exercises using synthetic slant column densities as described in the next sections. The ability of the algorithms to reconstruct vertical profiles is assessed based on the agreement between initial and retrieved quantities including tropospheric columns, surface concentrations and the overall agreement between initial ('true') and retrieved profile shapes. The results of these intercomparison exercises are presented in deliverable D5 and in Frieß et al. (2019).

It should be noted that we did not proceed to a Round-Robin selection for the total O_3 column and stratospheric NO_2 profile retrieval algorithms given their much higher level of maturity compared to the MAX-DOAS profiling tools and the already existing community consensus on the corresponding retrieval methods and settings.

4.1 Round-Robin strategy

The overall intercomparison strategy is depicted in Figure 5 and consisted of the following steps:

1. The first step of the intercomparison exercise was a comparison of the forward models on which the individual retrieval algorithms are based. A set of atmospheric scenarios and viewing geometries, for which all FRM_4DOAS participants simulated slant column densities (SCDs) of HCHO, NO_2 and O_4, has been prescribed as described in Section 4.1.2. This exercise allows for a quantitative comparison of the individual radiative transfer models (RTMs) on which the inversions are based.
2. A reference dataset of differential slant columns (dSCDs) of HCHO, NO_2 and O_4, calculated using the median of the ensemble of SCDs from step 1, has been compiled and distributed among the FRM_4DOAS participants.
3. The comparison of the profiles retrieved based on the reference dataset with the initial ('true') aerosol and trace gas profiles will allow for a quantitative quality assessment of the inverse models.
4. Benchmark tests allow for an assessment of the overall numerical performance of the individual retrieval algorithms.
5. Important information on aspects affecting the capability of the retrieval algorithms for being integrated into a community algorithm, including operating system, platform/programming language and licensing issues, have been collected from the participants using an assessment forms .

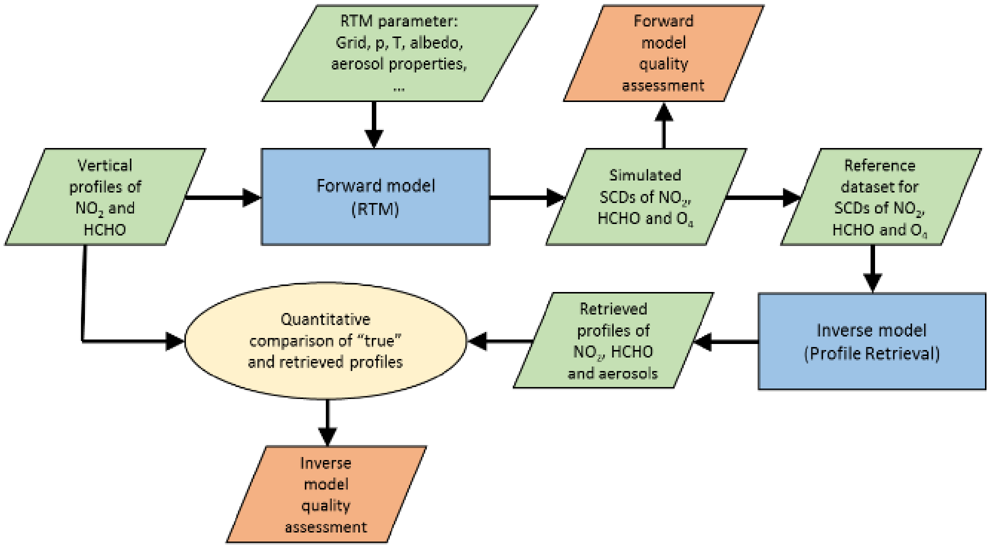


Figure 5: Flow diagram depicting the strategy for the retrieval algorithm intercomparison.

4.1.1 Participating algorithms

The retrieval algorithms available within the FRM_4DOAS team, together with the underlying RTM, are listed in Table 5. Two versions of the MPIC-MAPA algorithm have been involved in this intercomparison exercise, namely a version implemented in Matlab and a new version implemented in Python with a new numerical solver based on a Monte Carlo approach (referred to as MPIC_PARAM and MPIC_PAR_MC, respectively). A detailed description of each algorithm can be found in deliverable D5.

Table 5: List of available RTM and retrieval algorithms within the FRM_4DOAS consortium

Participant	Method	Algorithm	Forward Model
BIRA-IASB	OEM	BePro	LIDORT
BIRA-IASB	OEM	MMF	VLIDORT
IUPUB	OEM	Boreas	SCIATRAN
IUPHD	OEM	HEIPRO	SCIATRAN
KNMI	Parametrised		DAK
MPIC	Parametrised	MAPA	McArtim
MPIC	Parametrised	MAPA-MC	McArtim
MPIC	OEM	Priam	SCIATRAN
NASA	OEM	Quicklook	N/A

Algorithms involved in the Round-Robin intercomparison be separated into those that retrieve vertical profiles on a finite vertical grid using the optimal estimation method (OEM; Rodgers, 2000) and parametrised algorithms that use a small number of parameters (typically 2-4) to describe the shape of the atmospheric profile. Parametrised algorithms are typically faster than OEM algorithms, since they are usually based on precalculated lookup tables, while OEM algorithms rely on on-line radiative transfer modelling (RTM). Being based on Bayesian statistics, OEM algorithms have the advantage of providing a thorough error analysis as well as a quantitative characterization of the vertical resolution and the information content (Rodgers, 2000). In addition to OEM and parametrised approaches, a fast algorithm developed by NASA, which relies only on geometrical considerations and does invoke any radiative transfer modelling took also part of the Round-Robin intercomparison.

4.1.2 Round-Robin settings

The atmospheric scenarios for the modelling of SCDs of NO_2, HCHO and O_4 were mainly based on specifications agreed for the CINDI-2 profiling activities. The selected HCHO and NO_2 profiles and the aerosol vertical profiles are shown in the left and right panels of Figure 6. The profile shapes of Aer0…Aer7 are similar to the corresponding trace gas profile shapes. In addition, Aer8…Aer10 represent extreme cases with a 200 m fog layer as well as clouds at 1 km and 5 km altitude, respectively.

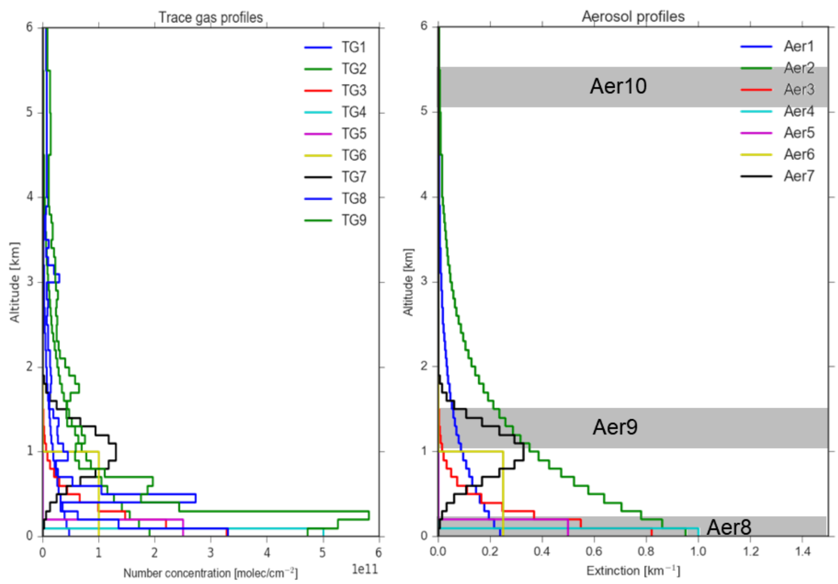


Figure 6: Trace gas (left) and aerosol extinction (right) profiles for the RTM intercomparison. Scenarios Aer8 – Aer10 (fog and clouds) each have an extinction of 10 km^{-1}.

The other settings (vertical altitude grid, wavelengths, viewing geometries (elevation, relative azimuth, and solar zenith angles), cross-section data sets, aerosol single scattering albedo and phase function asymmetry parameter, a priori profile and corresponding covariance matrices for OEM-based algorithms, etc) are extensively described in deliverable D5.

The following scenarios have been simulated:

- O_4 SCDs for each aerosol profile (Aer0…Aer10), i.e. 11 model atmospheres. Apart from O_4, the atmosphere should contain no trace gases (i.e., TG0).
- Trace gas SCDs (HCHO at 343 nm and NO_2 at 460 nm) for each combination of trace gas and aerosol profile. This yields 10 x 11 = 110 model atmospheres.

For profile retrieval, a reference dataset of O_4 (360 and 477 nm), NO_2 and HCHO dSCDs has been compiled based on these simulated SCDs. It contains the ensemble median dSCDs from all participants, together with values for typical retrieval errors as reported by the participants of the CINDI-2 campaign. The reference dataset contains dSCDs at 9 elevation angles for 990 different combinations of trace gas profiles, aerosol profiles and sun position (SZA and RAA). Initially, the participants had no knowledge on the true vertical profiles (blind intercomparison). This has been achieved by distributing a reference dataset where a random number has been assigned to the dSCDs of a particular atmospheric scenario. Trace gas and aerosol profiles were not specified in the initial reference data file and were thus not known to the participants.

Two reference datasets were created, one only containing the median dSCDs without any noise, and a second dataset with a normally distributed noise of 5% of the dSCDs. This noise is much higher than the typical noise from the DOAS analysis, with the aims to account for possible atmospheric variabilities, such as horizontal inhomogeneities and broken clouds.

4.2 Results

4.2.1 Comparison of simulated DSCDs

The ability of the forward models to realistically simulate trace gas SCDs has been assessed based on a comparison of the individual simulations with the ensemble median SCDs for each aerosol and trace gas scenario.

As an example, Figure 7 shows the comparison of the simulated NO_2 SCDs, with the ensemble mean for each participant except NASA, who did not report modelled SCDs. For all trace gases, most forward models yield equal SCDs under all conditions. IUPHD and MPIC-PRIAM, both using the (outdated) SCIATRAN version 2.1 as forward model, show deviations in case of fog and clouds (scenarios AER8 .. AER10), the shallow box profile (AER4), and in case of IUPHD also the exponential profile AER2.

Figure 7: Comparison of the modelled NO_2 SCDs from the individual participants to the ensemble median (x-axis). Colours indicate the aerosol profile as denoted in the legend.

Slope, intercept and regression coefficient for the comparison of O_4, NO_2 and HCHO to the ensemble median are listed in Table 6. For most cases, slope and regression coefficient deviate from unity by less than 1%.

Table 6: Slope, intercept and correlation coefficient for the different trace gas species and forward models (modelled SCDs versus ensemble median). Colours from yellow to red indicate the deviation from one and zero, respectively.

	NO_2			HCHO			Aerosol - 360 nm			Aerosol - 477 nm		
	Slope	Intercept [10^{17} cm^{-2}]	R	Slope	Intercept [10^{17} cm^{-2}]	R	Slope	Intercept [10^{43} cm^{-2}]	R	Slope	Intercept [10^{43} cm^{-2}]	R
BIRA	0.9863	-0.0060	0.9997	1.0008	0.0004	1.0000	0.9895	0.0302	0.9998	1.0002	-0.0067	0.9998
IUPB	1.0016	-0.0027	0.9999	1.0007	0.0003	1.0000	0.9890	0.0262	0.9999	0.9964	-0.0086	0.9999
IUPHD	0.9569	0.0158	0.9986	1.0087	-0.0003	0.9978	1.0377	-0.0404	0.9867	1.0665	-0.0509	0.9871
KNMI	1.0062	-0.0003	1.0000	1.0009	-0.0008	1.0000	1.0006	0.0067	1.0000	1.0078	-0.0233	1.0000
MPIC_PARAM	0.9964	0.0043	0.9999	0.9600	0.0164	0.9987	1.0008	-0.0025	0.9999	0.9565	0.1494	0.9994
MPIC_PRIAM	1.0439	-0.0093	0.9977	1.0519	-0.0131	0.9960	1.0227	0.0446	0.9897	1.0706	-0.1664	0.9782

4.2.2 Vertical profile intercomparison

Vertical profiles of aerosols and trace gases retrieved using the reference dataset described in Section 4.1.2 have been qualitatively compared to the 'true' profiles that were used for the simulation of the dSCDs. This comparison allowed for quantitatively assessing the ability of the individual algorithms to retrieve the atmospheric state from MAX-DOAS measurements.

4.2.2.1 Retrieval versions and available data

Retrievals were performed for modelled dSCDs with and without noise, and both for common settings as well as (optionally) for settings optimised individually by each participant. These retrieval runs were distinguished by a version number as detailed in Table 7. Optionally, each profile can be labelled with a Boolean validity flag which is defined individually by each participant.

Table 7: List of retrieval versions.

Version	Settings
v1	Common settings, SCDs without noise
v1n	Common settings, SCDs with random noise
v2	Free settings, SCDs without noise
v2n	Free settings, SCDs with random noise

Table 8 lists the data submitted by each participant. Only 4 out of 9 participants submitted data with free settings (v2 and v2n). No simulated dSCDs from KNMI and from both versions of the MPIC-PAR algorithm were submitted. NASA does not report errors on the retrieved profiles and only submitted HCHO and NO_2 profiles, but no aerosol extinction profiles.

Table 8: List of available data from the individual participants. Note that MPIC-PAR-MC is now called MAPA (see Beirle et al., 2018).

Algorithm	v1			v1n			v2			v2n		
	Profile	Error	SCD	Profile	Error	SCD	Profile	Error	SCD	Profile	Error	SCD
BIRA-BEPRO	✓	✓	✓	✓	✓	✓	✓	✓	✓	×	×	×
BIRA-MMF	✓	✓	✓	✓	✓	✓	×	×	×	×	×	×
IUPUB	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
IUPHD	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
KNMI	✓	✓	✓	✓	✓	✓	×	×	×	×	×	×
MPIC-PAR	✓	✓	×	✓	✓	×	×	×	×	×	×	×
MPIC-PAR-MC	✓	✓	×	✓	✓	×	×	×	×	×	×	×
MPIC-PRIAM	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
NASA	TG only	×	×	×	×	×	×	×	×	×	×	×

4.2.2.2 Comparison of vertical profiles

Retrieved aerosol and trace gas vertical profiles were compared to the true profiles. As an example, Figure 8 shows retrieved vertical profiles of NO_2 using common settings based on dSCDs without noise (v1). The corresponding plots for aerosols, HCHO can be found in deliverable D5.

Based on these figures and regression analysis of retrieved versus true aerosol extinction and trace gas concentrations at all altitude levels, the overall agreement between true and retrieved vertical profiles has been assessed and can be summarised as follows:

- PRIAM as well as BIRA-BEPRO at 477 nm have problems to retrieve the extinction profile in the aerosol-free case.
- Due to the lack of aerosol-free scenarios in its lookup table, the MPIC-PAR algorithm has problems with the AER0 scenario.
- The capability of the algorithms to retrieve a low-lying cloud (AER9) strongly varies, with best results by MPIC-PARAM as well as MPIC-PAR-MC (MAPA), and partly also by BIRA-MMF, IUPHD, IUPB and KNMI
- A cloud at 5 km altitude, i.e. above the retrieval domain (AER10) does not seem to have a significant impact on the resulting extinction profiles, except that BIRA-MMF retrieves some uplifted aerosol layers.
- In general, trace gas profiles exhibit a better agreement than aerosol extinction profiles. However, the OEM algorithms, and in particular BIRA-BEPRO, yield oscillating NO_2 profile shapes for some of the scenarios (in particular TG2).
- IUPHD NO2 profiles show oscillations at altitudes above ~1.5 km for scenarios TG2, TG7, and TG9 if high amounts of aerosols are present (in particular for AER9).
- In order to test how the retrievals deal with layers which are thinner than the retrieval vertical grid, the AER4 and TG4 scenarios consists of aerosol and trace gas surface layers of 100 m thickness, whereas the retrieval grid has a layer height of 200 m. Therefore deviations between retrieved and true profiles are to be expected. However, most algorithms retrieve about half the surface extinction/concentration, which yields the correct VCD.
- From a qualitative inspection of the profiles, it is obvious that the MPIC-PAR-MC (MAPA) and BIRA-MMF algorithms yields the most stable results among the parametrised and OEM algorithms, respectively.

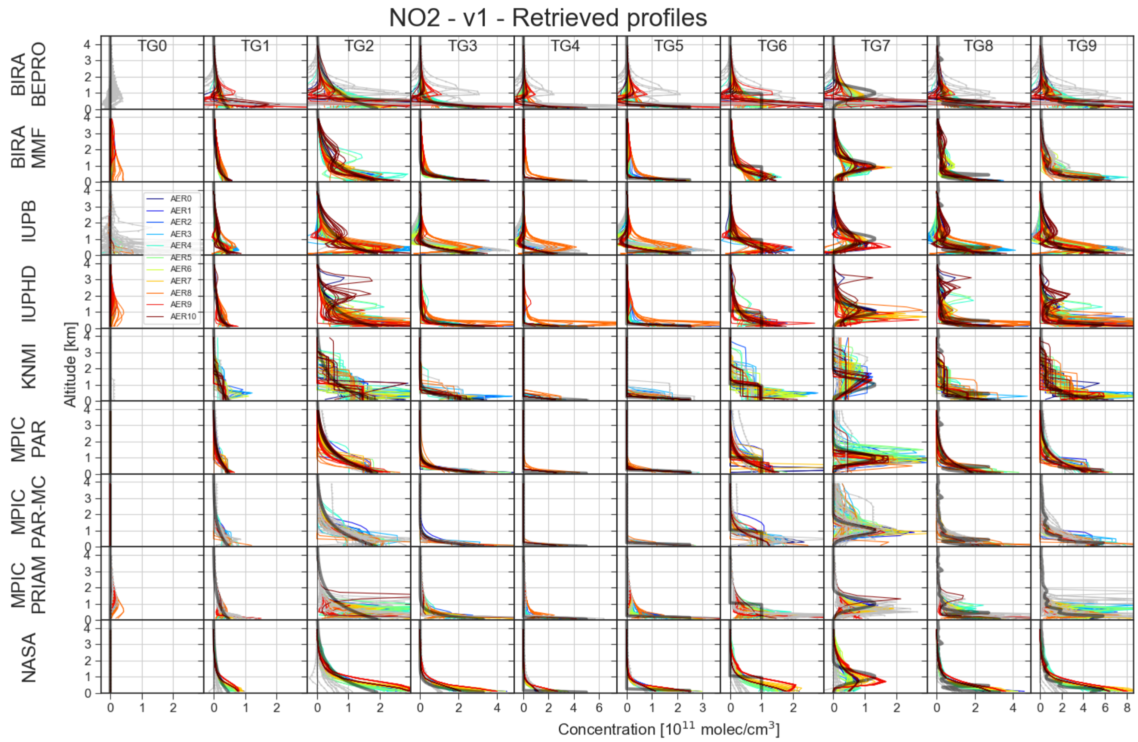


Figure 8: Comparison of retrieved (coloured lines indicating aerosol scenario as denoted in the legend) with true (black lines) NO_2 profiles for retrievals based on SCDs without noise (v1). Profiles flagged as invalid are shown as thin grey lines with coloured dots.

4.2.2.3 Comparison of surface extension/concentration

Since vertical profile retrievals from MAX-DOAS measurements are most sensitive near the surface, most accurate values are expected in the lowermost ~200 m of the atmosphere. Therefore, retrieved aerosol extinction and surface concentrations of HCHO and NO_2 were compared to the true values. As example, the box-whisker plots in Figure 9 shows NO_2 surface concentration comparison results for each trace gas scenario. For both aerosol and trace gases, a quantitative assessment has been performed through regression analyses (see Table 9).

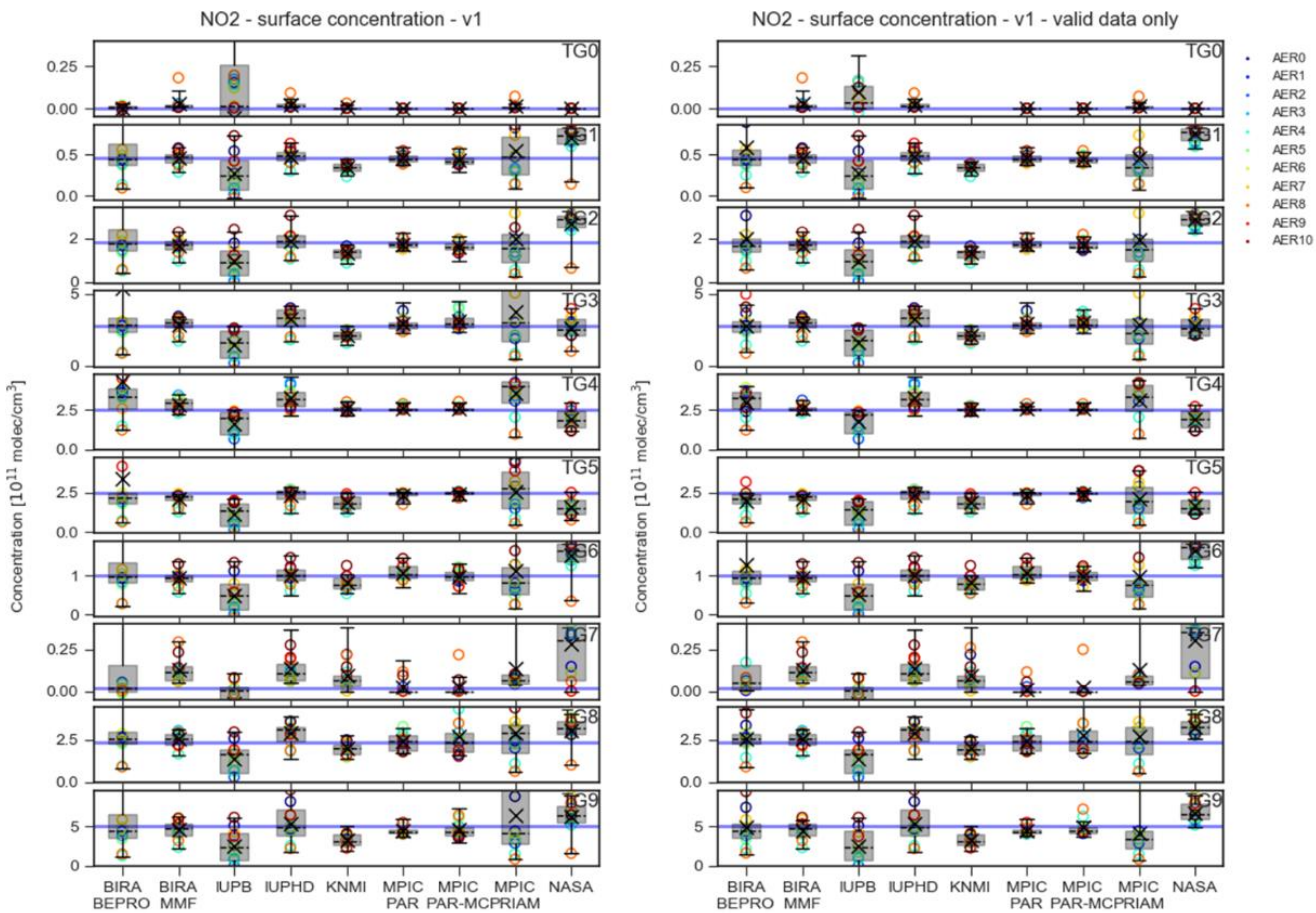


Figure 9: Comparison of NO_2 surface concentration. Thick blue lines indicate the true value. Grey boxes indicate the 25 – 75 % percentile, whiskers the 5% - 95% percentile. Black crosses show the median value and dashed horizontal lines the mean. Small coloured circles indicate the median for each aerosol scenario as denoted in the legend. The left and right panels show statistics based on all data and on data flagged as valid only, respectively.

Table 9: Regression analysis of retrieved versus true aerosol extinction and trace gas concentrations at surface level for the aerosol scenarios AER0 … AER7 (i.e., fog and cloud scenarios excluded).

		Aerosol - 360 nm					Aerosol - 477 nm					HCHO					NO_2				
		N	Slope	Intercept [km-1]	R	RMS [km-1]	N	Slope	Intercept [km-1]	R	RMS [km-1]	N	Slope	Intercept [10^{11} cm-3]	R	RMS [10^{11} cm-3]	N	Slope	Intercept [10^{11} cm-3]	R	RMS [10^{11} cm-3]
V1																					
BIRA-BEPRO	a	720	1.004	-0.001	0.977	0.065	720	-0.250	0.738	-0.078	1.060	720	0.912	0.070	0.955	0.443	720	1.717	0.464	0.280	8.802
	f	720	1.004	-0.001	0.977	0.065	610	0.952	0.013	0.962	0.082	629	0.896	0.117	0.941	0.474	556	0.877	0.210	0.845	0.780
BIRA-MMF	a	720	0.952	0.002	0.967	0.078	720	0.992	0.007	0.953	0.095	720	0.876	0.141	0.949	0.466	720	0.939	0.098	0.938	0.514
	f	720	0.952	0.002	0.967	0.078	720	0.992	0.007	0.953	0.095	720	0.876	0.141	0.949	0.466	676	0.930	0.072	0.945	0.489
IUPB	a	720	0.879	0.011	0.967	0.086	720	0.849	0.019	0.979	0.080	720	0.506	0.045	0.742	1.304	720	0.467	0.093	0.555	1.556
	f	680	0.893	0.008	0.970	0.082	670	0.869	0.015	0.981	0.072	680	0.503	0.055	0.723	1.342	614	0.504	0.060	0.681	1.374
IUPHD	a	720	1.205	-0.021	0.897	0.198	720	1.072	-0.010	0.922	0.139	720	1.043	0.154	0.908	0.739	720	1.067	0.085	0.896	0.805
	f	720	1.205	-0.021	0.897	0.198	720	1.072	-0.010	0.922	0.139	720	1.043	0.154	0.908	0.739	720	1.067	0.085	0.896	0.805
KNMI	a	720	1.157	-0.020	0.979	0.095	720	1.119	0.004	0.914	0.161	720	0.754	0.096	0.950	0.621	720	0.712	0.120	0.902	0.767
	f	600	1.164	-0.029	0.975	0.097	650	1.045	0.021	0.929	0.127	607	0.739	0.146	0.933	0.672	643	0.697	0.164	0.880	0.812
MPIC_PARAM	a	720	0.313	0.426	0.148	0.682	720	-0.949	1.206	-0.187	1.674	720	0.932	0.074	0.986	0.255	720	0.922	0.114	0.957	0.423
	f	720	1.124	-0.020	0.977	0.088	720	1.043	-0.003	0.967	0.084	720	0.897	0.064	0.978	0.341	720	0.968	0.089	0.906	0.659
MPIC_PAR_MC	a	700	1.132	-0.022	0.976	0.089	640	1.032	0.000	0.960	0.087	600	0.911	0.032	0.987	0.277	566	0.954	0.045	0.964	0.391
	f	675	0.480	0.323	0.288	0.505	620	1.024	-0.012	0.962	0.082	714	0.931	0.072	0.986	0.254	712	0.920	0.111	0.959	0.416
MPIC_PRIAM	a	720	0.875	-0.009	0.923	0.129	720	0.667	0.027	0.854	0.187	720	0.924	0.152	0.855	0.825	720	1.033	0.056	0.724	1.439
	f	700	0.877	-0.010	0.920	0.131	630	0.648	0.039	0.814	0.200	695	0.906	0.159	0.855	0.813	615	0.913	0.076	0.761	1.145
NASA	a	0					720	0.599	0.097	0.850	0.174	720	1.123	0.035	0.879	0.943	720	1.152	0.073	0.884	0.979
	f	0					720	0.599	0.097	0.850	0.174	720	1.123	0.035	0.879	0.943	690	1.146	0.070	0.881	0.983
V2																					
BIRA-BEPRO	a	720	0.954	0.021	0.932	0.112	720	0.902	0.002	0.873	0.158	720	0.886	0.133	0.959	0.425	690	1.346	0.379	0.259	7.400
	f	650	0.938	0.014	0.986	0.055	480	0.923	0.017	0.989	0.053	599	0.886	0.096	0.956	0.441	571	0.838	0.161	0.594	1.618
BIRA-MMF	a	0					0					0					0				
	f	0					0					0					0				
IUPB	a	720	0.928	0.003	0.972	0.075	720	0.998	-0.001	0.884	0.158	720	0.497	0.088	0.738	1.291	720	0.522	0.083	0.757	1.242
	f	680	0.937	0.001	0.972	0.074	670	0.901	0.011	0.985	0.061	679	0.492	0.104	0.718	1.330	674	0.522	0.092	0.739	1.270
IUPHD	a	720	1.154	-0.002	0.928	0.158	720	1.075	-0.009	0.937	0.125	720	1.065	0.149	0.924	0.703	720	1.075	0.074	0.897	0.804
	f	720	1.154	-0.002	0.928	0.158	720	1.075	-0.009	0.937	0.125	720	1.065	0.149	0.924	0.703	720	1.075	0.074	0.897	0.804
KNMI	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PARAM	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PAR_MC	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PRIAM	a	720	0.916	0.018	0.947	0.098	720	0.979	-0.017	0.927	0.122	720	1.006	0.211	0.883	0.808	720	1.042	0.111	0.808	1.124
	f	720	0.916	0.018	0.947	0.098	550	0.997	-0.023	0.914	0.135	719	1.005	0.212	0.883	0.809	517	0.952	0.190	0.847	0.878
NASA	a	0					0					0					0				
	f	0					0					0					0				
V1n																					
BIRA-BEPRO	a	720	1.000	0.010	0.959	0.089	720	-0.312	0.796	-0.098	1.074	720	0.950	0.074	0.937	0.523	720	1.304	1.250	0.238	7.967
	f	717	1.000	0.010	0.959	0.090	571	0.946	0.023	0.953	0.091	618	0.931	0.130	0.915	0.563	559	0.957	0.143	0.760	1.125
BIRA-MMF	a	720	0.981	-0.010	0.929	0.119	720	0.975	0.009	0.925	0.121	720	0.895	0.111	0.929	0.545	720	0.933	0.115	0.847	0.859
	f	690	0.983	-0.011	0.926	0.121	613	0.973	0.013	0.910	0.130	720	0.895	0.111	0.929	0.545	622	0.897	0.106	0.901	0.587
IUPB	a	720	0.890	0.009	0.941	0.107	720	0.833	0.026	0.966	0.092	720	0.511	0.045	0.730	1.310	720	0.470	0.091	0.544	1.575
	f	672	0.903	0.006	0.942	0.105	646	0.849	0.023	0.967	0.086	660	0.518	0.038	0.711	1.346	590	0.473	0.137	0.523	1.584
IUPHD	a	720	1.188	-0.019	0.876	0.211	720	1.043	0.000	0.904	0.150	720	1.036	0.146	0.890	0.803	720	1.049	0.125	0.824	1.074
	f	720	1.188	-0.019	0.876	0.211	720	1.043	0.000	0.904	0.150	720	1.036	0.146	0.890	0.803	720	1.049	0.125	0.824	1.074
KNMI	a	720	1.189	-0.022	0.834	0.248	720	1.261	-0.027	0.792	0.312	720	0.762	0.101	0.921	0.674	720	0.724	0.119	0.875	0.808
	f	601	1.214	-0.036	0.801	0.267	635	1.243	-0.030	0.779	0.305	610	0.747	0.150	0.897	0.728	643	0.710	0.163	0.848	0.854
MPIC_PARAM	a	720	-0.010	0.676	-0.002	1.303	720	-1.574	1.619	-0.227	2.265	711	0.999	0.082	0.873	0.815	711	0.883	0.215	0.823	0.901
	f	720	1.160	-0.029	0.864	0.211	720	1.030	0.009	0.928	0.127	720	0.980	0.010	0.854	0.868	720	0.967	0.117	0.766	1.182
MPIC_PAR_MC	a	574	1.224	-0.049	0.845	0.231	527	1.120	-0.012	0.917	0.131	423	1.017	-0.011	0.806	1.047	511	1.014	0.101	0.742	1.326
	f	675	0.647	0.268	0.295	0.636	620	1.033	0.009	0.868	0.170	705	0.999	0.082	0.873	0.818	704	0.881	0.213	0.823	0.902
MPIC_PRIAM	a	720	0.824	0.003	0.906	0.143	720	0.650	0.034	0.842	0.192	720	0.911	0.162	0.822	0.927	720	0.956	0.186	0.693	1.452
	f	676	0.828	0.001	0.898	0.147	630	0.626	0.049	0.798	0.205	664	0.833	0.218	0.832	0.839	660	0.843	0.235	0.714	1.191
NASA	a	0					720	0.599	0.097	0.851	0.174	720	1.125	0.025	0.873	0.965	720	1.151	0.075	0.884	0.979
	f	0					720	0.599	0.097	0.851	0.174	687	1.122	0.034	0.865	0.987	649	1.141	0.082	0.869	1.012
V2n																					
BIRA-BEPRO	a	0					0					0					0				
	f	0					0					0					0				
BIRA-MMF	a	0					0					0					0				
	f	0					0					0					0				
IUPB	a	720	0.941	0.000	0.940	0.107	720	0.989	0.006	0.875	0.165	720	0.504	0.087	0.723	1.299	720	0.514	0.119	0.698	1.299
	f	673	0.952	-0.002	0.939	0.107	650	0.893	0.018	0.970	0.077	668	0.497	0.108	0.697	1.346	698	0.491	0.151	0.690	1.298
IUPHD	a	720	1.178	-0.008	0.883	0.205	720	1.058	-0.002	0.907	0.151	720	1.077	0.126	0.883	0.881	720	1.071	0.107	0.816	1.133
	f	720	1.178	-0.008	0.883	0.205	720	1.058	-0.002	0.907	0.151	720	1.077	0.126	0.883	0.881	720	1.071	0.107	0.816	1.133
KNMI	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PARAM	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PAR_MC	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PRIAM	a	720	0.878	0.023	0.881	0.148	720	0.861	0.011	0.859	0.165	720	0.961	0.210	0.798	1.068	720	0.951	0.304	0.665	1.571
	f	690	0.875	0.022	0.880	0.148	631	0.863	0.009	0.829	0.176	683	0.952	0.204	0.814	0.986	663	0.925	0.252	0.718	1.243
NASA	a	0					0					0					0				
	f	0					0					0					0				

In general, the surface values can be retrieved accurately by most algorithms, with regression coefficients and slopes close to unity. Exceptions are:

- cases of extremely high extinction (AER8 .. AER10)

- the BIRA-BEPRO NO_2 concentration and aerosol extinction at 477 nm, where good agreement is only achieved after filtering out a significant amount of data points.
- MPIC-PARAM for aerosol-free conditions (AER0).
- the IUPB HCHO and NO_2 surface concentrations, as well as the MPIC-PRIAM NO_2 surface concentrations, which show a significantly larger scatter than the other algorithms.

4.2.2.4 Comparison of total columns

The ability of the retrieval algorithms to retrieve aerosol and trace gas total columns has been assessed through comparisons between retrieved and true values. As example, the box-whisker plots in Figure 10 shows NO_2 vertical column density (VCD) comparison results for each trace gas scenario.

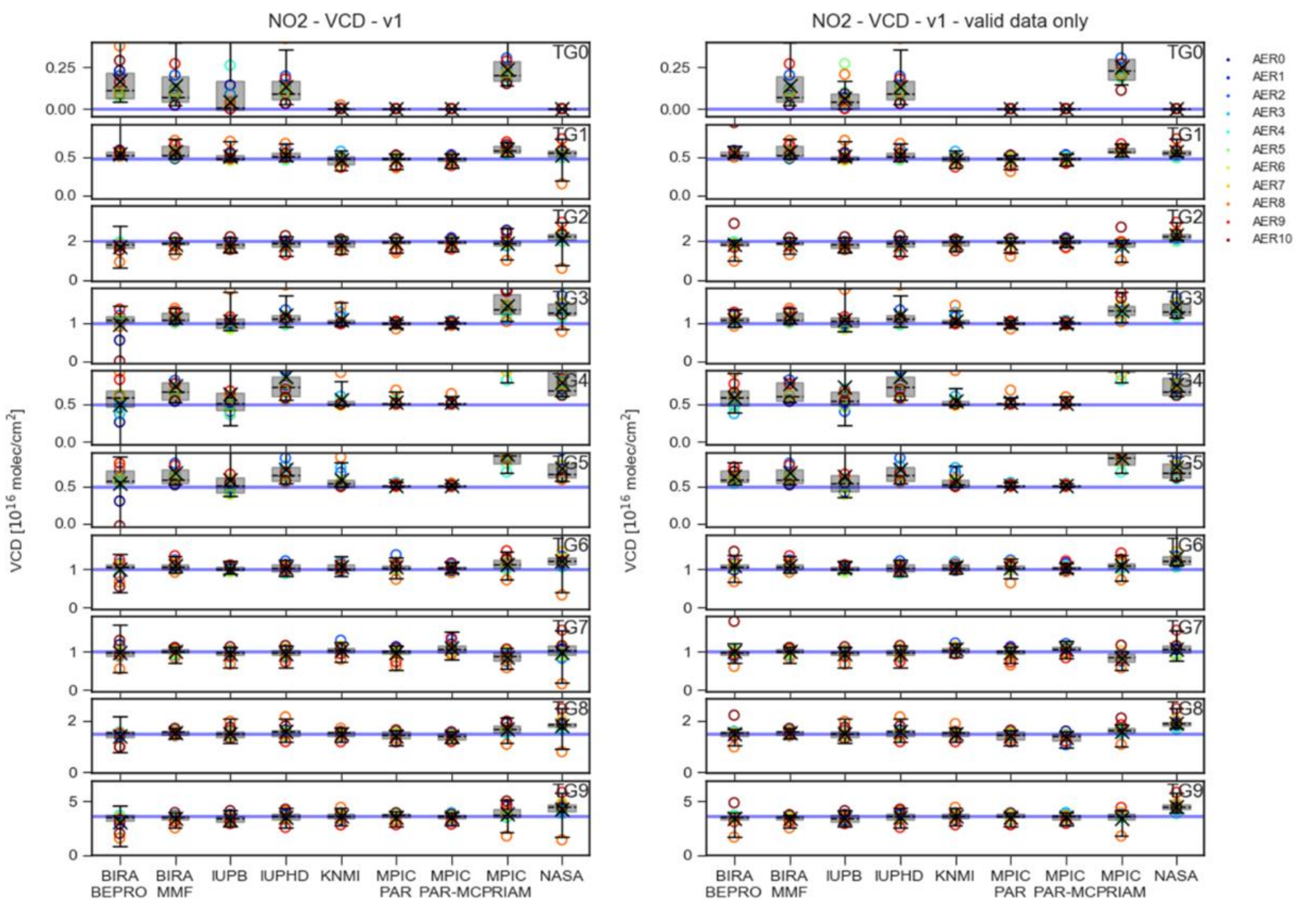


Figure 10: Comparison of NO_2 VCDs. Thick blue lines indicate the true value. Grey boxes indicate the 25 – 75 % percentile, whiskers the 5% - 95% percentile. Black crosses show the median value and dashed horizontal lines the mean. Small coloured circles indicate the median for each aerosol scenario as denoted in the legend. The left and right panels show statistics based on all data and on data flagged as valid only, respectively.

As can been seen from the box-whisker plots and regression analysis results (see Table 10), the integrated column can be retrieved accurately by most algorithms. As for the surface concentration, the following cases are problematic:

- cases of extremely high extinction, in particular the fog scenario (AER8).

- the BIRA bePRO NO_2 VCD and AOD at 477 nm, where good agreement is only achieved after filtering out a significant amount of data points.
- the MPIC-PARAM AOD for aerosol-free conditions (AER0).
- the IUPB HCHO and NO_2 VCDs, as well as the MPIC-PRIAM NO_2 surface concentrations, which show a significantly larger scatter than the other algorithms.

Table 10: Regression analysis of retrieved versus true total columns for the aerosol scenarios AER0 … AER7 (i.e., fog and cloud scenarios excluded).

		Aerosol - 360 nm					Aerosol - 477 nm					HCHO					NO_2				
		N	Slope	Intercept [Unitless]	R	RMS [Unitless]	N	Slope	Intercept [Unitless]	R	RMS [Unitless]	N	Slope	Intercept [10^{16} cm^{-2}]	R	RMS [10^{16} cm^{-2}]	N	Slope	Intercept [10^{16} cm^{-2}]	R	RMS [10^{16} cm^{-2}]
		V1																			
BIRA-BEPRO	a	720	0.668	0.084	0.958	0.110	720	0.303	0.278	0.289	0.356	720	0.893	0.140	0.988	0.172	720	0.922	0.088	0.707	0.907
	f	720	0.668	0.084	0.958	0.110	610	0.604	0.105	0.946	0.129	629	0.892	0.143	0.987	0.176	556	0.940	0.090	0.990	0.140
BIRA-MMF	a	720	0.695	0.073	0.958	0.105	720	0.670	0.078	0.943	0.115	720	0.840	0.273	0.981	0.242	720	0.931	0.126	0.990	0.156
	f	720	0.695	0.073	0.958	0.105	720	0.670	0.078	0.943	0.115	720	0.840	0.273	0.981	0.242	676	0.934	0.115	0.990	0.151
IUPB	a	720	0.714	0.048	0.945	0.111	720	0.859	0.040	0.886	0.133	720	0.960	0.158	0.969	0.267	720	0.889	0.074	0.874	0.499
	f	680	0.742	0.044	0.951	0.101	670	0.808	0.049	0.883	0.125	680	0.947	0.184	0.967	0.274	614	0.899	0.058	0.978	0.222
IUPHD	a	720	0.719	0.079	0.960	0.099	720	0.643	0.090	0.946	0.119	720	0.815	0.278	0.968	0.282	720	0.919	0.140	0.979	0.208
	f	720	0.719	0.079	0.960	0.099	720	0.643	0.090	0.946	0.119	720	0.815	0.278	0.968	0.282	720	0.919	0.140	0.979	0.208
KNMI	a	720	1.100	-0.006	0.820	0.220	720	0.960	0.023	0.892	0.139	720	0.958	0.057	0.927	0.381	720	0.967	0.052	0.981	0.189
	f	600	0.979	0.003	0.838	0.175	650	0.923	0.030	0.874	0.142	607	0.956	0.046	0.945	0.324	643	0.959	0.067	0.979	0.196
MPIC_PARAM	a	720	0.793	0.088	0.688	0.245	720	0.680	0.171	0.514	0.344	720	0.964	0.022	0.992	0.123	720	0.999	0.011	0.987	0.161
	f	720	1.015	-0.004	0.937	0.107	720	1.066	0.014	0.855	0.186	720	0.957	0.028	0.993	0.121	720	0.982	0.022	0.985	0.172
MPIC_PAR_MC	a	700	0.988	-0.005	0.940	0.098	640	0.994	-0.002	0.920	0.115	600	0.959	0.036	0.993	0.114	566	0.983	0.027	0.985	0.172
	f	675	0.821	0.072	0.715	0.233	620	1.000	-0.010	0.957	0.086	714	0.965	0.019	0.994	0.114	712	1.001	0.006	0.989	0.150
MPIC_PRIAM	a	720	0.606	0.111	0.923	0.132	720	0.442	0.158	0.899	0.169	720	0.793	0.367	0.950	0.351	720	0.947	0.251	0.954	0.350
	f	700	0.601	0.113	0.921	0.134	630	0.452	0.152	0.916	0.165	695	0.789	0.366	0.950	0.349	615	0.914	0.245	0.967	0.288
NASA	a	0					720	0.286	0.229	0.593	0.232	720	1.182	0.032	0.985	0.363	720	1.177	0.034	0.978	0.383
	f	0					720	0.286	0.229	0.593	0.232	720	1.182	0.032	0.985	0.363	690	1.182	0.027	0.981	0.376
		V2																			
BIRA-BEPRO	a	720	0.647	0.175	0.598	0.276	720	-52.817	31.254	-0.280	55.876	720	0.779	0.353	0.964	0.317	690	1.214	0.327	0.431	2.558
	f	650	0.777	0.079	0.941	0.105	480	0.750	0.113	0.875	0.144	599	0.802	0.299	0.965	0.285	571	0.930	0.245	0.899	0.463
BIRA-MMF	a	0					0					0					0				
	f	0					0					0					0				
IUPB	a	720	0.838	0.026	0.956	0.088	720	1.158	0.052	0.642	0.405	720	1.096	0.066	0.967	0.348	720	1.029	0.056	0.956	0.323
	f	680	0.849	0.026	0.954	0.086	670	0.986	0.039	0.856	0.170	679	1.091	0.076	0.964	0.358	674	1.029	0.063	0.954	0.326
IUPHD	a	720	0.757	0.071	0.962	0.092	720	0.690	0.084	0.958	0.105	720	0.886	0.195	0.983	0.206	720	0.955	0.101	0.981	0.194
	f	720	0.757	0.071	0.962	0.092	720	0.690	0.084	0.958	0.105	720	0.886	0.195	0.983	0.206	720	0.955	0.101	0.981	0.194
KNMI	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PARAM	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PAR_MC	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PRIAM	a	720	0.852	0.071	0.946	0.098	720	0.829	0.095	0.970	0.090	720	0.903	0.256	0.965	0.295	720	1.016	0.158	0.962	0.335
	f	720	0.852	0.071	0.946	0.098	550	0.879	0.065	0.978	0.069	719	0.912	0.250	0.971	0.279	517	1.016	0.149	0.958	0.338
NASA	a	0					0					0					0				
	f	0					0					0					0				
		V1n																			
BIRA-BEPRO	a	720	0.644	0.100	0.949	0.117	720	0.059	0.421	0.019	0.939	720	0.896	0.137	0.985	0.184	720	0.892	0.050	0.713	0.867
	f	717	0.643	0.100	0.949	0.118	571	0.584	0.120	0.943	0.135	618	0.895	0.143	0.984	0.189	559	0.950	0.090	0.981	0.189
BIRA-MMF	a	720	0.660	0.091	0.854	0.149	720	0.664	0.086	0.929	0.121	720	0.869	0.249	0.980	0.237	720	0.953	0.111	0.982	0.193
	f	690	0.652	0.096	0.849	0.152	613	0.622	0.110	0.924	0.131	720	0.869	0.249	0.980	0.237	622	0.933	0.117	0.976	0.185
IUPB	a	720	0.713	0.053	0.936	0.113	720	0.868	0.041	0.879	0.138	720	0.945	0.188	0.966	0.283	720	0.901	0.051	0.973	0.239
	f	672	0.740	0.050	0.943	0.105	646	0.820	0.050	0.876	0.131	660	0.925	0.214	0.964	0.281	590	0.895	0.068	0.968	0.249
IUPHD	a	720	0.724	0.083	0.953	0.102	720	0.645	0.095	0.935	0.122	720	0.826	0.268	0.964	0.288	720	0.937	0.134	0.970	0.247
	f	720	0.724	0.083	0.953	0.102	720	0.645	0.095	0.935	0.122	720	0.826	0.268	0.964	0.288	720	0.937	0.134	0.970	0.247
KNMI	a	720	0.924	0.052	0.727	0.249	720	1.020	0.026	0.842	0.188	720	0.964	0.045	0.934	0.362	720	0.949	0.067	0.953	0.300
	f	601	0.923	0.037	0.776	0.211	635	1.023	0.024	0.832	0.193	610	0.956	0.050	0.929	0.370	643	0.946	0.070	0.973	0.219
MPIC_PARAM	a	720	0.743	0.107	0.601	0.291	720	0.634	0.219	0.374	0.471	711	0.935	0.076	0.818	0.646	711	1.021	0.005	0.955	0.310
	f	720	0.896	0.124	0.588	0.362	720	1.405	0.001	0.802	0.336	720	0.945	0.047	0.861	0.549	720	0.985	0.023	0.969	0.245
MPIC_PAR_MC	a	574	0.862	0.021	0.918	0.106	527	1.201	-0.038	0.852	0.154	423	0.920	0.054	0.987	0.176	511	0.982	0.018	0.983	0.180
	f	675	0.851	0.045	0.802	0.184	620	1.063	-0.024	0.905	0.143	705	0.936	0.075	0.817	0.648	704	1.022	0.003	0.956	0.311
MPIC_PRIAM	a	720	0.585	0.115	0.918	0.137	720	0.439	0.160	0.884	0.172	720	0.797	0.363	0.947	0.354	720	0.945	0.251	0.955	0.348
	f	676	0.573	0.122	0.913	0.141	630	0.448	0.155	0.904	0.167	664	0.772	0.373	0.947	0.351	660	0.909	0.266	0.958	0.315
NASA	a	0					720	0.287	0.229	0.593	0.232	720	1.182	0.031	0.981	0.378	720	1.177	0.034	0.978	0.384
	f	0					720	0.287	0.229	0.593	0.232	687	1.175	0.046	0.980	0.385	649	1.179	0.036	0.979	0.389
		V2n																			
BIRA-BEPRO	a	0					0					0					0				
	f	0					0					0					0				
BIRA-MMF	a	0					0					0					0				
	f	0					0					0					0				
IUPB	a	720	0.824	0.036	0.943	0.096	720	1.180	0.045	0.655	0.399	720	1.090	0.087	0.960	0.377	720	1.014	0.082	0.952	0.334
	f	673	0.839	0.035	0.944	0.094	650	0.999	0.030	0.891	0.144	668	1.092	0.085	0.956	0.389	698	1.018	0.082	0.952	0.327
IUPHD	a	720	0.755	0.080	0.949	0.100	720	0.697	0.089	0.936	0.113	720	0.897	0.188	0.974	0.239	720	0.963	0.101	0.969	0.250
	f	720	0.755	0.080	0.949	0.100	720	0.697	0.089	0.936	0.113	720	0.897	0.188	0.974	0.239	720	0.963	0.101	0.969	0.250
KNMI	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PARAM	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PAR_MC	a	0					0					0					0				
	f	0					0					0					0				
MPIC_PRIAM	a	720	0.833	0.088	0.921	0.118	720	0.801	0.143	0.698	0.255	720	1.029	0.158	0.972	0.311	720	1.033	0.169	0.954	0.380
	f	690	0.836	0.087	0.920	0.118	631	0.912	0.080	0.835	0.179	683	1.028	0.157	0.972	0.306	663	1.068	0.125	0.960	0.348
NASA	a	0					0					0					0				
	f	0					0					0					0				

4.2.2.5 Comparison of simulated and true dSCDs

The agreement between initial DSCDs serving as measurement vector and dSCDs simulated for the retrieved profiles is crucial for the overall assessment of the ability to reproduce the observations and to simulate the atmospheric radiative transfer appropriately. Large discrepancies indicate a lack of convergence of the retrieval.

Correlation plots for NO_2 are shown in Figure 11 as an example, and the regression analysis results for all aerosol and trace gases presented in Table 11 provides a quantitative comparison.

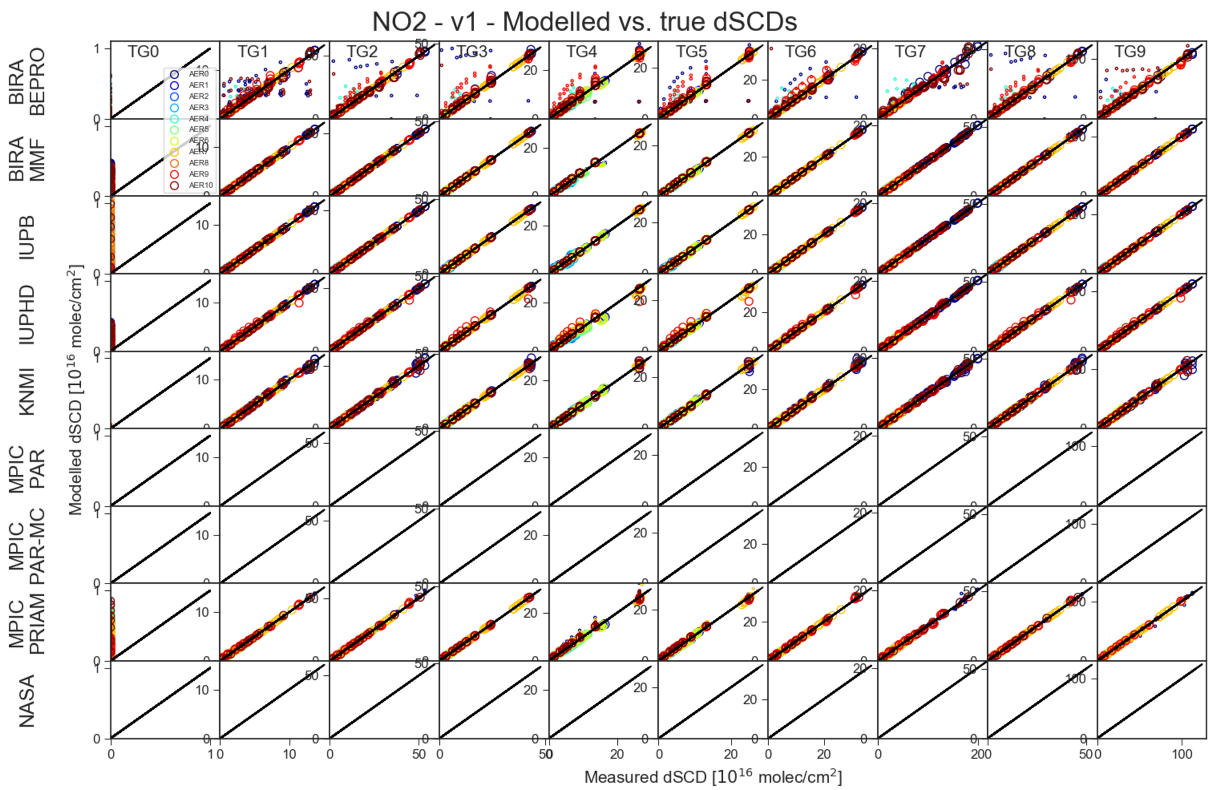


Figure 11: Comparison of initial NO_2 dSCDs and NO_2 dSCDs simulated for the retrieved profiles for the v1 run. The colour scale indicates the aerosol scenario as indicated in the legend. Small circles indicate data points flagged as invalid. No data is available for MPIC_PARAM and NASA.

Table 11: Regression analysis of simulated versus true SCDs for the aerosol scenarios AER0 … AER7 (i.e., fog and cloud scenarios excluded). Rows marked with 'a' and 'f' show data for all data and data marked as valid, respectively. The red shading indicates the level of deviation from the optimal values. No data is available for MPIC_PARAM, MPIC_PAR_MC (MAPA) and NASA.

		Aerosol - 360 nm					Aerosol - 477 nm					HCHO					NO_2				
		N	Slope	Intercept [10^{43} cm^{-5}]	R	RMS [10^{43} cm^{-5}]	N	Slope	Intercept [10^{43} cm^{-5}]	R	RMS [10^{43} cm^{-5}]	N	Slope	Intercept [10^{17} cm^{-2}]	R	RMS [10^{17} cm^{-2}]	N	Slope	Intercept [10^{17} cm^{-2}]	R	RMS [10^{17} cm^{-2}]
		V1																			
BIRA-BEPRO	a	6480	0.950	0.046	0.995	0.170	6480	0.461	0.957	0.698	2.014	6480	0.994	0.004	0.999	0.023	6480	0.913	0.082	0.221	3.740
	f	6480	0.950	0.046	0.995	0.170	5490	0.891	0.182	0.985	0.393	5661	0.994	0.004	0.999	0.025	5004	0.987	0.009	0.999	0.030
BIRA-MMF	a	6480	0.989	0.020	1.000	0.042	6480	0.993	0.029	1.000	0.075	6480	0.985	0.020	0.998	0.040	6480	0.996	0.009	1.000	0.028
	f	6480	0.989	0.020	1.000	0.042	6480	0.993	0.029	1.000	0.075	6480	0.985	0.020	0.998	0.040	6084	0.998	0.007	1.000	0.016
IUPB	a	6480	0.986	0.002	0.999	0.066	6480	0.922	0.146	0.996	0.312	6480	0.992	0.001	0.997	0.049	6480	0.989	0.019	0.945	0.318
	f	6120	0.985	0.005	0.999	0.066	6030	0.919	0.163	0.996	0.323	6120	0.992	0.002	0.997	0.051	5526	0.998	0.003	1.000	0.030
IUPHD	a	6480	0.981	0.034	0.999	0.055	6480	0.968	0.074	0.999	0.157	6480	0.984	0.019	0.998	0.041	6480	0.995	0.009	1.000	0.027
	f	6480	0.981	0.034	0.999	0.055	6480	0.968	0.074	0.999	0.157	6480	0.984	0.019	0.998	0.041	6480	0.995	0.009	1.000	0.027
KNMI	a	6479	1.007	-0.043	1.000	0.047	6479	1.004	-0.040	1.000	0.065	6479	0.954	0.016	0.988	0.102	6479	0.996	0.001	0.999	0.042
	f	5399	1.006	-0.042	0.999	0.047	5849	1.004	-0.042	1.000	0.067	5462	0.951	0.021	0.987	0.111	5786	0.996	0.002	0.999	0.044
MPIC_PARAM	a																				
	f																				
MPIC_PAR_MC	a																				
	f																				
MPIC_PRIAM	a	6480	0.872	0.170	0.991	0.275	6480	0.597	0.771	0.938	1.293	6480	0.984	0.019	0.998	0.044	6480	0.994	0.013	0.999	0.043
	f	6300	0.886	0.145	0.991	0.245	5670	0.844	0.276	0.985	0.337	6255	0.985	0.017	0.998	0.042	5535	0.994	0.009	1.000	0.021
NASA	a																				
	f																				
		V2																			
BIRA-BEPRO	a	6480	0.929	0.015	0.976	0.346	6480	0.229	1.147	0.451	2.644	6480	0.978	0.041	0.993	0.084	6210	0.846	0.076	0.915	0.351
	f	5850	0.946	0.045	0.994	0.193	4320	0.993	0.001	0.998	0.085	5391	0.990	0.013	0.997	0.050	5139	0.977	0.027	0.997	0.066
BIRA-MMF	a																				
	f																				
IUPB	a	6480	0.992	-0.009	0.999	0.062	6480	0.927	0.139	0.996	0.306	6480	0.990	0.003	0.996	0.060	6480	0.996	0.000	0.998	0.056
	f	6120	0.992	-0.008	0.999	0.062	6030	0.924	0.154	0.996	0.316	6111	0.990	0.003	0.996	0.061	6066	0.996	0.000	0.998	0.056
IUPHD	a	6480	0.987	0.025	1.000	0.044	6480	0.968	0.072	0.999	0.164	6480	0.990	0.012	0.999	0.029	6480	0.997	0.006	1.000	0.024
	f	6480	0.987	0.025	1.000	0.044	6480	0.968	0.072	0.999	0.164	6480	0.990	0.012	0.999	0.029	6480	0.997	0.006	1.000	0.024
KNMI	a																				
	f																				
MPIC_PARAM	a																				
	f																				
MPIC_PAR_MC	a																				
	f																				
MPIC_PRIAM	a	6480	0.878	0.132	0.993	0.273	6480	0.602	0.677	0.948	1.291	6480	0.991	0.011	0.999	0.028	6480	0.983	0.013	0.999	0.047
	f	6480	0.878	0.132	0.993	0.273	4950	0.898	0.116	0.989	0.245	6471	0.991	0.011	0.999	0.025	4653	0.997	0.004	1.000	0.008
NASA	a																				
	f																				
		V1n																			
BIRA-BEPRO	a	6480	0.877	0.155	0.984	0.317	6480	0.376	1.105	0.667	2.144	6480	0.991	0.006	0.998	0.045	6480	1.042	0.080	0.235	3.978
	f	6453	0.882	0.146	0.985	0.303	5139	0.907	0.128	0.985	0.299	5562	0.990	0.007	0.997	0.048	5031	0.977	0.016	0.998	0.056
BIRA-MMF	a	6479	0.975	0.049	0.996	0.133	6479	0.984	0.052	0.998	0.185	6479	0.983	0.021	0.997	0.053	6479	0.994	0.010	0.999	0.046
	f	6209	0.977	0.044	0.996	0.121	5516	0.987	0.037	0.997	0.127	6479	0.983	0.021	0.997	0.053	5598	0.994	0.009	0.999	0.028
IUPB	a	6480	0.960	0.050	0.995	0.160	6480	0.891	0.213	0.991	0.440	6480	0.988	0.003	0.996	0.062	6480	0.998	0.001	0.999	0.046
	f	6048	0.957	0.059	0.994	0.165	5814	0.893	0.230	0.992	0.428	5940	0.989	0.003	0.995	0.063	5310	0.998	0.001	0.999	0.044
IUPHD	a	6480	0.970	0.059	0.996	0.137	6480	0.965	0.084	0.997	0.228	6480	0.980	0.022	0.997	0.055	6480	0.993	0.010	0.999	0.047
	f	6480	0.970	0.059	0.996	0.137	6480	0.965	0.084	0.997	0.228	6480	0.980	0.022	0.997	0.055	6480	0.993	0.010	0.999	0.047
KNMI	a	6479	0.986	-0.002	0.995	0.153	6479	0.993	-0.011	0.997	0.194	6479	0.951	0.022	0.977	0.140	6479	0.998	0.001	0.999	0.043
	f	5408	0.986	-0.003	0.993	0.141	5714	0.995	-0.017	0.997	0.172	5489	0.947	0.029	0.974	0.152	5786	0.998	0.001	0.999	0.045
MPIC_PARAM	a																				
	f																				
MPIC_PAR_MC	a																				
	f																				
MPIC_PRIAM	a	6480	0.852	0.215	0.986	0.325	6480	0.585	0.804	0.931	1.338	6480	0.978	0.022	0.996	0.059	6480	0.969	0.026	0.997	0.070
	f	6084	0.882	0.165	0.986	0.257	5670	0.839	0.294	0.977	0.377	5976	0.977	0.021	0.996	0.053	5940	0.987	0.013	0.998	0.039
NASA	a																				
	f																				
		V2n																			
BIRA-BEPRO	a																				
	f																				
BIRA-MMF	a																				
	f																				
IUPB	a	6480	0.962	0.038	0.997	0.135	6480	0.885	0.223	0.991	0.454	6480	0.987	0.004	0.995	0.069	6480	0.989	0.004	0.997	0.074
	f	6057	0.960	0.045	0.997	0.137	5850	0.895	0.218	0.993	0.403	6012	0.987	0.004	0.994	0.070	6282	0.985	0.007	0.997	0.072
IUPHD	a	6480	0.984	0.029	0.999	0.075	6480	0.969	0.070	0.998	0.190	6480	0.988	0.013	0.998	0.044	6480	0.990	0.010	0.998	0.055
	f	6480	0.984	0.029	0.999	0.075	6480	0.969	0.070	0.998	0.190	6480	0.988	0.013	0.998	0.044	6480	0.990	0.010	0.998	0.055
KNMI	a																				
	f																				
MPIC_PARAM	a																				
	f																				
MPIC_PAR_MC	a																				
	f																				
MPIC_PRIAM	a	6480	0.870	0.143	0.989	0.307	6480	0.571	0.744	0.928	1.400	6480	0.975	0.018	0.997	0.051	6480	0.904	0.056	0.984	0.175
	f	6210	0.892	0.107	0.992	0.252	5679	0.839	0.210	0.981	0.394	6147	0.990	0.012	0.998	0.037	5967	0.988	0.010	0.999	0.038
NASA	a																				
	f																				

4.2.2.6 Comparison of averaging kernels

The averaging kernels, which quantify the sensitivity of the retrieval to the atmospheric state, provide an important diagnostic tool for the characterization of the OEM-based retrievals, in terms of vertical sensitivity and information content through the trace of the averaging kernels, also called degrees of freedom for signal (DOFS; see Rodgers, 2000).

As an example, Figure 12 and Figure 13 show the averaging kernels from the OEM algorithms for aerosol at 477 nm and NO_2. The shapes of the averaging kernels from the different models have a high degree of similarity, except for BOREAS aerosols; not shown here). The information content usually range from dofs < 0.5 during foggy conditions (AER8) to DOFS > 3 for aerosol-free atmospheres (AER0), with the information content being generally higher for aerosols than for trace gases.

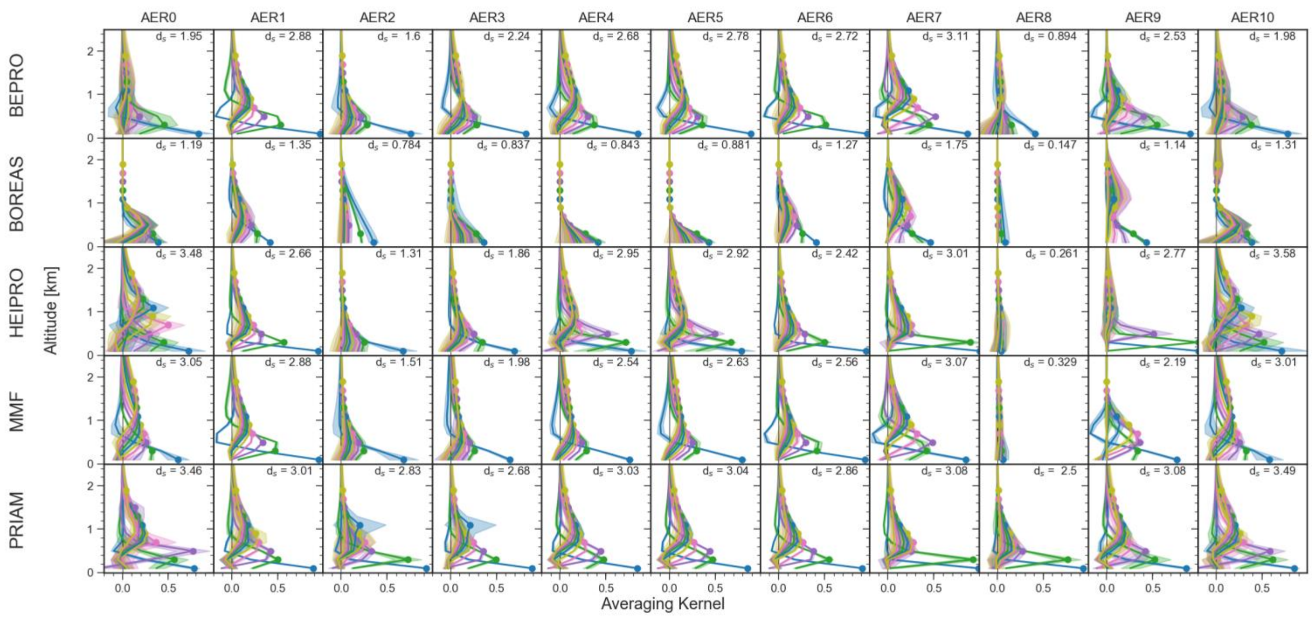


Figure 12: 477 nm aerosol averaging kernels. Each subplot shows the mean averaging kernel over all SZA and RAA for a specific aerosol scenario (columns) and retrieval algorithm (rows). Filled circles indicate the nominal altitude of the corresponding averaging kernel plotted in the same colour. Colour shaded areas indicate the standard deviation of the averaging kernels, i.e. the variation for different SZA and RAA. Also shown are the degrees of freedom for signal. Averaging kernels stem from retrievals from measurements without noise (v1).

Apart from the fog scenario (AER8), there is only a moderate dependency of vertical resolution and information content on the aerosol content of the atmosphere. The variability of the averaging kernels with the position of the Sun (shown as shaded areas in Figure 12 and Figure 13) is very small.

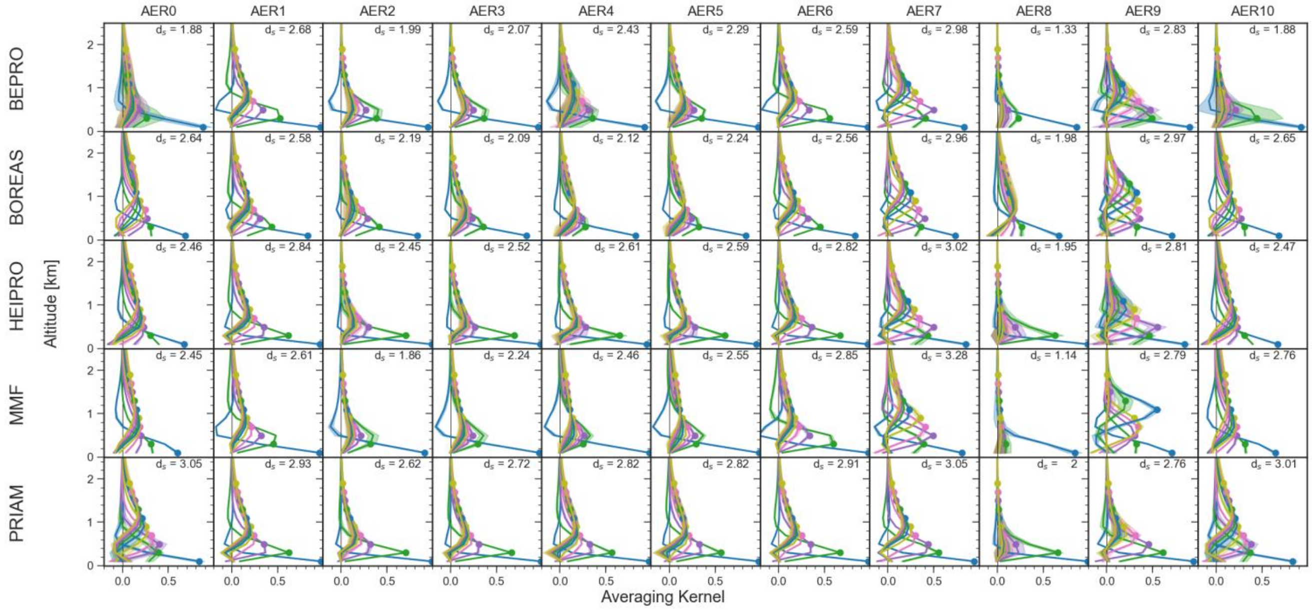


Figure 13: Same as Figure 12, but for NO_2.

4.2.2.7 Comparison of profiles based on CINDI-2 measurements

In order to assess the performance of the retrieval algorithms under real conditions, aerosol, NO_2 and HCHO profile retrievals were also performed for two selected days with high and low aerosol load (15 and 23 September 2016, respectively) using median dSCDs from all instruments operational during the CINDI-2 campaign. The retrievals were performed using the same settings as for the synthetic dSCDs. They were compared to Sun Photometer and LP-DOAS measurements (measured along a light beam from the surface to the top of the Cabauw mast at ~200 m, thus representative for the lowermost MAX-DOAS retrieval layer) as independent reference data sets. The following conclusions have been drawn from the correlation plots for MAX-DOAS AOD and surface concentrations with the reference measurements (not shown here; see deliverable D5 for more details):

- In general, the RMS difference between retrieved and reference AODs and surface concentrations are similar to the synthetic exercise based on dSCDs with noise, and even better for HCHO. A possible reason for this finding is that the noise added to the dSCDs in the synthetic exercise (3% of the dSCD in order to account for possible horizontal inhomogeneities) might be unrealistically high. Furthermore, the range of different trace gas amounts is much higher for the synthetic comparisons than during the two days of the CINDI-2 campaign considered here.
- All algorithms, except MPIC_PAR_MC (MAPA) in the UV, significantly underestimate the AOD for AOD > 0.3.
- The HCHO surface concentration is generally overestimated by the MAX-DOAS retrievals. Interestingly, the RMS difference between true and retrieved HCHO surface concentrations is better than for the synthetic exercise for most algorithms.
- The BIRA-BEPRO algorithm seems to suffer from the problems in the visible to a much lesser extent than during the synthetic exercise.

4.2.2.8 Numerical performance

Numerical performance is an important aspect which needs to be considered for the selection of a retrieval code as part of the community algorithm. Figure 14 shows the average time to retrieve a single aerosol and trace gas profile for each retrieval algorithm normalised to a single CPU core (the IUPB retrievals are performed on a computer grid with 10 – 30 processors). Specific properties of the computers, such as CPU speed and other factors influencing the performance, are not considered.

The numerical performance varies strongly between the individual algorithms, ranging from only 4.5 milliseconds for the NASA retrieval to more than 10 minutes for an aerosol profile at 477 nm by IUPB.

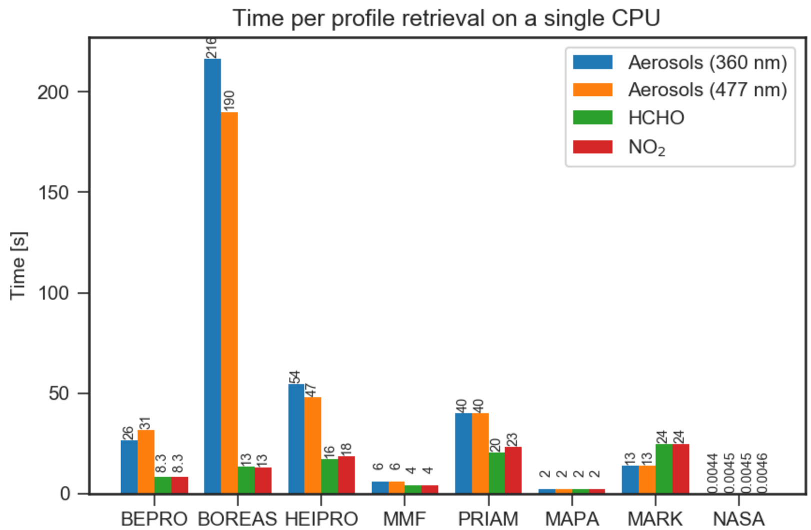


Figure 14: Average time for the retrieval of a vertical aerosol and trace gas profile on a single CPU for the v1 run. The numbers on top of the bars indicate the time in seconds. Hardware properties, such as different CPU speeds, have not been considered.

4.3 Selection of the FRM$_4$DOAS community algorithms

4.3.1 MAX-DOAS profiling algorithms

The overall performance of the different MAX-DOAS profile retrieval algorithms that participated in this study is summarized in Table 12. It shows the evaluation of these algorithms in terms of (1) simulated dSCDs, (2) trace gas concentrations/extinctions at all height levels, (3) surface concentration/extinction, (4) total column (VCD/AOD), and (5) execution speed. The evaluation is based on the RMS difference between retrieved and true quantities. The linear regression parameters are less good indicators for the overall performance of the algorithms since the scenarios TG2 and AER2, which have a significantly higher integrated column than the other scenarios, have a dominant impact on the slope of the regression line.

To a certain extent, the algorithms from all participants were able to reproduce the true aerosol and trace gas profiles. However, it can be seen from Table 12 that MPIC-PAR-MC (MAPA; Beirle et al., 2019) and BIRA-MMF (Friedrich et al., 2019) yield on overall the most accurate results of the parameterized and OEM algorithms, respectively. Therefore they were selected as community algorithms to be implemented in the FRM$_4$DOAS centralized processing system. A full description of both algorithms can be found in the deliverable D6 (‘Algorithm ATBD’).

Table 12: Table showing the assessment results of the individual participants (1 = best appropriate for inclusion in a central processing system, 9 = worst appropriate for inclusion in a central processing system) for the different aerosol and trace gas species in terms of RMS between retrieved and true values for (1) simulated dSCDs, (2) trace gas concentrations/extinctions at all height levels, (3) surface concentration/extinction, (4) total column (VCD/AOD), and (5) execution speed. Columns labelled 'a' and 'f' show the rank for all data and data flagged as valid. The top part shows the assessment for aerosol scenarios 0..7 (i.e., fog and cloud scenarios excluded). Top and bottom parts of the table show evaluations for retrievals based on SCDs without (v1) and with (v1n) random noise. No data on computational performance is available for the v1n runs. Note that MPIC_PARAM_MC is now called MAPA.

		Aerosol - 360 nm						Aerosol - 477 nm						HCHO						NO_2					
		dSCD	Profile	Surface	Column	Speed	Valid	dSCD	Profile	Surface	Column	Speed	Valid	dSCD	Profile	Surface	Column	Speed	Valid	dSCD	Profile	Surface	Column	Speed	Valid
		v1																							
BIRA-BEPRO	a	5	1	1	4	5	100%	6	8	8	9	5	85%	1	4	3	3	3	87%	6	9	9	9	3	77%
	f	5	2	1	5	5		6	2	2	6	5		1	4	4	3	3		4	5	4	1	3	
BIRA-MMF	a	1	4	2	2	4	100%	2	1	3	1	4	100%	2	2	4	4	6	100%	2	1	2	1	6	94%
	f	1	4	2	4	4		2	4	5	3	4		2	3	3	4	6		1	1	3	3	6	
IUPB	a	4	2	3	5	9	94%	4	2	1	3	9	93%	5	8	9	5	4	94%	5	8	8	8	4	85%
	f	4	1	3	3	9		4	1	1	5	9		5	8	9	5	4		5	9	9	7	4	
IUPHD	a	3	5	7	1	8	100%	3	3	4	2	8	100%	3	5	6	6	7	100%	1	4	5	5	8	100%
	f	3	5	7	2	8		3	6	7	4	8		3	5	6	6	7		3	4	5	6	8	
KNMI	a	2	7	5	7	3	83%	1	6	5	4	3	90%	6	9	5	9	8	84%	3	6	4	4	7	89%
	f	2	7	5	7	3		1	9	6	7	3		6	9	5	7	8		6	8	6	5	7	
MPIC_PARAM	a		8	8	8	6	94%		9	9	8	6	86%		1	1	2	9	99%		2	1	2	9	99%
	f		8	8	8	6			3	3	1	6			2	1	1	9			3	2	2	9	
MPIC_PAR_MC	a		3	4	3	2	97%		7	2	6	2	89%		3	2	1	2	83%		3	3	3	2	79%
	f		3	4	1	2			5	4	2	2			1	2	2	2			2	1	4	2	
MPIC_PRIAM	a	6	6	6	6	7	97%	5	4	7	5	7	88%	4	7	7	7	5	97%	4	7	7	6	5	85%
	f	6	6	6	6	7		5	7	9	8	7		4	7	7	8	5		2	7	8	8	5	
NASA	a								5	6	7	1	100%		6	8	8	1	100%		5	6	7	1	96%
	f								8	8	9	1			6	8	9	1			6	7	9	1	
		v1n																							
BIRA-BEPRO	a	5	1	1	3		100%	6	8	8	9		79%	1	1	1	1		86%	6	9	9	9		78%
	f	6	1	1	4			4	2	2	4			1	2	2	2			6	5	6	3		
BIRA-MMF	a	1	5	3	5		96%	1	2	2	1		85%	2	2	2	2		100%	3	4	2	1		86%
	f	1	6	3	6			1	4	3	2			2	1	1	3			1	1	1	2		
IUPB	a	4	2	2	2		93%	4	1	1	3		90%	5	8	9	3		92%	2	8	8	2		82%
	f	4	2	2	2			6	1	1	3			5	8	9	4			3	9	9	6		
IUPHD	a	2	4	6	1		100%	3	3	4	2		100%	3	3	4	4		100%	4	6	5	4		100%
	f	2	5	5	1			3	5	5	1			4	3	4	5			5	6	5	5		
KNMI	a	3	6	7	6		83%	2	6	7	5		88%	6	9	3	6		85%	1	5	1	5		89%
	f	3	7	7	8			2	9	9	8			6	9	3	7			4	7	2	4		
MPIC_PARAM	a		8	8	7		94%		9	9	8		86%		5	5	9		98%		2	3	6		98%
	f		8	8	7				6	6	5				5	5	9				3	3	7		
MPIC_PAR_MC	a		7	5	8		80%		7	3	7		73%		4	6	8		59%		3	6	3		71%
	f		3	6	3				3	4	6				6	8	1				4	8	1		
MPIC_PRIAM	a	6	3	4	4		94%	5	4	6	4		88%	4	7	7	5		92%	5	7	7	7		92%
	f	5	4	4	5			5	7	8	7			3	7	6	6			2	8	7	8		
NASA	a								5	5	6		100%		6	8	7		95%		1	4	8		90%
	f								8	7	9				4	7	8				2	4	9		

4.3.2 Total O_3 column and stratospheric NO_2 profiling algorithms

The FRM4DOAS partners decided that a Round-Robin exercise was not needed for the selection of the total O_3 column and stratospheric NO_2 profile retrieval algorithms, given their much higher level of maturity compared to the MAXDOAS profiling tools. Instead, a review of the already existing community consensus on the corresponding retrieval methods and settings has been performed. Those methods are the following (see deliverable D6 for their full description):

- Total O_3 column: Standard approach using NDACC O_3 AMF look-up tables generated from the TOMS version 8 (TV8) ozone and temperature profile climatology (see also Hendrick et al., 2011).

- Stratospheric NO_2 profile: BIRA profiling tool based on the OEM for linear case (see also Hendrick et al., 2004).

4.3.3 Spectral fitting software

There was a consensus between the FRM_4DOAS partners to use the well-established QDOAS package developed at BIRA as community spectral fitting software. QDOAS is also described in deliverable D6.

5. FRM_4DOAS central processing system

In this Section, we describe the FRM_4DOAS centralised processing system as it is at the end of CCN03, including the technical requirement, architecture design, processing system code, and processing system validation.

5.1 Technical requirements

The technical requirements for the FRM_4DOAS centralised processing system cover the following domains: functional, operational, resources, security, and quality. All the specific requirements corresponding to these domains are listed and shortly described in the requirements versus architecture design compliance matrix presented in Table 13 (version updated at the end of CCN03; see also deliverable D20). An extended description of all those requirements can be found in deliverable D7 ('Processing System Technical Requirements Document').

Table 13: Requirements versus architecture design compliance matrix (status at the end of CCN03; see also deliverable D20). Green, orange, and red boxes indicate that the architecture design of the processing system fully complies, partly complies, or does not comply with the technical requirements.

Requir. domain	Requir. type	Requir. ref.	Short description	Archi. ref.	Comply ?	Notes
Functional	Level-1 data	[FUN-3.1.1-01]	Instrument registration before submitting Level-1 data	2.2.3	Green	Instrument number attribution
		[FUN-3.1.1-02]	Only new or modified files should be submitted on the FTP server.	2.2.3	Green	
		[FUN-3.1.1-03]	Level-1 data should consist in one day of dark-current and offset corrected radiance spectra in the predefined netCDF format.	2.2.1	Green	
		[FUN-3.1.1-04]	Level-1 data file name should satisfy the predefined format.	2.2.7	Green	
		[FUN-3.1.1-05]	The names of input/output files and metadata should feed databases. Requests should be possible from python scripts or dashboards through APIs (JSON)	N/A		For the follow-up project
		[FUN-3.1.1-06]	Mechanisms should be setup to follow the processing steps and to ensure the traceability of the processing chain.	3.3	Green	
	Scientific modules	[FUN-3.1.2-01]	QDOAS is the selected algorithm for slant columns retrieval. It should be adapted to read netCDF Level-1 data files.	3.2.1	Green	
		[FUN-3.1.2-02]	QDOAS generates output files in netCDF format.	3.2.1	Green	
		[FUN-3.1.2-03]	The algorithms to retrieve tropospheric profiles from MAXDOAS measurements should be the ones selected by the Round Robin exercise (MAPA+MMF)	3.2.4 3.2.5	Green	
		[FUN-3.1.2-04]	The algorithm for stratospheric NO_2 profile retrieval should be the BIRA-IASB OEM-based algorithm (Hendrick et al., 2004)	3.2.3	Green	
		[FUN-3.1.2-05]	Total O_3 column retrieval should be based the standard NDACC approach.	3.2.2	Green	
		[FUN-CCN02-01]	QC flagging in MMF and MAPA and their consistency should be improved wrt Phase I versions.	N/A	Green	
		[FUN-CCN02-02]	The cloud flagging routine should be made operational.	N/A	Orange	CI statistics implemented but conversion to a cloud flagging approach postponed to the ESA FRM_4DOAS 2.0 project.
		[FUN-CCN02-03]	A correction for the temperature dependence of the NO_2 cross sections should be included in the stratospheric NO_2 profiling algorithm.	N/A	Green	

		[FUN-CCN02-04]	Wrappers should be improved/optimized in terms of structure, content, and management of the input/output data, settings, config, and log files.	N/A		
	Level-2 data	[FUN-3.1.3-01]	Granularity should be one file per instrument, per day and per supported product.	2.2.2		
		[FUN-3.1.3-02]	The processing system should be able to deliver the following products: • Total O_3 columns (two values per file) • Stratospheric NO_2 vertical profiles (two profiles per file) • Tropospheric NO_2, HCHO and aerosols profiles from MAXDOAS scans (one profile per scan)	3.2.2 3.2.3 3.2.4 3.2.5		
		[FUN-3.1.3-03]	Delivered data should include the main measurement quantities, uncertainty information and quality flags, as well as useful ancillary metadata.	2.2.2		
		[FUN-3.1.3-04]	Level-2 files will be generated per instrument, per day and per product and should be automatically catalogued.	2.2.2		
		[FUN-3.1.3-05]	The Level-2 files should contain the information necessary to track back the process and identify the Level-1 sources.	2.2.2		
		[FUN-CCN02-05]	A working data stream should be established between the central processing system level-2 files database and the NDACC and EVDC databases.	N/A		
	Ancillary data	[FUN-3.1.4-01]	Regarding T, p profiles, the system should be able to use those provided by default or to extract them from external sources (ECMWF analysis) or to use those provided by Level-1 data submitters.	2.2.2		
	QA/QC	[FUN-3.1.5-01]	Specific quality flags will be assigned to all records and will be updated if necessary throughout the processing chain.	2.2.7		Possibility to filter the records based on error masks and fill values
		[FUN-3.1.5-02]	The name and the content of Level-1 data files have to be validated in order to be catalogued in the Level-1 database.	2.2.3		
		[FUN-3.1.5-03]	Standardized and automated QA tests will be performed to validate the output at each step of the process.	2.2.7		Possibility to filter the records based on error masks and fill values
		[FUN-3.1.5-04]	According to the type of anomalies/issues (warning, fatal error), the system should be able to continue the processing chain with eventually flagged data or stop it on fatal error.	2.3 2.5.3		
	Configuration	[FUN-3.1.6-01]	The configuration of the system will consist in a collection of files specific to the modules and/or to the instruments. The selected format should offer a high level of modularity and flexibility.	2.1 2.3		
	Monitoring	[FUN-3.1.7-01]	A log analyzer should be developed to collect, parse, and format individual log files from all modules and generate processing reports	2.5		
		[FUN-3.1.7-02]	Alert messages should be delivered to the operators and the data submitters in case of severe errors or QA/QC issues reported by one of the processing module.	2.5		

		[FUN-3.1.7-03]	The system should be able to detect stop of data stream.	2.5.2		
		[FUN-3.1.7-04]	Statistics indicators should be generated (number of processed files per day, evolution of the database, average processing time for a module, maximum number of simultaneous processes,…)	2.5.4		Indicators already included in log files.
		[FUN-CCN02-06]	The reporting routine should be improved in terms of error message content.	N/A		
	Dashboard	[FUN-3.1.8-01]	The processing status of the submitted files will be made available through web-based dashboards. Regular reports (weekly or monthly) could be sent by mail to the data submitters.	2.6		Simplified reports are sent to data submitters on a daily basis. Development of web-based dashboard will be done as part of the Copernicus follow-up project
Operational	Operational level	[OPE-3.2-01]	The FRM$_4$DOAS processing chain shall be fully automated but as demonstration system, it is not intended to be an operational system stricto sensu.	1.2		
		[OPE-CCN02-01]	The processing system should run in an operational mode for a selection of NDACC-certified MAXDOAS stations.	N/A		
Resources	Compute servers	[RES-3.3.1-01]	In order to increase the data processing performances, the system should be designed to exploit as much as possible the execution in parallel of the different jobs.	2.3.2		
	HPC	[RES-CCN02-01]	The framework and scripts for running the processing system on HPC should be created.	N/A		
	FTP server	[RES-3.3.2-01]	A dedicated FTP server will be made available at BIRA-IASB for incoming for Level-1 radiance data. Control access rules should be setup.	2.2.3		
		[RES-3.3.2-02]	Instead of a FTP repository, Level-2 files could be delivered through dedicated web pages.	2.2.6		
	Dynamic web pages	[RES-3.3.3-01]	In order to present dashboards with the status of submitted files, web pages should be linked to the different databases and logbooks.	2.2.6		Postponed to the Copernicus follow-up project
	S/W	[RES-3.3.4-01]	All modules should be open source and developed in languages that do not require licenses : C/C++, Fortran, Python, bash	3.1		
		[RES-3.3.4-02]	Scientific modules might be adapted/improved for automatic processing and QA/QC procedures	N/A		
		[RES-CCN02-02]	MAPA algorithm should be optimized in terms of working memory	N/A		Not be done as

			management.			part of CCN02 and CCN03. Postponed to the Copernicus follow-up project
Security	**FTP/HTTP**	[SEC-3.4.1-01]	A password will be attributed to upload Level-1 data on FTP.	2.2.3		
		[SEC-3.4.1-02]	Users who want to download Level-2 data should register first.	2.2.6		
	Data and codes backup	[SEC-3.4.2-01]	Regular backup mechanisms should be set up in order to avoid any loss of data and to keep the history of the code.	3.5 4.1		
	Dashboards web pages	[SEC-3.4.3-01]	Data submitters should access only to the password-protected web pages related to their instruments.	2.2.6		Postponed to the Copernicus follow-up project
Quality	**Modularity and flexibility**	[QUA-3.5.1-01]	Code changes in one module should not affect the operation of the processing chain.	2.4		
		[QUA-3.5.1-02]	The system should be able to manage different module versions.	2.4		
		[QUA-3.5.1-03]	Possibility to restart an interrupted process from the step where the interruption occurred or a previous step.	2.4		
		[QUA-3.5.1-04]	Possibility to reprocess partially or totally the Level-1 database.	2.4		
		[QUA-3.5.1-05]	The system shall be able to test new releases of existing programs/algorithms in parallel with the automatic operation of existing ones.	2.4		
	Performance	[QUA-3.5.2-01]	The automatized processing chain shall deliver Level-2 output files between 6h and 24h after the spectra file submission.	2.3.2		
		[QUA-3.5.2-02]	If the process cannot successfully terminate, alerts should be sent to the appropriate people.	2.5.3		
		[QUA-CCN02-01]	The operational processing system should be validated in terms of response and performance for a limited period of time (2-4 weeks).	N/A		
	Scalability	[QUA-3.5.3-01]	The demonstration of the processing system should be based on a selection of 11 sites available from project partners + CINDI-2 data.	N/A		
		[QUA-3.5.3-02]	The system should be designed to be able to ingest more instruments and to support other products in a transparent way in the future.	2.3.1		
	Portability	[QUA-3.5.4-01]	The FRM_4DOAS processing system should be designed for Linux machines only.	3.1 4		
		[QUA-3.5.4-02]	The configuration files should be in a portable format (plain text, XML…).	2.4		
	Availability	[QUA-3.5.5-01]	Possibility to stop/restart the whole system with a general flag	2.4		
		[QUA-3.5.5-02]	Possibility to stop/restart a module with a flag	2.4		

		[QUA-3.5.5-03]	Possibility to suspend the process at any step for one instrument	2.4		
		[QUA-3.5.5-04]	Possibility to detect the best server to execute a job in order to optimize the processing time	2.4		
		[QUA-3.5.5-05]	Possibility to reprocess a given data set from or until a given module as far as input files have the required format and after providing the appropriate configuration files	2.4		
	Reliability	[QUA-3.5.6-01]	The integrity of the database should be kept in case of stop/restart of one or several modules or the reprocessing of a data set.	2.4		
		[QUA-CCN02-02]	The operational processing system should be validated through comparison with independent data sources.	N/A		
	Maintainability	[QUA-3.5.7-01]	The FRM4DOAS processing system will be fully maintained and continuously improved until the end of the project.	N/A		
		[QUA-3.5.7-02]	A technical documentation of the FRM4DOAS processing system will be provided at the end of the project. The code is open source and will be documented as far as possible.	N/A		
		[QUA-CCN02-03]	The technical documentation of the FRM4DOAS processing system should be updated in the course of the CCN02.	N/A		
		[QUA-CCN02-04]	DOI procedure and data policy approach should be implemented for the traceability and appropriate usage of the NDACC MAXDOAS service data products.	N/A		

5.2 Architecture design

5.2.1 Processing chain overview

Files of calibrated radiance spectra (the so-called Level-1 files) are uploaded by instrument data submitters on a dedicated BIRA-IASB incoming FTP server that is pooled every 15 minutes. After QA/QC test, Level-1 files are catalogued in a specific database. Then these files are processed by the core of the processing system which consists in the QDOAS spectral analysis software and the stratospheric and tropospheric profiling algorithms selected through the FRM$_4$DOAS Round-Robin exercise (see Section 4). QDOAS retrieves the slant column densities (SCDs) from the measured spectra and the resulting SCDs are then used as input by the profiling algorithms for retrieving vertical profiles and/or vertical column densities of the target trace gases. The final Level-2 products are delivered in two different file formats: netCDF master output files which contain all the variables used and generated by the retrieval and the standard GEOMS HDF4 files. Both file types are catalogued in a dedicated database. Only the GEOMS HDF4 files are made publicly available through the NDACC and EVDC DHFs while the access of the netCDF master output files are restricted to instrument data submitters through a password-protected web page available from the FRM$_4$DOAS web site.

The data flow in the processing chain is illustrated in Figure 15. As can be seen, an independent log analyzer is implemented to pool and analyze the collection of log files generated by the different modules. It allows on one hand, to detect anomalies in the processing chain and deliver e-mail alerts to the appropriate recipients (processing system administrators and data submitters); and on the other hand to generate various reports (status on processed files, statistics, list of anomalies). Data submitters are regularly informed about the status of the processing of their files though an automatic e-mail reporting service.

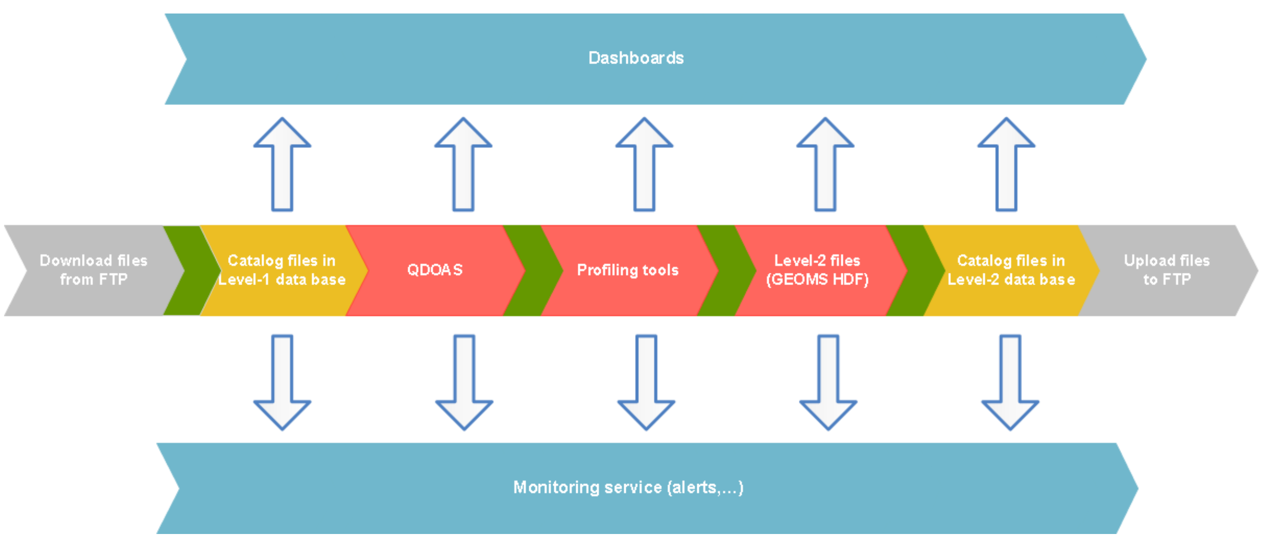


Figure 15: The FRM4DOAS processing chain

5.2.2 Architecture design overview

In order to exploit multi-processors environments and to satisfy flexibility and performance quality requirements, the processing system is based on an event-driven architecture which uses a succession of asynchronous modules (e.g. QDOAS or stratospheric NO_2 profiling modules) triggered by the presence of queues of input files. These queues, called trigger lists, are created by the preceding module(s) and contain a list of files to process. The execution of a module is controlled by an independent entity called wrapper. Its role is to check the presence of new input files in the corresponding trigger list, and for each new file in the list, to execute the module with the appropriate settings extracted from the metadata and configuration files. For example, the QDOAS module waits for validated Level-1 files and QDOAS output files could be used as input for one or several profiling modules.

A timestamp is assigned to each trigger list at its creation in order that the wrapper that pools the folder always selects only the most recent lists. Once the process is successfully executed, the names of output files are added in new trigger lists to be processed by the next modules. A new timestamp is assigned to these new trigger lists and, in order to track a file across the chain, their extension are the same as the original one completed with the extension specific to the next module. An example of the use of trigger lists is described in Figure 16.

Figure 16: Example of the use of trigger lists. Module M (e.g. QDOAS) will be executed three times with the files File_1.ext1,… if the timestamps A is higher than the last registered timestamp. After execution, according to the options associated with the files, two new trigger lists are created with the timestamp C to respectively trigger modules X (e.g. tropospheric profiling module) and Y (e.g. stratospheric NO_2 profiling). In this example, the same output File_2 is used as input for the two modules X and Y.

The processing system includes five wrappers controlling the main modules and their respective trigger list: Level-1 (incoming Level-1 files checking + dispatching in the Level-1 file database), QDOAS (DOAS spectral analysis), MAPA/MMF (NO_2 and/or HCHO tropospheric profiling), Total O_3 (total O_3 column retrieval), and Stratospheric NO2 (stratospheric NO_2 profiling). A flow chart presenting these wrappers and their interaction is shown in Figure 17.

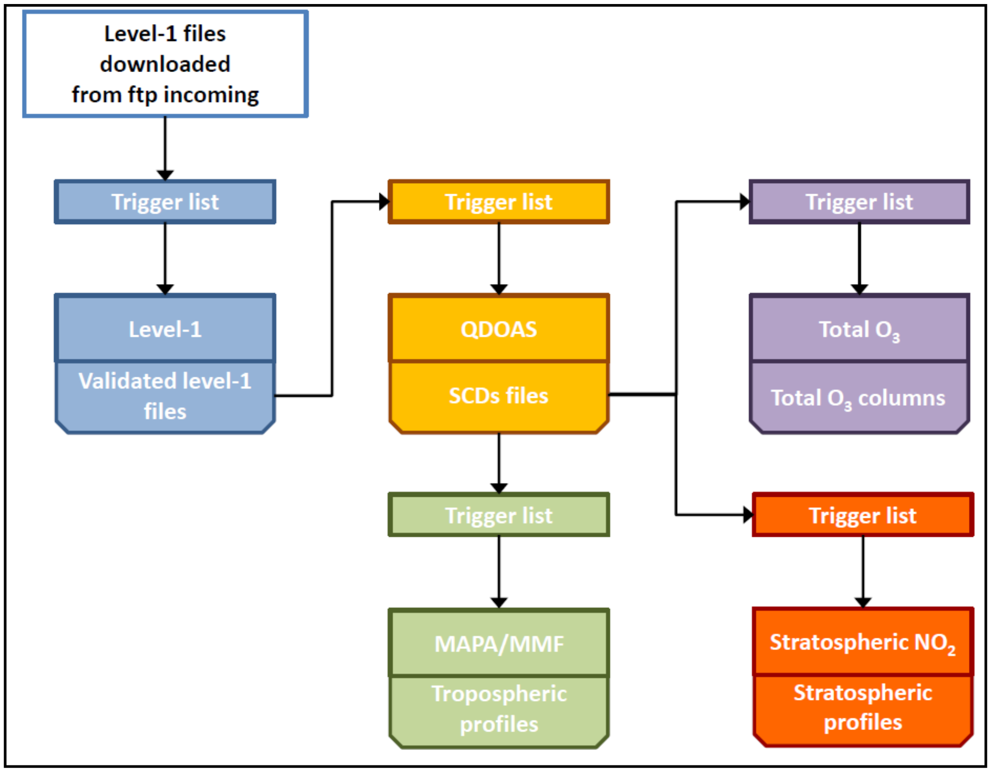


Figure 17: The five main wrappers of the processing system and their interaction through the corresponding trigger lists.

As mentioned above, the execution of a module should be performed using the appropriate settings and options. Those are provided through dedicated configuration files which can be divided in four categories:

- General (equal for all instruments) module specific configuration files (configuring the output files, quality check, etc)
- Instrument and module specific configuration files (specific retrieval settings for a certain module and instrument)
- System configuration files (e.g. tables including the different variables for validating the input file names (institutes, instrument number, stations names,…))
- General configuration files (such as template tables for creating the GEOMS files)

When a new Level-1 file is submitted, the file name has to be validated using system tables. The metadata file is selected with the instrument number. The file names includes information such as the instrument (identified by an instrument number and a channel number), the institute, the station, the process version… This information allows selecting the right configuration file from a table that associates the instruments to their corresponding configuration files to execute.

This first configuration file (instrument_config_file) indicates which modules to activate in the processing chain and redirects to configuration files with the settings appropriate to the instrument for these modules.

On the system side, there are configuration files with general options for the supported modules (paths where to find the module, paths of ancillary data, available values for given options, …)

Such a system is flexible enough to allow:

- To describe the path to follow for a file in the different modules of the processing chain
- Redirecting the output to different trigger lists (for example, the output of QDOAS can be redirected to the stratospheric and the tropospheric profiling tools or only one of these modules).
- That a same file is executed twice by the same module but with different configuration settings (two lines in the configuration file or two configuration files).
- To enable/disable the execution of a module or the processing of an instrument.
- Reprocessing of a large amount of files is possible at any point of the processing chain by generating the starting triggers lists. Data to reprocess must have satisfied QA/QC procedure before. If necessary, temporary configuration files can be inserted in the system.
- Processing of data set that are not part of the automatic process is possible after creating the appropriate configuration files
- To test a new version of the existing modules or a new module in parallel to the processing chain

In order not to interfere with the real-time processing when a reprocessing is performed, the availability of the processors is accounted for and a priority level is assigned to each process (or processes are distributed in time by generating lists with different timestamps).

The meta-languages XML and YAML are a logical choice for master configuration files because they offer a high level of flexibility and modularity. The format is plain text, i.e. human readable in any text editor; the syntax is very simple: options can be presented as pairs of attributes/values eventually embedded in tagged sections. The number of options can be progressively extended with needs without revising the complete file. In addition, most programming languages already include functions to read XML and YAML files. Specific tools have been developed to generate and handle these configuration files.

An important aspect of such central processing systems like FRM$_4$DOAS is to monitor the status of the processed files and to keep the data submitters informed on a regular basis about this status. This has been achieved through the development of a combined log analyzer, reporting and mailing service system. In brief, in this system, each wrapper generates log files and history files per processed job (see Figure 18). Log files contain all messages output by the processing system including errors, warnings, debug... while history files is a kind of log book (list of processed files and per file entry, the process id, the start and end times, the return code).

The content of history files and log files is summarized per Level-1 file by the history processor and the log processor, respectively. From the log files, only the errors lines are retrieved. The report processor browses the last individual reports generated within a given period and output the information of the last wrappers processing (history and log errors). If some options are enabled (for example, warnings), the report processor retrieves the request information from the log files if available.

The mail delivery service calls the report processor to generate a report and sends it automatically to the data submitter. The log analyzer is able to:

- Filter the information (processing errors only, system errors, warning)
- Deliver report on a daily, weekly, monthly basis
- Deliver alerts in case of a flux interruption
- Store the information needed to make statistics (for example, execution time average, etc) in the history files (however, this information has not been exploited so far)

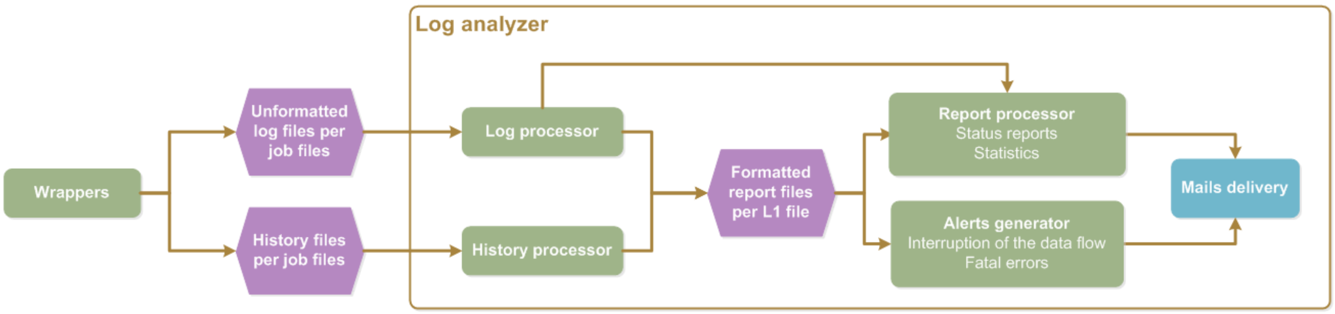


Figure 18: Overview of the log analyzer, reporting and mailing service system.

Further details of the architecture design of the FRM$_4$DOAS centralised processing system can be found in deliverable D8 ('Processing System Architecture') and D20 ('MAXDOAS Network Operational Processing System Architecture Design Document').

5.3 Processing system code description

A thorough description of the processing system code can be found in the documentation provided with the software package. This documentation consists in html pages as illustrated in Figure 19. This format was chosen because it offers a high level of flexibility and comfort in terms of reading: a general description of the processing system software including how to install and run it can be found, but also detailed information on all individual routines, input/output file formats, etc through simple clicks on dedicated html links. Moreover, since this documentation is automatically generated from the code itself using the Doxygen software, the preparation of the successive updates has been made easier.

Figure 19: Main html page of the FRM_4DOAS processing system software documentation.

5.4 Data processing status at the end of the project main phase

After conversion to the netCDF format specifically developed for FRM_4DOAS and format testing by BIRA-IASB, project partners have started in June 2018 to submit in NRT (24h latency) their level-1 spectra files from their stations to the centralised processing system. Table 14 gives the status of the level-1 files submission and processing in November 2018 (end of the project phase I).

Table 14: Status of the station data processing at the end of project phase I. At that time, no data were available from Thessaloniki, Izana, La Réunion Maïdo, Lauder, and Neumayer stations (see Table 1 in the Introduction Section).

STATION	Instr. id nr.	Geometry	Channel nr.	Daily submission start date	MMF/MAPA	Total O_3	Stratospheric NO_2
XIANGHE	1669	maxdoas	2	01/07/2018	x	x	x
UCCLE	1670	maxdoas	2	01/07/2018	x	x	x
HARESTUA	1671	zenith	2	29/06/2018	N/A	x	x
NY.ALESUND	1672	maxdoas	2	01/07/2018	x	No twilight	No twilight
BREMEN	1673	maxdoas	2	01/07/2018	x	x	x
ATHENS	1674	maxdoas	1	01/07/2018	x	x	x
DEBILT	1675	maxdoas	1	19/07/2018	x	x	x
MAINZ	1676	maxdoas	4	31/07/2018	x	N/A	x
HEIDELBERG	1679	maxdoas	2	01/08/2018	x	x	x

In addition, the CINDI-2 spectra files from the following instruments and corresponding to the semi-blind intercomparison period (12-28 September 2016) have been processed for the validation of the processing system (see Table 15):

Table 15: Status of the CINDI-2 campaign data processing.

Instrument number	Affiliation	Geometry	Channel nr.	MMF/MAPA	Total O_3	Stratospheric NO_2
aiofm-1	Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Hefei	maxdoas	1	x	N/A	x
auth-3	Aristotle University of Thessaloniki, Thessaloniki	maxdoas	2	x	N/A	x
bira-4	Royal Belgian Institute for Space Aeronomy, Brussels	maxdoas	2	x	x	x
boku-6	Universität für Bodenkultur Wien, Wien	maxdoas	2	x	x	x
cma-7	China Meteorological Administration, Beijing	maxdoas	1	x	N/A	x
cma-8	China Meteorological Administration, Beijing	maxdoas	1	x	x	x
dlrustc-13	Deutsches Zentrum fuer Luft- und Raumfahrt, Wessling; University of Science and Technology of China, Hefei	maxdoas	2	x	x	x
dlrustc-14		maxdoas	2	x	x	x
iupb-18	University of Bremen, Bremen	maxdoas	2	x	x	x
iuph-19	University of Heidelberg, Heildelberg	maxdoas	1	x	N/A	x
knmi-23	Royal Netherlands Meteorological Institute, De	maxdoas	2	x	N/A	x

	Bilt					
luftblick-27	LuftBlick, Mutters	maxdoas	2	x	x	x
mpic-28	Max-Planck Institute for Chemistry, Mainz	maxdoas	1	x	N/A	x
lmumim-35	Ludwig-Maximilians-Universität München, Munich	maxdoas	2	x	x	x

5.5 Processing system validation during project phase I

Validation of the Processing System is complex and can include many different aspects like the validation of the profiles retrieved by the system using independent profile information but also the validation of technical aspects such as timeliness of processing of data, consistency of formats, feedback to the users and proper reaction to error situations.

The validation approach adopted in FRM_4DOAS was the following:

1. Perform a technical verification of the processing system, e.g. are the log and history files corresponding to the different steps of the processing successfully and timely created? Are the appropriate output files successfully created after each processing step?
2. Apply the processing system to data from a subset of instruments operating during the CINDI-2 campaign, evaluate the data for consistency, plausibility and coverage, and compare the results to validation data available from the campaign. This part is based on work performed in the framework of the CINDI-2 profile intercomparison exercise that has been published in Tirpitz et al. (2021).
3. Apply the processing system to a full month of data of the Bremen MAX-DOAS instruments in Bremen, Athens and Ny-Alesund, to evaluate these data for plausibility, consistency and coverage, and to compare the results with those obtained from the standard data evaluation performed at IUP Bremen.

Since the FRM_4DOAS validation results are extensively described in deliverable D10, only the main outcomes of the study are given here.

5.5.1 Technical verification

The technical verification has mainly consisted in checking the presence and content of the log and history files for all processing steps starting from the checking and cataloguing of Level-1 spectra files to the generation of profiling results. For each wrapper, the presence of corresponding output files in the correct format (e.g. QDOAS analysis result netCDF file after the QDOAS processing of Level-1 spectra files) has been also carefully verified.

These verification tests have been performed for a selection of example Level-1 spectra files from FRM_4DOAS stations that cover different scenarios (success or failure at different steps of the processing chain).

Regarding the timing of a process, the start time and end time of individual modules are registered in the log and history files. The total duration time for the processing of a radiance file depends on the overhead time (i.e. the time between the update of a trigger list with new entries and the execution of the corresponding wrapper for these new entries based on the wrappers schedule presented Table 16) but also on the number of files to process and the availability of the CPU reserved to the wrapper.

Table 16: Check frequency of the input trigger lists for all wrappers. In addition, the log analyser and reporting mail processor are pooled every hh:28 and hh:12, respectively.

Wrapper name	Check frequency
L1_incoming	hh:01 to hh:59 every 15 min
qdoas	hh:03 to hh:59 every 15 min
MMF/MAPA	hh:07 to hh:59 every 5 min
no2strato	hh:09 to hh:59 every 15 min
o3strato	hh:07 to hh:59 every 15 min

As can be seen in Table 16, there is a staggering of the check start time of the different wrappers. This time lag between successive wrappers (e.g. L1_incoming -> qdoas -> MMF/MAPA) allows to reduce the processing time: e.g. delaying the check of the qdoas wrapper by two minutes with respect to the L1_incoming wrapper gives the possibility to start the QDOAS process of the level-1 files catalogued between hh:01 and hh:03, which is not possible if the QDOAS process would have started at the same time of the L1_incoming wrapper. In that later case, the qdoas wrapper would have to wait hh:01 + 15 min before starting to process the catalogued level-1 files. Regarding the log analyser and reporting mail processor, they are pooled once per hour (check start times arbitrarily chosen at hh:28 and hh:12, respectively).

The report e-mailing system has been also verified, in particular the consistency between the status of a process (failed/successful + error source(s) if any) reported in the log file and the corresponding message reported in the e-mail sent to the processing system administrators and/or data submitters.

5.5.2 Validation using CINDI-2 data

Data from four selected instruments (see Table 17) was processed by the prototype processing system for NO_2, HCHO and aerosol in the UV and visible spectral ranges. For NO_2, only the visible retrievals were evaluated. As the AUTH instrument only has an UV channel, no NO_2 and visible aerosol data from that instrument are available. All data was then converted to the format required for the profile intercomparison study for CINDI-2, and run through the software and analysis package developed by J.-L. Tirpitz (IUPHD) for this exercise (see Tirpitz et al., 2021).

One important part of the output of the processing system are quality flags for each profile. As the CINDI-2 intercomparison software does not allow for warnings, only for valid and in-valid profiles, the QA flags from the FRM_4DOAS processor have been mapped onto the CINDI-2 flags by making all MMF retrievals valid unless an error was flagged while MAPA retrievals were flagged as valid only if neither errors, nor warnings were raised. This different treatment of the two retrieval types was recommended by the retrieval developers. In the following comparisons, often results are shown for both, the complete data set and only those data which were flagged as valid. In addition, sometimes cloudy and clear-sky scenarios are reported separately.

Table 17: Selected CINDI-2 MAX-DOAS instruments for the FRM_4DOAS processing system validation.

Algorithm	Institute	Code	Symbol
MAPA (0.8)	BIRA	BIR	●
	IUP-Bremen	IUP	▲
	AUTH	AUT	■
	DLR/USTC	DLR	▲
MAPA (1.0)	BIRA	BIR	●
	IUP-Bremen	IUP	▲
	AUTH	AUT	■
	DLR/USTC	DLR	▲
MAPA (free)	BIRA	BIR	●
	IUP-Bremen	IUP	▲
	AUTH	AUT	■
	DLR/USTC	DLR	▲
MMF	BIRA	BIR	●
	IUP-Bremen	IUP	▲
	AUTH	AUT	■
	DLR/USTC	DLR	▲

For the validation of the retrievals on MAX-DOAS data, different measurements are available, including:

- Long-path-DOAS (LP-DOAS) measurements of NO_2 and HCHO for the surface concentrations
- Direct sun DOAS measurements for NO_2 tropospheric columns
- Sun-photometer measurements for AOD at different wavelengths
- Ceilometer data for relative extinction profiles
- NO_2 vertical profiles from the KNMI NO_2 sondes and the RIVM NO_2 lidar observations for a few short time periods during the campaign

5.5.2.1 Overview of the results

As a first step, overview figures on the profiles retrieved by the 4 algorithms on the data from the 4 (3 in the visible range) instruments were created as illustrated in Figure 20 for NO_2 visible. Where available, the lowest rows also show independent comparison data. The corresponding figures for aerosols and HCHO can be found in deliverable D10. From all these figures, the following observations can be made:

- **Aerosol at 360 nm:**
  1. On average, all three MAPA retrievals create vertical profiles with more aerosol in higher layers than MMF
  2. MMF has a tendency to retrieve elevated layers, both in the presence and absence of clouds
  3. In the presence of clouds, the elevated layers retrieved by MMF are often in similar altitudes as shown in the ceilometer data
  4. From the visual impression, MAPA AOD appears to be larger than that retrieved by MMF.

5. Differences between MMF and the three MAPA variants are larger than difference between different MAPA versions
6. While results for the two high SNR instruments BIRA and IUP-UB are often similar, results for AUTH are often different (less vertical structure) while the DLR instrument is often in between
7. A large number of profiles are flagged as invalid, in particular for MAPA. Flagging between MAPA and MMF appears to be not consistent

- **Aerosol at 477 nm**: similar observations as for 477 nm are made with some differences. In particular

1. There are even more defined layers in MMF at visible wavelengths
2. The consistency between instruments is better in the vis
3. There are less profiles flagged as invalid
4. Flagging between UV and visible is not consistent

- **HCHO:**

1. The temporal evolution of HCHO over the campaign is similar in all retrievals
2. There are quite a few profiles where unrealistically large HCHO values are retrieved by MAPA, in particular if the O_4 factor is free to vary. However, all these cases are flagged as invalid.
3. As was the case for aerosols, MMF often retrieves elevated layers, which is not the case in any of the MAPA variants.
4. HCHO profiles retrieved from BIRA and IUP-UB data are quite consistent, while AUTH profiles seem to be confined more to the lowest layers. DLR profiles are somewhere in between.
5. In MAPA retrievals, many if not most profiles are flagged; for example, not a single HCHO profile of the AUTH instrument was flagged as valid.

- **NO_2:**

1. Most profiles are confined to the lowest layers
2. There is much better overall consistency between different retrievals as for the other three products
3. There still are some failed profiles for MAPA free
4. Also for NO_2, a large fraction of the profiles is flagged as invalid
5. The DLR retrievals show consistently elevated layers for NO_2 for MMF during the first days

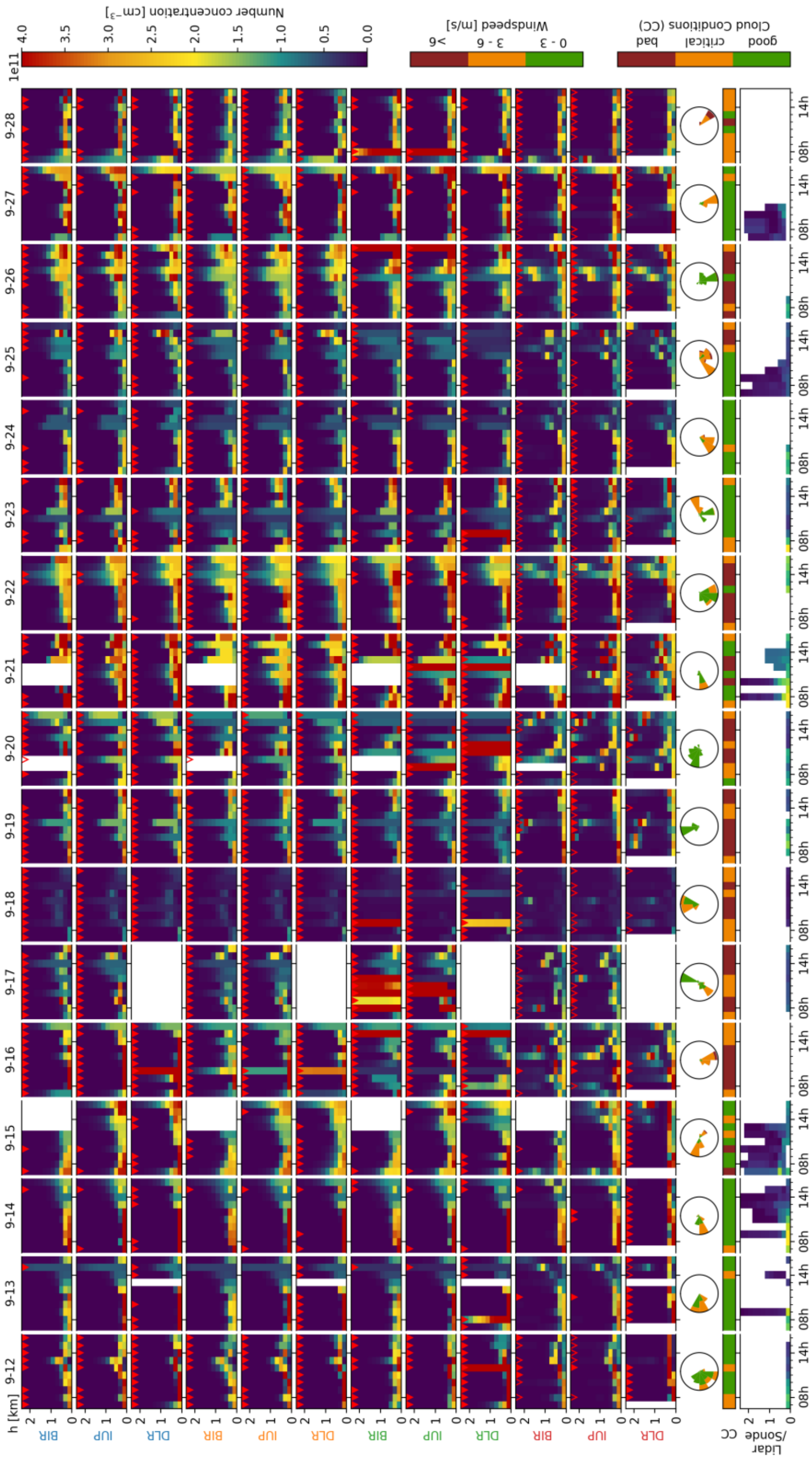


Figure 20: Overview on NO_2 retrieval results for 460 nm. Results for the 4 algorithms are shown from top to bottom (MAPA 0.8, MAPA 1.0, MAPA free, MMF, each for the three instruments providing visible radiances. The lowest three lines show wind direction, cloud conditions (derived from MAX-DOAS data with the MPIC cloud classification algorithm as described in Wagner et al., 2014) and LP-DOAS surface concentrations as well as NO_2 sonde and NO_2 lidar profiles where available. Red triangles indicate flagged results.

5.5.2.2 Retrieval quality check

The quality of MAX-DOAS profile retrievals is usually evaluated through the level of agreement between measured and modelled DSCDs and in case of the OEM-based retrievals, also through the averaging kernels and DOFS (see also Sections 4.2.2.5 and 4.2.2.6, respectively).

Figure 21 shows the correlation of measured and modelled DSCDs for O_4 in both the UV and visible, HCHO, and NO_2. On overall, measured DSCDs are reproduced very well by all retrievals. However, mainly the different noise levels are apparent, resulting in more scatter in the UV than the visible, in particular for data from the AUTH instrument. There also are some clear outliers in the BIRA UV data (some even out of the plotting range) because of instrumental problems on some days. Comparing the four retrievals among each other, MAPA with fitting of the O_4 scaling parameter often produces the smallest scatter, with the exception of the NO_2 retrievals. This should probably be expected as it adds another free parameter to the fit which can adapt the modelled values better to the measured ones. This does however not automatically imply better profiles.

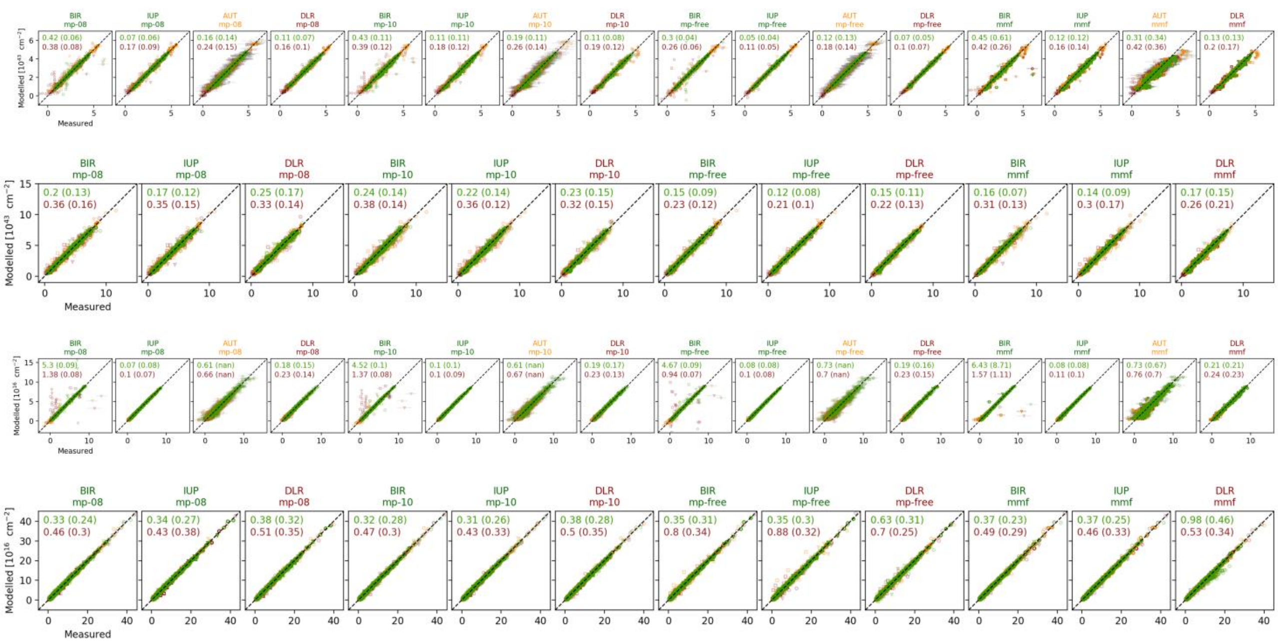


Figure 21: Correlations of measured and modelled DSCDs for O_4 in the UV, O_4 in the visible, HCHO, and NO_2. In each line, again results from all contributing instruments are shown, grouped by retrieval (MAPA 0.8, MAPA 1.0, MAPA free, and MMF). The numbers indicate the RMS of the DSCDs, for clear sky (green) and cloudy (red) conditions as well as for all data and in brackets results only for valid data.

An overview of the MMF averaging kernels can be found in Figure 22. As can be seen, most of the information is very close to the surface and the gain in information and the vertical resolution drop quickly with altitude (AVKs quickly broaden and are less peaked). The visible AVKs are also nearly identical for the instruments in the visible spectral range while in the UV, the DLR instrument has less vertical information than BIRA and IUP-UB but more than AUTH which has clearly lower information content, presumably because of higher noise levels.

In terms of DOFs, the profiles from the different instruments have very similar information content in the visible aerosol retrievals as well as in the NO_2 profiles, while for the UV retrievals, AUTH has lower information content for both aerosols and HCHO. It is also worthy to note that the temporal evolution of the DOFS over the course of the CINDI-2 campaign (not shown here) remains surprisingly constant, even during days with full or broken cloud coverage.

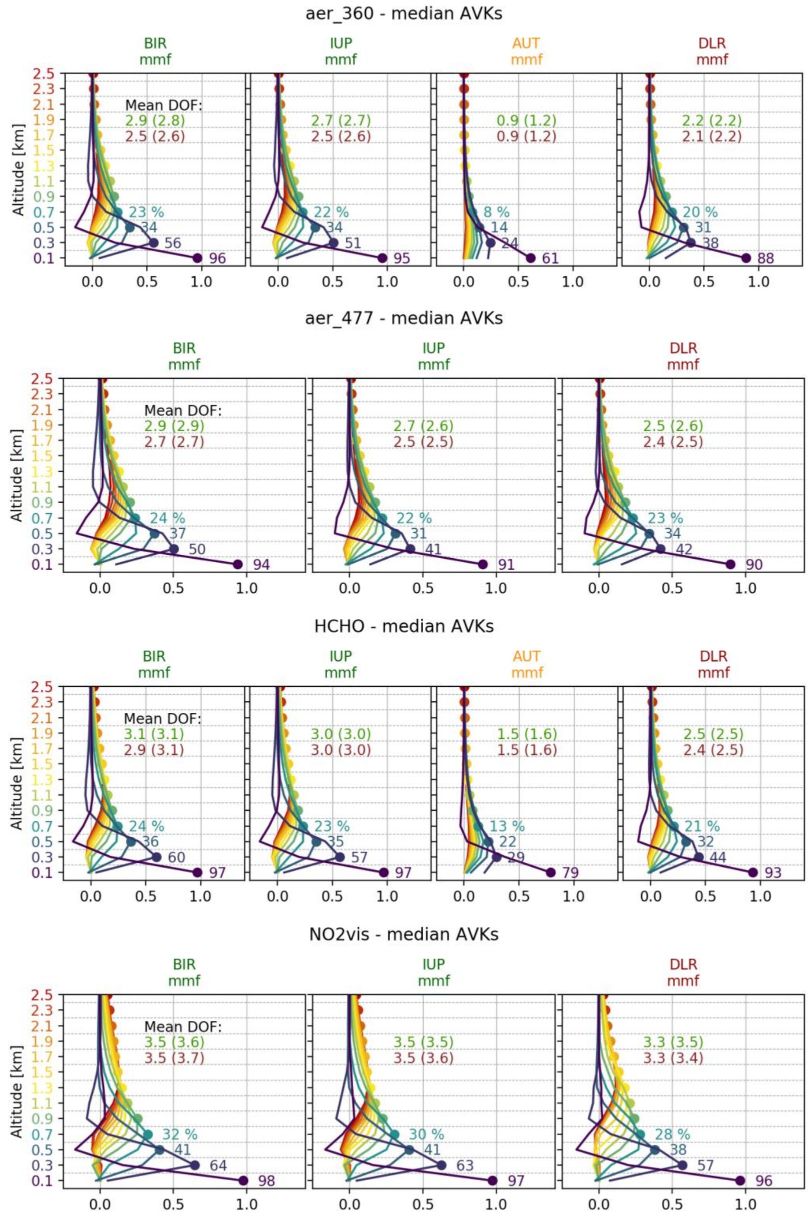


Figure 22: Median averaging kernels of the MMF profiles for aerosols at 360 nm and 477 nm, HCHO, and NO_2 (from top to bottom). Also indicated are the degrees of freedom (DOF) for clear sky (green) and cloud scenes (red). Values in parenthesis are for valid data only.

5.5.2.3 Validation of surface concentration and columns

In addition to the profile itself, there are two additional important outputs of a profile retrieval: the tropospheric vertical column/AOD and the surface concentration/extinction. Both can be compared to independent measurements, which makes them easier to validate than the profiles and also they are important for air quality applications and satellite data validation.

For the CINDI-2 campaign, surface concentrations of NO_2 and HCHO can be compared to results from continuous LP-DOAS measurements taken during the campaign in a measurement volume close to that observed by the MAX-DOAS instruments. However, it has to be kept in mind that even the lowest point in the retrieved profiles is sensitive to absorption in higher atmospheric layers (see AVKs in Figure 22), meaning that MAX-DOAS instruments might see the real surface concentration smeared into layers above.

In Figure 23, the results are shown for both the median of the data sets and for individual data points. For HCHO, the median of the retrievals fits excellently to the LP-DOAS values throughout the campaign, regardless of weather conditions. This is surprising as HCHO absorption is small and HCHO profiles are usually subject to noise. Possible explanations are less vertical variability making the a priori used more representative of the real situation or less horizontal variability when compared to NO_2. When comparing individual results then MAPA 1.0 and MMF have a tendency to be lower than the other two MAPA versions on many days. Overall the scatter of individual results is not insignificant but mostly of the order of 30%.

For NO_2, there is more variability, and at least on some days, there is a clear underestimation of the surface NO_2 in the morning by the median of the MAX-DOAS retrievals, mainly driven by MAPA results. When looking at individual values, the scatter is large, partly more than 100% and no clear pattern of which algorithm or instrument yields lower or higher values can be identified. The large variability is a surprise given the much stronger absorption signal when compared to HCHO, but the confinement of the NO_2 to a shallow layer close to the surface makes retrieval of surface concentrations from MAX-DOAS measurements difficult. As is the case for HCHO, there is no clear indication for better results under clear-sky conditions.

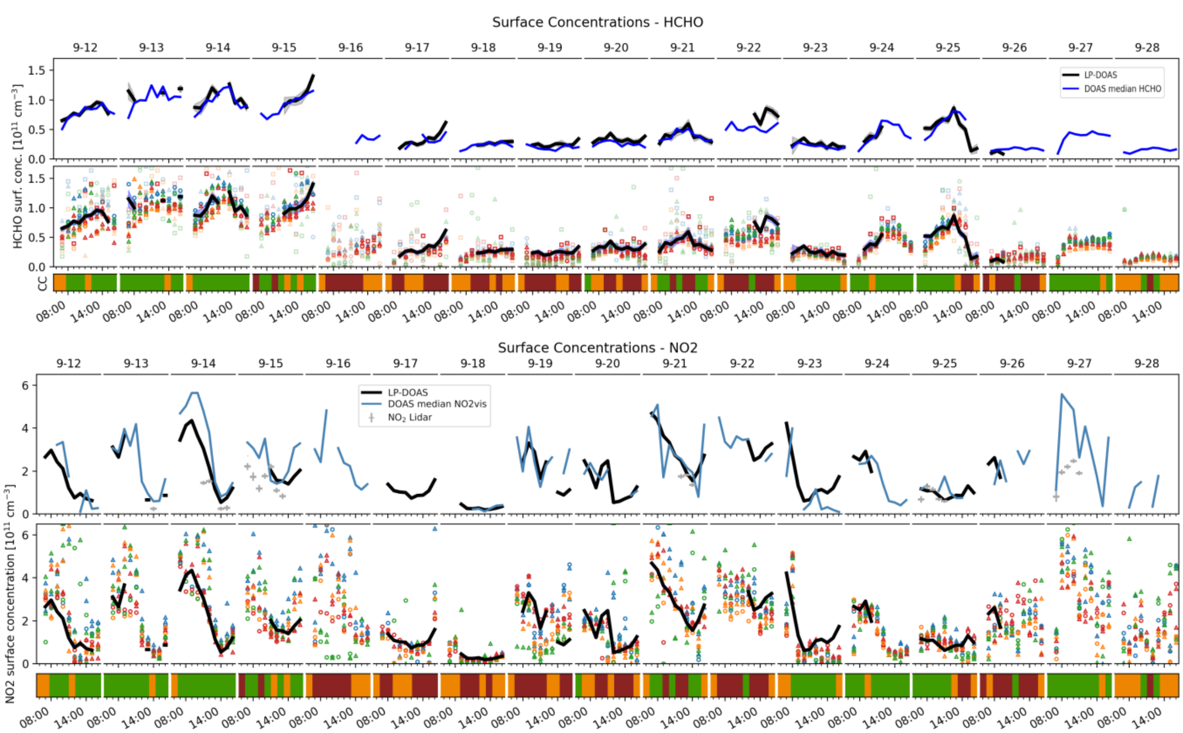


Figure 23: Comparison of retrieved surface concentrations of HCHO (top) and NO_2 (bottom) with LP-DOAS observations. The upper panel in each figure shows the comparison between LP-DOAS (black) and the median of the data (blue), the lower panel includes all individual values. Results from invalid profiles are shown in washed out colours. For NO_2, lidar data are included as grey crosses where available.

In addition to surface concentrations, also vertical tropospheric columns of NO_2 and aerosol (AOD) were compared to independent measurements from direct sun observations and a CIMEL sunphotometer, respectively. In this report, only the MAX-DOAS/CIMEL AOD comparison will be discussed.

Figure 24 shows that MAX-DOAS AOD retrievals follow the temporal evolution of sun-photometer AOD well. In particular during the first days, agreement with the partial AOD (evaluated by combining the vertical extinction profile from the ceilometer with the MAX-DOAS averaging kernels to compute that part of the sun-photometer observed AOD that is accessible to MAX-DOAS measurements; see Tirpitz et al., 2021) is clearly better than with the full AOD as expected. When evaluating individual retrievals, there is a clear tendency for MMF and MAPA 1.0 retrievals to underestimate sun-photometer results whereas the other two MAPA retrievals are closer to the validation data. This could be interpreted as support for the need of an O_4 scaling factor as for example discussed in Wagner et al. (2019).

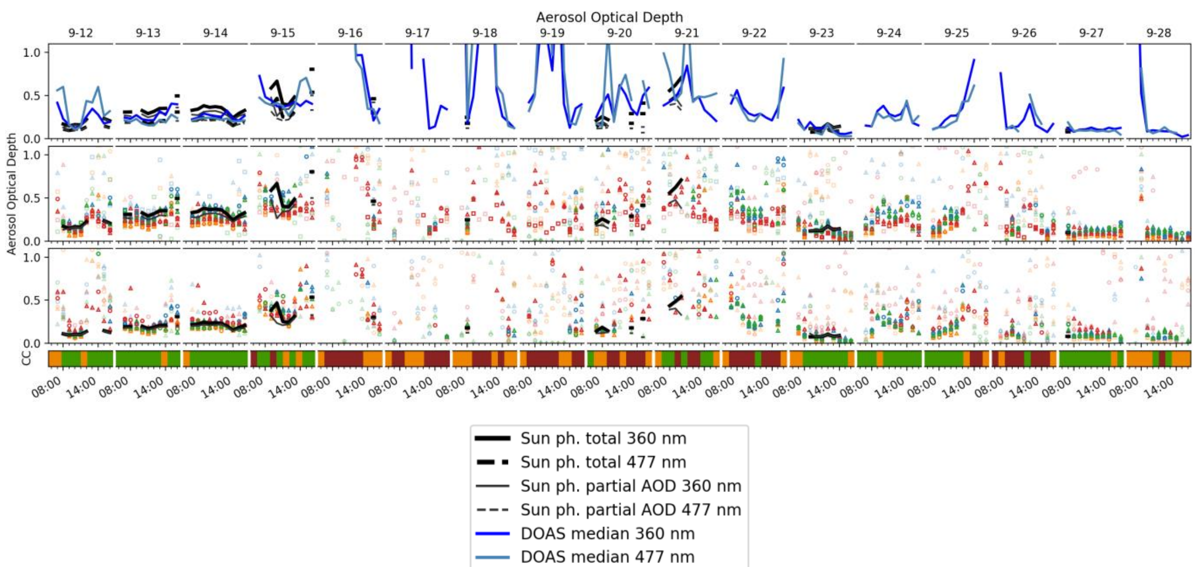


Figure 24: Comparison of MAX-DOAS and sun-photometer retrieved AOD during the campaign. The first row shows the comparison between medians and total and partial sun-photometer AOD, the second and third row show all results for the UV and the visible aerosol retrievals.

As expected, AOD values vary widely during cloudy days, and no validation data is available as sun-photometer measurements are only possible during clear-sky periods.

5.5.2.4 Validation of surface concentration and columns

In addition to the quality of the retrieved profiles, also the number of successful retrievals is an important parameter. This number depends on the robustness of the retrieval, the assumed uncertainties in the measurements but most importantly on the flagging applied. If the criteria for removing profiles are too weak, poor profiles will be included in the output which users will not appreciate. If filtering is too aggressive, only few profiles remain and users have nothing to work with.

A statistical evaluation was performed for the number of profiles retrieved for the different quantities, instruments and retrievals (not shown here, see deliverable D10). The main observations were that

- Overall, roughly 50% of all profiles are flagged as valid.
- For the UV retrievals, MMF is much more generous than MAPA. For example, MAPA does not retrieve a single HCHO profile for the AUTH instrument while MMF reports 53% of successful retrievals. Also for the DLR instrument, much more MMF results are present than MAPA results.
- For MAPA, the number of successful retrievals is low if an O_4 scaling factor of 1.0 is used. The other two MAPA variants result in comparable numbers of profiles.

The differences are mainly linked to the way the flagging is implemented in the two different retrievals and therefore can potentially be tuned once more data is available. There is however also a basic difference in that MMF uses a priori information, which may result in reasonable profiles even if little information is in the measurements while MAPA has no such regularisation. This may also explain why MMF results were found to be of comparable quality as MAPA results, although much less data is removed by flagging.

5.5.3 Validation using IUP-UB retrievals

In addition to the comparison of CINDI-2 results, also one month of FRM$_4$DOAS retrievals for the IUP-UB MAX-DOAS stations in Bremen, Ny-Alesund and Athens was evaluated. These data were produced in the automated NRT mode, where spectra files were transferred from the instruments at night and automatically ingested and processed in the FRM$_4$DOAS demonstration processor. Data for August 2018 were selected as this month had few data gaps in the IUP-UB data and had many days with favourable observing conditions. It should be noted that the selected three stations differ in their characteristics with

- Bremen being a moderately polluted mid-latitude site.
- Athens being a polluted site with a complex topography and an instrument position on a hill close to the city at 350m altitude.
- Ny-Alesund being an Arctic clean air site with some local NO_2 pollution from cruise ships and a power generator which occasionally is blown into the viewing direction of the MAX-DOAS instrument. During summer, there is polar day with round the clock observations

The FRM$_4$DOAS results have been compared to retrievals from the IUP-UB BOREAS algorithm (Bösch et al., 2018) and to the standard NDACC retrievals. The BOREAS retrieval is an Optimal Estimation based profiling retrieval which for the aerosol retrieval applies additional Tikhonov regularisation. It also differs from other retrievals in that it applies pre-scaling of the a priori profiles using a quick pre-analysis and can apply dynamic weights to the regularisation but the latter option is not used here. BOREAS is still a rather new retrieval and has only been tested in detail on synthetic and CINDI-2 data (see Frieß et al., 2019 and Tirpitz et al., 2021) as well as on Bremen data (Bösch, 2019). The standard NDACC retrieval is based on a simple AMF approach applied to the 30° elevation measurements without profile retrieval. As climatological assumptions are applied to the vertical profile as well as the aerosol load, one would expect that the results are less accurate than the results from the profile inversions. On the other hand, the simple AMF retrieval is more robust and does not suffer from the instabilities sometimes found in profile retrievals. In this report, only the comparison results with BOREAS will be discussed (see deliverable D10 for the complete evaluation).

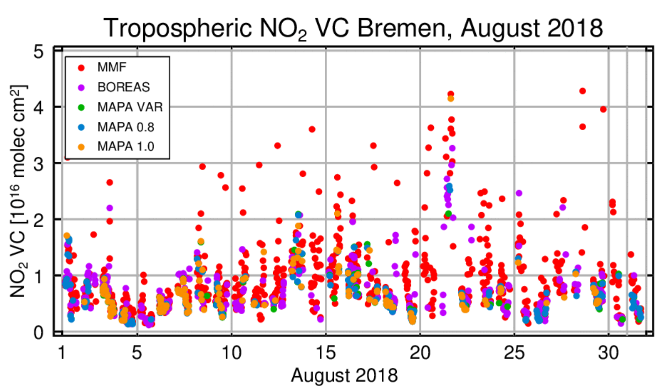
The time evolution of the MAPA/MMF and BOREAS tropospheric NO_2 columns is shown in Figure 25 for the three stations. Different scales are used for better representation of the variability. The main findings are that

- In Bremen, the overall evolution is similar between the retrievals, but MMF has a tendency of high outliers
- In Athens, there is very good agreement in the time evolution, but BOREAS results appear to be higher than the FRM_4DOAS retrievals
- In Ny-Alesund, there is more scatter, and MMF has many unexpectedly high results. For the three MAPA algorithms, very few retrievals are flagged as successful.

How different the five algorithms treat flagging is summarised in Table 18 – MMF is very generous, while MAPA is very strict. BOREAS is somewhere in between. Close inspection of the data shows, that not only the number of retrievals flagged as successful varies, but also the times for which profiles are deemed valid. As a result, a direct comparison of NO_2 vertical columns between the retrievals leads to only a relatively small number of coincidences even for a full month of data.

Table 18: Number of successful NO_2 retrievals for August 2018 data from the three IUP-UB stations

Station	MMF	MAPA 1.0	MAPA 0.8	MAPA VAR	BOREAS
Bremen	803	155	201	163	311
Athens	1272	349	306	480	1217
Ny-Alesund	1694	31	68	67	906



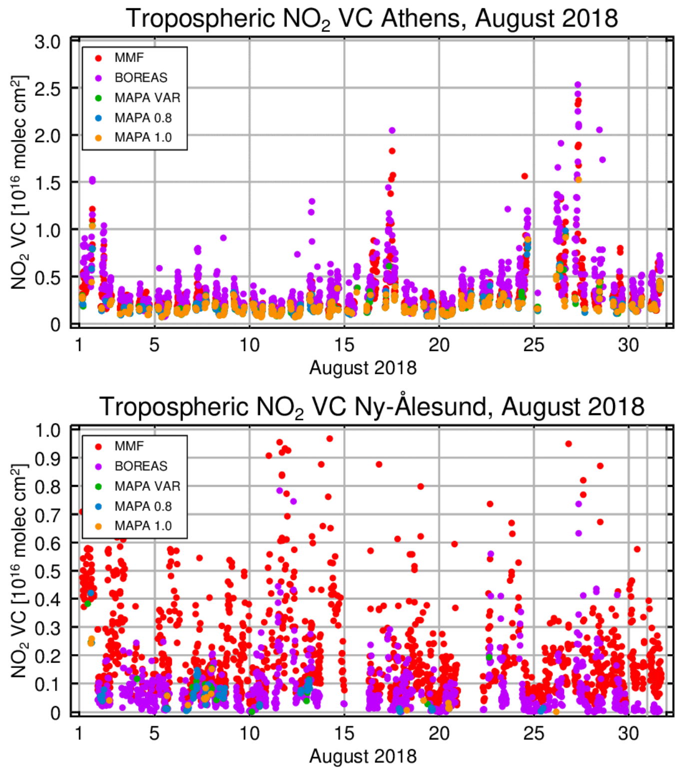


Figure 25: Comparison of tropospheric NO_2 vertical columns for the three IUP-UB stations in August 2018. All flagged data is shown with the exception of some MMF outliers, which are off scale.

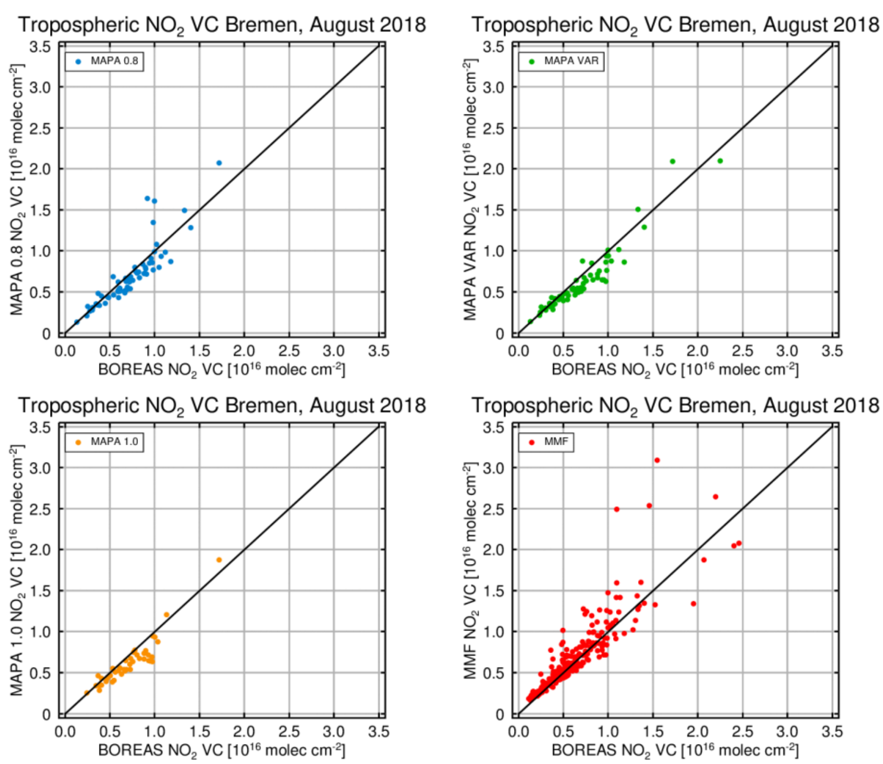


Figure 26: Correlation plots between tropospheric NO_2 columns for the four FRM$_4$DOAS retrievals and the UB BOREAS results. All flagged data from August 2018 is used. Results of the linear regression are summarised in Table 19.

Comparisons of matching tropospheric NO_2 VCDs for all three stations are shown as scatter plots in Figure 26, Figure 27, and Figure 28, always with the BOREAS retrievals as x-axis. The results from a linear regression on these data are given in Table 19. The main conclusions from the comparison are:

- For Bremen, the agreement of the tropospheric NO_2 columns is good, with MAPA underestimating and MMF overestimating the BOREAS columns. The scatter of MMF columns is larger, but also more values are provided.
- For Athens, the correlation is good for all three FRM_4DOAS retrievals, but BOREAS columns appear to be systematically higher. This can be explained by the approach taken in BOREAS for the high altitude station: It is modelled as a flying instrument. This choice was made as clearly much of Athens' pollution is below the measurement altitude, and from the viewing point of the instrument, photons from below the station altitude will also contribute to the scatter light observed. Therefore, BOREAS profiles extend below the instrument's altitude, resulting in larger columns.
- In Ny-Alesund, very few MAPA retrievals are flagged as successful, but those agree very well with BOREAS: MMF on the other side is systematically much larger than BOREAS and MAPA and shows much scatter. This indicates that MMF still has problems for scenarios with low trace gas content (see deliverable D5), as for the Arctic clean air site Ny-Alesund. This issue could eventually be fixed by assuming, as part of FRM_4DOAS, more location-specific a priori profiles.

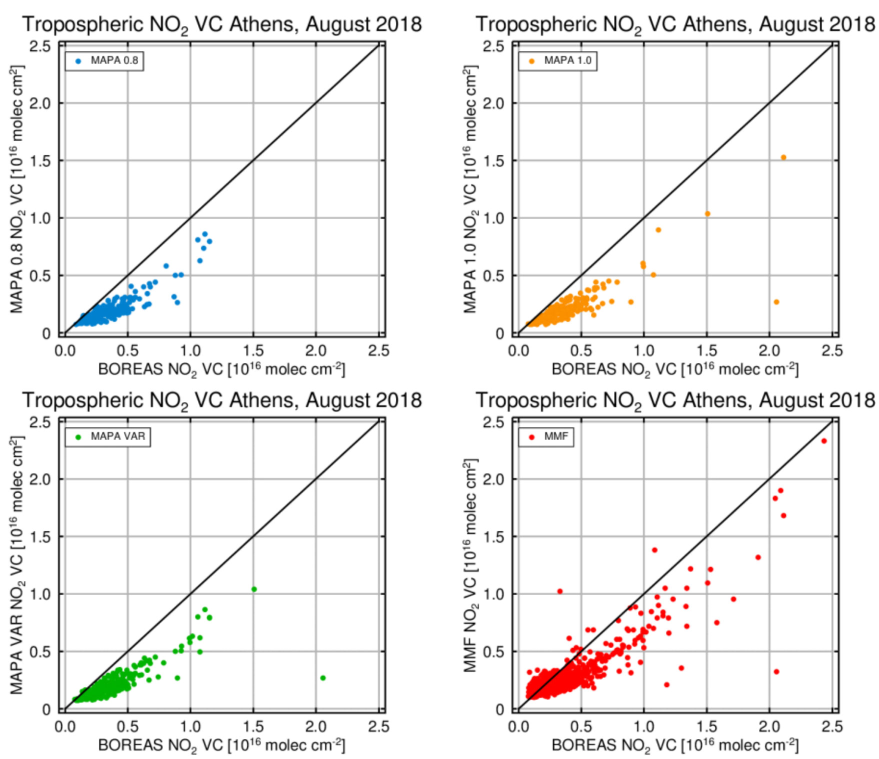


Figure 27: As Figure 26 but for Athens

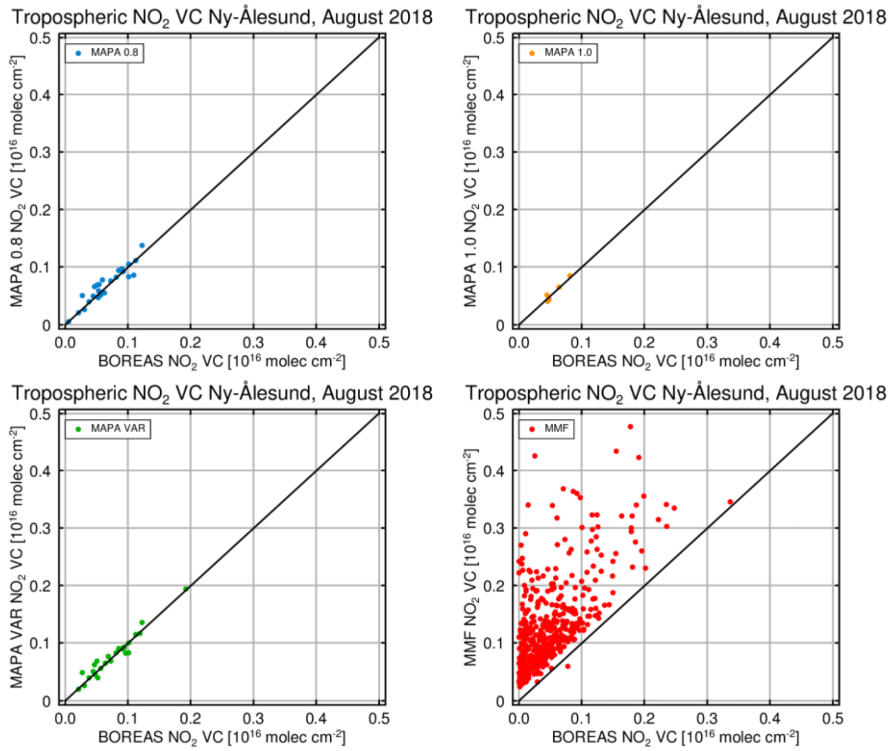


Figure 28: As Figure 26 but for Ny-Alesund

Table 19: Linear regression results of NO_2 retrievals for August 2018 relative to BOREAS results. The offset is given in units of molec cm^{-2}.

Station	MMF	MAPA 1.0	MAPA 0.8	MAPA VAR
Bremen correlation	0.91	0.93	0.88	0.95
slope	1.05	0.93	1.06	0.96
offset	3.2×10^{14}	-5.4×10^{14}	-7.3×10^{14}	-5.1×10^{14}
Athens correlation	0.89	0.84	0.89	0.86
slope	0.63	0.48	0.55	0.48
offset	3.7×10^{14}	2.3×10^{14}	1.1×10^{13}	2.6×10^{14}
Ny-Alesund	0.79	0.97	0.93	0.97
correlation	1.59	1.1	1.02	0.96
slope	5.3×10^{14}	-5.8×10^{13}	-6.2×10^{11}	4.3×10^{13}
offset				

A similar comparison as for the NO_2 vertical columns has also been performed for the AOD from the five algorithms (not shown here, see deliverable D10). The main features were that AOD values vary strongly, with MMF showing the largest spread and including very high values while BOREAS cuts off at AOD of 1.0 and MAPA is mostly but not always limited to small values. In Bremen, little correlation between the different retrievals can be seen by eye within the scatter, while in Athens, the overall temporal evolution is picked up by all retrievals in a similar way. In Ny-Alesund, MMF values appear to be much larger than those of the other retrievals.

5.5.4 Main outcomes of the FRM$_4$DOAS Phase I processing system validation

Overall, the FRM$_4$DOAs profiling algorithms worked well on both the CINDI-2 and the University of Bremen data sets. In particular for NO_2 columns, the agreement is very reasonable, both within the data set, with external validation data and with the independent BOREAS results. Surface concentrations of NO_2 and HCHO during CINDI-2 showed good agreement with independent data sets, as did the AOD retrievals. For the University of Bremen stations, AOD results are less consistent, probably because of the high probability of cloud contamination and the different treatment of these in the different algorithms.

While not in all cases the agreement between results of different retrievals is good, this is not a problem linked to the FRM$_4$DOAS retrievals alone, but rather reflects the current state of the art in profile retrievals on MAX-DOAS data. Because of the ill posed retrieval problem, different assumptions made in the algorithms are bound to lead to different results, and no clear decision can be taken on which approach is generally better unless a large and robust validation data set becomes available.

One of the most important aspects of MAX-DOAS profiling is flagging of valid data, and clearly the algorithms take very different approaches here, leading to large differences in the number of profiles flagged as valid and also in surprisingly small overlap of the valid time series for individual stations. In particular, the MAPA valid data seem to be of pretty good quality. However, when the measurement conditions are not ideal the results quickly become unreasonable and are flagged. In contrast, MMF seems much more robust under worse measurement conditions.

From this validation exercise, it is recommended that for the implementation of the FRM$_4$DOAS processing system, reasons for cases with large differences between results from the different algorithms should be investigated, and flagging should be revisited and if possible harmonised more in order to come to more consistent results. For instance, the possibility to select more location-specific a priori profiles for the MMF retrievals could be investigated.

B. FRM_4DOAS CCN02 and CCN03 main achievements

The main achievements/results of the FRM_4DOAS CCN02 (08/2019-05/2020) and CCN03 (10/2020-03/2021) are described 'bullet-wise' in the present section. For more details, the reader is invited to have a look at the corresponding deliverables which are on the FRM_4DOAS web site.

1. CCN02

The main achievements of CCN02 are the following (see CCN02 final presentation available at https://frm4doas.aeronomie.be/ProjectDir/frm4doas_ccn02_overview_hendrick_v3_website.pdf):

- Algorithmic optimization:
  - Improvement of the QC flagging consistency between MMF and MAPA through four different checks (VCD consistency, profile consistency, at least one code is not flagged as error by its own flagging system, extra-DSCD error smaller than 3 times the DOAS fit error).
  - Implementation of a scan-based flagging system for broken cloud situations
  - Implementation of a correction for the temperature dependence of the NO_2 cross sections in the stratospheric NO_2 profiling algorithm
  - Development or refinement of the flagging approaches implemented for the stratospheric products (total O_3 VCDs and stratospheric NO_2 vertical profiles)
- Central Processing System (CPS) operationalization on both technical and administrative aspects:
  - Improvement and optimization of all system programs (structure, content, input/output data, settings, configuration, and log files), resulting in a clean code with a high level of modularity for easier service upscaling
  - Creation of the framework and scripts for running the CPS on High Performance Computer (HPC)
  - Agreement on the final tropospheric data products to be delivered on NDACC and EVDC: MMF retrieval products quality-controlled with MAPA results
  - Establishment of a working data stream between the FRM_4DOAS database and the NDACC and EVDC DHFs: One single submission to NDACC and then mirroring on EVDC.
  - Creation of the new NDACC Appendix VII protocol about the NDACC MAX-DOAS instrument and data retrieval certification procedures (see http://www.ndaccdemo.org/data/protocols/appendix-vii-uv-vis-instruments and deliverable D18 'NDACC MAXDOAS Certification Procedures document')
  - Establishment of a DOI (Digital Object Identifier) and data policy for NDACC MAX-DOAS Service products. The granularity of the DOIs and corresponding landing pages is one per group. The data policy is based on the Creative Commons (CC) licensing system. More details on both aspects can be found in Deliverable D17 'NDACC MAXDOAS DOI and procedure document'

- Preparation of an updated version of the NDACC MAX-DOAS Service questionnaire for a new consultation of the community in the perspective of the Service upscaling that will be part of the future FRM_4DOAS operational activity.

- Central Processing System verification and validation (see deliverable D21 'Operational Processing System and Test Report document'):
  - Retrieval results verification of tropospheric products (HCHO, NO_2, aerosols) through MAPA versus MMF correlation (VCD/AOD and surface concentration/extinction), comparisons of MAPA and MMF data to ancillary observations (CINDI-II data sets) and to retrieval results from the IUP-Bremen BOREAS algorithm.
  - Retrieval results verification of stratospheric products (total O_3 VCDs and stratospheric NO_2 vertical profiles and VCDs) through comparisons to co-located satellite observations (GOME-2 WFDOAS and OMI TEMIS, respectively)
  - Based on the above verification exercises, the FRM_4DOAS tropospheric NO_2 and total O_3 product have been assessed as mature enough for submission on NDACC. The other products (tropospheric HCHO and aerosols and stratospheric NO_2 vertical profiles) need to be further consolidated.
  - Technical verifications: Successful Central processing system response to various level-1 spectra file contents; Successful verification of the GEOMS files content and their compliance with the GEOMS UV-vis DOAS templates.
  - CPS performance assessment in terms of computing time corresponding to the different processing steps and the size of the corresponding input/output files using example level-1 spectra files.

- Assessment of NDACC MAX-DOAS Service readiness for the mature FRM_4DOAS products at the selected partners' stations (see https://frm4doas.aeronomie.be/ProjectDir/frm4doas_ccn02_NDACC_MD_service_readiness.pdf)

2. CCN03

The main achievements of CCN03 are the following (see deliverable D26 'NDACC MAXDOAS Service performance assessment report'):

- Upgrade of the NRT branch of the Central Processing System with the last version of the code and run it for measurements from partners' stations.
- Since November 2020, set up of the automatic daily submission of tropospheric NO_2 and total O_3 GEOMS files to NDACC Rapid Delivery repository with mirroring to EVDC. The status of the FRM_4DOAS GEOMS HDF4 files submission to the NDACC and EVDC DHFs is presented in Table 20.

Table 20: Status of FRM_4DOAS GEOMS HDF data files submission to NDACC and EVDC at the end of FRM_4DOAS CCN03 (March 2021). 'w' stands for ~weekly submission of level-1 spectra files to the CPS, 'N/A' for algorithm not applicable, and 'MTS' for mountain-top station (only total O_3 files submitted to NDACC). Due to the low tropospheric NO_2 content, NO_2 retrievals at Ny-Alesund and Neumayer are still under testing (-> orange instead

of green crosses in the Table). The data stream for the last 4 stations (1670, 1675, 1684, 1688) are currently stopped.

Instr #	Station name	affiliation	Geometry	Level-1 daily sub. status	TOTAL O3	TROPO NO2	Level-2 sub. to NDACC RD (+ mirroring on EVDC)
1669	XIANGHE	BIRA.IASB	Maxdoas	X	X	X	Daily since 10/2020
1671	HARESTUA	BIRA.IASB	Zenith	X	X	N/A	Daily since 10/2020
1672	NY.ALESUND	IUP	Maxdoas	X	X	X	Daily since 02/2021
1673	BREMEN	IUP	Maxdoas	X	X	X	Daily since 12/2020
1674	ATHENS	IUP	Maxdoas	X	X	X	Daily since 12/2020
1676	MAINZ (x4)	MPIC	Maxdoas	X	N/A	X	Daily since 12/2020
1677	LAUDER	NIWA	Maxdoas	w	X	X	Bi-weekly since 01/2021
1678	NEUMAYER	UHEIDELBERG	Maxdoas	X	X	X	Daily since 01/2021
1679	HEIDELBERG	UHEIDELBERG	Maxdoas	X	X	X	Daily since 12/2020
1683	THESSALONIKI_AUTH	AUTH	Maxdoas	X	N/A	X	Daily since 12/2020
1684	DEBILT	KNMI	Maxdoas	X	N/A	X	Daily since 03/2020
1686	THESSALONIKI_AUTH	AUTH	Maxdoas	X	X	X	Daily since 12/2020
1698	IZANA	INTA	Maxdoas	w	X	MTS	Bi-weekly since 12/2020
1670	UCCLE	BIRA.IASB	Maxdoas	-	X	X	07/2018-02/2020
1675	DEBILT	KNMI	Maxdoas	-	X	X	07/2018-11/2019
1684	LA.REUNION.MAIDO	BIRA-IASB	Maxdoas	-	X	MTS	07/2018-12/2019
1688	DEBILT	KNMI	Maxdoas	-	N/A	X	12/2020-01/2021

As stated in the previous Section, only the MAX-DOAS tropospheric NO_2 and twilight zenith-sky total O_3 products have been assessed to be mature enough for their public release on NDACC and EVDC

- Establishment of data streams for level-2 output files between the FRM_4DOAS database and instrument PIs through a password-protected data download web page made available on the project website (see https://frm4doas.aeronomie.be/index.php/partners/registered-users-login).
- General maintenance of the NDACC MAX-DOAS Service including central processor, level-1 and -2 data streams, data storage, QA/QC monitoring, and support to instrument PIs
- Continuation of the NDACC affiliation procedure for partners' instruments/stations

C. Outreach

1. Oral and poster presentations at conferences, workshops

The FRM_4DOAS project results were presented at the following conferences/meetings:

- **3rd ACTRIS-2 General Meeting, Granada (Spain), 1-2 February 2017:**

  F. Hendrick et al., MAX-DOAS trace gas observations in NDACC: Towards harmonized and quality-assessed multi-year data products – Oral

- **European Geosciences Union General Assembly, Vienna (Austria), 23-28 April 2017:**

  Poster:

  K. Kreher et al., First results of the CINDI-2 semi-blind MAX-DOAS intercomparison

  U. Frieß et al., Comparison of MAX-DOAS profiling algorithms during CINDI-2 - Part 1: aerosols

  F. Hendrick et al., Comparison of MAX-DOAS profiling algorithms during CINDI-2 - Part 2: trace gases

  F. Hendrick et al., The ESA FRM_4DOAS project: Towards a quality-controlled MAXDOAS Centralized Processing System

- **CEOS Atmospheric Composition Virtual Constellation Meeting #13, Paris (France), 28-30 June 2017:**

  M. Van Roozendael et al., Status and future plans of the air-quality FRM projects - Oral

- **S5P-MAG #16, ESA/ESTEC, Noordwijk (The Netherlands), 29-30 August 2017:**

  M. Van Roozendael et al., Status of CINDI-2 and FRM_4DOAS activities - Oral

- **8th International DOAS Workshop, Yokohama (Japan), 4-6 September 2017:**

  Oral:

  U. Frieß et al., Comparison of algorithms for the retrieval of aerosol and trace gas vertical profiles using synthetic MAX-DOAS data

  K. Kreher et al., CINDI-2 semi-blind MAX-DOAS intercomparison – data analysis and results

  J.-L. Tirpitz et al., Comparison of MAX-DOAS and ancillary profiling results during CINDI-2

  Poster:

  S. Beirle et al., The MAinz Profile Algorithm (MAPA)

  M. Van Roozendael et al., Central reprocessing of CINDI-2 MAX-DOAS NO_2, O_4, HCHO and O_3 slant column data

  F. Hendrick et al., The ESA FRM_4DOAS project: Towards a quality-controlled MAXDOAS Centralized Processing System

- **8th GEMS Science Team Meeting, Seoul (Korea), 25-27 September, 2017:**

M. Van Roozendael et al., Intercalibration and harmonization of MAX-DOAS measurements as part of the CINDI-2 and FRM_4DOAS projects – Oral

- **Second Sentinel-5 Precursor (S5P) Validation Team Meeting and First Results Workshop, Noordwijk (The Netherlands), 5-6 February 2018:**

  Oral:

  F. Hendrick et al., The ESA FRM_4DOAS project: Towards a quality-controlled MAXDOAS Centralized Processing System

- **2nd CINDI-2 Workshop, Mutters (Austria), 14-15 March 2018:** See all oral and poster presentations at https://frm4doas.aeronomie.be/index.php/documents/2nd-cindi-2-workshop.

- **European Geosciences Union General Assembly, Vienna (Austria), 8-13 April 2018:**

  Oral:

  S. Beirle et al., Empirical O_4 scaling factors derived with the MAinz Profile Algorithm (MAPA)

  M. Van Roozendael et al., Central reprocessing of CINDI-2 MAX-DOAS slant column data sets

  Poster:

  U. Frieß et al., Comparison of algorithms for the retrieval of aerosol and trace gas vertical profiles using synthetic MAX-DOAS data

  J.-L. Tirpitz et al., Consistency of MAX-DOAS aerosol and trace gas profiling results during the CINDI-2 intercomparison campaign

  S. Beirle et al., The MAinz Profile Algorithm (MAPA)

- **ATMOS-2018, Salzburg (Austria), 26-29 November 2018:**

  Oral:

  F. Hendrick et al., ESA FRM_4DOAS: Towards a Quality-Controlled MAXDOAS Centralized Processing System in Support of Air Quality Satellite Sensors Validation

- **AGU Fall Meeting 2018, Washington (USA), 10-14 December 2018:**

  Oral:

  F. Hendrick et al., ESA FRM_4DOAS: Towards a Quality-Controlled MAXDOAS Centralized Processing System in Support of Air Quality Satellite Sensors Validation

- **3rd CINDI-2 Workshop, ESA/ESRIN, Frascati (Italy), 26-27 February 2019:** See all oral and poster presentations at https://frm4doas.aeronomie.be/index.php/documents/3rd-cindi-2-workshop.

2. Peer-reviewed papers

The following papers related to the FRM_4DOAS activities have been published during the timeframe of the project:

Beirle, S., et al., The Mainz profile algorithm (MAPA), Atmos. Meas. Tech., 12, 1785–1806, 2019, https://doi.org/10.5194/amt-12-1785-2019, 2019.

Donner, S., et al., Evaluating different methods for elevation calibration of MAX-DOAS instruments during the CINDI-2 campaign, Atmos. Meas. Tech., 13, 685–712, https://doi.org/10.5194/amt-13-685-2020, 2020.

Frieß, U., et al., Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies using synthetic data, Atmos. Meas. Tech., 12, 2155–2181, 2019, https://doi.org/10.5194/amt-12-2155-2019, 2019.

Kreher, K., et al., Intercomparison of NO_2, O_4, O_3 and HCHO slant column measurements by MAX-DOAS and zenith-sky UV-Visible spectrometers during the CINDI-2 campaign, Atmos. Meas. Tech., 13, 2169–2208, https://doi.org/10.5194/amt-13-2169-2020, 2020.

Peters, E., Full-azimuthal imaging-DOAS observations of NO_2 and O_4 during CINDI-2, Atmos. Meas. Tech., 12, 4171–4190, https://doi.org/10.5194/amt-12-4171-2019, 2019.

Tirpitz, J.-L., et al., Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies on field data from the CINDI-2 campaign, Atmos. Meas. Tech., 14, 1–35, https://doi.org/10.5194/amt-14-1-2021, 2021.

Wang, Y., et al., Inter-comparison of MAX-DOAS measurements of tropospheric HONO slant column densities and vertical profiles during the CINDI-2 campaign, Atmos. Meas. Tech., 13, 5087–5116, https://doi.org/10.5194/amt-13-5087-2020, 2020.

D. Overall conclusions and perspectives

The NDACC MAX-DOAS Service, developed in the framework of the FRM_4DOAS project and its successive CCNs, has been launched in November 2020. This service includes the first central NRT (24h latency) processing system for MAX-DOAS instruments, which includes state-of-the-art retrieval algorithms for tropospheric and stratospheric NO_2 vertical profiles, tropospheric HCHO vertical profiles, and total O_3 columns. In addition to the CPS development, significant effort was also put in the creation of the administrative framework for future service upscaling, like the assignment of DOIs to the different data sets, the set-up of a user data policy based on the Creative Commons licensing approach, and the drafting of a new NDACC affiliation and certification procedure protocol for MAX-DOAS instruments.

So far, only GEOMS HDF files of tropospheric NO_2 and total O_3 products for the selected 14 partners' stations are daily submitted to the NDACC rapid delivery database, with mirroring on EVDC. The other FRM_4DOAS products (tropospheric HCHO and stratospheric NO_2 vertical profiles) have been assessed as not mature enough for public release and need further consolidation. Several months of operation of the service have allowed to learn some important lessons in the perspective of the service upscaling (see deliverable D26 'NDACC MAX-DOAS_Service performance assessment report'), like the need for (1) continuous monitoring of the DOAS fit RMS and errors to detect possible instrumental degradation, (2) further refinements of the QA/QC flagging approach for the tropospheric NO_2 product, and (3) a thorough characterization of the measurement site (like e.g. the presence of potential obstacle in any of the viewing directions) by the PIs before submitting first spectra files to the Central Processing System.

The NDACC MAX-DOAS Service will be further consolidated and upgraded (more products and stations) as part of the ESA FRM_4DOAS-2.0 R&D and Copernicus operational follow-up projects.

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