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

ESA Contract No. 4000135355/21/I-DT-lr

Deliverable D3.4

MAX-DOAS Aerosol Algorithm revised ATBD

Date: June 4, 2025
Version: 1.0

Authors

Martina M. Friedrich[1]
Udo Frieß[2]
Steffen Beirle[3]
Thomas Wagner[3]
Alexis Merlaud[1]
Caroline Fayt[1]
Gaia Pinardi[1]
Michel Van Roozendael[1]
Steffen Ziegler[3]
Ankie Piters[4]
Alkis Bais[5]
Dimitris Karagkiozidis[5]
Dimitris Nikolis[5]
Andreas Richter[6]
Kai Krause[6]
Cristina Prados Roman[7]
Laura Gomez Martin[7]
Margarita Yela[7]
Monica Navarro[7]
Olga Puentedura Rodriguez[7]

[1] Belgian Institute for Space Aeronomy (BIRA-IASB)
[2] Institut für Umweltphysik, Universität Heidelberg (IUPHD)
[3] Max Planck Institute for Chemistry (MPIC)
[4] Koninklijk Nederlands Meteorologisch Instituut (KNMI)
[5] Aristotle University of Thessaloniki (AUTH)
[6] Institut für Umweltphysik, Universität Bremen (IUPB)
[7] Instituto Nacional de Técnica Aeroespacial (INTA)

Document change record

issue	date	item	comments
0.1	June 4, 2025	all	Initial version

Contents

1 Introduction

1.1 Purpose of the ATBD

This Algorithm Theoretical Basis Document (ATBD) describes the changes proposed to the algorithms that are implemented in the Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observations (FRM4DOAS) centralised processing system for the retrieval of tropospheric vertical aerosol extinction profiles and Aerosol Optical Depth (AOD). It is part of Work package (WP)-3 of FRM4DOAS-2.0. As an extension to FRM4DOAS, this ATBD is an amendment to the tropospheric retrieval description part of ATBD **[RD-1]** of FRM4DOAS.

The aerosol extinction profile retrieval is not a stand-alone-product in FRM4DOAS, it only constitutes the first step for the tropospheric trace gas profile retrieval in each of the two retrieval algorithms.

The goal in this WP of the follow-up project FRM4DOAS-2.0 was to include aerosol tropospheric AOD together with the vertical aerosol extinction profile as a stand-alone product, as described in **[AD-1]**. The performance of the algorithms, MAinz Profile Algorithm (MAPA) and Mexican MAX-DOAS Fit (MMF) that were chosen during the initial FRM4DOAS project based on a Round-Robin exercise, see **[RD-2]** were further investigated and optimized.

The initial description of the algorithms implemented in FRM4DOAS, MAPA and MMF, is given in **[RD-1]**, and Beirle et al. (2019) and Friedrich et al. (2019), respectively.

1.2 Document overview

Following the sections on applicable and reference documentation (Sect. 2) and acronyms and abbreviations (Sect. 3), a short reminder of the two profile retrieval algorithms implemented in FRM4DOAS is given in Sect.4.

Sect.5 describes the changes to MAPA and MMF that were implemented to mitigate these shortcomings. Sect.6 outlines the future plans and sets this ATBD in context with the associated deliverables of WP-3

2 Applicable and reference documents

2.1 Applicable documents

[AD-1] ESA/ESRIN. *Statement of Work*. ESA Express Procurement - [EXPRO] ESA-EOPG-EOPGMQ-SOW-23. FRM4DOAS-2.0, Mar. 2021.

2.2 Refrence documents

[RD-1] FRM4DOAS. *MAXDOAS Algorithm ATBD*. Deliverable D6. Deliverable D6. FRM4DOAS, Jan. 2018.

[RD-2] FRM4DOAS. *MAXDOAS Algorithm Round-Robin Definition and Results Document*. Deliverable D5. Deliverable D5. FRM4DOAS, Jan. 2018.

[RD-3] FRM4DOAS-2.0. *Technical Note*. Deliverable D3.2. Deliverable D3.2. FRM4DOAS-2.0.

[RD-4] FRM4DOAS-2.0. *Validation Report*. Deliverable D3.3. FRM4DOAS-2.0.

[RD-5] FRM4DOAS-2.0. *Technical Implementation Plan*. Deliverable D3.5. FRM4DOAS-2.0.

3 Acronyms and abbreviations

AOD Aerosol Optical Depth.

ATBD Algorithm Theoretical Basis Document.

AUTH Aristotle University of Thessaloniki.

BIRA-IASB Belgian Institute for Space Aeronomy.

DOF Degrees of Freedom.

DSCD Differential Slant Column Density.

DSP Demonstration software package.

FRM4DOAS Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observations.

INTA Instituto Nacional de Técnica Aeroespacial.

IUPB Institut für Umweltphysik, Universität Bremen.

IUPHD Institut für Umweltphysik, Universität Heidelberg.

KNMI Koninklijk Nederlands Meteorologisch Instituut.

LUT Look-up table.

MAPA MAinz Profile Algorithm.

MAX-DOAS Multi-Axis DOAS.

MMF Mexican MAX-DOAS Fit.

MPIC Max Planck Institute for Chemistry.

RAA relative azimuth angles.

RMS Root mean square.

SF O_4 scaling factor.

TIP Technical implementation plan.

TN Technical note.

UV ultraviolet.

VLIDORT Vector LInearized Discrete Ordinate Radiative Transfer.

VR Validation report.

WF weighting function.

WP Work package.

4 Aerosol retrieval algorithms in FRM4DOAS

Aerosol extinction vertical profiles are derived for each Multi-Axis DOAS (MAX-DOAS) elevation scan based on the corresponding measured oxygen dimer (O_4) Differential Slant Column Densities (DSCDs).

The retrieval relies on the well-known and nearly constant vertical distribution of O_4, which is proportional to the square of the O_2 concentration. Therefore, O_4 DSCDs provide valuable insights into the vertical distribution of aerosols (Wagner et al. (2004), Friess et al. (2006)).

4.1 MAPA

MAPA (Beirle et al. (2019)) makes use of an aerosol extinction profile shape parametrization with just a few free parameters:

- layer height h
- total AOD
- shape parameter s.

Additionally the measured O_4 DSCDs can be scaled by

- a O_4 scaling factor (SF) applied on measured O_4 DSCDs,

as found necessary by some studies (c.f. Wagner et al. (2019) and references therein). This parameter can be also treated as a fitting parameter in MAPA. Technically, it is however implemented as the inverse and applied on the simulated DSCD instead. In FRM4DOAS, three versions are implemented, one where this parameter is treated as a fitting parameter (SF VAR), one where it is set to 1 (SF 1) and one where it is set to 0.8 (SF 0.8).

Look-up tables (LUTs) are created starting with sets of profiles varying those parameters using off-line forward simulations using McArtim (Deutschmann et al. (2011)), as described in Beirle et al. (2019).

The main reason to use a low number of free parameters is that the information content of MAX-DOAS observations with respect to the vertical distribution of aerosols (and trace gases) is limited, and therefore, a sufficiently wide range of possible profile shapes can be retrieved with a limited but appropriate choice of free parameters, see Vlemmix et al. (2015).

Regarding the minimization strategy for MAPA, DSCDs from LUTs based on forward model simulations are compared to the measured DSCDs by use of a Monte-Carlo scheme (testing approximately 10000 different random combinations of the three parameters) where vertical aerosol profiles are derived from the best matching (smallest Root mean square (RMS) of the difference between simulated and measured DSCD) sets of parameters. In FRM4DOAS, the profile weighted mean is returned for the aerosol extinction profile, while the best match for the AOD parameter is returned as AOD.

The following set of flags is implemented in MAPA with the following limits configured:

- RMS: 1 for warning 3 for error (c.f. eq.3 in Beirle et al. (2019))
- normalized RMS: 0.05 for warning and 0.3 for error (cf. eq. 12 in Beirle et al. (2019))

- consistence in AOD (cf. eq. 13 in Beirle et al. (2019))
  - relative AOD tolerance: 0.2 for warning, 0.5 for error
  - absolute AOD tolerance: 0.05 for warning, 0.2 for error with $\epsilon = 0.05$ as the default for aerosols already incorporated
- upper threshold for h: 3 km for warning, 4.5 km for error
- lower threshold for percentage of total AOD that is located in the lowest 4km (only if total AOD is larger than 0.05 for warning or 0.2 for error): 80% for warning, 50% for error.
- upper limit for aod: 2 for warning, 3 for error
- upper limit of scale factor: 1.2 for warning, 1.4 for error
- lower limit of scale factor: 0.6 for warning, 0.4 for error
- special AOD limit for low relative azimuth angles (RAA) (below 15 °): 0.5 for warning, 3 for error.

In FRM4DOAS, warnings are considered invalid. While all three SF versions (1, 08 and VAR) are performed in FRM4DOAS, SF VAR is considered the default and the results with those settings are used further.

4.2 MMF

MMF (Friedrich et al. (2019)) uses constrained least-squares fitting and optimal estimation as regularization. For the forward model, it uses Vector LInearized Discrete Ordinate Radiative Transfer (VLIDORT)[1] (Spurr (2013)). It works in linear observation space and in logarithmic state space. As iteration scheme, it uses Levenberg-Marquardt minimisation scheme, see Rodgers (2000), implemented in MMF is given by Eq.1

$$\mathbf{x}_{i+1} = \mathbf{x}_i + \left((1+\gamma)\mathbf{S}_a^{-1} + \mathbf{K}_i^{\mathrm{T}}\mathbf{S}_\epsilon^{-1}\mathbf{K}_i\right)^{-1}\left[\mathbf{K}_i^{\mathrm{T}}\mathbf{S}_\epsilon^{-1}(\mathbf{y} - \mathbf{F}(\mathbf{x}_i)) - \mathbf{S}_a^{-1}(\mathbf{x}_i - \mathbf{x}_a)\right], \tag{1}$$

see eq. A1 in Friedrich et al. (2019), and K as in Eq.2

$$\mathbf{K} = \frac{\partial \mathbf{y}}{\partial \mathbf{x}} \tag{2}$$

where F is the forward model operator, K_i is the sequentially updated weighting function (WF) matrix at step i, and the superscripts T and -1 denote the transposed and inversed matrix, respectively. γ is a factor that is decreased if the cost function diminished in an iteration step and it increased (and the newly calculated x_i is disregarded) if the cost function increased. Since MMF uses VLIDORT as the forward model, no consecutive perturbations of each layer of the *a priori* profile and recalculation of the set of measurements is necessary, since the weighting functions are a direct output of VLIDORT, if configured accordingly, see Spurr (2013).

[1] however, only the intensity part of it, so effectively LIDORT

The *a priori* aerosol extinction profile x_a and covariance matrix S_a where constructed as follows as in Eq.3 and Eq.4, respectively.

$$x_i = \frac{\text{AOD}_a}{h_s} e^{(-h_i/h_s)} \tag{3}$$

where h_s is the scale height and was taken to be 1000 m, and AOD_a is the desired *a priori* AOD value, taken as 0.18 and h_i the middle layer height.

$$S_{a_{ij}} = \alpha\, x_{a_i}\, \alpha\, x_{a_j}\; e^{-\ln(2)\left(\frac{z_i - z_j}{\gamma}\right)^2}, \tag{4}$$

where γ is the correlation length and was taken as 200 m and α, representing the relative variance w.r.t the *a priori* profile, in FRM4DOAS is 0.5.

MMF implements the following flags:

- upper limit on AOD: no limit for warning, 8 for error
- lower limit on Degrees of Freedom (DOF): 1.6 for warning, 1.3 for error
- limit on RMS between measured and simulated DSCDs in units of measured error: 2 for warning, 4 for error.
- limit on stability (each retrieval is run twice): 1% difference is warning, 20% difference is error.

In FRM4DOAS warnings are considered as valid. No scaling factor on the measured or simulated O_4 DSCDs was applied, which corresponds to MAPA's SF 1 case.

5 Changes to the Aerosol retrieval algorithms in FRM4DOAS

As detailed in **[RD-3]**, the implementation of the aerosol retrieval algorithms in FRM4DOAS, especially that of MMF show a sub-optimal performance. The main shortcomings that were identified for the two codes are:

- MAPA
  - sometimes the integrated extinction up to 4 km exceeds the reported AOD.
- MMF
  - MMF systematically underestimates the AOD, especially in the UV.
  - The MMF flagging scheme is too strict, only a very small amount of MMF profiles is flagged as no warning.

Hence, work was carried out to optimize retrieval settings for the algorithms and to develop, similar to the tropospheric NO_2 product, a unified tropospheric aerosol product. These tests, together with a unified validation of the final proposed settings were presented in **[RD-4]**.

5.1 MAPA

The reason that occasionally the reported AOD is larger than the integrated extinction up to 4 km is that, as outlined in Sect.4, the AOD is reported as the best matching parameter, while the profile is reported as a weighted mean. To have more consistent values in the FRM4DOAS level 2 output, it is proposed to report two sets of results that match each other: one reporting the best match (both for the AOD as well as for the profile resulting from the best matching parameters) and a weighted average. For comparison with MMF, the integrated extinction profile up to 4 km from the weighted average profile is taken.

5.2 MMF

As detailed in **[RD-4]**, changes in the *a priori* settings (both for the profile as well as in the covariance matrix) are not sufficient for closing the gap between the AOD measured by sun-photometers and MMF retrieved AOD. Accounting for a higher variability in layers above 1 km, by means of a higher a-priori and hence a larger covariance matrix at those altitudes, can only account for roughly 10% of the gap.

Furthermore, **[RD-4]** showed that it is the measurements themselves that drive the retrieval to values much below the *a priori* profile in the lower 2 km. Although the reason for the need of the aforementioned SF applied to the measured O_4 DSCDs is yet unknown, MAPA results, c.f. **[RD-3]**, show that applying such a SF results in retrieved AODs that are in better agreement with sun-photometer measured AODs. Hence, tests for MMF applying different SFs were performed. It was shown that in the ultraviolet (UV)-wavelength range, a fixed scaling factor of the order 0.85 - 0.95 is sufficient to close the gap. It was shown that with a choice of a SF=0.85, the profile in the lowermost 1.5 km is independent of the choice of *a priori* settings and the gap in AOD is closed.

However, it was also demonstrated that a fixed SF does not perform well in the visible wavelength region. A comparison of MAPA-fitted SF values in Uccle between the visible and UV ranges, revealed a significant different SF distribution in the visble compared to the UV. While the nature of the need for the SF is unknown, Beirle et al. (2019) and also **[RD-3]** and **[RD-4]** showed that MAPA SF VAR always outperforms MAPA SF 1 and often outperforms MAPA SF 0.8.

Hence, tests, both in the UV and visible were carried out at different stations pre-applying the MAPA fitted SF to the measured DSCDs of O_4. These tests showed that MMF with MAPA's fitted variable SF performs equally good in terms of AOD regression slope and often slightly better in terms of AOD correlation, compared to sun-photometer AOD. Hence, it was decided to choose this as the MMF implementation for aerosol extinction profile and AOD retrieval.

Additionally it was found that changing the *a priori* settings from the default ones as presented in Sect.4 to ones that give more freedom in the upper layers, but at the same time counteract oscillations by enforcing a larger correlation length, slightly improve the correlation at altitudes above 1.5 km. The new set of parameters is as follows:

- $h_s = 2000$m
- $\alpha = 0.6$
- $\gamma = 600$m

with no changes to AOD_a, which stays at 0.18. The result is a different distribution of the DOF over altitude, giving more freedom to higher altitudes by reducing the penality in the cost function for deviations from the *a priori* profile at higher altitudes.

To address the second point, refinements and additions to the flagging scheme were introduced for MMF within FRM4DOAS, as detailed below.

- upper limit on AOD: 2 for warning, 5 for error
- lower limit on DOF: 1.6 for warning, 1.3 for error
- limit on RMS between measured and simulated DSCDs in units of measured error: 2 for warning, 3 for error.
- limit on stability[2]: 10% difference is warning, 30% difference is error (unless the AOD is below 0.03, then this flag is inactive).
- new component: filter out heavy oscillations by putting a limit on the third derivative: no limit for warning, 0.5 for error.
- new component: filter out top-heavy profiles: if more than 75% of the AOD is located above 3 km, this results in an error.
- flag based on MAPA fitted SF that is pre-applied on measured DSCDs before MMF retrieval: a SF outside of [0.7, 1.2] is considered an error.

6 Future plans

The changes outlined in Sect.5 have been applied in a post processing in an off-line ad-hoc version on existing data. That sofware package will be delivered as Demonstration software package (DSP) deliverable D3.6.

The Technical note (TN) **[RD-3]** listed as a lesser important change to be considered in a new implementation the change of the retrieval grid to include a finer grid close to the surface. The Validation report (VR) **[RD-4]** did not work on a finer grid. However, the Technical implementation plan (TIP) **[RD-5]** includes the implementation of such a finer grid as an optional component.

7 Refrences

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