IDEAS-QA4EO
WPs 2250-2251: “DOAS-BO: Towards a new FRM4DOAS-compliant site”

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WPs 2250-2251: DOAS-BO- Phase I

A step backward …. in the previous contract

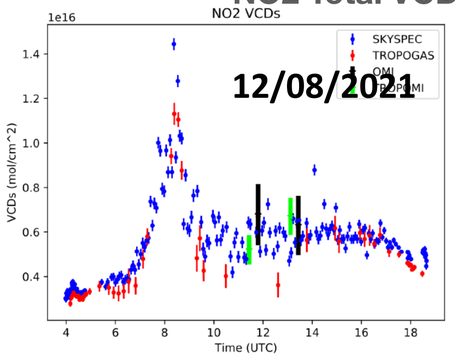
- The objectives were: 1) take a step to close the lack of DOAS measurements within the Po valley and demonstrate the value and importance of ground based DOAS measurements in this particular region. 2) Re-enforce the Italian know-how on DOAS technique

- We assess the performances of a custom-built research-grade MAX-DOAS instrument developed at CNR-ISAC (named TROPOGAS) with respect of FRM4DOAS requirements and update its measurement set-up.

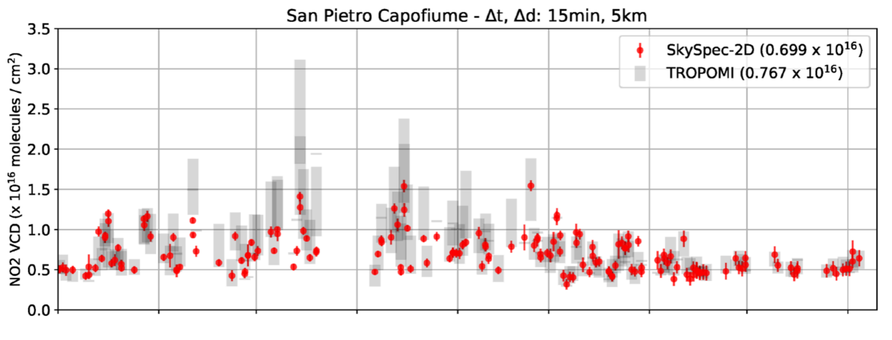
- In the meanwhile, CNR-ISAC acquired (in the frame of a national funded project) two MAX-DOAS systems (SkySpec-2D by Airyx) fully compliant with FRM4DOAS requirements. We performed two inter-comparison campaigns: one in Bologna with TROPOGAS one in BAQUNIN with Pandora.

- Then the SkySpec-2D was moved to the “Giorgio Fea” observatory at San Pietro Capofiume. Its spectra have been selected by FRM4DOAS community for the inclusion in their centralized processing. **We are still in a testing phase, waiting for evolutions.**

TROPOGAS vs SkySpec-2D vs TROPOMI and OMI
NO2 Total VCDs



Skyspec-2D system
at San Pietro Capofiume



WPs 2250-2251: DOAS-BO- Phase II

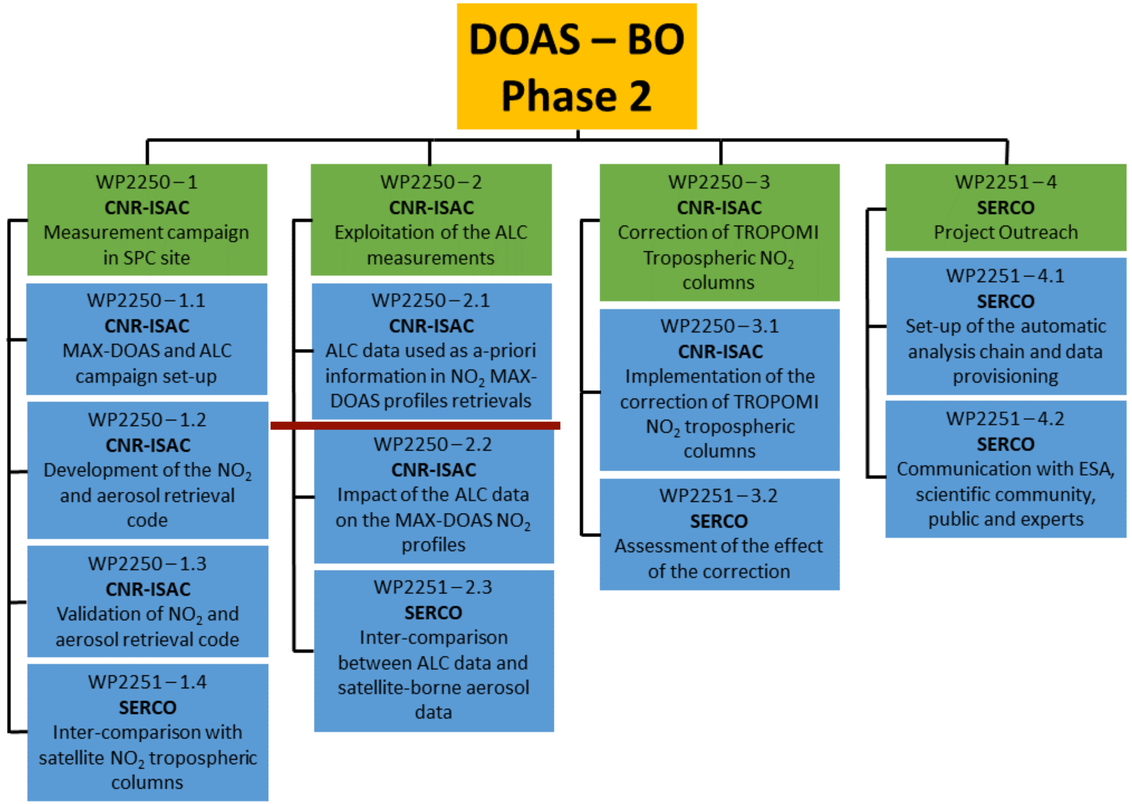
- Exploit the synergies between MAX-DOAS and aerosol remote sensing data in the Po Valley for satellite validation.
- Development of a retrieval code for profiles retrievals from MAX-DOAS measurements (starting from the raw one developed in Phase I), validation with FRM4DOAS specifications
- Comparison of retrieved NO_2 tropospheric VCDs with TROPOMI.
- Use of collocated aerosol profiles from ground-based instrumentation :

1) as initial guess for MAX-DOAS retrieval - new comparison with TROPOMI

2) for comparison with aerosol satellites products

- Correction of TROPOMI NO_2 Tropospheric VCD to account for NO2 profile - comparison with ground based MAX-DOAS profiles.
- 18 months - Serco and CNR-ISAC

WPs 2250-2251: DOAS-BO- Phase II



We are here

Month	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Delivery				D-1v1			D-2					D-3				D-4		D-1v2
WP 2250-1																		
WP 2250-1.1																		
WP 2250-1.2																		
WP 2250-1.3																		
WP 2251-1.4																		
WP 2250-2																		
WP 2250-2.1																		
WP 2250-2.2																		
WP 2251-2.3																		
WP 2250-3																		
WP 2250-3.1																		
WP 2251-3.2																		
WP 2251-4																		
WP 2251-4.1																		
WP 2251-4.2																		

- We set up the MAX-DOAS and the ALC systems
- Developed and validated the NO_2 and aerosol profile retrieval code (DEAP)
- Started processing SPC MAX-DOAS data and comparing tropospheric NO_2 VCDs with TROPOMI
- Start ingesting ALC data as initial guess for DEAP
- Deliverables : D-1v1 and D-2 delivered

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.1 MAX-DOAS and ALC Campaign set-up

Exploitation of synergy of remote sensing instruments at SPC

"Giorgio Fea" observatory at San Pietro Capofiume

(https://www.isac.cnr.it/it/node/7803, Latitude: 44.65° N, Longitude: 11.62° E, Altitude: 11 m a.s.l.)

Ceilometer VAISALA LD40 (ALC)

MAX-DOAS SkySpec 2D

Raymetrics LIDAR

The site, founded in the early 1980s, is managed by the Agenzia Regionale per la Prevenzione, l'Ambiente e l'Energia (Arpae, https://www.arpae.it/it/arpae/arpae) of Emilia Romagna, while CNR-ISAC operates in the field under the umbrella of a long-term agreement with ARPAE.

The station is equipped for in-situ monitoring of trace gases and particulate matter sampling for atmospheric chemical speciation.

ARPAE also runs radar measurements, radio soundings and operates a phenological station.

The station is part of the European Research Infrastructure ACTRIS, as Mt. Cimone - Po Valley facility (CMN-PV, https://atmo-access.isac.cnr.it).

Currently, a MAX-DOAS instrument and an Automatic LIDAR/Ceilometer are operating on the site, a Raymetrics LIDAR is in the setting up phase.

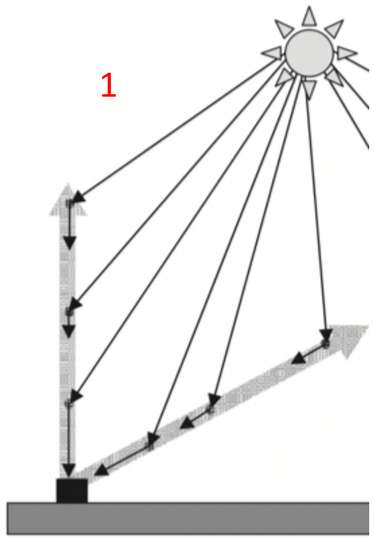
Trans-National Access at CMN-PV opportunities are provided by H2020 ATMO-ACCESS.

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.2 Development of the NO_2 and aerosol retrieval code

DOAS technique: Slant Column Densities (SCDs) retrieval from diffuse solar spectra

1



The light path relative to a spectrum is not well-defined!

Measured spectrum **SCD of jth gas** **Absorption cross section** **Instrumental function**

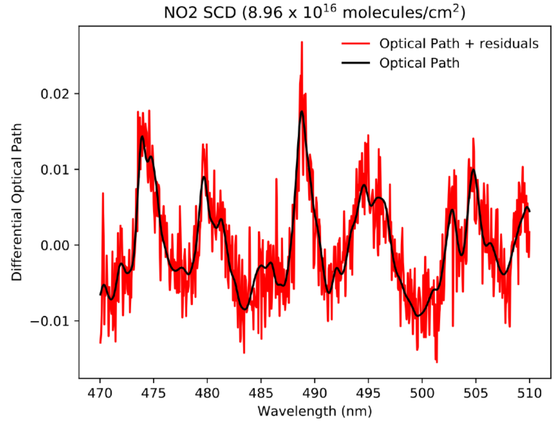
$$ln\left(\frac{I^*(\lambda,L)}{I_0'^*(\lambda,L)}\right) \approx \sum_j \bar{c}_j L ln\left[\int_{-\Delta\lambda}^{\Delta\lambda} e^{-\sigma'(\lambda-\lambda')} H(\lambda')d\lambda'\right]$$

Reference spectrum

2

3

SCDs fitted with QDOAS (developed at BIRA, Bruxelles)
SCDs intensity-weighted averaged over all the possible light paths.

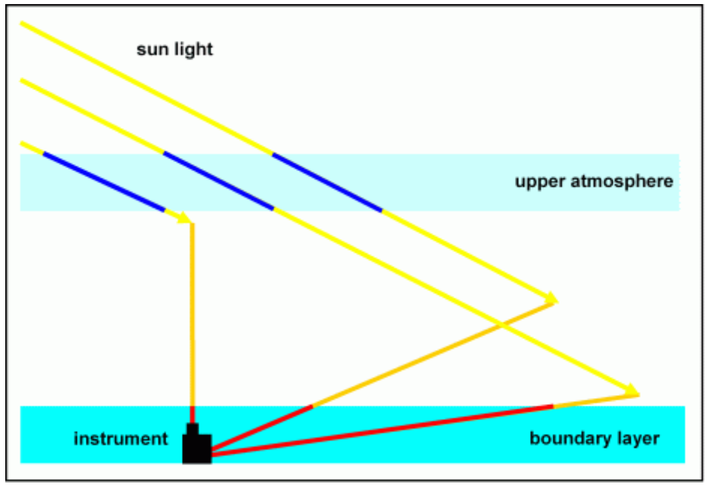


ASSUMPTIONS:

- Single scattering.
- Low absorption approximation.

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.2 Development of the NO_2 and aerosol retrieval code

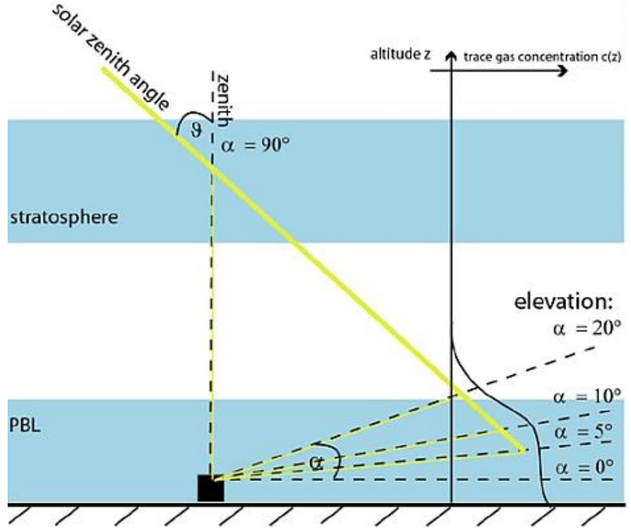


from "Introduction to Measurement Techniques in Environmental Physics-":Summer term 2009
Differential Optical Absorption Spectroscopy (DOAS)
Andreas Richter

- long optical path in troposphere
- path lengths in troposphere depends on elevation angle
- almost constant path in the stratosphere
- tropospheric measurements affected by clouds

- Zenith measurement of each scan used as reference (avoiding problems of residual contribution in reference spectra)

Need of retrieval codes for profile determinations



SPC Skyspec	ZENITH ACQUISITION	MAX-DOAS ACQUISITION
SZA range (°)	94-85	<85
Azimuth directions (°)	/	135, 250, 315
Elevation angles (°)	90	1, 2, 3, 5, 10, 30, 90
Channels	UV/VIS	UV/VIS

- Automatic chain from spectra acquisition to NO_2 VCDs.

- Spectra provided to FRM4DOAS network.

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.2 Development of the NO_2 and aerosol retrieval code

DEAP (DOAS optimal Estimation Atmospheric Profile retrieval) algorithm.

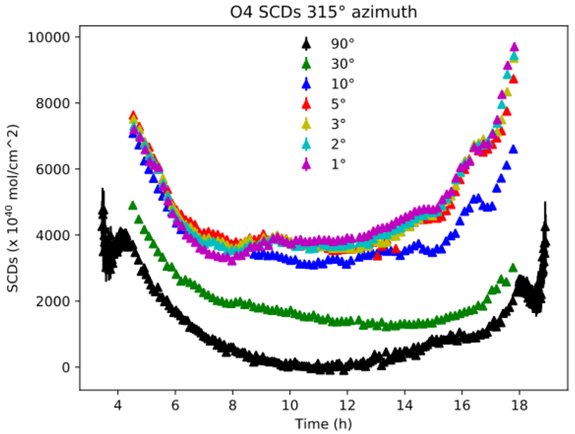
The DEAP code is an OE algorithm that exploits the SCIATRAN code, as FM and a two-step approach to retrieve tropospheric profiles from SCDs.

Why O_4 SCDs for aerosol retrievals?

1st step: aerosol extinction profile retrieval from O4 SCDs

$$x_{i+1} = x_i + (K^T S_y^{-1} K + S_0^{-1} + g K^T S_y^{-1} K)^{-1} (K^T S_y^{-1}(y - y_i) - S_0^{-1}(x_i - x_0))$$

y=O_4 SCD
K=d O_4 SCD/ d aer_ext
x =aer_ext



WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.2 Development of the NO_2 and aerosol retrieval code

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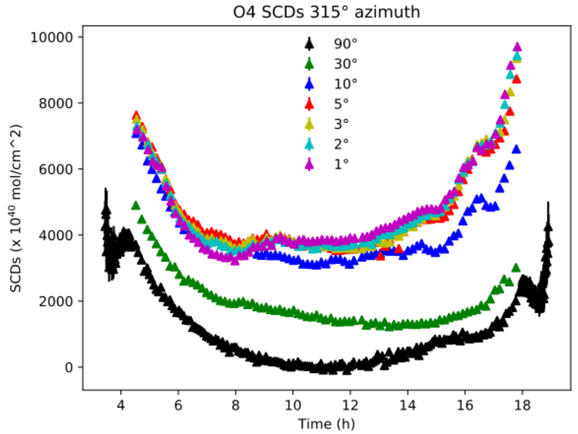
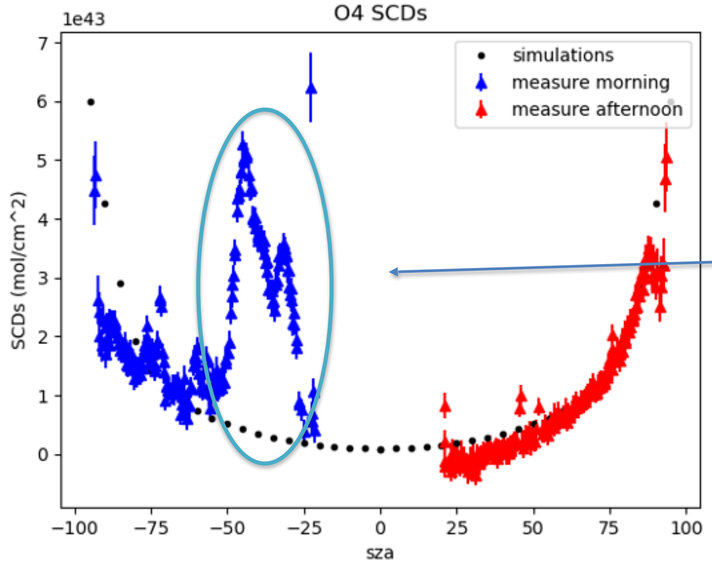


Why O_4 SCDs for aerosol retrievals?

Clouds and aerosol impact on O4 SCDs

O_4 SCD $\propto$ Atmospheric path

~~Atmospheric concentration~~



Assumption: constant O_4 distribution in time and space

Effects of clouds

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.2 Development of the NO_2 and aerosol retrieval code

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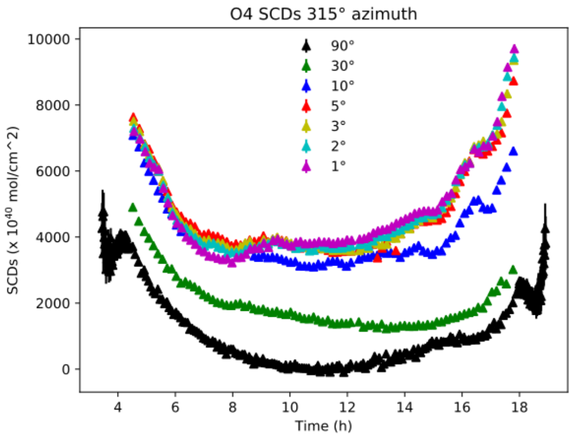
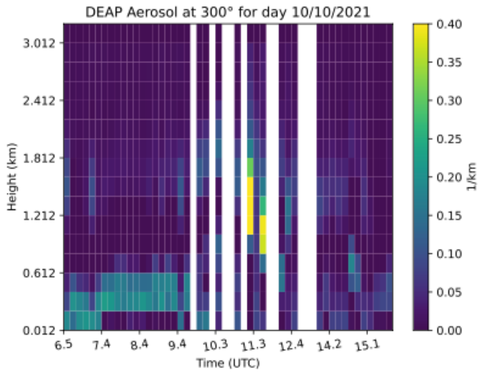
y=O_4 SCD
K=d O_4 SCD/ d aer_ext
x =aer_ext



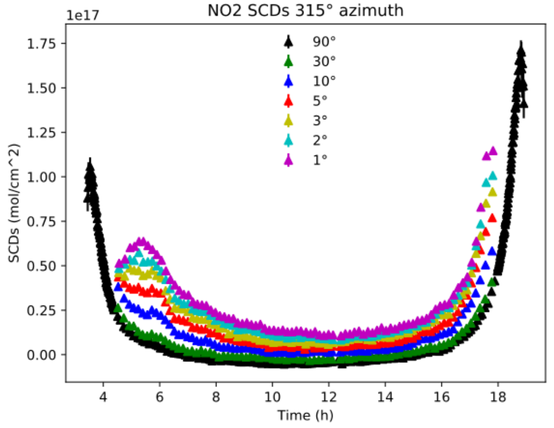


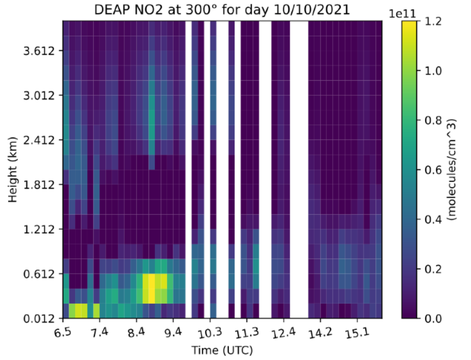
Box-AMF (K)

2nd step: gaseous profile retrieval

$$x_i = x_0 + S_0 K^T (K S_0 + S_0 K^T + g S_y)^{-1} (y - K x_0)$$

y=NO_2 SCD
K=NO_2 box-AMF
x =NO_2_conc





WPs 2250-2251: DOAS-BO- Phase II

- WP2250-1.3 Validation of NO_2 and aerosol retrieval code

DEAP has been validated using the synthetic SCDs provided by FRM4DOAS community

Those SCDs have been used in a round-robin exercise among different retrieval codes in the frame of FRM4DOAS activities

Atmos. Meas. Tech., 12, 2155–2181, 2019
https://doi.org/10.5194/amt-12-2155-2019
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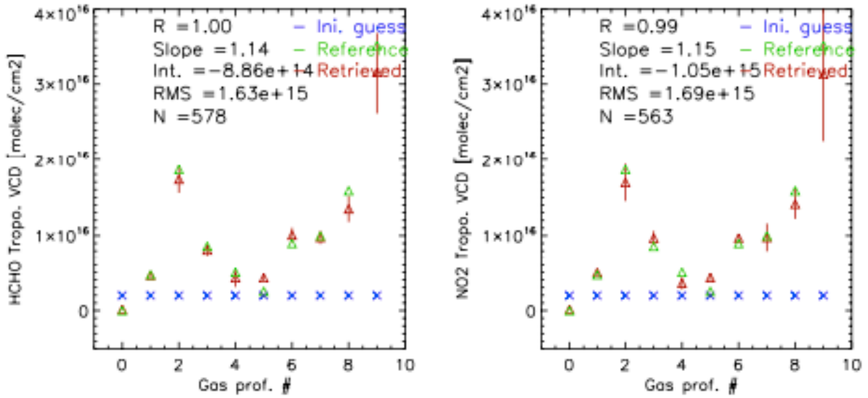
Atmospheric Measurement Techniques
Open Access
EGU

FRM4DOAS

Intercomparison of MAX-DOAS vertical profile retrieval algorithms: studies using synthetic data

Udo Frieß[1], Steffen Beirle[2], Leonardo Alvarado Bonilla[3], Tim Bösch[3], Martina M. Friedrich[4], François Hendrick[4], Ankie Piters[5], Andreas Richter[3], Michel van Roozendael[4], Vladimir V. Rozanov[3], Elena Spinei[6,a], Jan-Lukas Tirpitz[1], Tim Vlemmix[5], Thomas Wagner[2], and Yang Wang[2]

DEAP performances are comparable to MMF and MAPA reference algorithms at least on synthetic data



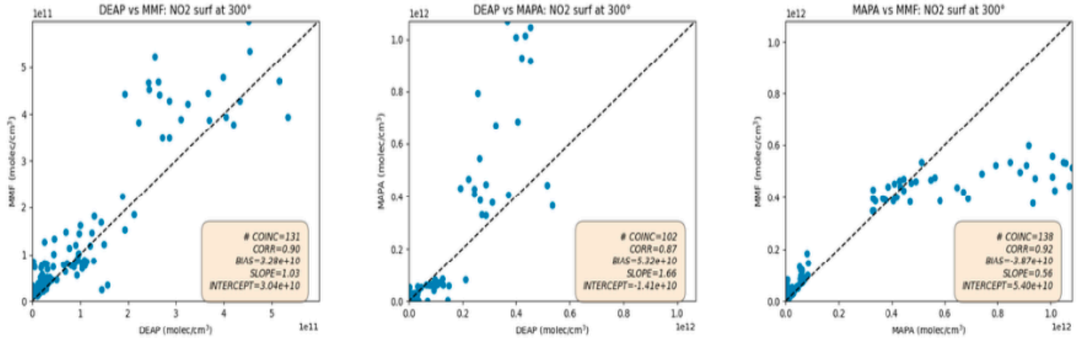
WPs 2250-2251: DOAS-BO- Phase II

- WP2250-1.3 Validation of NO_2 and aerosol retrieval code

The SkySpec-2D SPC spectra have been provided to the FRM4DOAS community for centralized processing. Although we are still in the testing phase, some profiles retrievals performed with MAPA and MMF codes, are already available. The FRM4DOAS team kindly provided us the NO_2 and aerosol extinction (preliminary product) profiles and columns retrieved from SkySpec-2D SPC spectra in the VIS range using the two official retrieval codes*.

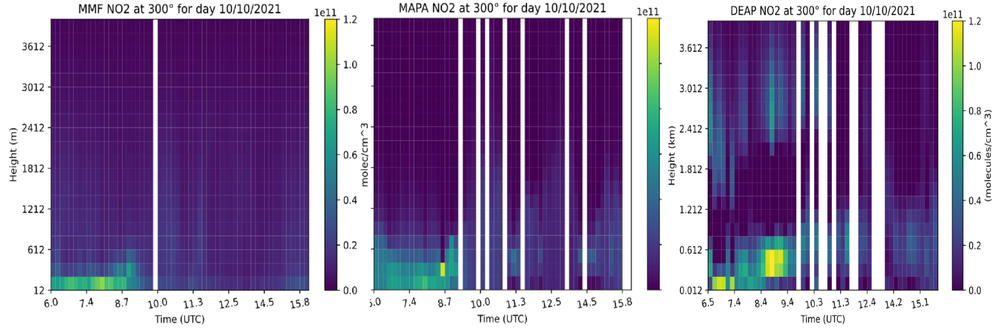
We apply the DEAP algorithm to SkySpec-2D SPC MAX-DOAS observations obtained on 1, 7 and 10 October 2021 and 14 December 2021.

DEAP performs more similarly to MMF than to MAPA. MMF and MAPA are very similar in tropospheric columns retrievals, differences are found for high NO2 surface values



NO_2 Values @ surface

Figure 5.11: Scatterplots of NO_2 surface values retrievals from DEAP, MMF and MAPA.



NO_2 profiles from MMF, MAPA, DEAP for the 10 October 2021.

NO_2 Tropospheric Columns

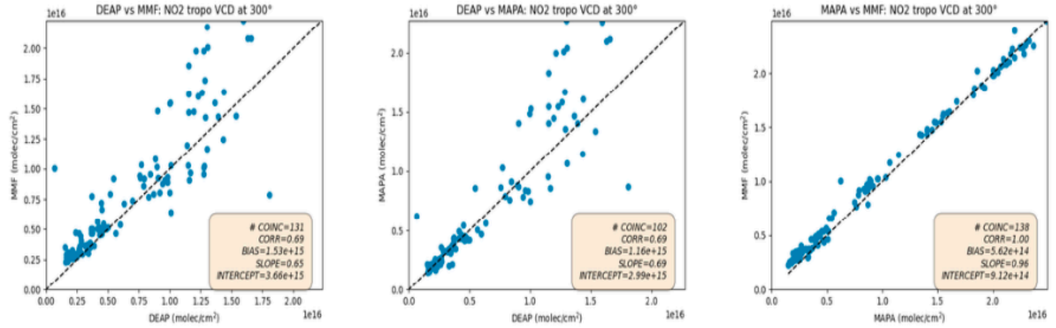


Figure 5.9: Scatterplots of NO_2 tropospheric column retrievals from DEAP, MMF and MAPA.

*For the MAPA and MMF retrievals we kindly acknowledge the FRM4DOAS and FRM4DOAS-2.0 projects (ESA contracts n˘r4000118181/16/I-EF and 4000135355/21/I-DT-Ir) and, in particular Caroline Fayt, Martina M. Friedrich, François Hendrick (IASB-BIRA) and Steffen Beirle (MPIC).

WPs 2250-2251: DOAS-BO- Phase II

- WP2250-1.3 Validation of NO_2 and aerosol retrieval code

NO2 Values @ surface

ALC data

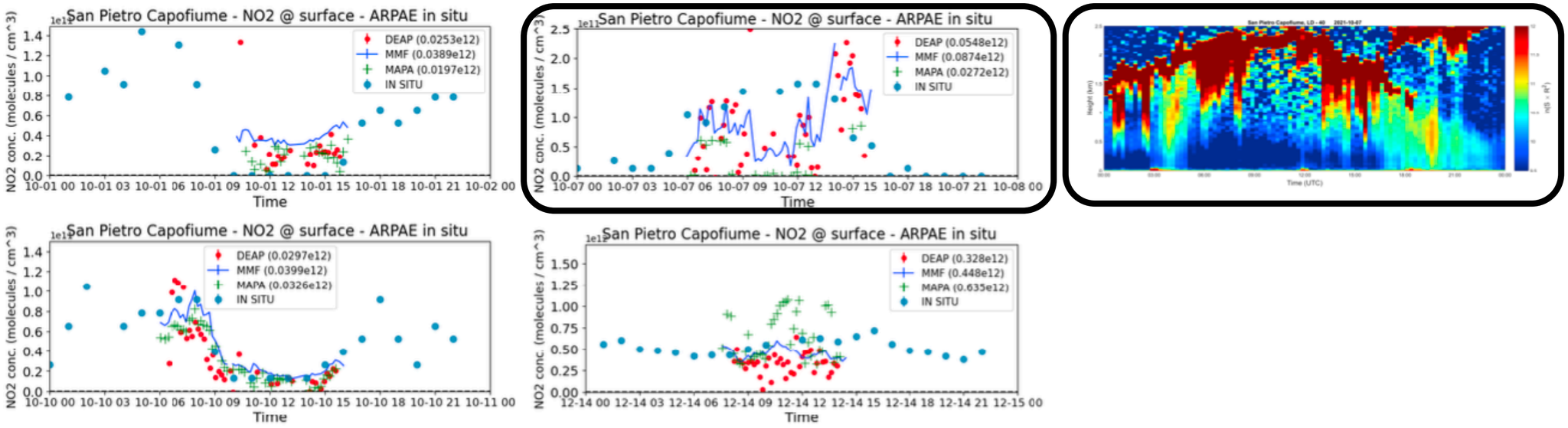


Figure 5.10: NO_2 at surface retrievals for 1, 7, 10 October 2021 and 14 December 2021 from DEAP (red), MMF (blue), and MAPA (green) together with Arpae hourly mean values (blue dots).

Exploitation of synergy of in situ data from ARPAE and remote sensing measurements at SPC

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.3 Validation of the NO_2 and aerosol retrieval code / WP2250-1.4 Inter-comparisons with satellite NO_2 tropospheric columns

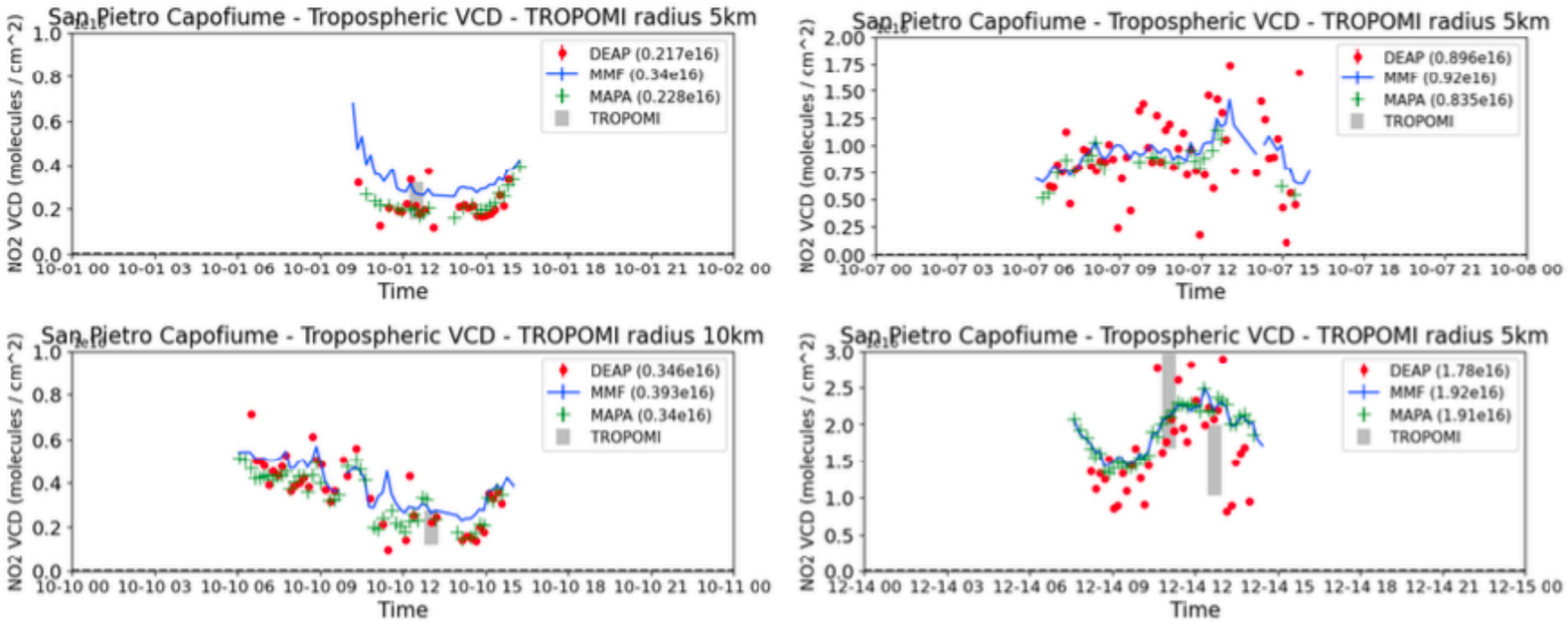
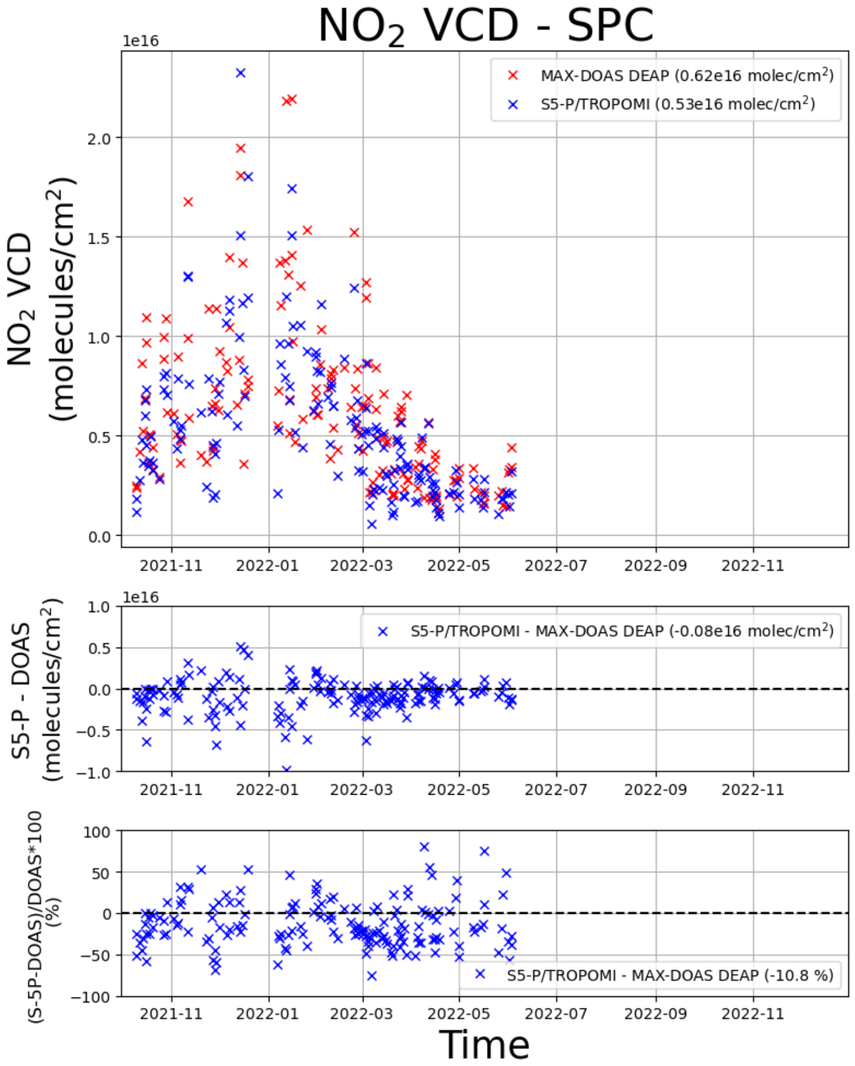


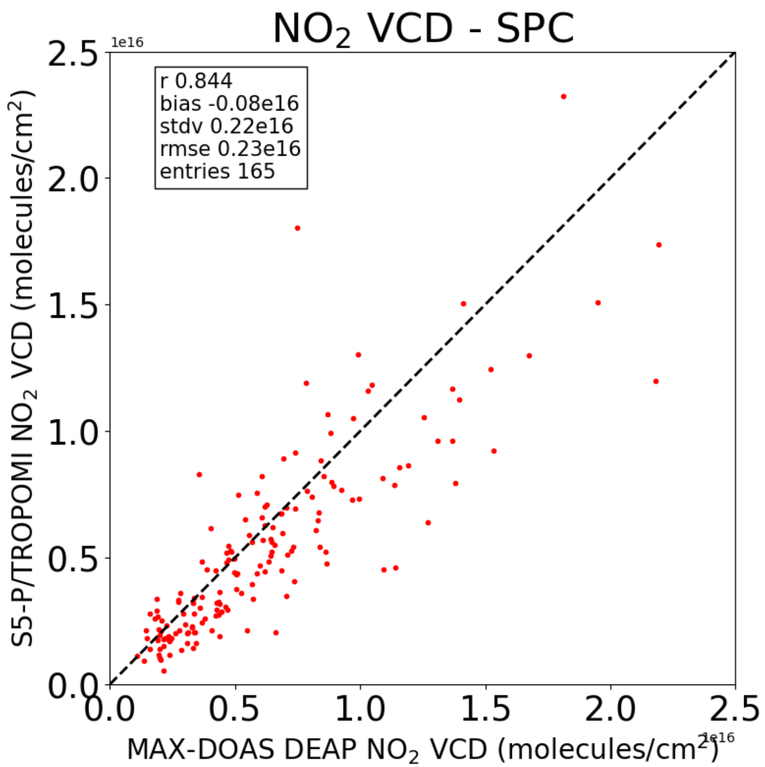
Figure 5.8: NO_2 tropospheric column retrievals for 1, 7, 10 October 2021 and 14 December 2021 from DEAP (red), MMF (blue), and MAPA (green) together with TROPOMI coincident values (when available) in grey.

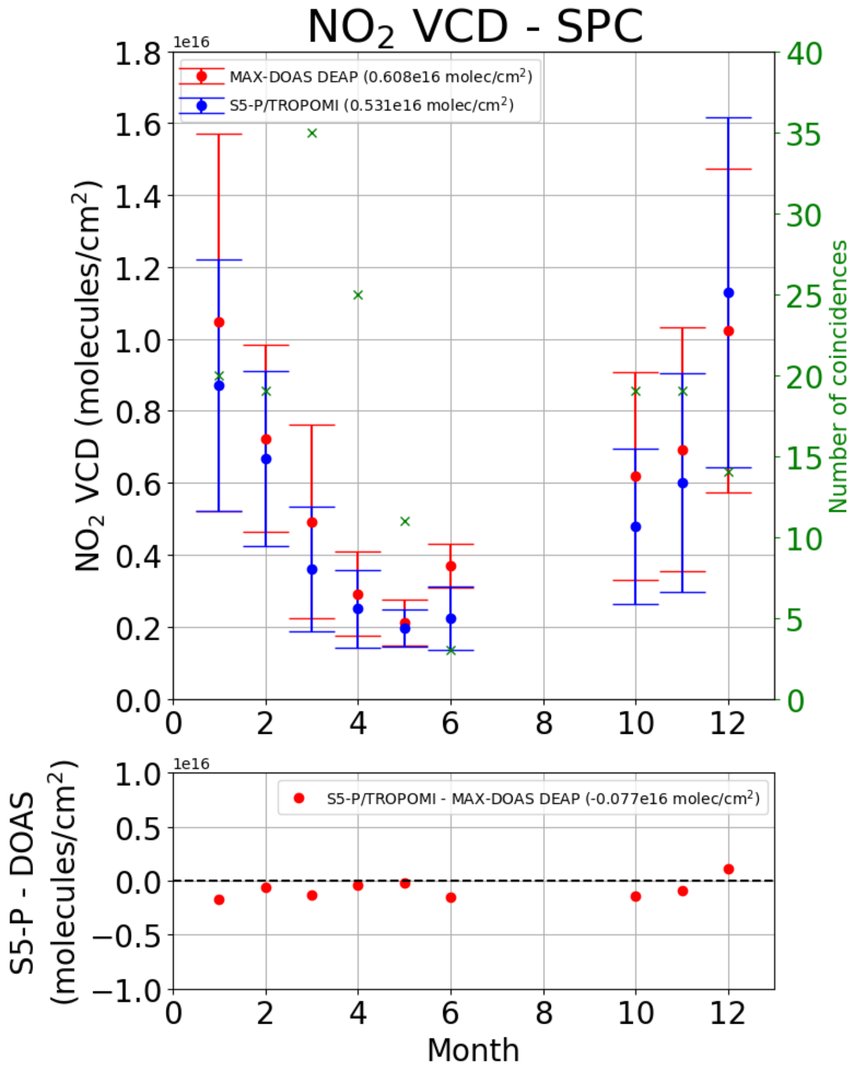
Exploitation of synergy of ground based remote sensing measurements at SPC and satellite data

WPs 2250-2251: DOAS-BO- Phase II

WP2250-1.4 Inter-comparisons with satellite NO_2 tropospheric columns





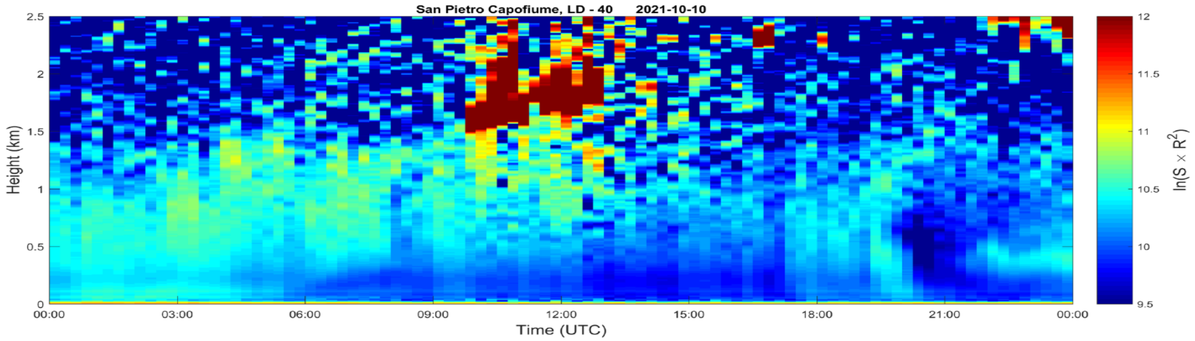


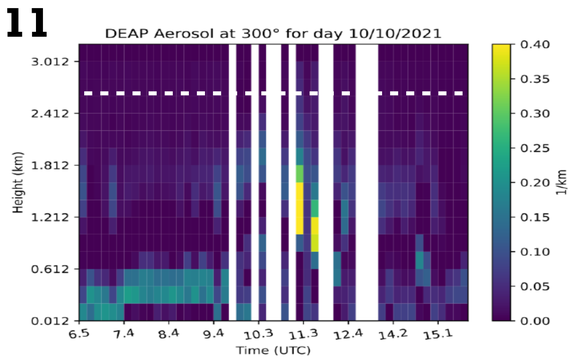
WPs 2250-2251: DOAS-BO- Phase II

WP2250-2.1 ALC data used as a-priori informations in NO_2 MAX-DOAS profiles retrievals

Exploitation of synergy of MAX-DOAS and ALC measurements at SPC

ln (SxR2) vs altitude and time from Ceilometer VAISALA LD40 on 10 October 2021





Extinction profiles retrieved with DEAP from SkySpec-2D MAX-DOAS measurements at 300 azimuth degrees on the 10 October 2021

Aerosol extinction profiles compares quite well with ALC signal

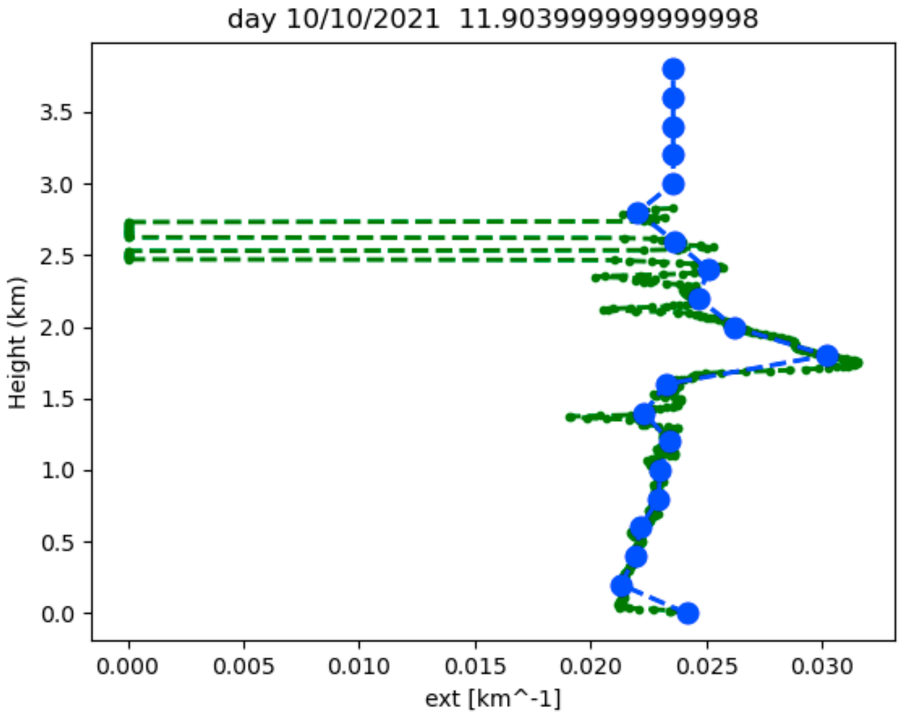
The retrieval of aerosol extinction is critical for a “good” NO_2 profile retrieval

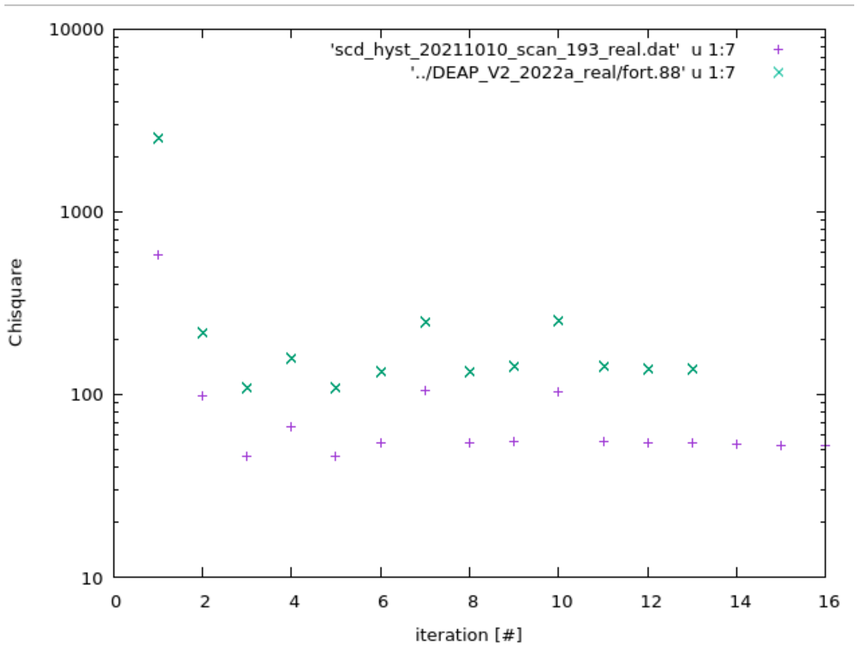
The use of ALC profile shape as initial guess should improve the NO_2 profile retrieval from MAX-DOAS measurements

WPs 2250-2251: DOAS-BO- Phase II

WP2250-2.1 ALC data used as a-priori informations in NO_2 MAX-DOAS profiles retrievals

The use of ALC profile shape as initial guess should improve the NO_2 profile retrieval from MAX-DOAS measurements





Chisquare= agreement between modeled and measured O_4 SCDs weighted by the noise

Using ALC aerosol extinction profile shape as initial guess produce a lower Chi-square value -> better agreement between measurements and simulations

WPs 2250-2251: DOAS-BO- Phase II

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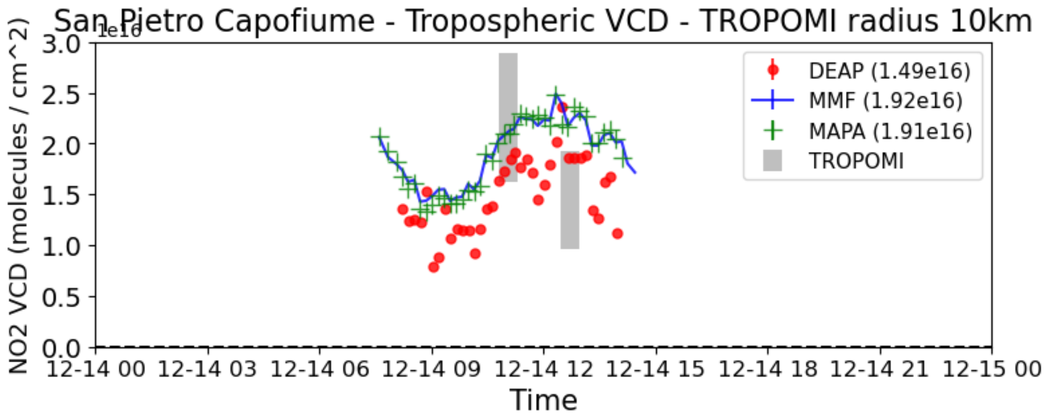
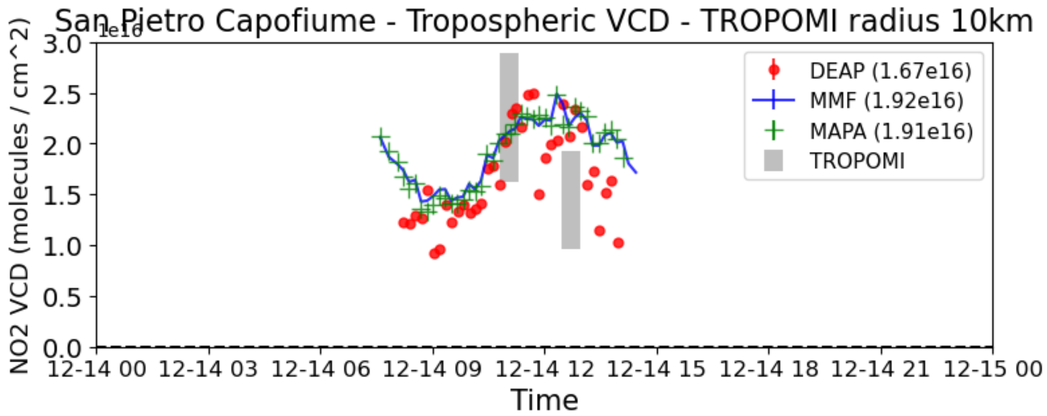
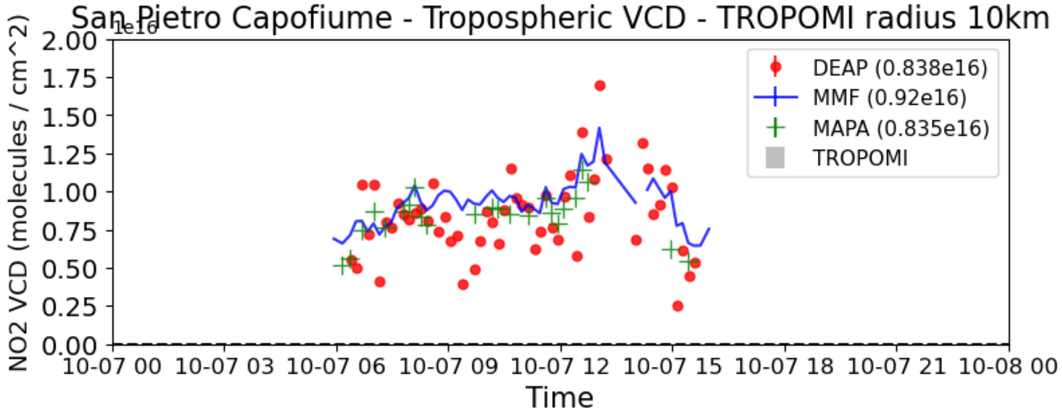
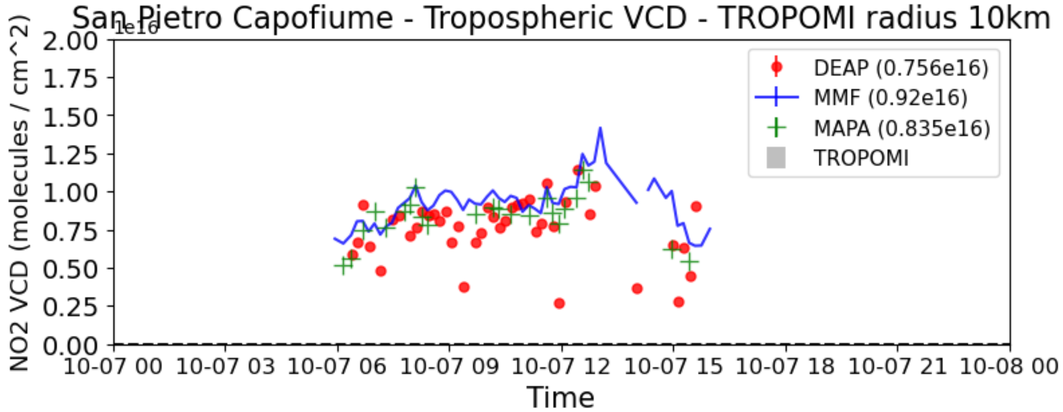
Without LD



With LD







WPs 2250-2251: DOAS-BO- Phase II

Conclusions

- Exploit the synergies between MAX-DOAS and aerosol remote sensing data in the Po Valley for satellite validation.

  We set-up the MAX-DOAS and the ALC systems

- Development of a retrieval code for profiles retrievals from MAX-DOAS measurements validation with FRM4DOAS specifications

  We Developed and validated the NO_2 and aerosol profile retrieval code (DEAP)

- Comparison of retrieved NO2 tropospheric VCDs with TROPOMI.

  We Start processing SPC MAX-DOAS data and comparing tropospheric NO_2 VCDs with TROPOMI

- Use of collocated aerosol profiles from ground-based instrumentation as initial guess for MAX-DOAS retrieval - new comparison with TROPOMI

  We Start ingesting ALC data as initial guess for DEAP

- Deliverables : D-1v1 and D-2 delivered
- Outreach: Poster presentation at “Sentinel 5p 5 years anniversary”

WPs 2250-2251: DOAS-BO- Phase II

Following WPs (end October 2023)

- Use of collocated aerosol profiles from ground-based instrumentation for comparison with aerosol satellites products

The “Giorgio Fea” observatory at SPC has a strong potential for exploitation of synergies among in-situ, remote sensing ground-based data and satellite data

Future availability of Raymetrics aerosols profiles is an added value of this observatory

A CIMEL has been recently installed in SPC, data available through AERONET since 6 February 2023 (only Level 1 available)

- Correction of TROPOMI NO_2 Tropospheric VCD to account for NO_2 profile - comparison with ground based MAX-DOAS profiles.

Future improvements and ideas

Up to now we concentrate on NO_2 for code development. However, Formaldehyde (HCHO) plays a key role in air quality.

We plan to:

1) At SPC : retrieve HCHO profiles and Tropospheric VCDs and use them to study the behavior of $HCHO/NO_2$ ratio vs O_3 in the Po Valley

2) In Rome Tor Vergata: retrieve HCHO profiles and Tropospheric VCDs and compare with Pandora data