



O3M SAF

**Study of the interpixel variability of NO₂ from GOME-2
using Mobile-DOAS measurements**

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**VISITING SCIENTIST ACTIVITY 2015/2016
FINAL REPORT**

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Acronyms and abbreviations

AMF	Air Mass Factor
AROMAT	Airborne ROmanian Measurements of Aerosols and Trace gases
BIRA-IASB	Royal Belgian Institute for Space Aeronomy
DOAS	Differential Optical Absorption Spectroscopy
DSCD	Differential Slant Column Density
ESA	European Space Agency
EUMETSAT	European Organisation for the Exploitation of Meteorological Satellites
GDP	GOME Data Processor
GOME-2	Global Ozone Monitoring Experiment-2
MAD-CAT	Multi Axis Doas - Comparison campaign for Aerosols and Trace gases
MAX-DOAS	Multi-Axis Differential Optical Absorption Spectroscopy
MetOp	Meteorological Operational satellite programme
NO ₂	Nitrogen Dioxide
O3M-SAF	Ozone and Atmospheric Chemistry Monitoring Satellite Application Facility
SCD	Slant Column Density
UGAL	Dunarea de Jos University of Galati
VCD	Vertical Column Density
VSA	Visiting Scientist Activity



Introduction

Scope of this document

This report summarizes the Visiting Scientist Activity (VSA) realized by Daniel-Eduard Constantin, that has been realized in Romania and Belgium during September 2015-October 2016. This activity is based on the collaboration between Dunarea de Jos University of Galati (UGAL) and the royal Belgian Institute for Space Aeronomy (BIRA-IASB) in the framework of O3M-SAF project.

The main objective of the activity was to study the interpixel variability of tropospheric NO₂ VCD derived from GOME-2 (A&B) observations on both MetOp-A&B platforms, from the GOME Data Processor (GDP version 4.7) against ground-based mobile-DOAS observations performed by UGAL and BIRA-IASB.

Preliminary notes

The first task of the proposed VSA was to generate a database of tropospheric NO₂ VCD deduced from mobile DOAS measurements and to extract coincident GOME-2 observations. For this purpose, tropospheric NO₂ VCDs derived from GOME-2 observations on both MetOp-A and MetOp-B were extracted from the latest available version of the GOME Data Processor (GDP version 4.7). All coincidences between the MetOp A&B satellite overpasses and existing mobile DOAS measurements performed by UGAL and by BIRA-IASB since 2010 were considered and the corresponding data sets extracted (see first intermediate VSA report). The second task was focused on the investigation of the averaging effect created inside the satellite pixels through analysis of the NO₂ VCD variability as sampled by the mobile-DOAS measurements within the GOME-2 satellite pixel (see second intermediate VSA report).

This report summarizes all findings and provides a more comprehensive discussion of the results. A statistical analysis was performed according to a classification related to the NO₂ content, as seen by GOME-2. The mobile-DOAS measurements were selected using different time-coincidence criteria, in order to evaluate the impact of this parameter on the validation results.

During the VSA several airborne-DOAS experiments were also analysed, but due to the small number of DOAS flights and the limited matches with GOME-2 overpasses, the airborne DOAS experiments have been integrated in the data base of ground-based DOAS observations. We found only four days of airborne-DOAS measurements matching GOME-2 overpasses, from a total of eight airborne-DOAS experiments.



Background

The GOME-2 instruments [1, 2] on board of MetOp-A/B satellite platforms are based on nadir-viewing scanning spectrometer dedicated to observation of atmospheric trace gases. Both MetOp platforms fly on polar sun-synchronous orbits with an overpass time at the equator of 09:30 local time. MetOp-A was launched in October 2006 and was followed by MetOp-B in September 2012. The main trace gases detected by GOME-2 are: ozone, nitrogen dioxide, sulphur dioxide, water vapour, bromine oxide and formaldehyde, as well as aerosols. The ground-pixel size of GOME-2 on MetOp-A is 80 x 40 km² before 15 July 2013 and 40 x 40 km² after 15 July 2013. The footprint size of the GOME-2 instrument on MetOp-B is 80 x 40 km².

Methodology

During the VSA car and airborne DOAS measurements performed in Romania and other countries from Europe were examined.

The mobile DOAS observations used are part of three important campaigns: AROMAT 2014/2015 (Airborne Romanian Measurements of Aerosols and Trace gases- [<http://uv-vis.aeronomie.be/aromat>] performed in Romania and MAD-CAT (Multi Axis Doas - Comparison campaign for Aerosols and Trace gases [http://joseba.mpch-mainz.mpg.de/mad_cat.htm] held in Mainz, Germany – June 7th/July 5th 2013. In addition, routine measurements performed in 2010-2011 by BIRA-IASB, mainly inside Belgium, were included in this work [3].

The mobile DOAS instrument used for the ground-based DOAS measurements consists of a compact Czerny-Turner spectrometer, fiber optic, telescope, GPS and a PC [4]. The car-DOAS measurements were performed using zenith-sky [4] and MAX-DOAS geometries [5]. The MAX-DOAS system is based on a compact double spectrometer, where the scattered light spectra are recorded simultaneously in the zenith direction and 30° above the horizon.

Tropospheric NO₂ VCDs (VCD_{tropo}) derived from GOME-2 observations on both Metop-A and Metop-B were extracted from GDP version 4.7 [6, 7]. All coincidences between the Metop A&B satellite overpasses and existent mobile DOAS measurements performed by UGAL and by BIRA-IASB since 2010 were considered and the corresponding data sets extracted. This resulted in 143 days of mobile DOAS observations selected as potential useful data for the space-ground intercomparison. After data analysis and quality control, this number was reduced to 111 days of useful measurements ready for comparison with GOME-2. In a second stage, the geo-coincidence between mobile DOAS observations and GOME-2 observations was checked pixel by pixel. A number of 125 pixels (GOME-2A=106; GOME-2B=19) were identified as having a useful geo-coincidence with the mobile DOAS observations. Only cloud free pixels (cloud fraction < 30%) and positive VCD_{tropo} were selected.

The comparison between GOME-2 pixels and mobile DOAS observations was performed using a classification based on several parameters: satellite pixel coverage by the mobile DOAS observations, time window, distance to the main source of pollution, and level of pollution observed by GOME-2.

An estimation of the *pixel coverage* was obtained by defining the 25%, 50% and 100% coverage of the mobile DOAS measurements inside of a satellite pixel. Figure 1 illustrates the three types of pixel classifications as a function of the trajectory and distance travelled inside a GOME-2 pixel.

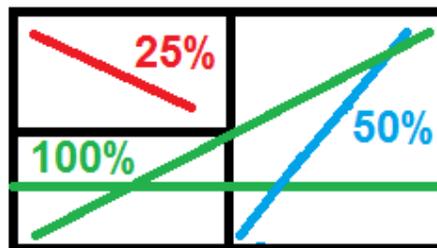


Figure 1. Classification of pixel coverage by trajectory and distance travelled inside a GOME-2 pixel

The *time window* selection was performed in our investigation using only mobile DOAS observations available in a time window of ± 1 , ± 2 , and ± 3 hours of the GOME-2 overpass.

The *pollution level* conditions were analysed as a function of the tropospheric NO_2 column amounts observed by GOME-2. The pollution level was classified as: Low (1 to 5×10^{15} molec/cm²), Medium (5 to 10×10^{15} molec/cm²) and High ($> 10 \times 10^{15}$ molec/cm²),

The *distance to the main source of pollution* was calculated as the distance between the ground-based station (e.g. Bruxelles, Mainz, etc) and the center of the GOME-2 pixel selected for comparison.

Results and discussions

To achieve the purpose of the present VSA, Pearson correlation coefficients were calculated between GOME-2 observations and mobile DOAS measurements. The analysis follows the methodology presented in the previous chapter, focusing on pixel cover, time window, distance to the main source of pollution, and level of pollution.



GOME-2 A

Figure 2 presents the time series (including errors) of the GOME-2A and mobile DOAS observations using measurements performed during 2010-2015, without any filtering. This corresponds to a correlation of $R=0.514$

Table 1 presents correlations between GOME-2A and mobile DOAS observations as a function of the pixel coverage definition (Figure 1). It was found that the correlation has a higher coefficient when the distance travelled inside the pixel covers a larger area (see Table 1). For a coverage larger than 50% we find a correlation coefficient $R=0.56$. For the next level of coverage (100%) the correlation coefficient increases less but the slope and intercept reinforce the idea that a larger coverage leads to a better agreement between space and ground-based observations. Figures 3 and 4 present scatter plots between GOME-2A and mobile DOAS observations. Analysing Figure 4, one can see that the lowest scatter between space-based and ground-based observations is obtained in the case of largest coverage (100%).

Table 2 presents the correlation between GOME-2A and mobile DOAS observations as a function of the time window (before or after satellite overpass). The correlation coefficient is directly influenced by the time window of mobile DOAS observations around the satellite overpass. In addition to the correlation coefficient, we observe that the slope and intercept also depend on the selected time window. The slope increases and the intercept decreases when the time window increases. We found that the best correlation coefficient ($R=0.667$) is obtained for a time window ± 3 hours according to the satellite overpass, here a slope of 0.82 and an intercept of 3.7 was calculated. Figure 5 shows a map of the tropospheric NO_2 VCD observed by GOME-2A and mobile DOAS measurements corresponding to the use of a time window ± 3 hours around the satellite overpass.

Table 3 introduces the correlations between GOME-2A and mobile DOAS observations as a function of the distance to the main source of pollution (distance measured from the center of the pixel used for comparisons). The best agreement ($R=0.55$) is found when pixels centered at less than 30km from the source are used. A higher correlation coefficient ($R=0.66$) is even determined when only pixels centered at 10-30km from the source are used.

The smallest correlation coefficient ($R=0.41$) is obtained when the ground observations are performed close to highly polluted sources (less than 10km). In this case it is likely that the NO_2 field is very heterogeneous and therefore car-DOAS measurements are not representative of satellite measurements anymore.

Note however that generally the correlation coefficient increases inversely to the distance to the source of pollution. The best correlation coefficient was obtained for observations in a range of 10-20km to the sources of pollution. In this case, besides a good correlation coefficient ($R=0.67$), also a slope very close to 1 was obtained.

Table 4 shows the correlations between GOME-2A and mobile DOAS observations as a function of the level of pollution. We find the best correlation ($R=0.43$) between GOME-2A and mobile DOAS observations when GOME-2A observations are classified as medium polluted (tropospheric NO_2 VCD = 5 to 10×10^{15} molec./ cm^2). For the two other types of classification (low and high) a correlation coefficient $R < 0.36$ was obtained. For pixels with a tropospheric NO_2 content higher than 1×10^{16} molec./ cm^2 a correlation coefficient $R=0.18$ was determined. In this case a slope of 0.22 and a very large intercept ($I=14.41$) were calculated. This small correlation coefficient may be caused by the fact that GOME-2A "sees" NO_2 sources which are not in the range of the detection of mobile DOAS observations. In the opposite case, when the ground DOAS observations detect more NO_2 than GOME-2A a low correlation coefficient ($R=0.36$) is obtained. Here one can mention the case of the Turceni area, Romania, where large power plants are located. GOME-2A detects a low tropospheric NO_2 content ($< 5 \times 10^{15}$ molec./ cm^2) while mobile DOAS measurements, which were performed nearby the sources, detect the entire NO_2 amount released by the power plant ($> 1 \times 10^{16}$ molec./ cm^2). The correlation coefficient calculated for the medium level of pollution ($R=0.43$) is probably related to the homogeneous NO_2 fields which is generally encountered in such conditions.

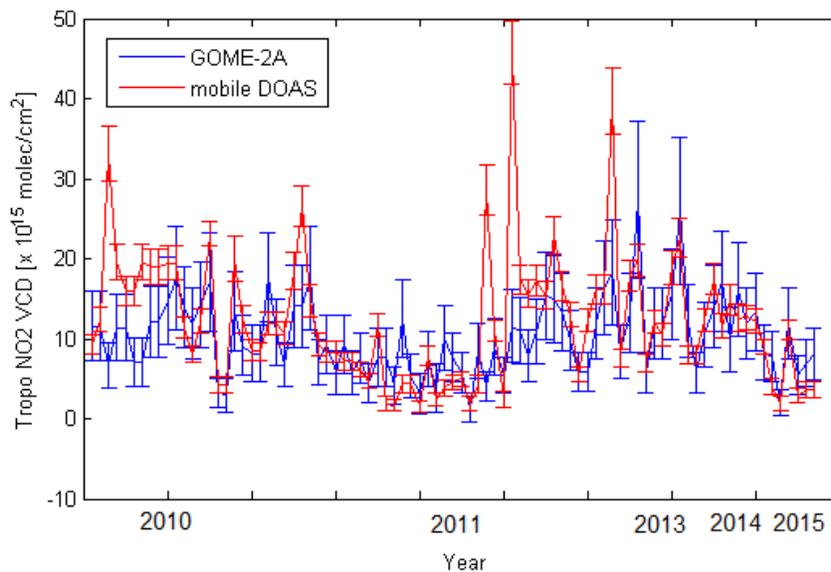


Figure 2. Time series (including errors) of the GOME-2A & mobile DOAS observations using measurements performed during 2010-2015

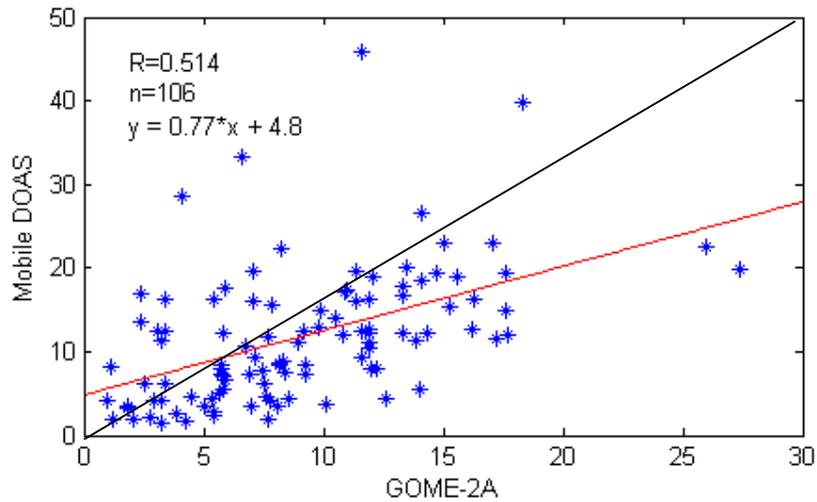


Figure 3. Scatter plot and linear regression analysis of the tropospheric NO₂ VCD observed by GOME-2A and mobile DOAS observations

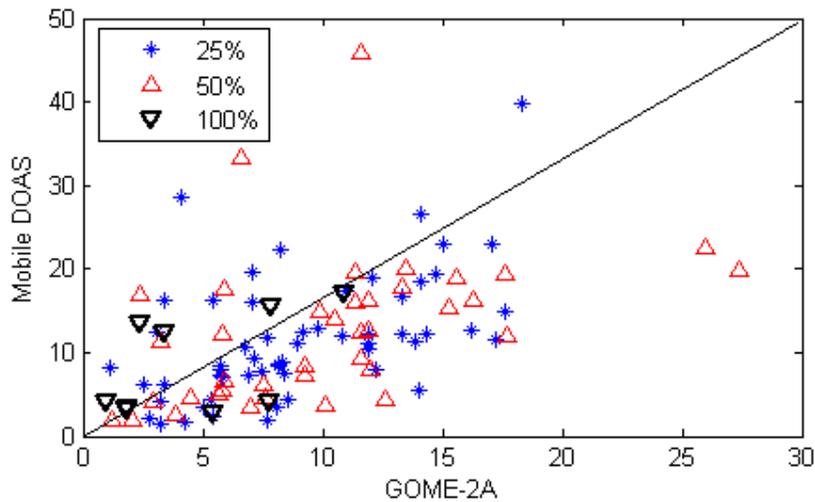


Figure 4 Scatter plot and linear regression analysis of the tropospheric NO₂ VCD observed by GOME-2A and mobile DOAS observations highlighting the pixel coverage

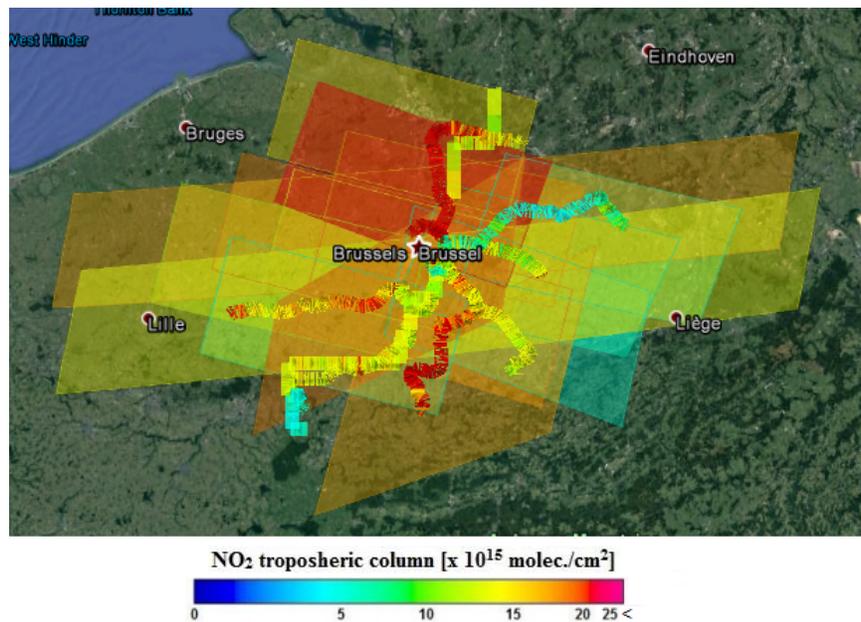


Figure 5. Color-coded tropospheric NO₂ VCD observed by GOME-2A and mobile DOAS measurements performed in the vicinity of Belgium in 2010 and 2011.

Table 1. Correlation between GOME-2A and mobile DOAS observations as a function of the pixel cover

Mobile data Coverage	R	N	S	I
25%	0.439	57	0.88	3.8
50%	0.567	39	0.67	5.9
100%	0.574	10	0.91	4.2

where: R= correlation coefficient; N=number of observations; S=slope; I=intercept

Table 2. Correlation between GOME-2A and mobile DOAS observations as a function of the time window around satellite overpass

Time window (around satellite overpass)	R	N	S	I
±1	0.394	24	0.68	5.9
±2	0.446	62	0.74	5
±2 (excluding ±1)	0.518	42	0.71	5.2
±3	0.494	83	0.76	4.4

± 3 (excluding $\pm 1, \pm 2$)	0.667	18	0.82	3.7
$\pm \geq 3$	0.506	38	0.81	5.6

Table 3. Correlations between GOME-2A and mobile DOAS observations as a function of the distance to the main source of pollution

Distance to the main source of pollution	R	N	S	I
<10km	0.41	11	0.74	6.9
<20km	0.52	33	0.82	5.3
<30km	0.55	50	0.82	4.1
10-20km	0.67	23	0.95	3.49
20-30km	0.65	17	0.67	2.8
30-40km	0.23	18	0.42	11.21

Table 4. Correlations between GOME-2A and mobile DOAS observations as a function of the level of pollution

Level of pollution	R	N	S	I
Low	0.36	25	0.13	2.68
Medium	0.43	23	0.16	6.46
High	0.18	58	0.22	14.41



GOME-2 B

Comparisons between GOME-2B observations and mobile DOAS observations were also performed. A Pearson correlation coefficient of $R=0.18$ was found between GOME-2B observations and mobile DOAS measurements. Figure 6 presents the time series (including errors) of the GOME-2B & mobile DOAS observations using measurements performed during 2013-2015. Most of the mobile DOAS measurements used for the GOME-2B comparisons were performed in areas where the NO_2 content was higher than 1×10^{16} molecules/cm², in Romania in the vicinity of large coal-burning power plants.

Figure 7 presents a map of the tropospheric NO_2 VCD observed by GOME-2B and mobile DOAS measurements in the area of Turceni and Rovinari power plants, Romania. These measurements were performed during the AROMAT-2014 and 2015 campaigns. The measurements were performed very close to exhaust plume of the power plant, being directly influenced by a large amount of NO_2 . The NO_2 emissions over these areas are highly localized, which leads to an average of tropospheric NO_2 observed by mobile DOAS observations very high. Considering the coarse resolution of the GOME-2B instrument ($40 \times 80 \text{ km}^2$) an important smoothing effect is expected to take place inside the pixel, which probably explains the low correlation coefficient $R=0.18$ (slope=0.61 and intercept=0.65).

Also for comparisons with GOME-2B, additional mobile DOAS measurements were performed in Bucuresti city, Romania. Figure 8 shows the tropospheric NO_2 VCD observed by GOME-2B and mobile DOAS measurements over Bucuresti., which shows very good agreement of the two datasets outside the city, and higher mobile columns in the city center.

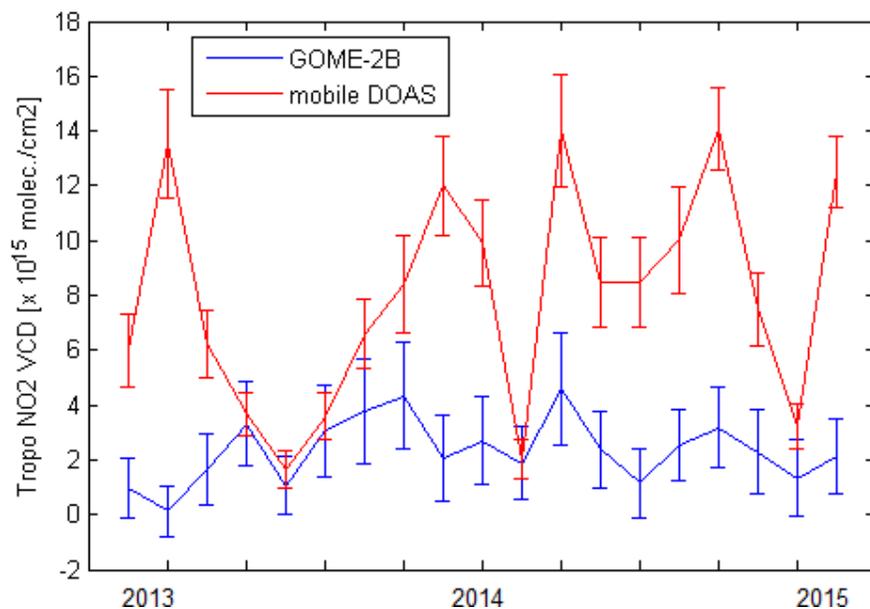


Figure 6. Time series (including errors) of the GOME-2B & mobile DOAS observations using measurements performed during 2013-2015

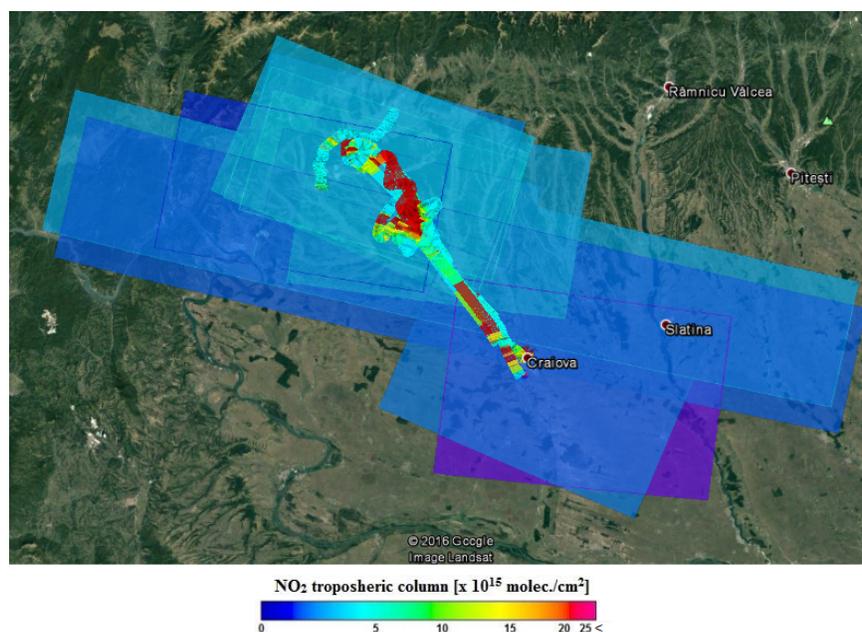


Figure 7. Color coded tropospheric NO_2 VCD observed by GOME-2B and mobile DOAS measurements performed in Romania on 2014 and 2015 (in this map also the airborne-DOAS measurements are represented)

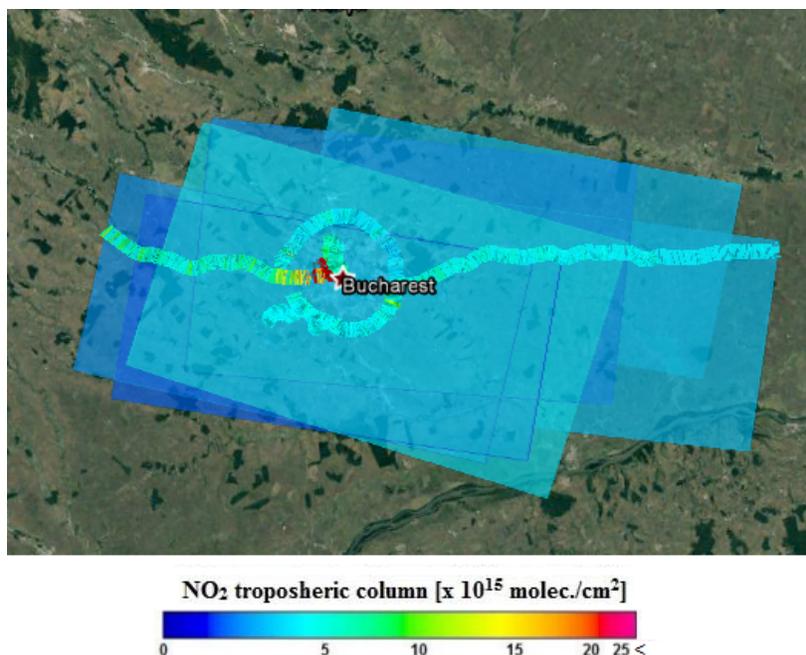


Figure 8. Color-coded tropospheric NO₂ VCD observed by GOME-2B and mobile DOAS measurements performed in Romania on 2014 and 2015

Conclusions and outlook

In the framework of this Ozone SAF Visiting Scientist project dedicated to the study of the interpixel variability of NO₂ from GOME-2 using Mobile-DOAS measurement several activities/tasks described in the proposed workplan have been achieved:

- A large database of mobile DOAS observations was used for the first time for the study of the interpixel variability of tropospheric NO₂ VCD observed by GOME-2 onboard Metop-A/B platforms. This database has included mobile DOAS measurements performed during three important campaigns: AROMAT 2014/2015 performed in Romania and MADCAT2013 performed in Germany. Also, other measurements performed in Romania and Belgium were included in this work.
- An improved retrieval algorithm for the tropospheric NO₂ VCD deduced from mobile MAX-DOAS observations, based on AMF lookup tables, was developed by BIRA-IASB and used in this VSA.



- We found a good agreement between mobile car-DOAS measurements and GOME-2A observations ($R=0.51$). Calculating correlations between GOME-2A and ground-based observations as a function of the pixel coverage we found the best agreement ($R=0.57$) when the mobile DOAS measurements crossed more than 50% of the satellite pixel. The correlation coefficient is directly influenced by the time window of mobile DOAS observations around the satellite overpass. A time interval which is longer than one hour and less than three hours leads to a good agreement ($R>0.45$) between GOME-2A and ground observations. Another important factor is the distance to the main source of pollution. The best correlation coefficient ($R=0.67$) was obtained when satellite observations were selected in a range of 10-20km from the pollution sources. Also, correlations between GOME-2A and mobile DOAS observations as a function of the level of pollution were calculated. In this case the correlation coefficient calculated for the medium level of pollution ($R=0.43$) provided the best results, which is probably related to the homogeneity of the NO_2 fields under medium pollution levels. For GOME-2B the agreement is lower ($R=0.18$), mostly due to the reduced number of ground-based observations and the different conditions sampled by GOME-2 and mobile-DOAS observations.
- After performing this VSA, some general recommendations could be formulated regarding satellite validation using mobile DOAS measurements:
 1. the mobile DOAS measurements should cover as much as possible the satellite pixel surface. The linear measurements which cross more than 50% of satellite pixel could lead to a good agreement between space and ground DOAS observations.
 2. a time window of maximum ± 3 hours according to the satellite overpass could be recommended for comparisons with satellite observations. This time interval of maximum ± 3 hours is related to the time needed to travel a long-enough distance inside the pixel ($40 \times 80 \text{ km}^2$ or $40 \times 40 \text{ km}^2$).
 3. when comparing satellite to ground-based DOAS observations, knowledge of the source of pollution is essential and a special attention should be paid to the magnitude of the source of pollution. The ground-based DOAS observations, selected for satellite comparisons, should be analysed considering the different cases of urban, rural and industrial areas.
- Results from the present VSA were presented during the 2016 EUMETSAT Meteorological Satellite Conference as a poster. Also, the results presented in this work will be the subject of a peer-reviewed publication.



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