The Airborne ROmanian Measurements of Aerosols and Trace gases (AROMAT) campaigns

Alexis Merlaud1, Livio Belegante2, Daniel-Eduard Constantin3, Mirjam Den Hoed4, Andreas Carlos Meier5, Marc Allart4, Magdalena Ardelean6, Maxim Arseni3, Tim Bösch5, Hugues Brenot1, Andreea Calcan6, Emmanuel Dekemper1, Sebastian Donner8, Steffen Dörner8, Carmelia Dragomir3, Lucian Georgescu3, Anca Nemuc2, Doina Nicolae2, Gaia Pinardi1, Andreas Richter5, Adrian Rosu3, Thomas Ruhtz7, Anja Schönhardt5, Dirk Schuetttemeyer10, Reza Shaiganfar8, Kerstin Stebel9, Frederik Tack1, Sorin Nicolae Vâjâiac6, Jeni Vasilescu2, Jurgen Vanhamel1, Thomas Wagner8, and Michel Van Roozendael1

1Royal Belgian Institute for Space Aeronomie (BIRA-IASB), Avenue Circulaire 3, 1180 Brussels, Belgium
2National Institute of R&D for Optoelectronics (INOE), Magurele, Street Atomistilor 409, Magurele 77125, Romania
3“Dunarea de Jos” University of Galati, Faculty of Sciences and Environment, Str. Domneasca, Nr. 111, Galati 800008, Romania
4Royal Netherlands Meteorological Institute (KNMI), De Bilt, The Netherlands
5Institute of Environmental Physics, University of Bremen (IUP-Bremen), Otto-Hahn-Allee 1, 28359 Bremen, Germany
6National Institute for Aerospace Research “Elie Carafoli” (INCAS), Bd. Iuliu Maniu no. 220, Bucharest, Romania
7Institute for Space Sciences, Free University of Berlin (FUB), Carl-Heinrich-Becker-Weg 6-10, 12165 Berlin, Germany
8Max-Planck-Institute for Chemistry (MPIC), Hahn-Meitner-Weg 1, 55128 Mainz, Germany
9Norwegian Institute for Air Research (NILU), Instituttveien 18, 2007 Kjeller, Norway
10European Space Agency (ESA-ESTEC), Keplerlaan 1, 2201 AZ Noordwijk, The Netherlands

Correspondence: A.Merlaud (alexism@oma.be)

Abstract.

The Airborne ROmanian Measurements of Aerosols and Trace gases (AROMAT) campaigns took place in Romania in September 2014 and August 2015. They focused on two sites: the Bucharest urban area and the power plants in the Jiu Valley. Their main objectives were to test recently developed airborne observation systems dedicated to air quality studies and to verify the concept of such campaigns in support of the validation of spaceborne atmospheric missions such as the TROPOspheric Monitoring Instrument (TROPOMI)/Sentinel-5 Precursor (S5P).

We show that tropospheric NO2 vertical column density (VCD) measurements using airborne mapping instruments are in principle valuable for satellite validation. The signal to noise ratio of the airborne NO2 measurements is one order of magnitude higher than its spaceborne counterpart when the airborne measurements are averaged at the TROPOMI pixel scale. A significant source of comparison error appears to be the time variation of the NO2 VCDs during a flight, which we estimated at about 4x1015 molec cm−2 in the AROMAT conditions. Considering the random error of the TROPOMI tropospheric NO2 VCD (σ), the dynamic range of the NO2 VCDs field extends from detection limit up to 37σ (2.6x1016 molec cm−2) or 29σ (2x1016 molec cm−2) for Bucharest and the Jiu Valley, respectively. For the two areas, we simulate validation exercises of the TROPOMI tropospheric NO2 product using airborne measurements. These simulations indicate that we can closely approach the TROPOMI optimal target accuracy of 25% by adding NO2 and aerosol profile information to the airborne mapping obser-
vations, which constrains the investigated accuracy to within 28%. In addition to NO₂, we also measured significant amounts of SO₂ in the Jiu Valley, as well as a hotspot of H₂CO in the center of Bucharest. For these two species, we conclude that the best validation strategy would consist in deploying ground-based measurement systems at key locations which the AROMAT observations help identify.

1 Introduction

Since the launch of the Global Ozone Monitoring Experiment (GOME, Burrows et al. (1999)) in 1995, spaceborne observations of reactive gases in the UV-visible range have tremendously improved our understanding of tropospheric chemistry. GOME mapped the large urban sources of NO₂ in North America and Europe, the SO₂ emissions from volcanoes and coal-fired power plants (Eisinger and Burrows, 1998), and the global distribution of H₂CO with its maxima above East Asia and the tropical forests (De Smedt et al., 2008). Subsequent air-quality satellite missions expanded on the observation capabilities of GOME. Table 1 lists the past, present, and near-future nadir-looking satellite instruments dedicated to ozone and air quality monitoring with their sampling characteristics in space and time. The pixel size at nadir has shrunk from 320x40 km² (GOME) to 3.5x5.5 km² (TROPOMI, Veefkind et al. (2012)). This high horizontal resolution enables for instance to disentangle contradictory trends in ship and continental emissions of NO₂ in Europe (Boersma et al., 2015) or to distinguish the different NO₂ sources in oil sand mines in Canada (Griffin et al., 2019). The satellite-derived air quality products are now reliable enough to improve the bottom-up emission inventories (e.g. Kim et al. (2009), Fioletov et al. (2017), Bauwens et al. (2016)) and to be used in operational services, for instance to assist air traffic control with the near-real time detection of volcanic eruptions (Brenot et al., 2014). The bottom lines of Table 1 presents the near-future perspective in spaceborne observation of the troposphere: a constellation of geostationary satellites will provide hourly observations of the troposphere above east Asia (GEMS, (Kim, 2012)), North America (TEMPO, Chance et al. (2013)), and Europe (Sentinel-4, Ingmann et al. (2012)). These new developments will open-up new perspectives for atmospheric research and air quality policies (Judd et al., 2018).

Validation is a key aspect of any spaceborne Earth observation mission. This aspect becomes even more important as the science matures and leads to more operational and quantitative applications. Validation involves a statistical analysis of the differences between measurements to be validated and reference measurements (von Clarmann, 2006). The aim of validation is to verify that the satellite data products meet their requirements in terms of accuracy and precision. Table 2 presents such requirements for the TROPOMI-derived tropospheric vertical column densities (VCDs) of NO₂, SO₂, and H₂CO (ESA, 2014). Richter et al. (2014) have discussed the challenges associated with the validation of tropospheric reactive gases. These challenges arise from the large variability in space and time of short-lived reactive gases, the dependency of the satellite products on different geophysical parameters (surface albedo, profile of trace gases and aerosols), the differences in vertical sensitivity between satellite and reference (ground-based or airborne) measurements, and the small signals. An ideal validation study would involve a reference dataset of VCDs whose well-characterized uncertainties would be small compared to those required for the investigated products. This reference dataset would cover a large amount of satellite pixels with adequate spatial and temporal representativeness at different seasons, places, and pollution levels. Beside the VCDs, the ideal validation exercise
would also quantify the geophysical parameters that impact the retrieval of the investigated satellite products. In the real world however, Richter et al. points out that "the typical validation measurement falls short in one or even many of these aspects".

The first validations of the tropospheric NO₂ and H₂CO VCD products of GOME involved in-situ samplings from aircraft (Heland et al., 2002; Martin et al., 2004). Such measurements may cover good fractions of satellite pixels but they miss the lower part of the boundary layer, where the trace gas concentrations often peak. Schaub et al. (2006) and Boersma et al. (2011) summarize other early validation studies for the tropospheric NO₂ VCDs retrieved from GOME, SCIAMACHY, and OMI. Several of these studies make use of the NO₂ surface concentration datasets from air quality monitoring networks. Compared to campaign-based data acquisition, operational in-situ networks provide long-term measurements, but their comparison with satellite products relies upon assumptions on the NO₂ profile. Other validation studies use remote-sensing from the ground and aircraft, in particular based on the Differential Optical Absorption Spectroscopy (DOAS) technique (Platt and Stutz, 2008), which is also the basis for the retrieval algorithms of the satellite-derived products. In comparison with in-situ measurements, DOAS has the benefit of being directly sensitive to the column density of a trace gas, i.e. the same geophysical quantity which is retrieved from space. Heue et al. (2005) conducted the first comparison between a satellite-derived product (SCIAMACHY tropospheric NO₂) and airborne DOAS data. Many validation studies also use ground-based DOAS measurements, in particular since the development of the Multi-AXis DOAS (MAX-DOAS) technique (Hönninger et al., 2004). MAX-DOAS measurements are valuable for validation due to their ability to measure integrated columns at spatial scales comparable to the satellite ground pixel size. Moreover, they broaden the scope of validation activities since they also provide limited profile information on both trace gases and aerosols (Irie et al., 2008; Brinksma et al., 2008; Ma et al., 2013; Kanaya et al., 2014; Wang et al., 2017; Drosoglou et al., 2018). The limitations of using the MAX-DOAS technique for validation arise from their still imperfect spatial representativeness compared to typical satellite footprints and to some extent from their limited sensitivity in the free troposphere. Spatial representativeness has often been invoked to explain the apparent low bias of the OMI tropospheric NO₂ VCDs in urban conditions (Boersma et al., 2018).

The unprecedented horizontal resolution enabled by the last generation of air-quality space-based instruments motivated preparatory field studies around polluted areas in North America (DISCOVER-AQ, https://discover-aq.larc.nasa.gov), Europe (AROMAT and AROMAPEX, Tack et al.,2019) and Korea (KORUS-AQ, https://www-air.larc.nasa.gov/missions/korus-aq/). These campaign activities quantified key pollutants (NO₂, SO₂, O₃, H₂CO, and aerosols) and assessed practical observation capabilities of future satellite instruments while preparing for their validation. They combined ground-based and airborne measurements. DISCOVER-AQ involved the deployment of the Geostationary Trace gas and Aerosol Sensor Optimization instrument (GEOTASO, Leitch et al.,2014; Nowlan et al.,2016) and of the Geostationary Coastal and Air Pollution Events (GEO-CAPE) Airborne Simulator (GCAS, Kowalewski and Janz,2014;Nowlan et al.,2018). In Europe, the two AROMAT campaigns, which took place in Romania in September 2014 and August 2015, demonstrated a suite of new instruments such as the Airborne imaging DOAS instrument for Measurements of Atmospheric Pollution (AirMAP, Schönhardt et al.,2015; Meier et al.,2017), the NO₂ sonde (Sluis et al., 2010), and the Small Whiskbroom Imager for atmospheric compositioN monitoring (SWING, Merlaud et al.,2018). Different airborne imagers were intercompared and further characterized during the AROMAPEX campaign in April 2016 (Tack et al., 2019).
In this work, we introduce the measurements performed during the AROMAT campaigns and analyze their relevance for the validation of air quality satellite products. The datasets collected during AROMAT fulfill several requirements of the ideal validation study, as described above. We further investigate the strengths and limitations of the acquired data sets.

The paper is structured as follows: Section 2 describes the two target areas and the deployment strategy. Section 3 characterizes the geophysical fields of the sampled areas and Section 4 presents a critical analysis of the strengths and limitations of the campaign results while elaborating on recommendations for future validation campaigns in Romania. The supplement lists the main instruments operated during the campaigns, gives practical details about their deployment, and presents additional information and measurements.

2 Target areas and deployment strategy

This section presents the two target areas of the AROMAT campaigns, Bucharest and the Jiu Valley. It also lists available studies on air quality at these two sites as well as logistical aspects of relevance.

Figure 1 presents a map of the tropospheric NO$_2$ vertical column densities (VCDs) above Romania, derived from OMI measurements (Levelt et al., 2006) and averaged between 2012 and 2016. The map also indicates the position of the 8 largest cities of the country. Compared to highly polluted areas in western Europe such as northern Belgium or the Netherlands, Romania appears relatively clean at the spatial resolution of the satellite data. There are however two major NO$_2$ sources clearly visible from space, which appear to be of similar magnitude with NO$_2$ columns around 2.5 x 10$^{15}$ molec cm$^{-2}$: the Bucharest area and the Jiu Valley, northwest of Craiova. For the latter, the NO$_2$ enhancement is due to a series of large coal-fired thermal power plants.

2.1 Bucharest

Bucharest (44.4° N, 26.1° E) is the capital and largest city (1.9 million inhabitants according to the 2011 census) of Romania. Within its administrative borders, the city covers an area of 228 km$^2$. Adding the surrounding Ilfov county, the total Bucharest metropolitan area numbers 2.3 million inhabitants in 1.583 km$^2$. The built-up areas are mainly located within a ring road whose diameter is around 20 km.

Iorga et al. (2015) described in detail the Bucharest Greater Area in the context of an extensive study of the air quality in the city between 2005 and 2010. Bucharest is located in a low-altitude plain, with a maximum altitude of 92 m a.s.l. The geographic configuration of the Carpathian Mountains explains the dominant northeast winds.

The NO$_2$ VCDs seen from space above Bucharest appear lower than over western European sites at the resolution of OMI (see Fig.S7 in the Supplement). However, this is partly due to the dilution effect for this relatively small and isolated source. Local studies based on the 8 air quality stations inside the city point out that, regarding local PM and NO$_x$ levels, Bucharest is amongst the most polluted cities in Europe (Alpopi and Colesca, 2010; Iorga et al., 2015). The city center is the most heavily polluted, with concentrations of pollutants well above the European thresholds. For instance, the annual mean concentration of NO$_2$ at the traffic stations was about 57 µ g.m$^{-3}$ in 2017 (EEA, 2019). Stefan et al. (2013) have shown the importance of local

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conditions and anthropogenic factors in air quality analysis in areas close to Bucharest, during two weeks of measurements in 2012. lorga et al. (2015) and Grigoraș et al. (2016) showed that the main NO$_x$ contributions came from traffic and production of electricity, spread over about 10 medium-size thermal power plants within the city.

Figure 2 shows the Bucharest metropolitan area and the flight tracks of the two scientific aircraft used during AROMAT-2 (the FUB Cessna-207 and the INCAS BN-2). Note that the BN-2 tracks are actually a good indication of the Bucharest ring road. We were not allowed to cross the ring road with the BN-2, except in the North of the city. The figure also pinpoints important locations for the AROMAT campaigns. The FUB Cessna took-off and landed at the Baneasa international airport, located 8 km north of Bucharest city center (44.502° N, 26.101° E). The INCAS BN-2 also used Baneasa airport during AROMAT-2, but this plane was mainly based at the Strejnicu airfield (44.924° N, 25.964° E), which lies 60 km north of Bucharest, near Ploiesti. The UAV operations in Bucharest during AROMAT-1 were performed at Clinceni airfield (44.359° N, 25.931° E). The latter is located in the southwest of the city, 7 km west of the INOE observatory in Magurele (44.348°N, 26.031°E).

2.2 The Jiu Valley between Targu Jiu and Craiova

The second NO$_2$ plume in Fig. 1 lies around 250 km west of Bucharest. It corresponds to a series of four thermal power plants located along the Jiu river between the cities of Targu Jiu (82,000 inhabitants, 45.03°N, 23.27°E) and Craiova (269,000 inhabitants, 44.31°N, 23.8°E). These plants were built in this area due to the presence of lignite (brown coal), which is burned to produce electricity.

The altitude of the valley ranges from 268 m a.s.l. in Targu Jiu to 90 m in Craiova. The valley is surrounded by moderately elevated hills (400 m a.s.l.). Due to the orography, the prevailing wind directions is from southwest to southeast.

Beside NO$_2$, the SO$_2$ emissions from these plants are also visible from space, as first reported by Eisinger and Burrows (1998) using GOME data. Since 2011, the OMI-derived trends above the area indicate that the emissions of SO$_2$ have been decreasing, while those of NO$_2$ are stable (Krotkov et al., 2016). This is related to the installation of flue gas desulfurization (FGD) systems, which was part of environmental regulations imposed on Romania following its entry in the European Union in 2007.

Figure 3 presents a map of the Jiu Valley area with the four power plants. The map also shows the tracks of the two airborne platforms (the FUB Cessna and an ultralight operated by UGAL) operated in this area during AROMAT-2. Table 3 presents the geographical positions, nominal capacities, and smokestack heights of the four power plants. From north to south, the plants are named according to their locations: Rovinari, Turceni, Isalnita and Craiova II.

During the AROMAT campaigns, we focused in particular on the emissions of the Turceni power plant (44.67°N, 23.41°E). With a nominal capacity of 1650 MW, it is the largest electricity producer in Romania. The Turceni power plant is located in a rural area, 2 km ESE of the village of Turceni. The plant emits aerosols, NO$\times$, and SO$$_2$$ from the 280 m high smokestacks.

Scientific studies on air quality inside the Jiu Valley are sparse. Previous measurements performed by INOE during a campaign in Rovinari in 2010 indicated elevated volume mixing ratios of NO$_2$ (up to 30 ppb) and of SO$_2$ (up to 213 ppb) (Nisulescu et al., 2011; Marmureanu et al., 2013). The maximum ground concentrations occurred in the morning, before the planetary
boundary layer development. Mobile-DOAS observations performed in 2013 revealed columns of NO$_2$ up to $1 \times 10^{17}$ molec cm$^{-2}$ (Constantin et al., 2015).

2.3 Groups, instruments, and platforms

The AROMAT consortium consisted of research teams from Belgium (BIRA-IASB), Germany (IUP-Bremen, FUB, MPIC), The Netherlands (KNMI), Romania (University “Dunarea de Jos” of Galati, hereafter UGAL, National Institute of R&D for Optoelectronics, hereafter INOE, and National Institute for Aerospace Research “Elie Carafoli”, hereafter INCAS), and Norway (NILU). The AROMAT consortium had a common focus on measuring the tropospheric composition using various techniques.

The supplement presents the main atmospheric instruments operated during the two campaigns, classified into airborne, ground-based, remote sensing, and in-situ sensors. The primary target species during AROMAT-1 were NO$_2$ and aerosols while the observation capacities expanded in AROMAT-2 through the improvements of the AirMAP and SWING sensors for SO$_2$ measurements and the deployments of other instruments such as SO$_2$ cameras, DOAS instruments targeted to H$_2$CO, and a PICARRO instrument to measure water vapor, methane, CO, and CO$_2$.

We used two small tropospheric aircraft: the Cessna-207 from FUB, and the Britten-Norman Islander (BN-2) from INCAS. The Cessna was dedicated to remote sensing. It mainly performed mapping flights at 3 km a.s.l. for the airborne imagers, while parts of the ascents and descents were used to measure aerosol extinction profiles with the FUBISS-ASA2 instrument. The BN-2, which was only used during AROMAT-2, was dedicated to in-situ measurements around Bucharest between surface and 3000 m a.s.l.. In AROMAT-2, there was also an ultralight aircraft used by UGAL for nadir-DOAS observations in the Jiu Valley. The ultralight aircraft typically flew between 600 and 1800 m a.s.l. Two UAVs, operated by INCAS and UGAL flew during AROMAT-1. These measurements were not repeated during AROMAT-2 since the coverage of the UAVs was too limited, both in horizontal and vertical direction. Finally, we also launched balloons carrying NO$_2$ sondes from Turceni and performed Mobile-DOAS measurements from several cars during both campaigns. The supplement provides more details about the practical deployments during the campaigns.

3 Geophysical results

This section presents selected findings related to the atmospheric structure and the geophysical fields of trace gases and aerosols in the two target areas. The Supplement gives details of the instruments involved in these observations. Tables S6 and S7 of the Supplement summarize the observed ranges of the main geophysical quantities for the two campaigns.

3.1 Bucharest

3.1.1 Aerosol optical properties

Figure 4 shows the lidar extinction profiles at 532 nm measured at INOE in Margurele (44.35° N, 26.03° E) during two days of AROMAT-2: 30 (red), and 31 (blue) August 2015. We retrieved these daytime extinction profiles from the backscatter profiles
using a column-averaged lidar ratio constrained with the AOD from collocated sunphotometer measurements. The extinction was larger on 30 August 2015. It reached 0.21 km$^{-1}$ in the boundary layer and up to 0.32 km$^{-1}$ in an elevated layer at 2 km altitude. A similar elevated layer also appeared in the FUBISS data (Meier et al., 2017, Fig.7).

During AROMAT-2, the AOD at 532 nm was around 0.2 most of the days, except on 30 August 2015 (0.35) and 1 September 2015 (0.05). Interestingly, the maximum AOD was seen on a Sunday, when the anthropogenic emissions of NO$_x$ from Bucharest were minimal, as indicated by Fig. 7, which compares the NO$_2$ above the city on Sunday 30 August and Monday 31 August 2015.

### 3.1.2 Boundary layer height

We estimated the boundary layer height (BLH) using a gradient method applied to the lidar range corrected signal (Belegante et al., 2014; Timofte et al., 2015). Figure 5 presents such BLH estimates using the RALI lidar observations during AROMAT-2 in Bucharest. The BLH varied between 700 m (morning of the 30 August 2015) and 2500 m (afternoon of the 24, 26, and 31 August 2015). During AROMAT-1, similar estimates resulted in BLH ranging between 1300 m (2 September 2014) and 2200 m (11 September 2014).

Figure 6 presents the PICARRO measurements of CO, CH$_4$, H$_2$O, and CO$_2$ volume mixing ratios (vmr) during two soundings of the BN-2: (i) between 11:24 and 11:41 UTC on 30 August 2015 (ii) between 12:20 and 12:39 UTC on 31 August 2015. All the species exhibit clear discontinuities in their vertical distribution at the altitude of the lidar-derived BLH: 800 m on 30 August 2015 and 1200 m a.s.l on 31 August 2015.

Except for CH$_4$ on 31 August 2015, the vmrs were relatively stable in the boundary layer, and rapidly decreased above. Both soundings exhibit more polluted layers aloft, probably linked with long-range transport. These vertical distributions give confidence in the lidar-derived BLH, under which convection and turbulences homogenize the distribution of aerosols and trace gases.

Unfortunately, the BN-2 did not vertically sample the exhaust plume of Bucharest due to flight restrictions. We thus have little access to the vertical distributions of the aerosols and NO$_2$ in the Bucharest plume.

### 3.1.3 Horizontal distribution of NO$_2$

Figure 7 presents two maps of the AROMAT NO$_2$ measurements performed with the AirMAP, CAPS, and MPIC mobile-DOAS instruments above Bucharest, on 30 (Sunday afternoon, left panel) and 31 (Monday afternoon, right panel) August 2015. AirMAP is a remote sensing instrument that mapped the NO$_2$ VCDs from the Cessna at 3 km a.s.l. and produced the continuous map. The CAPS is an in-situ instrument, it was operated on the BN-2 and sampled the air at 300 m a.s.l. and performed vertical soundings above Magurele. The MPIC Mobile-DOAS mainly drove along the Bucharest ring road.

The different datasets of Fig. 7 appear consistent. First, they reveal large differences of NO$_2$ amounts on Sunday 30 August 2015 compared to Monday 31 August 2015. On Sunday afternoon, the NO$_2$ VCDs peak around $1.5 \times 10^{16}$ molec cm$^{-2}$. On Monday, the NO$_2$ plume spread from the center to the northeast of the city. The observed NO$_2$ VCDs were smaller than the detection limit upwind and reach up to $3.5 \times 10^{16}$ molec cm$^{-2}$ inside the plume. The NO$_2$ vmr measured with the CAPS
was close to the detection limit on Sunday while it reached 5 ppb inside the plume on Monday 31 August 2015. Note that the systematic differences between AirMAP and the MPIC Mobile-DOAS at the eastern part of the ring road on 31 August 2015 were due to the time differences between both measurements. Meier (2018) compared the two instruments during the morning flights, which include more simultaneous observations. The comparison reveals a good agreement when averaging the forward and backward-looking Mobile-DOAS NO$_2$ VCDs, with a MPIC/AirMAP slope of 0.93 and a correlation coefficient of 0.94. Figure S6 in the Supplement presents this comparison.

Figure 8 presents collocated CAPS and AirMAP NO$_2$ measurements on 31 August 2015. The BN-2 carrying the CAPS flew from Magurele to the East of Bucharest, remaining outside the city ring at 300 m a.s.l. between 12:30 and 12:55 UTC, while AirMAP onboard the Cessna was mapping the city. We extracted the AirMAP NO$_2$ VCDs at the position of the CAPS observations. The figure confirms that the two instruments detected the plume at the same place. This suggests that along this portion of the flight, which was inside the plume but outside the city, the NO$_2$ vmr measured at 300 m a.s.l. may be used as a proxy for the NO$_2$ VCD. The BLH was about 1500m (Fig. 5) during these observations. Assuming a constant NO$_2$ vmr of 3.5 ppb leads to a NO$_2$ VCD of $1.4 \times 10^{16}$ molec cm$^{-2}$. This estimation is close to the AirMAP NO$_2$ VCD observed in the plume (Fig. 8). Future campaigns should include vertical soundings inside the plume to further investigate its NO$_2$ vertical distribution.

### 3.1.4 Horizontal distribution of H$_2$CO

Figure 9 shows the H$_2$CO and NO$_2$ VCDs measurements from the Avantes spectrometer operated onboard the Cessna on 31 August 2015 (morning flight), together with the time-coincident MPIC Mobile-DOAS measurements. The H$_2$CO VCDs range between $1 \pm 0.25 \times 10^{16}$ molec cm$^{-2}$ and $7.5 \pm 2 \times 10^{16}$ molec cm$^{-2}$, a maximum observed inside the city. We estimated the H$_2$CO reference column for the airborne data using the Mobile-DOAS measurements. Both NO$_2$ and H$_2$CO are in good agreement when comparing their distributions as seen from the airborne and ground-based instruments. However, if the highest H$_2$CO VCDs are found above the Bucharest city center, they are not coincident with the NO$_2$ maximum, as can be seen comparing the upper and lower panels of Fig. 9, for instance on the second Cessna flight line from the north.

The H$_2$CO hotspot observed above Bucharest is mainly anthropogenic. Indeed, biogenic emissions typically account for 1 to $2 \times 10^{16}$ molec cm$^{-2}$ (J.-F. Müller, personal communication), in agreement with the background VCDs measured by the Mobile-DOAS along the Bucharest ring. During the measurements, the wind was blowing from south and west. The difference between NO$_2$ and H$_2$CO spatial patterns may be explained by the different sources of NO$_x$ compared to H$_2$CO or by the formation time of H$_2$CO through the oxidation of VOCs.

Anthropogenic hotspots of H$_2$CO have already been observed, e.g. above Houston (Texas), an urban area which includes significant emissions from transport and petrochemical industry (Parrish et al., 2012; Nowlan et al., 2018). Nowlan et al. also deployed an airborne DOAS nadir instrument, they reported H$_2$CO VCDs up to $5 \times 10^{16}$ molec cm$^{-2}$ in September 2013.
3.2 The Jiu Valley

3.2.1 Spatial distribution of NO$_2$

Figure 10 presents the horizontal distribution of the NO$_2$ VCDs in the Jiu Valley measured with the MPIC Mobile-DOAS on 23 August 2015. The figure shows elevated NO$_2$ VCDs close to the four power plants listed in Table 3, with up to $8 \times 10^{16}$ molec cm$^{-2}$ downwind of Turceni and Rovinari. In comparison, the area East of Craiova is very clean, with typical NO$_2$ VCDs under $1 \times 10^{15}$ molec cm$^{-2}$.

The situation of Fig. 10 is characteristic of the conditions encountered in the Jiu Valley, with high NO$_2$ VCDs observed north and west of the plants due to the prevailing wind directions. During both campaigns, we observed maximum NO$_2$ VCDs reaching up to $1.3 \times 10^{17}$ molec cm$^{-2}$ close to the plants with Mobile-DOAS instruments.

Figure S3 in the Supplement shows the AirMAP and SWING NO$_2$ VCDs measured around the Turceni power plant on 28 August 2015. The two airborne instruments largely agree, detecting NO$_2$ VCDs up to $8 \times 10^{16}$ molec cm$^{-2}$ in the exhaust plume of the power plant. Figure S4 in the Supplement (upper panel) extracts the AirMAP and SWING NO$_2$ VCDs along the path of the simultaneous ground-based Mobile-DOAS measurements and compares the three datasets. This comparison confirms that there is a good agreement for the airborne instruments but indicates that comparing with Mobile-DOAS instruments is less straightforward. When observed with the Mobile-DOAS, the plume shows higher NO$_2$ VCDs and appears narrower than with the airborne instruments. This is partly related to air mass factor uncertainties, but probably also to 3-D effects as the plume is very thin and heterogeneous close the power plants, as discussed in Merlaud et al. (2018).

Figure 11 shows those AROMAT-1 NO$_2$ sonde measurements above Turceni which detected the plume. The NO$_2$ is not well-mixed in the boundary layer, with maxima aloft and lower vmrs close to the surface. This is understandable so close to the source, as high-temperature NO$_x$ is emitted from the 280 m high stack. In these balloon-borne datasets, the observed maximum NO$_2$ vmr is about 60 ppb inside the plume, and the NO$_2$ vmr vanishes above 1200 m a.s.l.. These results suggest that airborne measurements with the ULM-DOAS, which can fly safely at 1500 m a.s.l., can provide reliable measurements of the integrated column amount inside the plume. Note that we measured NO$_2$ vmrs of up to 95 ppb on the ground at the soccer field (see Fig. S11 in the Supplement) when the wind was blowing from the power plant.

3.2.2 Horizontal distribution of SO$_2$

Figure 12 presents the SO$_2$ horizontal distributions measured around Turceni with AirMAP (left panel) and SWING (right panel) on 28 August 2015. The maps show the plume from the Turceni plant transported in the northwest direction, and other areas with elevated SO$_2$ VCDs in the east and south of Turceni. Meier (2018) presents in detail these AirMAP SO$_2$ observations and compares them with SWING results. It is found that the AirMAP-derived SO$_2$ columns inside the plume SO$_2$ reach $6 \times 10^{17}$ molec cm$^{-2}$ and that the AirMAP and SWING SO$_2$ VCDs agree within 10%. Moreover, for these airborne data, the SO$_2$ horizontal distribution broadly follows that of NO$_2$. The discrepancies can be explained by the different lifetimes of the two species.
We have measured higher SO₂ levels during AROMAT-2 than during AROMAT-1, both in terms of VCDs and vmrs, as shown in Fig. S10 of the supplement material. This corresponds to a shutdown of the desulfurization unit of the power plant, which local workers reported during the campaign. On 26 August 2015 in particular, the SO₂ vmr went up to 250 ppb, above the WMO guidelines of 191 ppb for the 10 minutes mean, while it never exceeded 50 ppb in 2014.

As for NO₂, it appears difficult to quantitatively relate the airborne and Mobile-DOAS SO₂ VCDs observations in the close vicinity of the power plant. As shown in Fig. S4 of the supplemental material (lower panel), the maximum SO₂ VCD measured from the ground on the road close to the factory amounts to $1.3 \times 10^{18}$ molec cm$^{-2}$ while from the aircraft, the SO₂ VCD reached $8 \times 10^{17}$ molec cm$^{-2}$. Part of this difference can be explained by 3-D effects on the radiative transfer, as for NO₂. As discussed below, it seems easier to compare the SO₂ flux.

4 Discussion

This section addresses the lessons learned from our study for the validation of satellite observations of the three investigated tropospheric trace gases, namely NO₂, SO₂, and H₂CO. For each molecule, we discuss the added value of conducting such an airborne campaign as well as the choice of Romania as a campaign site. The last part of the section estimates the NOₓ and SO₂ emissions from Bucharest and from the power plants of the Jiu Valley, using the different datasets of the campaigns.

4.1 Lessons learned for the validation of space-borne NO₂ VCDs

4.1.1 Number of possible pixels and dynamic range at the TROPOMI resolution

Regarding Bucharest, the mapped area of Fig. 7 virtually covers 43 TROPOMI near-nadir pixels. Averaging the high spatial resolution AirMAP NO₂ VCDs within these 43 hypothetical TROPOMI measurements reduces the dynamic range of the observed NO₂ field. The latter decreases from $3.5 \times 10^{16}$ to $2.6 \times 10^{16}$ molec cm$^{-2}$ (37 $\sigma$ where $\sigma$ is the required precision on the tropospheric NO₂ VCD). Nevertheless, 33 of the 43 hypothetical TROPOMI pixels exhibits a NO₂ VCD above the required 2-$\sigma$ random error for TROPOMI ($1.4 \times 10^{15}$ molec cm$^{-2}$).

Regarding the Jiu Valley, a similar exercise based on our measurements on 28 August 2012 (Fig.S3 in the Supplement) leads to 48 near-nadir TROPOMI pixels, out of which 35 would have a NO₂ VCD above the 2-$\sigma$ TROPOMI error. The largest NO₂ tropospheric VCD seen by TROPOMI would be around $2 \times 10^{16}$ molec cm$^{-2}$ (29 $\sigma$ for TROPOMI).

4.1.2 Characterization of the reference measurements

Table 4 summarizes the NO₂ observations during the AROMAT campaigns. For each instrument, the table indicates the measured range of NO₂ VCDs (or vmr), the ground sampling distance and a typical detection limit and bias. Regarding DOAS instruments, we estimated the detection limits on the NO₂ VCDs from typical 1-$\sigma$ DOAS fit uncertainties divided by typical air mass factors (AMF). Table S1 in the Supplement presents these typical AMFs and detection limits. The 1-$\sigma$ DOAS fit uncertainty is instrument specific and an output of the DOAS fitting algorithms. The AMF depends on the observation’s geometry,
atmospheric and surface optical properties. Uncertainties on the AMF usually dominate the systematic part of the error for the DOAS measurements. Therefore, for these instruments, the bias given in Table 4 corresponds to the uncertainty in their associated AMF.

Combined with the ground sampling distance, the detection limit enables one to quantify the random uncertainty of a reference observation at the satellite horizontal resolution. Indeed, considering reference measurements averaged within a satellite pixel, the random error associated with the averaged reference measurements decreases with the square root of the number of measurements, following Poisson statistics. For instance, a continuous mapping performed with SWING at a spatial resolution of 300 x 300 m² inside a TROPOMI pixel of 3.5 x 5.5 km² would lead to 214 SWING pixels. Averaging the NO₂ VCDs of these SWING pixels would divide the SWING original uncertainty (1.2x10^{15} molec.cm^{-2}) by \sqrt{214}, leading to 8.2x10^{13} molec.cm^{-2}, about one tenth of the random error of TROPOMI (7x10^{14} molec.cm^{-2}) given in table 2.

However, the temporal variation of the NO₂ VCDs further adds uncertainty to the reference measurements when comparing them with satellite data. The validation areas typically extend over a few tens of kilometers. At this scale, satellite observations are a snapshot in time of the atmospheric state, while an airborne mapping typically takes one or two hours.

We investigated the temporal variation of the NO₂ VCDs comparing consecutive AirMAP overpasses above Bucharest from the morning flight of 31 August 2015. During this flight, the Cessna covered the same area three times in a row between 07:06 and 08:52 UTC. For each AirMAP overpass, we averaged the NO₂ VCDs at the horizontal resolution of TROPOMI (see previous section). The standard deviation of the differences between two averaged overpasses then indicates the random part of the NO₂ VCDs temporal variation during an aircraft overpass. This standard deviation is 3.7x10^{15} and 4.2x10^{15} molec cm^{-2}, respectively between the first and second, and second and third overpass. Hereafter, we used 4x10^{15} molec cm^{-2} as random error due to the temporal variation. Figure S10 in the Supplement illustrates these investigations on the temporal variation of the NO₂ VCDs at the TROPOMI resolution.

Clearly, the NO₂ VCD temporal variation depends on characteristics of a given validation experiments, such as the source locations and the wind conditions during the measurements. In the studied case however, this error source is larger for the reference measurements than the TROPOMI precision (7x10^{14} molec cm^{-2}). This is quite different from using static MAX-DOAS as reference. The latter are usually averaged within one hour around the satellite overpass. Compenolle et al. (paper in preparation) quantify the temporal error for MAX-DOAS NO₂ VCDs, typically ranging between 1 to 5x10^{14} molec cm^{-2}. Whether the reference is based on airborne or ground-based measurements, underestimating its temporal random error propagates in underestimating the slope between reference and satellite observations. For MAX-DOAS, this also happens when averaging the NO₂ VCDs within larger time windows around the satellite overpass (Wang et al., 2017).

4.1.3 Simulations of validation exercises in different scenarios

We simulated TROPOMI Cal/Val exercises with the spatially averaged AirMAP observations. We considered these averaged AirMAP NO₂ VCDs as the ground truth in simulated TROPOMI pixels, on which we added Gaussian noise to build synthetic satellite and reference NO₂ VCDs datasets. For the synthetic satellite observations, the noise standard deviation corresponded to the TROPOMI random error (the precision in Table 2). For the synthetic airborne observations, we added in quadrature
the aforementioned averaged airborne shot noise (e.g. $7 \times 10^{13}$ molec cm$^{-2}$ for SWING) and temporal error ($4 \times 10^{15}$ molec cm$^{-2}$, which we assumed to be also realistic around Turceni). We then applied orthogonal distance regressions to a series of such simulations to estimate the uncertainty on the regression slope. This led to slope uncertainties of about 6% and 10% in Bucharest and Turceni, respectively.

In a real-world validation experiment, this regression slope would quantify the combined biases of the two NO$_2$ VCDs datasets (satellite and reference). These biases mainly originate from errors in the AMFs, resulting in particular from uncertainties on the NO$_2$ and aerosol profiles, and on the surface albedo. To some extent, these quantities can be measured from an aircraft with the type of instrumentation deployed in the AROMAT activity. The ground albedo can be retrieved with the DOAS instruments by normalizing uncalibrated airborne radiances to a reference area with known albedo (Meier et al., 2017) or by using a radiometrically calibrated DOAS sensor (Tack et al., 2019). The NO$_2$ and aerosol profiles can be measured with in-situ instruments such as a CAPS NO$_2$ monitor and a nephelometer. For legal reasons, vertical soundings are difficult above cities. One can measure the NO$_2$ and aerosol profile further down in the exhaust plume, once the latter is above rural areas. The conditions inside the city can be different and this motivates the deployment of ground-based instruments, e.g. sunphotometers and MAX-DOAS, inside the city.

Regarding uncertainties on the references AMFs, the added value of knowing the aerosol and NO$_2$ profile appears when comparing the AMF error budget for airborne measurements above Bucharest (26%, Meier et al. (2017)) and above the Turceni power plant (10%, Merlaud et al. (2018)). In the latter case, there was accurate information on the local NO$_2$ and aerosol profiles thanks to the lidar and the balloon-borne NO$_2$ sonde, respectively. We used these two AMF uncertainties to estimate a total possible bias between reference and satellite observations.

Table 7 summarizes this section and presents total error budgets for different scenarios of validation exercises using reference airborne mapping to validate spaceborne tropospheric NO$_2$ VCDs. We estimated the random and systematic uncertainties between satellite and reference measurements with SWING and AirMAP, including (or not) profile information on the aerosols and NO$_2$ vmr, and for measurements over Bucharest or Turceni. Note that we considered 25% for the satellite accuracy. The temporal error of the airborne measurements clearly dominates the total random error, making the differences in detection limit between AirMAP and SWING irrelevant for this application. Adding the profile information on the other hand reduces the total multiplicative bias from 37% to 28% or 29% in Bucharest and Turceni. This quantifies the capabilities of such airborne measurements for the validation of the imaging capabilities of TROPOMI regarding the NO$_2$ VCDs above Bucharest and the Jiu Valley.

### 4.2 Lessons learned for the validation of space-borne H$_2$CO VCDs

Table 5 is similar to Table 4 but for H$_2$CO, which we only measured in significant amounts in and around Bucharest.

The background level of the H$_2$CO VCD around the city is around $1 \times 10^{16}$ molec cm$^{-2}$ and the anthropogenic increase in the city center is up to $7 \times 10^{16}$ molec cm$^{-2}$ (Fig. 9). The background falls within the TROPOMI H$_2$CO spread ($1.2 \times 10^{16}$ molec cm$^{-2}$), and Fig. 9 indicates that the extent of the urban hotspot only corresponds to a few TROPOMI pixels, with a maximum at 6 $\sigma$. This limits the relevance of individual mapping flights for the validation of H$_2$CO. The information on the
H$_2$CO horizontal variability is nevertheless useful, as it justifies the installation of a second MAX-DOAS in the city center, in addition to background measurements outside the city. Indeed, long-term ground-based measurements at two sites would be useful to investigate seasonal variations of H$_2$CO. Averaging the H$_2$CO over a season would reduce the random errors of the satellite measurements and it could reveal the horizontal variability of H$_2$CO from space. The H$_2$CO hotspot around Bucharest seems to be visible in the TROPOMI data of summer 2018 (I. De Smedt, personal communication).

Getting information on the profile of H$_2$CO during an airborne campaign may also help to understand the differences between ground-based and space-borne observations. This could be done by adding to the BN-2 instrumental set-up an in-situ H$_2$CO sensor such as the In Situ Airborne Formaldehyde instrument (ISAF, Cazorla et al. (2015)) or the COmpact Formaldehyde Fluorescence Experiment (COFFEE, St. Clair et al. (2017)).

### 4.3 Lessons learned for the validation of space-borne SO$_2$ VCDs

Table 6 is similar to Table 4 but for SO$_2$, which we only measured in significant amounts in the Jiu Valley. The higher bias of the airborne measurements for SO$_2$ compared to NO$_2$ is due to the albedo. The latter is lower in the UV where we retrieve SO$_2$, which leads, for the same albedo error, to a larger AMF uncertainty (e.g. Merlaud et al., 2018, Fig.10).

Averaging the SO$_2$ VCDs from the airborne mapping of Fig. 12 at the TROPOMI resolution leads to 30 near nadir TROPOMI pixels above a 2-$\sigma$ error of 5.4x10$^{16}$ molec cm$^{-2}$. The maximum SO$_2$ tropospheric VCD seen by TROPOMI would be 2.4x10$^{17}$ molec cm$^{-2}$ (7 $\sigma$). This tends to indicate that airborne mappings of SO$_2$ VCDs above large power plants could help to validate the horizontal variability of the SO$_2$ VCDs measured from space, to a limited extent in the AROMAT conditions due to the small dynamic range (7 $\sigma$).

However, it would be difficult to quantify the bias of the satellite SO$_2$ VCD with AROMAT-type of airborne measurements. Adding in quadrature the biases of the SO$_2$ VCDs for airborne measurements (40%, Table 6) and for TROPOMI (30%, Table 2) already leads to a combined uncertainty of 50%, without considering any temporal variation or regression error. This best-case scenario is already at the upper limit of the TROPOMI requirements for tropospheric SO$_2$ VCDs (Table 2).

As for H$_2$CO, the validation of the satellite-based SO$_2$ measurements should thus rely on ground-based measurements, enabling to improve the signal-to-noise ratio of the satellite and reference measurements by averaging their time series. An additional difficulty for validating SO$_2$ VCDs emitted by a power plant arise from the spatial heterogeneity of the SO$_2$ field around the point source, which renders ground-based VCDs measurements complicated. On the other hand, Fioletov et al. (2017) presented a method to derive the SO$_2$ emissions from OMI data and validated it against reported emissions. The SO$_2$ fluxes can be measured locally in several ways and we tested some of them during AROMAT-2 (see Sect. 4.4.2 below). To validate satellite-derived SO$_2$ products in Europe, it thus seems possible to compare satellite and ground-based reference SO$_2$ fluxes. Theys et al. (2019) already validated TROPOMI-derived volcanic SO$_2$ fluxes against ground-based measurements. In this context, a SO$_2$ camera pointing to the plant stack would be a valuable tool since it could be permanently installed and automated. One advantage of such a camera compared to the other tested remote-sensing instruments, beside its low operating cost, is that it derives the extraction speed from the measurements, avoiding dependence on low-resolution wind information.
Note that since the desulfurization unit of the Turceni power plant was not fully operational during AROMAT-2, the SO$_2$ VCDs detected above Turceni on 28 August 2015 are expectedly higher than for standard conditions, all the more so as the SO$_2$ VCDs in the area seem to have further decreased (D. Constantin, personal communication). The first reported TROPOMI SO$_2$ measurements above the area pinpoint other power plants in Serbia, Bosnia–Herzegovina, and Bulgaria (Fioletov et al., 2020). For validation studies, it would be worth to install automatic SO$_2$ cameras around these plants, until they are equipped with FGD units.

4.4 Emissions of NO$_x$ and SO$_2$ emissions from Bucharest and the Jiu Valley

4.4.1 NO$_x$ flux from Bucharest

We estimated NO$_x$ fluxes from the Bucharest urban area using the NO$_2$ VCDs measured with the Mobile-DOAS systems along the external ring and the wind data on 8 September 2014 and 31 August 2015. We derived the wind direction from the maxima of the NO$_2$ VCDs in the DOAS observations. For the wind speed, we took 1.1 m s$^{-1}$ on 8 September 2014, the value Meier (2018) used for the AirMAP-derived flux, which originates from meteorological measurements at Baneasa airport. On 31 August 2015, we used the ERA5 wind data (C3S, 2017) at the time when the Mobile-DOAS crossed the NO$_2$ plume (15:00 UTC). The ERA5 database indicates a constant windspeed between 1000 and 900 hPA of 1.2 m s$^{-1}$. Finally and similarly to Meier (2018), we took a ratio of 1.32 for the NO$_x$ to NO$_2$ ratio and estimated the chemical loss of NO$_x$ with a lifetime of 3.8h and an effective source location in the center of Bucharest.

Table 8 presents the AirMAP and Mobile-DOAS derived NO$_x$ fluxes from Bucharest, ranging between 12.5 and 17.5 mol s$^{-1}$. On 8 September 2014, the Mobile and airborne observations were coincident. Their estimated NO$_x$ fluxes agree within 20%. This gives confidence in the flux estimation yet one should keep in mind that the same wind data was used for both estimations. Meier (2018) estimated the uncertainties on the AirMAP-derived NO$_x$ flux to be around 63%, while the uncertainty of Mobile-DOAS derived NO$_x$ flux typically range between 30% and 50% (Shaiganfar et al., 2017).

We compared our measured NO$_x$ fluxes with the European Monitoring and Evaluation Programme inventory (EMEP, https://www.ceip.at/). In practice, we summed the EMEP gridded yearly NO$_x$ emissions between 44.2$^\circ$ and 44.6$^\circ$N and between 25.9$^\circ$E and 26.3$^\circ$E and we assumed the emissions are constant during one year. This led to NO$_x$ emissions of 6.14 and 6.33 mol s$^{-1}$ for 2014 and 2015. Studying the reported emissions from several European cities including Bucharest, Trombetti et al. (2018) mentions that the EMEP emissions are well below other inventories for all the pollutants. We thus also compared our flux with the Emissions Database for Global Atmospheric Research (EDGAR v4.3.2, Crippa et al. (2018)), which is only available until 2012. The same method led to a NO$_x$ flux of 18.4 mol s$^{-1}$, to compared with the 2012 EMEP NO$_x$ emissions of 7.1 mol s$^{-1}$. Based on summer measurements, the AROMAT-derived NO$_x$ emissions do not include residential heating. The latter ranges between 10 and 40% of the total NO$_x$ according to Trombetti et al. (2018). This tends to confirm that the EMEP inventory underestimates the NO$_x$ emissions for Bucharest.
4.4.2 NO\textsubscript{x} and SO\textsubscript{2} fluxes from the power plants in the Jiu Valley

We estimated the NO\textsubscript{x} and SO\textsubscript{2} flux from the power plants using several instruments: a Mobile-DOAS, the ULM-DOAS, and the SO\textsubscript{2} camera. For the DOAS instruments, we inferred the wind direction from the plume position and we retrieved the wind speed from the ERA5 database. Considering the observed vertical extent of the plume downwind of Turceni (Fig. 11), we took the wind speed at 950 hPa (ca. 500 m a.s.l.).

Figure 14 presents the ULM-DOAS-estimated fluxes of NO\textsubscript{x} and SO\textsubscript{2} from the power plants in Turceni, Rovinari, and Craiova for the flight on 26 August 2015. The figure also shows the reported emissions from the European Environment Agency (EEA) large combustion plants database (EEA, 2018), assuming constant emissions throughout the year. Turceni appears to be the largest SO\textsubscript{2} source (78 mol s\textsuperscript{-1}), while Rovinari is the largest NO\textsubscript{x} source (8 mol s\textsuperscript{-1}).

It is difficult to interpret the discrepancies between those measured fluxes and the yearly reported emissions since we observed large variations in the instantaneous emissions with the SO\textsubscript{2} camera (see below and Fig. 13). However, the ratio of the two fluxes appears interesting since we can assume its relative stability. This ratio for a given power plant depends on whether or not a desulfurization unit is operational at the plant. On Fig. 14, Turceni appears to have both the largest measured ratio and the largest discrepancy between the measured and reported ratios. This is consistent with a temporary shutdown of the desulfurization unit of the Turceni power plant, as was reported by the plant workers during the campaign. The ULM-DOAS measurements on 25 August 2015 (shown in Fig.S14 the Supplement), which also sampled the Isalnita plume, indicates that the desulfurization unit may not have been operational there either. The ULM-DOAS measurements on 25 August 2015, which covered the four power plants, enable to estimate total NO\textsubscript{x} and SO\textsubscript{2} fluxes to be about 22 and 147 mol s\textsuperscript{-1}, respectively.

Table 9 focuses on the Turceni power plant and lists all estimates of the NO\textsubscript{x} and SO\textsubscript{2} emissions from this source. Meier (2018) estimated the NO\textsubscript{x} flux from the Turceni power plant using the AirMAP measurements of 2014 and 2015. This leads to similar values for the two flights on 11 September 2014 and 28 August 2015, of about 8 mol.s\textsuperscript{-1}. On this second day, the UGAL Mobile-DOAS crossed the plume along the road in front of the power plant. These ground-based measurements lead to a NO\textsubscript{2} flux of 2.2 mol.s\textsuperscript{-1}, much lower than the aforementioned AirMAP-derived value. However, Meier (2018) calculated the latter based on AirMAP measurements at 3.5 km from the source. At shorter distances, the AirMAP estimated NO\textsubscript{2} flux is smaller and closer to the Mobile-DOAS observations. This is probably related to the fact that the NO/NO\textsubscript{2} ratio has not yet reached its steady state value above the road where we performed the Mobile-DOAS observations, which is only around 1 km from the stack. The agreement is better for SO\textsubscript{2} (25 and 32 mol.s\textsuperscript{-1}). On 25 August 2015, we had a coincidence of ULM-DOAS and Mobile-DOAS observations and we observed a similar range of values. This gives us confidence in our estimate of the NO\textsubscript{x} flux from the aircraft but confirms that the nearby road is too close to the plant to estimate a meaningful NO\textsubscript{x} flux from Mobile-DOAS NO\textsubscript{2} observations. Note that the conversion of NO into NO\textsubscript{2} is also visible right above the Turceni stack in the NO\textsubscript{2} imager data of 24 August 2015, as appears in Fig.6 of Dekemper et al. (2016).

Figure 13 presents a time series of the SO\textsubscript{2} emissions from the Turceni power plant between 9:00 and 10:50 UTC on 28 August 2015. We derived SO\textsubscript{2} fluxes at different altitudes above the stack using a UV SO\textsubscript{2} camera which is an updated version of the Envicam2 system, used during the SO\textsubscript{2} camera intercomparison described by Kern et al. (2015). We converted the
measured optical densities to SO₂ column densities using simultaneous measurements with an integrated USB spectrometer (Lübcke et al., 2013). We estimated the stack exit velocity from the SO₂ images, recorded with a time resolution of about 15 seconds, by tracking the spatial features of the plume. Dekemper et al. (2016) used a similar approach to derive the NO₂ flux from NO₂ camera imagery.

The SO₂ fluxes retrieved for transverses at 400 to 700 m vertical distances above the stack agree on average with each other within 20%. Emissions estimated 100 m above the stack are underestimated due to saturation (SO₂ column densities above 2 x 10¹⁸ molec.cm⁻²) and high aerosol concentration close to the exhaust.

The SO₂ emissions show large fluctuations. During the time of our observations they increased from 1 kg.s⁻¹ (15.6 mol.s⁻¹) to around 4 ± 1 kg⁻¹ (62.4 mol.s⁻¹). The images (Fig.S13 in the supplement) also show a second and weaker source that emits SO₂. This is probably the desulfurization unit, which was reported to be turned on again on this day, after the temporary shutdown. Indeed, as appears in Table 9, the SO₂/NO₂ ratio measured from AirMAP is lower than the ones measured from the ULM-DOAS during the previous days, and the same holds true for the Mobile-DOAS measurements.

5 Conclusions

The two AROMAT campaigns took place in Romania in September 2014 and August 2015. These campaigns combined airborne and ground-based atmospheric measurements and focused on air quality-related species (NO₂, SO₂, H₂CO, and aerosols). The AROMAT activity targeted the urban area of Bucharest and the power plants of the Jiu Valley. The campaigns aimed at testing new instruments, measuring the concentrations and emissions of key pollutants in the two areas, and investigating the concept of such campaigns for the validation of air quality satellite-derived products.

We have shown that the airborne mapping of tropospheric NO₂ VCDs above Bucharest is potentially valuable for the validation of current and future nadir-looking satellite instruments. These measurements agree with ground-based measurements and cover a significant part of the dynamic range of the NO₂ tropospheric VCDs at an appropriate signal to noise ratio, enabling to constrain the accuracy of the satellite NO₂ VCDs within 37 or 28%, with and without information on the aerosol and NO₂ profile, respectively. This points out the importance of the profile information to approach the TROPOMI optimal target accuracy for tropospheric NO₂ VCDs (25%).

A unique advantage of airborne mapping is its ability to validate the imaging capabilities of nadir-looking satellites. This feature becomes more important as the satellite horizontal resolutions reaches the suburban scale. Judd et al. (2019) pointed out the difficulty for static ground-based measurements to represent the NO₂ VCDs measured from space in polluted areas, due to the horizontal representativeness error. This error cancels out by mapping the full extent of satellite pixels. The caveat is the temporal error, which can be larger than with static ground-based measurements. We have estimated the temporal error to be about 4 x 10¹⁵ molec cm⁻² in our observations above Bucharest, but it varies with local conditions for a given experiment. This indicates the usefulness of simultaneous ground-based measurements, which may also be useful to estimate the reference NO₂ VCDs in the airborne observations.
We also detected a clear signal of \( \text{H}_2\text{CO} \) in and around Bucharest, with an anthropogenic hotspot in the city center. Due to the lower signal to noise ratio of the spaceborne \( \text{H}_2\text{CO} \) observations, this structure is not visible in daily satellite measurements. We thus propose considering long-term ground-based MAX-DOAS measurements in the city for the validation of \( \text{H}_2\text{CO} \).

In the Jiu Valley, \( \text{NO}_2 \) is clearly visible from both satellite and aircraft, and the VCDs are comparable in magnitude with the signal detected above Bucharest. However, it appears more complicated to quantitatively compare the \( \text{NO}_2 \) VCDs datasets in the thick exhaust plumes of the power plants. These plants also emit \( \text{SO}_2 \) but, as for \( \text{H}_2\text{CO} \), the low signal to noise ratio of satellite measurements reduces the validation relevance of individual airborne measurements. As the \( \text{SO}_2 \) emissions have been drastically reduced with the installation of flue-gas desulfurization units in the Jiu Valley, we propose targeting \( \text{SO}_2 \) emissions from other coal-fired power plants having higher emissions, e.g. in Serbia.

Considering the optimal validation study mentioned in the introduction, the validation relevance of an international airborne campaign is usually limited by its timespan of typically a couple of weeks, imposed in practice by cost considerations. To overcome this limitation, one might consider routine airborne mapping of \( \text{NO}_2 \) VCDs by local aircraft operators, close to a well-equipped ground-based observatory. Such a set-up would reduce the fixed costs of the observations, which could be allocated to flight hours in different seasons. This would combine the advantages of long-term ground-based and airborne measurements. In the longer term, high altitude pseudo-satellites (HAPS) could help to achieve such routine measurements above selected supersites, which would be particularly valuable to validate the observations from geostationary satellites.

**Competing interests.** The authors declare that they have no conflict of interest.

**Author contributions.** AM, LB, D-EC, MDH, ACM, LG, DN, and MVR planned and organised the campaign. All coauthors contributed to the campaign either as participants or during campaign preparation and/or follow up data analysis, including the writing of this manuscript, which was coordinated by AM and MVR with feedback and contributions from all the coauthors.

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References


Figure 1. The tropospheric NO$_2$ VCD field seen from space with the OMI/AURA instrument above Romania (OMNO2d product, averaged for 2012-2016 with Giovanni, NASA GES DISC). The black stars pinpoint the largest cities of Romania.

Figure 2. The Bucharest area with important locations for the AROMAT campaigns: the INOE atmospheric observatory in Magurele, the Baneasa airport, and the Clinceni airfield. Buit-up areas appear in grey. The red and black lines, respectively, show the BN-2 and Cessna flight tracks during AROMAT-2. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.
Figure 3. The Jiu Valley and its four power plants between Targu Jiu and Craiova. The scientific crew was based in Turceni during the AROMAT campaigns. The green and red lines, respectively, show the ultralight and Cessna flight tracks during AROMAT-2. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

Figure 4. RALI extinction profiles at 532 nm above Magurele during two days of AROMAT-2. The different lines correspond to the different lidar measurements between 07:33 and 12:32 UTC (30 August 2015) and 07:29 and 15:01 UTC (31 August 2015).
Figure 5. Lidar-derived boundary layer heights above Magurele in August 2015 during AROMAT-2.
Figure 6. Vertical distributions of gases measured from the BN-2 during AROMAT-2 with the PICARRO. The upper panels shows the sounding at 08:30 UTC on 30 August 2015, the lower panel shows the sounding at 12:30 UTC on 31 August 2015.
Figure 7. Measurements of NO$_2$ VCDs and volume mixing ratios in Bucharest on 30 (Sunday) and 31 (Monday) August 2015 with AirMAP (continuous map), the CAPS (black-rimmed circles), and the MPIC Mobile-DOAS (plain colour circles). © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

Figure 8. Volume mixing ratio and VCDs of NO$_2$ in and out of the pollution plume of Bucharest, as measured with the CAPS (on the BN-2) and AirMAP (on the Cessna) during the afternoon flights on 31 August 2015. Note that the two measurements are not exactly time coincident and that the plot shows the VCDs extracted at the position of the CAPS measurements.
Figure 9. Horizontal distribution of tropospheric H$_2$CO and NO$_2$ VCDs measured on 31 August 2015 with the IUP-UB nadir-only compact spectrometer from the Cessna (flight tracks) and with the MPIC Mobile-DOAS (coloured circles). © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.

Figure 10. Tropospheric vertical column densities of NO$_2$ measured with the MPIC Mobile-DOAS instruments in the Jiu Valley on 23 August 2015. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.
Figure 11. Examples of NO$_2$ sondes data from Turceni during AROMAT-1 (11 and 12 September 2014).

Figure 12. AirMAP (left panel) and SWING (right panel) measurements of SO$_2$ from the Cessna above the Jiu Valley on 28 August 2015. © OpenStreetMap contributors 2019. Distributed under a Creative Commons BY-SA License.
Figure 13. $\text{SO}_2$ fluxes from the Turceni power plant on 28 August 2015. They were estimated with the Envicam2 $\text{SO}_2$ camera for 4 transverses at vertical altitudes above the stack of 100, 400, 500 and 700 m. The red line shows the estimated plume speed (m.s$^{-1}$).

Figure 14. $\text{SO}_2$ and $\text{NO}_x$ fluxes from three power plants of the Jiu Valley as (1) measured with the ULMDOAS on 26 August 2015 (green bars) and (2) estimated from the reported emissions of 2015 assuming constant emissions throughout the year (blue bars). Uncertainties on the ULMDOAS-derived fluxes are around 60%.
Table 1. Past and near-future space missions focused on air quality: coverage, pixel size, and temporal sampling.

<table>
<thead>
<tr>
<th>Launch year</th>
<th>Instrument</th>
<th>Pixel size at nadir (km$^2$)</th>
<th>Coverage</th>
<th>Revisit time</th>
</tr>
</thead>
<tbody>
<tr>
<td>1995</td>
<td>GOME</td>
<td>320 x 40</td>
<td>Global</td>
<td>3 days</td>
</tr>
<tr>
<td>2002</td>
<td>SCIAMACHY</td>
<td>60 x 30</td>
<td>Global</td>
<td>6 days</td>
</tr>
<tr>
<td>2004</td>
<td>OMI</td>
<td>13 x 24</td>
<td>Global</td>
<td>1 day</td>
</tr>
<tr>
<td>2006</td>
<td>GOME-2</td>
<td>80 x 40</td>
<td>Global</td>
<td>1 day</td>
</tr>
<tr>
<td>2011</td>
<td>OMPS</td>
<td>50 x 50</td>
<td>Global</td>
<td>1 day</td>
</tr>
<tr>
<td>2017</td>
<td>TROPOMI</td>
<td>3.5 x 5.5</td>
<td>Global</td>
<td>1 day</td>
</tr>
<tr>
<td>2023 (planned)</td>
<td>Sentinel-5</td>
<td>7 x 7</td>
<td>Global</td>
<td>1 day</td>
</tr>
<tr>
<td>2020 (planned)</td>
<td>GEMS</td>
<td>7 x 8</td>
<td>East Asia</td>
<td>1 hour</td>
</tr>
<tr>
<td>2022 (planned)</td>
<td>TEMPO</td>
<td>2 x 4.5</td>
<td>North America</td>
<td>1 hour</td>
</tr>
<tr>
<td>2023 (planned)</td>
<td>Sentinel-4</td>
<td>8 x 8</td>
<td>Europe</td>
<td>1 hour</td>
</tr>
</tbody>
</table>

Table 2. Data quality targets for the S5P TROPOMI data products relevant in the AROMAT context (extracted from ESA (2014)).

<table>
<thead>
<tr>
<th>Product</th>
<th>Accuracy</th>
<th>Precision</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tropospheric NO$_2$</td>
<td>25-50%</td>
<td>7x10$^{14}$ molec cm$^{-2}$</td>
</tr>
<tr>
<td>Tropospheric SO$_2$</td>
<td>30-50%</td>
<td>2.7-8.1x10$^{16}$ molec cm$^{-2}$</td>
</tr>
<tr>
<td>Total H$_2$CO</td>
<td>40-80%</td>
<td>0.4-1.2x10$^{16}$ molec cm$^{-2}$</td>
</tr>
</tbody>
</table>

Table 3. Main characteristics of the power plants of the Jiu Valley.

<table>
<thead>
<tr>
<th></th>
<th>Craiova II</th>
<th>Islandita</th>
<th>Turceni</th>
<th>Rovinari</th>
</tr>
</thead>
<tbody>
<tr>
<td>Latitude (°N)</td>
<td>44.343</td>
<td>44.39</td>
<td>44.67</td>
<td>44.91</td>
</tr>
<tr>
<td>Longitude (°E)</td>
<td>23.81</td>
<td>23.72</td>
<td>23.41</td>
<td>23.135</td>
</tr>
<tr>
<td>Nominal capacity (MW)</td>
<td>300</td>
<td>630</td>
<td>1650</td>
<td>1320</td>
</tr>
<tr>
<td>Smokestack height (m)</td>
<td>149</td>
<td>200</td>
<td>280</td>
<td>280</td>
</tr>
</tbody>
</table>
### Table 4. Summary of the AROMAT measurements of NO₂.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Type</th>
<th>Ground Sampling</th>
<th>Observed range (molec cm⁻² / ppb)</th>
<th>Detection limit (molec cm⁻² / ppb)</th>
<th>Bias (%)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>AirMAP Imager</td>
<td>Imager</td>
<td>100</td>
<td>0-8 x 10¹⁶</td>
<td>1.5 x 10¹⁵</td>
<td>25%</td>
<td>Meier et al. (2017)</td>
</tr>
<tr>
<td>SWING Imager</td>
<td>Imager</td>
<td>300</td>
<td>0-8 x 10¹⁶</td>
<td>1.2 x 10¹⁵</td>
<td>25%</td>
<td>Merlaud et al. (2018)</td>
</tr>
<tr>
<td>ULMDOAS Nadir</td>
<td>Nadir</td>
<td>400</td>
<td>0-1.7 x 10¹⁷</td>
<td>5 x 10¹⁴</td>
<td>25%</td>
<td>Constantin et al. (2017)</td>
</tr>
<tr>
<td>Tube MAX-DOAS car-based</td>
<td>500</td>
<td>0-1.3 x 10¹⁷</td>
<td>1.3 x 10¹⁴</td>
<td>20%</td>
<td></td>
<td>Donner et al. (2015)</td>
</tr>
<tr>
<td>Mini Max-DOAS car-based</td>
<td>500</td>
<td>0-1.3 x 10¹⁷</td>
<td>6 x 10¹⁴</td>
<td>20%</td>
<td></td>
<td>Wagner et al. (2010)</td>
</tr>
<tr>
<td>UGAL Mobile car-based</td>
<td>500</td>
<td>0-2.5 x 10¹⁷</td>
<td>4 x 10¹⁴</td>
<td>25%</td>
<td></td>
<td>Constantin et al. (2013)</td>
</tr>
<tr>
<td>BIRA Mobile car-based</td>
<td>500</td>
<td>0-1.3 x 10¹⁷</td>
<td>8 x 10¹⁴</td>
<td>20%</td>
<td></td>
<td>Merlaud (2013)</td>
</tr>
<tr>
<td>KNMI sonde in-situ</td>
<td>n.a.</td>
<td></td>
<td>0-60</td>
<td>1</td>
<td>40%</td>
<td>Sluis et al. (2010)</td>
</tr>
<tr>
<td>CAPS in-situ</td>
<td>n.a.</td>
<td></td>
<td>0-20</td>
<td>0.1</td>
<td>40%</td>
<td>Kebabian et al. (2005)</td>
</tr>
</tbody>
</table>

### Table 5. Summary of the AROMAT measurements of H₂CO.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Type</th>
<th>Ground Sampling</th>
<th>Observed range (molec cm⁻²)</th>
<th>Detection limit (molec cm⁻²)</th>
<th>Bias (%)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bremen nadir Airborne nadir</td>
<td>1800</td>
<td>1-7 x 10¹⁶</td>
<td>6 x 10¹⁵</td>
<td>25%</td>
<td></td>
<td>Bösch et al. (2016)</td>
</tr>
<tr>
<td>Tube MAX-DOAS car-based</td>
<td>500</td>
<td>1-7.5 x 10¹⁶</td>
<td>8 x 10¹⁴</td>
<td>20%</td>
<td></td>
<td>Donner et al. (2015)</td>
</tr>
</tbody>
</table>

### Table 6. Summary of the AROMAT measurements of SO₂.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Type</th>
<th>Ground Sampling</th>
<th>Observed range (molec cm⁻²)</th>
<th>Detection limit (molec cm⁻²)</th>
<th>Bias (%)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>AirMAP Imager</td>
<td>Imager</td>
<td>100</td>
<td>0-6 x 10¹⁷</td>
<td>1.7 x 10¹⁶</td>
<td>40%</td>
<td>Meier et al. (2017)</td>
</tr>
<tr>
<td>SWING Imager</td>
<td>Imager</td>
<td>300</td>
<td>0-4 x 10¹⁷</td>
<td>2 x 10¹⁶</td>
<td>40%</td>
<td>Merlaud et al. (2018)</td>
</tr>
<tr>
<td>ULMDOAS Nadir</td>
<td>Nadir</td>
<td>400</td>
<td>0-2.5 x 10¹⁸</td>
<td>3 x 10¹⁵</td>
<td>40%</td>
<td>Constantin et al. (2017)</td>
</tr>
<tr>
<td>Tube MAX-DOAS car-based</td>
<td>500</td>
<td>0-1x10¹⁸</td>
<td>5 x 10¹⁵</td>
<td>20%</td>
<td></td>
<td>Donner et al. (2015)</td>
</tr>
<tr>
<td>Mini Max-DOAS car-based</td>
<td>500</td>
<td>0-2.2x10¹⁸</td>
<td>1 x 10¹⁶</td>
<td>20%</td>
<td></td>
<td>Wagner et al. (2010)</td>
</tr>
<tr>
<td>UGAL Mobile car-based</td>
<td>500</td>
<td>0-4x10¹⁸</td>
<td>4 x 10¹⁵</td>
<td>25%</td>
<td></td>
<td>Constantin et al. (2013)</td>
</tr>
</tbody>
</table>
Table 7. Total simulated error budget for the validation of spaceborne NO\textsubscript{2} VCDs validation using airborne mapping at different resolution, with or without profile informations.

<table>
<thead>
<tr>
<th>Place</th>
<th>Shot noise</th>
<th>Time error</th>
<th>Tot. error</th>
<th>Ref. error</th>
<th>Fit error</th>
<th>Tot. error</th>
</tr>
</thead>
<tbody>
<tr>
<td>AirMAP B</td>
<td>3x10\textsuperscript{13}</td>
<td>4x10\textsuperscript{15}</td>
<td>4.1x10\textsuperscript{15}</td>
<td>26%</td>
<td>6%</td>
<td>37%</td>
</tr>
<tr>
<td>SWING B</td>
<td>7x10\textsuperscript{13}</td>
<td>4x10\textsuperscript{15}</td>
<td>4.1x10\textsuperscript{15}</td>
<td>26%</td>
<td>6%</td>
<td>37%</td>
</tr>
<tr>
<td>AirMAP + profile B</td>
<td>3x10\textsuperscript{13}</td>
<td>4x10\textsuperscript{15}</td>
<td>4.1x10\textsuperscript{15}</td>
<td>10%</td>
<td>6%</td>
<td>28%</td>
</tr>
<tr>
<td>AirMAP T</td>
<td>3x10\textsuperscript{13}</td>
<td>4x10\textsuperscript{15}</td>
<td>4.1x10\textsuperscript{15}</td>
<td>26%</td>
<td>10%</td>
<td>37%</td>
</tr>
<tr>
<td>AirMAP + profile T</td>
<td>3x10\textsuperscript{13}</td>
<td>4x10\textsuperscript{15}</td>
<td>4.1x10\textsuperscript{15}</td>
<td>10%</td>
<td>10%</td>
<td>29%</td>
</tr>
</tbody>
</table>

Table 8. NO\textsubscript{x} emissions from Bucharest estimated from the AROMAT measurements.

<table>
<thead>
<tr>
<th>Date</th>
<th>AirMAP Mobile-DOAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>8 September 2014</td>
<td>14.6 mol.s\textsuperscript{-1} 12.5 mol.s\textsuperscript{-1}</td>
</tr>
<tr>
<td>9 September 2014</td>
<td>13.1 mol.s\textsuperscript{-1} n.a.</td>
</tr>
<tr>
<td>31 August 2015</td>
<td>n.a. 17.5 mol.s\textsuperscript{-1}</td>
</tr>
</tbody>
</table>

Table 9. NO\textsubscript{x} and SO\textsubscript{2} emissions from the Turceni power plant estimated from the AROMAT measurements.

<table>
<thead>
<tr>
<th>Date</th>
<th>Instrument</th>
<th>Distance</th>
<th>SO\textsubscript{2} flux</th>
<th>NO\textsubscript{x} flux</th>
<th>SO\textsubscript{2}/NO\textsubscript{2}</th>
</tr>
</thead>
<tbody>
<tr>
<td>11 September 2014 - 09:00 UTC</td>
<td>AirMAP</td>
<td>7 km</td>
<td>n.a.</td>
<td>8 mol.s\textsuperscript{-1}</td>
<td>n.a.</td>
</tr>
<tr>
<td>25 August 2015 - 07:45 UTC</td>
<td>Mobile-DOAS</td>
<td>1 km</td>
<td>105 mol.s\textsuperscript{-1}</td>
<td>4 mol.s\textsuperscript{-1}</td>
<td>15.4</td>
</tr>
<tr>
<td>25 August 2015 - 08:30 UTC</td>
<td>Mobile-DOAS</td>
<td>1 km</td>
<td>52 mol.s\textsuperscript{-1}</td>
<td>2 mol.s\textsuperscript{-1}</td>
<td>26</td>
</tr>
<tr>
<td>25 August 2015 - 08:30 UTC</td>
<td>ULMDOAS</td>
<td>10 km</td>
<td>85 mol.s\textsuperscript{-1}</td>
<td>10 mol.s\textsuperscript{-1}</td>
<td>8.5</td>
</tr>
<tr>
<td>26 August 2015 - 10:00 UTC</td>
<td>ULMDOAS</td>
<td>5 km</td>
<td>78 mol.s\textsuperscript{-1}</td>
<td>6 mol.s\textsuperscript{-1}</td>
<td>13</td>
</tr>
<tr>
<td>27 August 2015 - 07:45 UTC</td>
<td>ULMDOAS</td>
<td>8.5 km</td>
<td>145 mol.s\textsuperscript{-1}</td>
<td>17 mol.s\textsuperscript{-1}</td>
<td>8.5</td>
</tr>
<tr>
<td>27 August 2015 - 07:55 UTC</td>
<td>Mobile-DOAS</td>
<td>1 km</td>
<td>77 mol.s\textsuperscript{-1}</td>
<td>5 mol.s\textsuperscript{-1}</td>
<td>16</td>
</tr>
<tr>
<td>28 August 2015 - 07:00 UTC</td>
<td>Mobile-DOAS</td>
<td>1 km</td>
<td>24.8 mol.s\textsuperscript{-1}</td>
<td>1.7 mol.s\textsuperscript{-1}</td>
<td>14.7</td>
</tr>
<tr>
<td>28 August 2015 - 10:00 UTC</td>
<td>AirMAP</td>
<td>7 km</td>
<td>25 mol.s\textsuperscript{-1}</td>
<td>8 mol.s\textsuperscript{-1}</td>
<td>3.1</td>
</tr>
<tr>
<td>28 August 2015 - 10:15 UTC</td>
<td>Mobile-DOAS</td>
<td>1 km</td>
<td>32 mol.s\textsuperscript{-1}</td>
<td>4 mol.s\textsuperscript{-1}</td>
<td>8</td>
</tr>
<tr>
<td>28 August 2015 - 09:00-11:00 UTC</td>
<td>SO\textsubscript{2} camera</td>
<td>Above stack</td>
<td>15.6-62.4 mol.s\textsuperscript{-1}</td>
<td>n.a</td>
<td>n.a.</td>
</tr>
</tbody>
</table>