SATELLITE VALIDATION OF TROPOMI-SO_2 OVER THE BALKAN REGION BY AIRBORNE SO_2 MEASUREMENTS OF COAL-FIRED POWER PLANTS

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Abstract

The first airborne in situ measurements of sulphur dioxide (SO_2) emissions (plumes) from two coal-fired power plants in Bosnia-Herzegovina (Tuzla) and Serbia (Nikola Tesla) were carried out with the German research aircraft *Falcon-20* in cooperation with local partners during the METHANE-To-Go field experiment in autumn 2020. Downwind of the power plants, SO_2 mixing ratios exceeding 100 ppb were measured in a distance ~20-40 km from the sources. The plumes were trapped in well-defined inversion layers between ~500-1000 m altitude. Our airborne measurements can be used to validate synchronously, spaceborne SO_2 measurements from the TROPOspheric Monitoring Instrument (TROPOMI) onboard the Sentinel-5P satellite. A first intercomparison indicates some problems with dense smoke clouds frequently covering these countries in winter. However, one part of the Nikola Tesla flight is well suited for TROPOMI-SO_2 validation, since it was obtained during cloud-free conditions with a well-defined vertical extension of the probed SO_2 plume (needed to estimate the Vertical Column Density, VCD). These airborne measurements and model simulations can also be used to determine the SO_2 emission strength of the power plants. First estimates (mass balance approach) show that the SO_2 mass flux from Tuzla is about twice as high as indicated by common emission inventories.

Keywords: sulphur dioxide emissions, power plants, airborne measurements, satellite validation, Balkans

INTRODUCTION

Recent results from spaceborne measurements indicate that the Balkans is a hot spot region for anthropogenic SO_2 emissions in Europe (Theys et al., 2015; Fioletov et al., 2017, 2020; Liu et al., 2018). These emissions mainly originate from a few coal-fired power plants located in Bosnia-Herzegovina and Serbia. The Serbian power plants, Nikola Tesla, are ranked on position number 18 of the world´s strongest sources of SO_2 pollution according to Dahiya and Myllyvirta (2019), whereas the Bosnia-Herzegovina power plant Tuzla ranks on position number 25.

Here we report on the first airborne in situ measurements in this region carried out by the German Aerospace Center (Deutsches Zentrum für Luft- und Raumfahrt, DLR).

FIELD CAMPAIGN AND DATA

The DLR-funded field experiment METHANE-To-Go-Europe, with the objective to investigate anthropogenic methane (CH_4) and sulfur dioxide sources (SO_2), was carried out in October and November 2020 together with local partners in the Balkans (https://www.dlr.de/pa/en/desktopdefault.aspx/tabid-2342/6725_read-70275/). The focus of the CH_4 measurements was on emissions from off-shore gas facilities in the Adriatic Sea, whereas the focus of the SO_2 measurements was on emissions from coal-fired power plants in the Balkans. Here we only report on results from the latter objective.

Airborne in situ measurements were carried out with the German research aircraft *Falcon-20* of the DLR, which was equipped with a Chemical Ionization Ion-Trap Mass Spectrometer (CI-ITMS) (accuracy: ~15%; detection limit ~50 ppt; temporal resolution: ~3 s; Speidel et al., 2007) and meteorological instrumentation (Huntrieser et al., 2016). Two flights were dedicated to power plant emissions, a flight to Bosnia-Herzegovina (Tuzla) on 2nd November and a flight to Serbia (Nikola Tesla near Belgrade) on 7th November 2020. Here we mainly concentrate on the latter flight, however in the oral presentation also results from the first flight will be shown. **Figure 1** gives a first glance of the flight to the Nikola Tesla power plants on a typical autumn day with a strong inversion layer trapping the emissions from the power plant in a thin layer.

One of the main objectives of this study is to validate spaceborne SO_2-measurements by the TROPOMI/S5P instrument with our airborne measurements, which is the first study of its kind in the Balkans. TROPOMI/S5P is a novel passive imaging spectrometer which combines different methods/algorithms to retrieve SO_2 concentrations and cloud properties with a daily passage around local noon time (Theys et al., 2017; Loyola et al., 2018). In this work, we use two different sets of TROPOMI-SO_2 products, the official operational product and the COBRA product (Theys et al., 2021), which will replace the current operational product in the future. The variables extracted from the TROPOMI products are i.a. the Vertical Column Density (VCD) SO_2, the quality assurance value (QA), and the Cloud Fraction (CF).

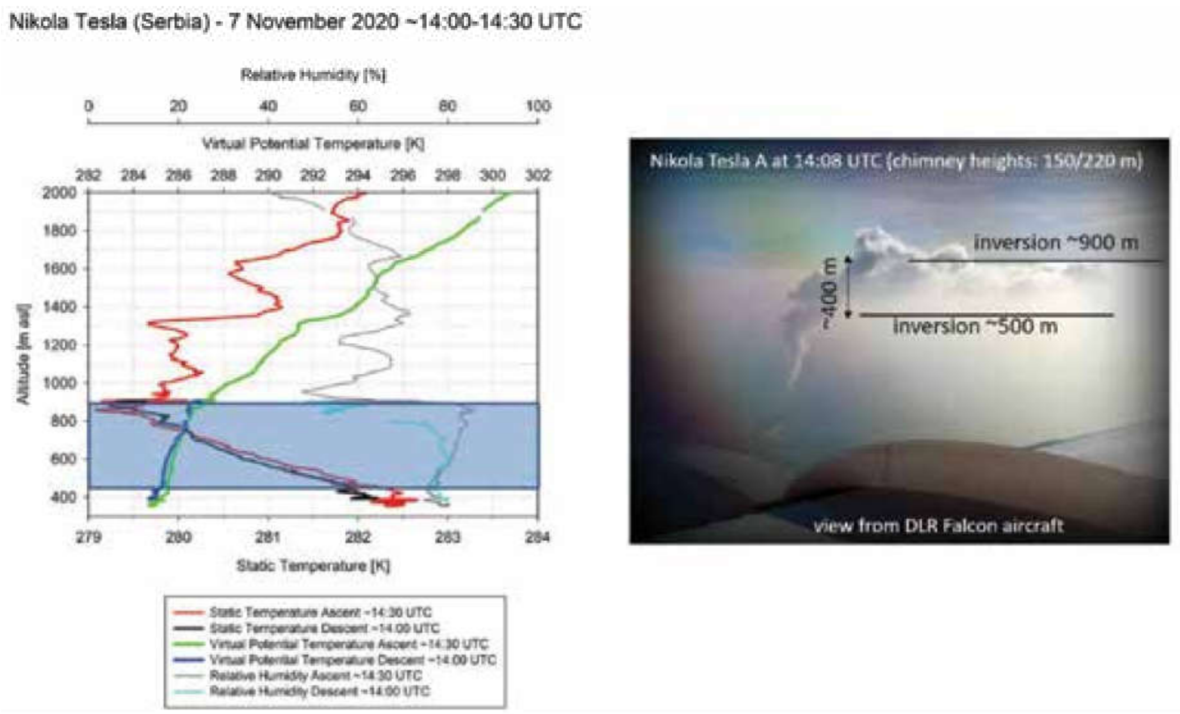

*Figure 1. Vertical temperature and humidity profiles from the DLR Falcon-20 flight to the Nikola Tesla A power plant on 7th November 2020 (**left**). The emissions from the coal-fired power plant were trapped in an inversion layer located between ~500 and ~900 m asl (**right**).*

METHODS

For the validation of the spaceborne measurements with airborne measurements, the first task is to select reliable TROPOMI/S5P pixels. However, this task turned out to be more difficult than expected. The inspection of daily TROPOMI-SO_2 images showed a very noisy situation on many days, especially at the end of the campaign in November. The TROPOMI measurements along the long light path at this time of the year were influenced by the low position of the sun in the winter months and the unexpected high frequency of dense smoke clouds over the Balkans. Aerosols in the smoke clouds have the capability to also absorb light in the same wave length range as SO_2 and therefore might disturb the SO_2 signal (Bergstrom et al., 2007). **Figure 2** shows one example of how the strong TROPOMI-SO_2 signals from the power plants in Bosnia-Herzegovina and Serbia deteriorate from October 2019 to November 2019 and simultaneously the noise from unspecified SO_2 sources in the surroundings increases. Similar noisy observations were made for the months December and January. First in February less disturbed TROPOMI-SO_2 signals are available as for the entire summer season.

The mission flights to Bosnia-Herzegovina on November 2nd and to Serbia on November 7th were influenced differently by the noise in the SO_2 retrieval. A number of nearby local fires affected the first flight (Tuzla power plant) significantly, it is questionable if a satellite validation will be possible. However, the second flight to the Nikola Tesla power plants in Serbia was less affected by noise in the SO_2 retrieval in the envisaged area and was found to be suitable for a validation.

To select reliable SO_2 pixel from the Serbian flight, the SO_2 data was combined with additional TROPOMI products (i.a. QA and CF) as mentioned in Section 2. Reliable SO_2 pixels were defined when $QA \geq 50$ and $CF \leq 0.10$. As will be shown later in Section 4, only one part of the Serbian flight is suitable for a validation.

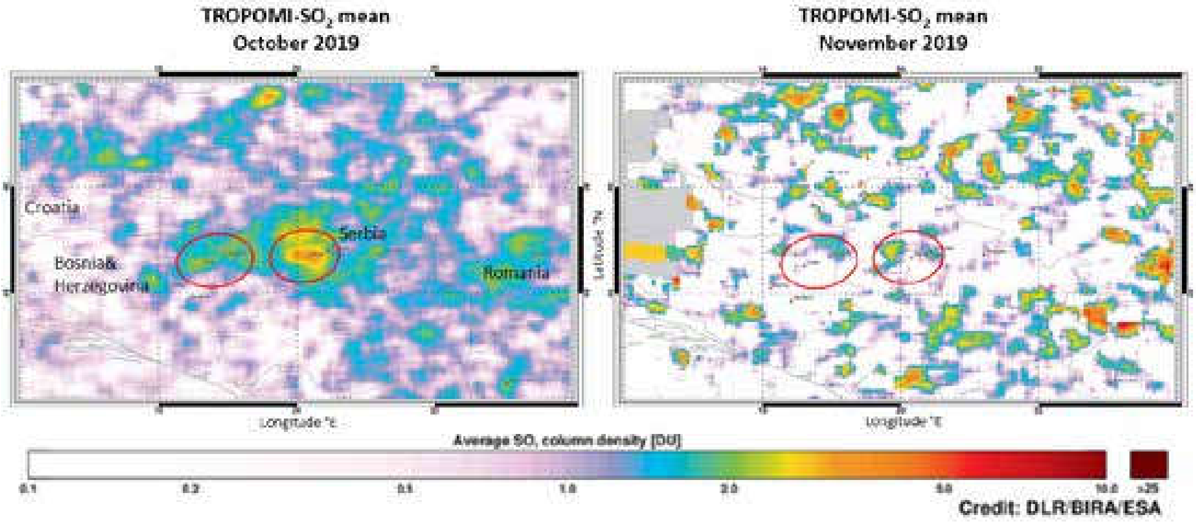

*Figure 2. Sulphur dioxide (SO_2) measurements from the Sentinel-5P satellite obtained with the TROPOspheric Monitoring Instrument (TROPOMI). Mean SO_2-values in Dobson Units (DU) for the Balkans are shown for October 2019 (**left**) and November 2019 (**right**). The red circles highlight the two SO_2 hot spot areas of the Balkans investigated in this study (power plants Tuzla in Bosnia-Herzegovina and Nikola Tesla in Serbia).*

Presently the TROPOMI/S5P pixel size is 3.6 x 5.6 km^2. For the selected validation area, our airborne SO_2 measurements were averaged over the area of each pixel. For the calculation of the VCD from the airborne measurements, also information about the vertical SO_2 distribution is necessary. For 7 selected pixels, measurements were performed at three different altitudes (~400, 540 and 900 m), just below, inside and at the top of the strong inversion layer (**Fig. 1**).

From **Fig. 3** it is clear, that the main amount of SO_2 contributing to the VCD was located in this inversion layer between ~450 and 900 m (due to the elevated emission height from the tall chimneys). Therefore, our airborne measurements were vertically interpolated between these layers and then integrated over the inversion layer to receive VCD values comparable with satellite measurements. Both our airborne measurements and the ground-based measurements from the nearby site Obrenovac indicate that the contribution to the VCD from the layer below ~400 m was less important on this day (note the logarithmic scale).

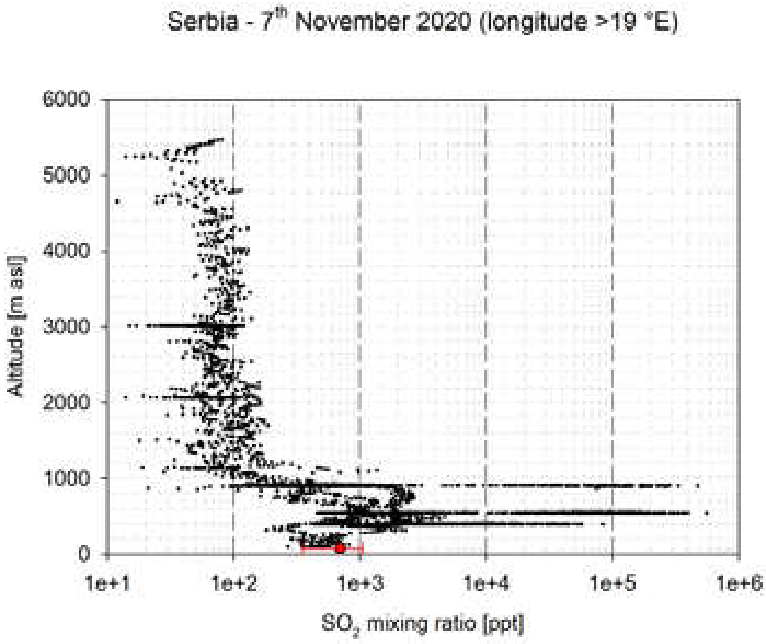

Figure 3. Vertical SO_2 profile from the DLR Falcon-20 flight over Serbia on 7th November 2020 (black dots). Superimposed is the daily mean value and standard deviation (red dot with bar) of the ground-based SO_2 measurements in Obrenovac (76 m asl), located near the Nikola Tesla power plants, on 7th November 2020. The SO_2 mixing ratios are given in parts per trillion (ppt).

RESULTS

The horizontal distribution of SO_2 mixing ratios along the flight path in vicinity of the Nikola Tesla A (NTA) and B (NTB) power plants is shown in **Fig. 4 (left)** in the unit parts per billion (ppb). Areas with highly elevated SO_2 mixing ratios are highlighted in green, yellow and red colors. It is clear, that the plumes from the NTA and NTB power plants are advected with the wind to the northwest and are distinguishable from each other, as also observed by the aircraft crew. Unfortunately, the CI-ITMS instrument had problems to measure the unexpectedly high SO_2 mixing ratios inflight and the uncertainty of the measurements could be in the range of several hundred percent, which still has to be determined by more laboratory tests.

In **Fig. 4 (right)**, several TROPOMI/S5P products have been added to the airborne measurements from **Fig. 4 (left)**. Color-coded in traffic light colors are the TROPOMI-SO_2 VCD values in Dobson Units (DU). Noticeable is the agreement of the location of the highest SO_2 values for the airborne and spaceborne measurements in the westernmost part of the flight track capturing mainly the NTB plume. In comparison, the NTA plume is not well captured in TROPOMI-SO_2 signal compared to the airborne measurements. The reason is a nearby cloud remnant from the morning fog in the river valley, as indicated schematically in the figure according to the TROPOMI cloud product CF. This result shows that, as previously noticed, only the westernmost part of the flight (including the NTB, but not NTA plume) is suitable for a validation. Fortunately, this part of the flight was also flown at three different vertical levels as mentioned in Section 3 and the VCD-SO_2 can be determined from the airborne measurements and compared to the spaceborne TROPOMI-SO_2 measurements as discussed in the next section.

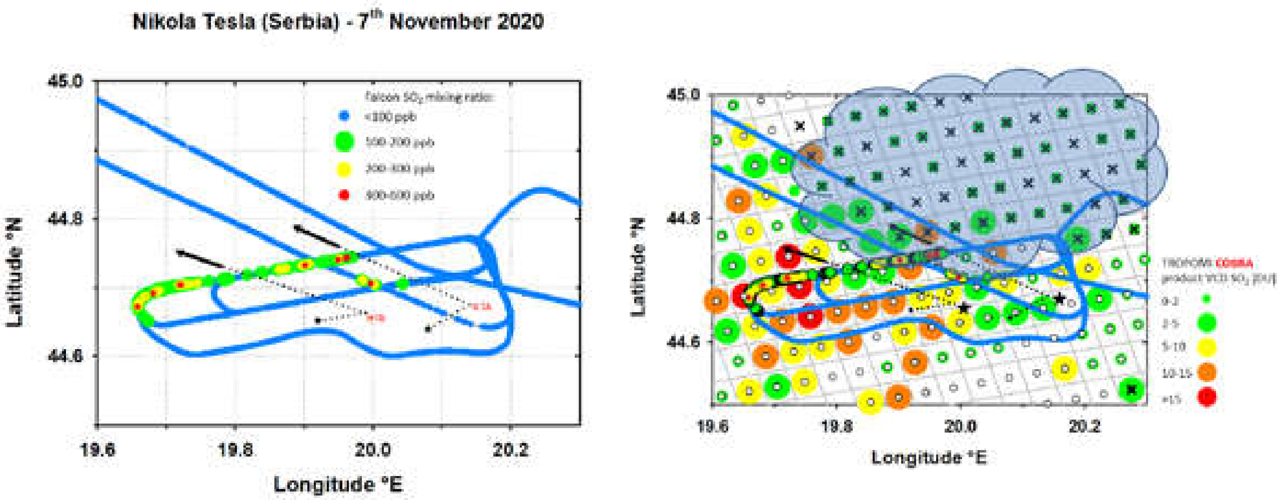

*Figure 4. SO_2 mixing ratios in ppb (color-coded) along the DLR Falcon-20 flight track in vicinity of the power plants Nikola Tesla A (NTA in red) and B (NTB also in red) on 7th November 2020 (**left**). The dotted lines with a star at the end indicate the measured wind direction at the height of the power plant chimneys (~150-300 m); varying between northeasterly to easterly direction. The dotted lines with an arrow at the end indicate the wind direction measured at the Falcon flight levels downwind of the power plants (~540-900 m); predominantly from southeasterly direction. Underlaid below the flight track is the corresponding TROPOMI-SO_2 COBRA product in DU (also color-coded) (**right**). In addition, the cloud fraction (CF) is superimposed as black crosses (CF ≥0.20) and as white circles (CF<0.20), and the position of a cloud remnant from the morning fog is schematically indicated.*

DISCUSSION

In **Fig. 5 (left)**, both VCD-SO_2 values obtained from airborne (*Falcon-20*) and spaceborne (TROPOMI) measurements (COBRA retrieval) are shown, indicating a positive correlation with a high correlation coefficient (r>0.9). However, the number of data points is unfortunately rather limited. In **Fig. 5 (right)** the SO_2 mean values of all 7 data points in **Fig. 5 (left)** were determined and in addition for the TROPOMI operational product. The TROPOMI COBRA product value is slightly lower (~10%) than the value of the operational TROPOMI product. The airborne measurements are presently a factor of 2.5-3 lower than the obtained TROPOMI values, which might be caused by the standard (instead of adjusted) averaging kernel used.

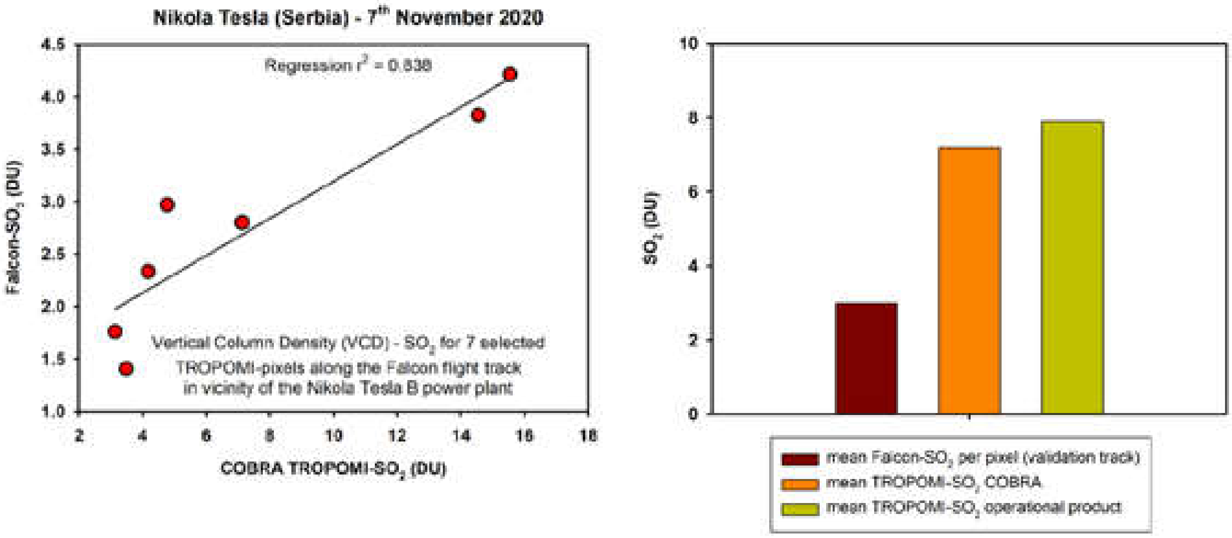

*Figure 5. Preliminary validation of the TROPOMI-SO_2 retrieval based on the COBRA algorithm with vertical column density (VCD) estimates in DU from airborne SO_2-measurements obtained from the DLR Falcon-20 in vicinity of the Nikola Tesla B power plant on 7th November 2020 (**left**). Preliminary comparison of mean VCD SO_2 values obtained from two different TROPOMI-SO_2 retrievals (COBRA and operational product) and from estimates based on the airborne SO_2-measurements (**right**).*

CONCLUSION

Our measurements during the METHANE-To-Go-Europe field experiment are:

- the first airborne in situ measurements of SO_2 plumes from coal-fired power plants in Serbia and Bosnia-Herzegovina,
- and the first try to conduct a TROPOMI-SO_2 validation with airborne measurements for this region (~2h time shift present between the two measurements).

We found that:

- a validation is difficult in the winter months due to the low sun position and the frequent and widespread occurrence of smoke clouds causing noise in the retrieval,
- however, a validation is partly simplified due to pronounced inversion layers,
- for the Serbian flight a validation of the SO_2 plume from the Nikola Tesla B power plant seems to be feasible, despite of a number of uncertainties and only few data points.

Outlook:

- tests with different averaging kernels are planned,
- CI-ITMS performance tests in the laboratory are planned → more airborne SO2 measurements of this kind are needed in the region to improve satellite algorithms.

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