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Emissions of Sulphur Dioxide (SO_2) from Coal-Fired Power Plants in Serbia and Bosnia-Herzegovina: First Attempts of a Validation of TROPOMI Satellite Products with Airborne In Situ Measurements

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Abstract: *The Western Balkan region is known for emitting alarmingly high sulphur dioxide amounts from coal-fired power plants. Though a number of environmental regulations have been introduced in recent years (e.g. desulphurisation installations, construction of modern power plants), the pollution burden is still much higher than recommended by the authorities. A number of different montoring systems are required to observe the growing pollution situation in the Western Balkan region, partly caused by a high energy demand from outside (e.g. Western Europe).*

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Цитирање: Хунтризер Х, Клаузнер-Харлаш Т, Ауфмхоф Х, Бауман Р, Фин А, Гетшалт К-Д, Хеделт П, Луц Р, Mrazovac Курилић С, Подрашчанин З, Илић П, Тејс Н, Јекел П, Лојола Д, Макрум И, Мертенс М, Ројгер А (2023) Емисије сумпор-диоксида (SO_2) из термоелектрана на угаљ у Србији и Босни и Херцеговини: први покушаји валидације резултата сателитског in situ мјерења у ваздуху помоћу TROPOMI. У: Илић П, Говедар З, Пржуљ Н (уредници) Животна средина. Академија наука и умјетности Републике Српске, Бања Лука, Монографија LV:169-201

Several of the top ten SO_2 polluters in Europe are located in Bosnia-Herzegovina and Serbia. Here we present the first in situ measurements of sulphur dioxide in this region conducted with a German research aircraft in cooperation with local scientists in Bosnia-Herzegovina and Serbia. Two of the strongtest emitting coal-fired power plants were selected for the measurements in autumn 2020: Tuzla in Bosnia-Herzegovina and Nikola Tesla in Serbia (Nikola Tesla). The measurements were mainly conducted in the boundary layer (below ~1 km altitude in winter). Downwind of the power plants, extremely high SO_2 mixing ratios exceeding 100 parts per billion (ppb = nmol mol^{-1}) were measured at a distance of ~20-40 km from the sources. The SO_2 plumes from the power plants were trapped in well-defined inversion layers between ~500-1000 m altitude. The airborne measurements can be used to validate synchronous 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 the winter months. However, it turned out that the Nikola Tesla flight is to some extent suited for a 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, measured by the satellite). In addition, these airborne measurements accompanied by model simulations can be used to determine the SO_2 emission strength of the power plants and to compare it to the source strength reported by the power plant operators. The results indicate a reasonable agreement between the airborne measurements, model results, emission inventories, and satellite measurements for the Nikola Tesla power plants.

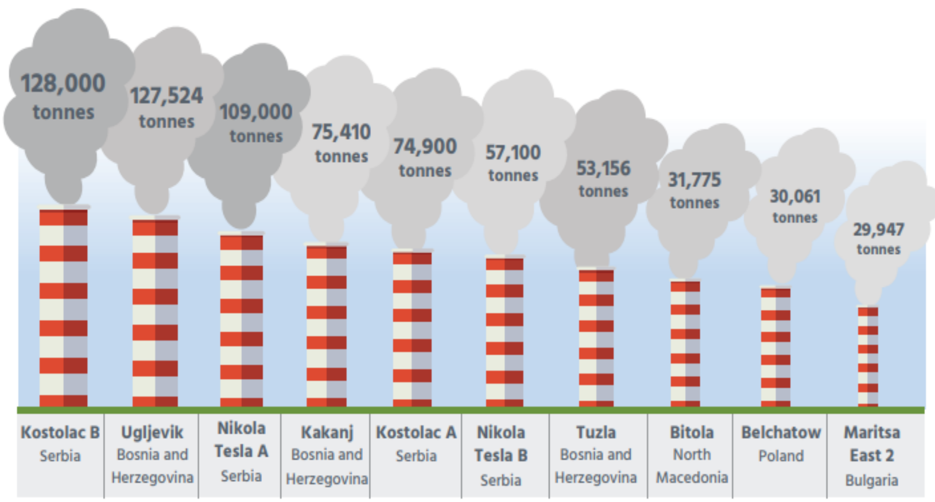
Keywords: Sulphur dioxide emissions, coal-fired power plants, airborne measurements, satellite validation, Balkans

6.1. Introduction

The Western Balkan region belongs to the most polluted regions in Europe significantly impacting the health of the population (e.g. Lammel et al 2011; Podrascanin and Mihailovic 2013; Ilić et al 2018; Mrazovac Kurilić et al 2019; Podrascanin 2019; Banja et al. 2020; Ilić et al 2020; Mrazovac Kurilić et al 2020; Ulniković et al 2020; Milošević and Mrazovac Kurilić 2021; Mimić et al. 2021; Bellis et al. 2022; Radović et al. 2022; Šikoparija et al. 2022). It is well-known that especially pollution from coal-fired power plants damages health, impacting particularly the respiratory system, causing bronchitis, asthma, a number of cardiovascular diseases and premature deaths (Lin et al. 2018; Matkovic et al. 2020).

In Europe, the Western Balkan emits alarmingly high levels of pollutants as sulphur dioxide (SO_2), since a number of obsolescent and ineffective coal-fired power plants are still in use (Puljic et al. 2019). Even though the active power plants in this region account for only 6% of all European coal-fired power plants, they emit more SO_2 than all remaining 94% European plants (Chronic coal pollution report, 2019).

The average power plant in the Western Balkans emits 20 times more SO_2 and concurrently produces less energy compared to its European opponents. In the Western Balkan region, mainly the power plants in Serbia (Nikola Tesla, Kostolac) and Bosnia-Herzegovina (Ugljevik, Kakanj, Tuzla) are responsible for most SO_2 emissions (e.g. Jacimovski et al. 2016) (Scheme 6.1).



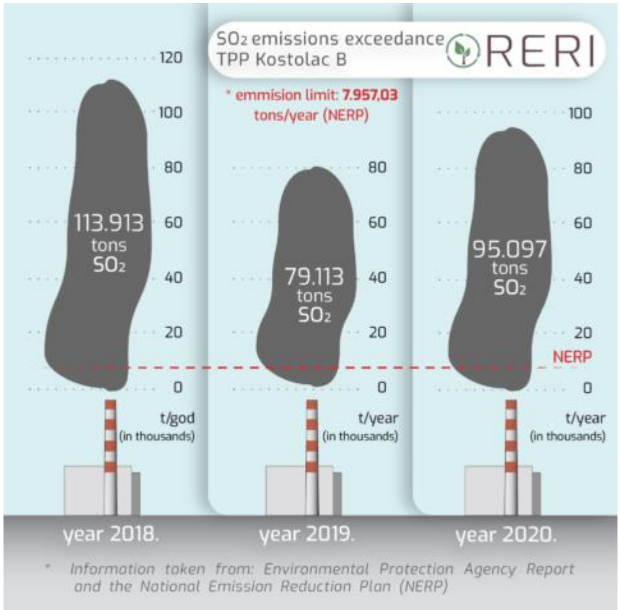
Scheme 6.1. Showing the top 10 polluting power plants in Europe by SO_2 emissions in 2016 (in tons SO_2 per year) (Chronic coal pollution 2019)

Схема *6.1. Приказ 10 електрана које највише загађују у Европи по емисијама SO_2 у 2016. (у тонама SO_2 годишње) (Chronic coal pollution 2019)*

Desulphurisation installations are rare or not properly applied in this region. Though such an installation was added to Kostolac B in 2017, the SO_2 emissions have only slightly reduced from 114.000 tons in 2018, to 95.000 tons in 2020 (*Scheme* 6.2).

According to the National Emission Reduction Plan (NERP), the total SO_2 emissions from the twelve thermal power plants covered by NERP in Serbia are limited to

54.575 tons per year. This means that Kostolac B by itself still exceeded this number by a factor of almost two.



Scheme 6.2. Showing the temporal evolution (2018-2020) of SO_2 emissions (in tons per year) from the power plant Kostolac B in Serbia (www.reri.org.rs/en/untrue-allegations-of-zorana-mihajlovic-emissions-of-sulfur-dioxide-from-the-kostolac-b-thermal-power-plant-are-almost-12-times-higher-than-the-values-allowed-by-the-national-emission-reduction-plan/)

Схема *6.2. Приказ временске евалуције (2018-2020) емисије SO_2 (у тонама годишње) из термоелектране Костолац Б у Србији (www.reri.org.rs/en/untrue-allegations-of-zorana-mihajlovic-emissions-of-sulfur-dioxide-from-the-kostolac-b-thermal-power-plant-are-almost-12-times-higher-than-the-values-allowed-by-the-national-emission-reduction-plan/)*

In addition, recent results from spaceborne measurements support the finding that the Balkans is a hot spot region for anthropogenic SO_2 emissions in Europe (Theys et al. 2015; Fioletov et al. 2017; Liu et al. 2018; Fioletov et al. 2020; Theys et al. 2021a). These emissions mainly originate from a few coal-fired power plants located in Bosnia-Herzegovina and Serbia, as mentioned before (see also Fig. 6.1). The Serbian power plants, Nikola Tesla A and B, are ranked as 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 as number 25.

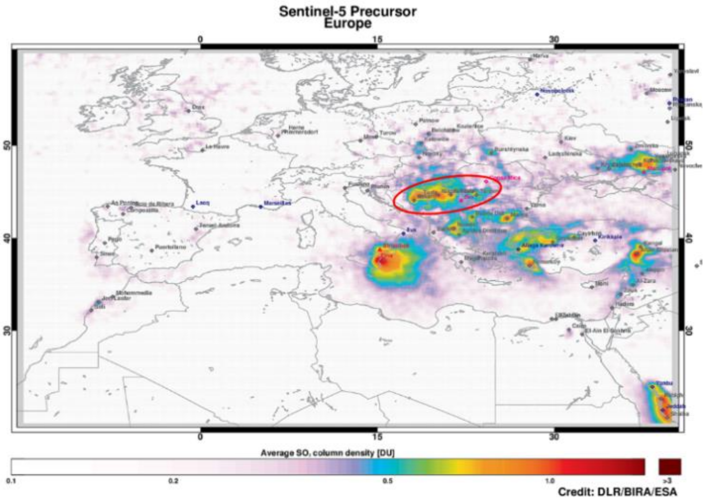


Fig. 6.1. Sulphur dioxide (SO_2) measurements from the Sentinel-5P satellite obtained with the TROPOspheric Monitoring Instrument (TROPOMI). Mean SO_2-values in Dobson Units (DU) for Europe are shown for May 2018 – December 2019 for the operational TROPOMI product. The red circle highlights the SO_2 hot spot area of the Western Balkans investigated in this study (composed by emissions from the power plants Ugljevik, Kakanj, and Tuzla in Bosnia-Herzegovina plus Kostolac and Nikola Tesla in Serbia). The SO_2 hot spot over Sicilia and southern Italy is related to natural SO_2 emissions from the volcano Etna

Сл. 6.1. Мјерења сумпор-диоксида (SO_2) са сателита Sentinel-5P са инструментом TROPOspheric Monitoring Instrument (TROPOMI). Средње вриједности SO_2 у Добсоновим јединицама (Dobson Units, DU) за Европу су приказане за мај 2018 – децембар 2019 оперативни производ TROPOMI. Црвени круг наглашава жариште SO_2 западног Балкана које је истражено у овој студији (састављено од емисија из електрана Угљевик, Какањ и Тузла у Босни и Херцеговини, плус Костолац и Никола Тесла у Србији). Жариште SO_2 изнад Сицилије и јужне Италије повезана је са природним емисијама SO_2 из вулкана Етна

The SO_2 emissions from these two coal-fired power plants were targeted in 2020, within the framework of the METHANE-To-Go-Europe project, when scientists from the German Aerospace Center (DLR: Deutsches Zentrum für Luft- und Raumfahrt) started a collaboration with local scientists in Bosnia-Herzegovina and

Serbia to carry out first airborne in situ measurements of these strong SO_2 emitters. The team has a wide experience in measuring trace gases from natural sources (Huntrieser et al. 2016) and from air pollution (Huntrieser et al. 2004, 2005; Roiger et al. 2011a-b; 2012, 2015, 2017, 2018a-b; Klausner et al. 2018, 2020a-c; Fiehn et al. 2020a-c, 2021), as e.g. sulphur dioxide (Aufmhoff et al. 2011, 2012; Huntrieser et al. 2020, 2021a-c; Klausner et al. 2021), furthermore in performing airborne field campaigns (Roiger et al. 2011a, 2015; Huntrieser et al. 2016; Fiehn et al. 2020a; Klausner et al. 2020b; Huntrieser et al. 2021a-c) accompanied by model simulations (Jöckel et al. 2005, 2006a-b, 2008, 2010; Baumann and Schlager, 2012; Gottschaldt et al. 2013; Jöckel et al. 2016; Mertens et al. 2016; Gottschaldt et al. 2017, 2018; Mertens et al. 2018, 2020a-b, 2021).

6.2. 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 (www.dlr.de/pa/en/desktopdefault.aspx/tabid-2342/6725_read-70275/; last visit 26 July 2022). 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 (max. flight altitude ~13 km) (Fig. 6.2), which was equipped with a non-commercial Chemical Ionization Ion-Trap Mass Spectrometer (CI-ITMS) (accuracy: ~15%; detection limit ~50 ppt; temporal resolution: ~3 s) (Speidel et al. 2007; Aufmhoff et al. 2011, 2012) and a commercial gas analyzer based on pulsed fluorescence technology (Thermo Scientific 43i, less temporal resolution: 10 s, higher response time and less accuracy compared to CI-ITMS) to measure the amount of SO_2 in the air. Furthermore, meteorological instrumentation measuring e.g. wind speed and direction was installed (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 report on the latter flight to Serbia and only briefly touch upon the first flight to Bosnia-Herzegovina.

To support the flight planning, a variety of SO_2 model forecasts (HYSPLIT, WRF-Chem and MECO(n)) were used to forecast the position of the SO_2 pollution plumes over the Balkan region a couple of days in advance (Baumann and Schlager

2012; Mertens et al. 2016). The source strength was derived from various inventories and reports as:

1) the European Pollutant Release and Transfer Register (E-PRTR) (www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-22) and
2) OMI-HTAP which includes emissions from OMI-detected sources obtained from satellite measurements (Theys et al. 2015; Liu et al. 2018) and
3) EDGAR (https://edgar.jrc.ec.europa.eu/emissions_data_and_maps).

Fig. 6.2. The research aircraft Falcon-20 of the German Aerospace Center (DLR: Deutsches Zentrum für Luft-und Raumfahrt) was equipped with trace gas instrumentation to measure e.g. sulphur dioxide in-situ

Сл. 6.2. Истраживачки авион Falcon-20 Њемачког ваздухопловног центра (DLR: Deutsches Zentrum für Luft-und Raumfahrt) био је опремљен са опремом за мјерење гаса у траговима, нпр. сумпор-диоксида in-situ

Here only details on the HYSPLIT simulations are given due to the limited space. In the Lagrangian particle dispersion model (LPDM) HYSPLIT (Draxler et al. 1997, 1998, 1999; Stein et al. 2015) a set of continuously emitting, point-like sources in three distinct geographical regions were simulated for the METHANE-To-Go-Europe field experiment (Fig. 6.3).

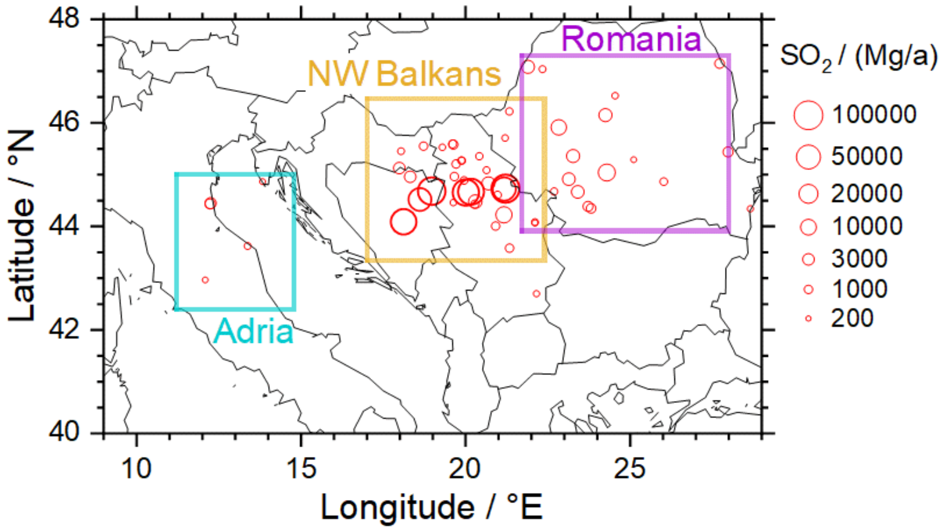


Fig. 6.3. Location and strength of all SO_2 point sources used in the HYSPLIT dispersion forecasts for the METHANE-To-Go-Europe field experiment

Сл. 6.3. Локација и јачина свих тачкастих извора SO_2 који се користе у прогнозама дисперзије HYSPLIT за теренско истраживање METHANE-To-Go-Europe

All SO_2-sources were simulated in HYSPLIT as squared patches with diameters between 10 m (discrete stacks) and 15 km (some cities and large oil and gas facilities). The injection height was simulated according to the known height of the power plant stacks.

HYSPLIT was driven by meteorological data from ECMWF (European Centre for Medium-Range Weather Forecasts) (horizontal resolution of 0.1°×0.1°, hourly forecasts up to 3 days in advance possible). An example of a typical HYSPLIT-SO_2 forecast for the region of interest is shown in Fig. 6.4.

One of the main objectives of this study is to validate spaceborne SO_2-measurements by the TROPOMI/S5P instrument with the airborne measurements focusing on SO_2 emissions from coal-fired power plants, which is the first study of its kind in the Balkans.

Previous validation studies were more focused on volcanic SO_2 (Theys et al. 2019; Hedelt et al. 2019, 2020a-b, 2021; Theys et al. 2021b), which is released at higher altitudes where the sensitivity of the satellite measurements is higher compared to ground-emitted SO_2.

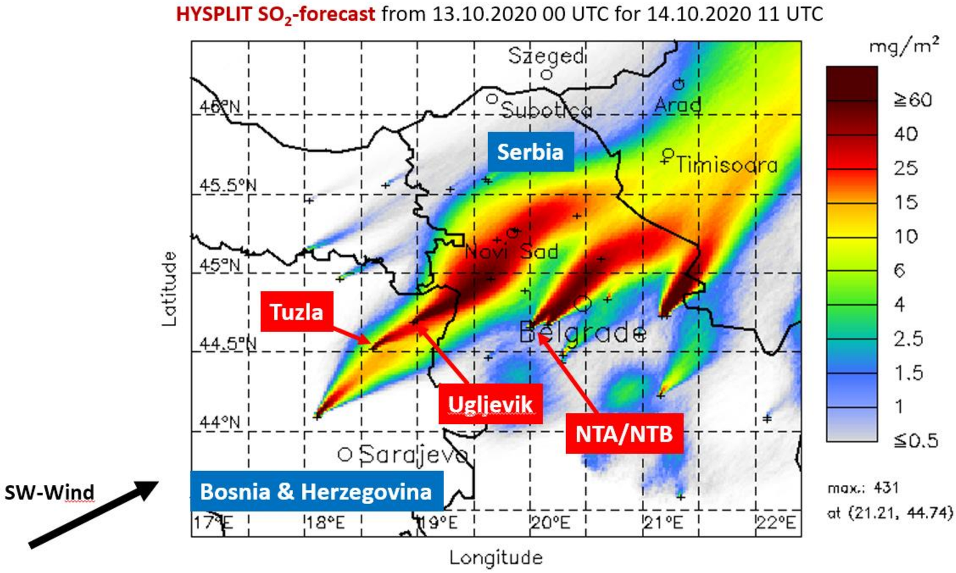


Fig. 6.4. Example of a typical HYSPLIT-SO_2 forecast for the region of interest indicating the direction of the SO_2 plumes from the coal-fired power plants depending on the prevailing wind direction (color-coded: total vertical SO_2 column mass per surface area in mg m^{-2})

Сл. 6.4. Примјер типичне HYSPLIT-SO_2 прогнозе за регион од интереса који указује на правац SO_2 облака из електрана на угаљ у зависности од преовлађујућег правца ветра (кодирано бојом: укупна вертикална маса стуба SO_2 по површини у mg m^{-2})

For specific studies on volcanic SO_2 emissions, neural network methods and inverse learning machines have been developed in the past to handle the high amount of data gained by satellite measurements (e.g. Loyola et al. 2006, 2010, 2011, 2016; Hedelt et al. 2018 a-c; Loyola et al. 2020).

TROPOMI/S5P is a novel passive imaging spectrometer which combines different methods/algorithms to retrieve SO_2 concentrations and cloud properties from a near-polar, sun-synchronous orbit with a daily passage around local noon time (Theys et al. 2017; Lutz et al. 2017, 2018a-b; Theys et al. 2018; Lutz et al. 2019a-b; Theys et al. 2020). In this work, we use two different sets of TROPOMI-SO_2 products, the official operational DOAS retrieval (Loyola et al. 2018) and the COBRA retreival (Theys et al. 2021a), which will replace the current operational product in the future.

The variables extracted here in this study from the TROPOMI products are e.g. the Vertical Column Density (VCD) SO_2 (here "pollution-VCD" used), and in addition the quality assurance value (QA) and the Cloud Fraction (CF) from the TROPOMI cloud product.

Satellite meaurements of SO_2 require cloud-free conditions, which were rarely present during the second half of the METHANE-To-Go-Europe field campaign in November 2020, when flights to the Western Balkan were realised. However, on 7th November, during the flight to the Nikola Tesla power plants in Serbia, the weather conditions were suitable for a first attempt of a TROPOMI validation with airborne SO_2 measurements.

The boundary layer was dominated by a strong inversion layer trapping the emissions from the power plants in a thin layer (Fig. 6.5), which is of advantage for the calculation of the vertical SO_2 column density (SO_2-VCD) from airborne measurements, as needed for the TROPOMI-validation.

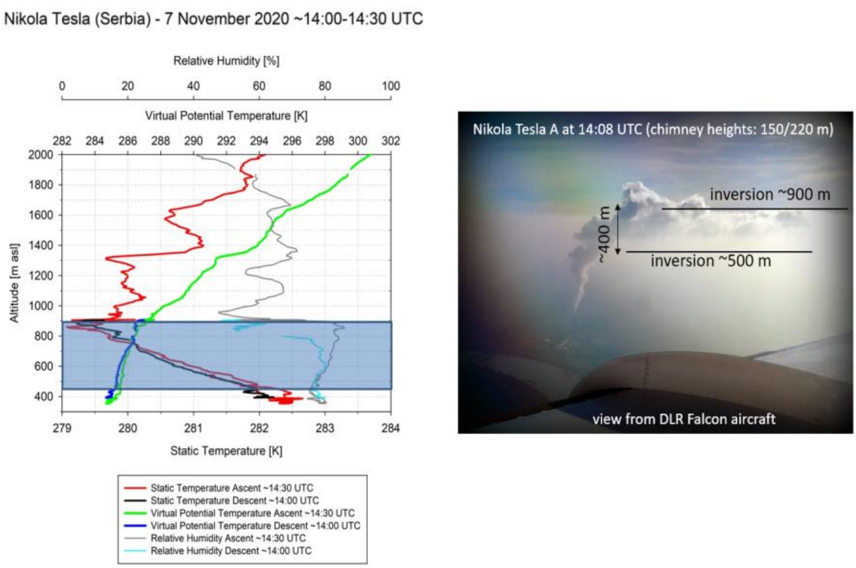


Fig. 6.5. Vertical temperature and humidity profiles from the DLR Falcon-20 flight to the Nikola Tesla A power plant on 7th November 2020 (left). The emissions from the coal-fired power plant were trapped in an inversion layer located between ~500 and ~900 m asl, as seen from Falcon (right)

Сл. 6.5. Вертикални профили температуре и влажности са лета DLR Falcon-20 до електране Никола Тесла А 7. новембра 2020 (лијево). Емисије из електране на угаљ биле су заробљене у инверзионом слоју који се налази између ~500 и ~900 m надморске висине, као што се види са Фалкона (десно)

6.3. Methods

This subsection describes the different steps and methods that were used to determine the VCD SO_2 from airborne measurements in detail. In summary:

1) Selection *of reliable TROPOMI pixels according to the TROPOMI product Cloud Fraction (here CF≤0.10 chosen) and Quality Assurance Index (here QA≥50 chosen).*
2) *Averaging of the airborne SO_2 mixing ratios measured over the area of each selected TROPOMI pixels (e.g. $P1_{SO2}$). Repetition for all available vertical levels (e.g. $P1_{SO2@950\ hPa}$ and $P1_{SO2@900\ hPa}$).*
3) *Vertical interpolation of these averaged measurements ($P1_{SO2@950\ hPa}$, $P1_{SO2@900\ hPa}$ and so on) for each selected pixel (e.g. P1, P2 and so on) up to the next inversion layer. Multiplication of this interpolated vertical profile with the Averaging Kernel.*
4) *Integration over the entire vertical column (here only up to the next inversion layer) for each TROPOMI pixel area (e.g. P1 and P2 and so on) to determine the VCD.*

For the validation of spaceborne with airborne measurements, first reliable TROPOMI/S5P pixels (size 3.6 x 5.6 km^2) were selected which turned out to be more difficult than expected. The inspection of daily TROPOMI-SO_2 images showed a very noisy situation on many campaign days, especially 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 wavelength range as SO_2 and therefore might disturb the SO_2 signal (e.g. Bergstrom et al. 2007). Fig. 6.6. indicates how the strong TROPOMI-SO_2 signals from the Balkan power plants deteriorate from October 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/January. First in February, less disturbed TROPOMI-SO_2 signals are available as for the entire summer season. By using the COBRA retrieval instead of the operational TROPOMI DOAS retrieval, the noise and biases in the signal could be reduced, as also illustrated in Fig. 6.6. (comparable to Figs. 5-6 in Fioletov et al. 2020 and Fig. 4 in Theys et al. 2021a).

The mission flights to Bosnia-Herzegovina on November 2nd and to Serbia on November 7th 2020 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 and it was found that a reliable satellite validation is not possible due to the high density of noisy SO_2 signals, as indicated in Fig. 6.7. (top). In contrast, Fig. 6.7. (bottom) shows a textbook example from January 2020 (prior to the field experiment), when an extended snow cover caused very clear and less noisy SO_2 signals over Serbia (due to the constant albedo of the snow cover)

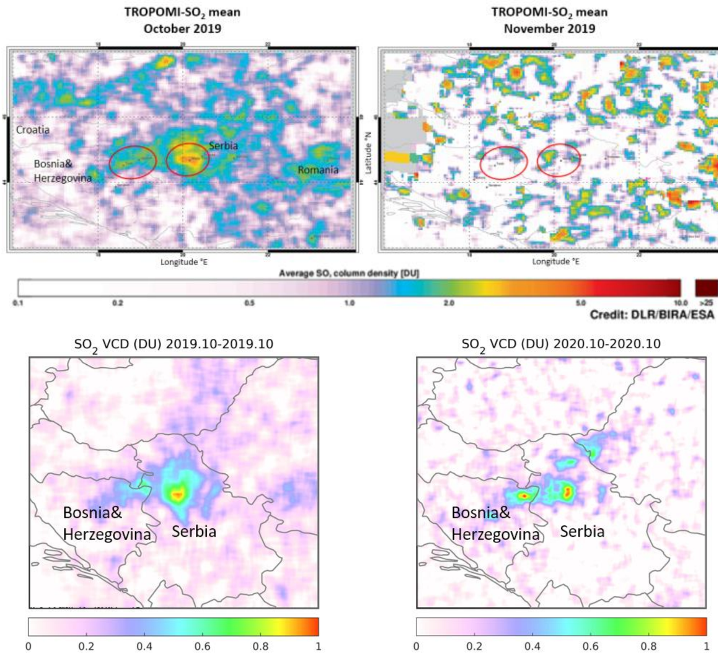


Fig. 6.6. Satellite measurements of SO_2 obtained with TROPOMI. Mean SO_2-values in Dobson Units (DU) for the Balkans are shown for October (upper left) and November 2019 (upper right) for the operational TROPOMI product. The red circles highlight the two SO_2 hot spot areas of the Balkans investigated in this study (containing SO_2 emissions from the power plants Ugljevik, Kakanj, and Tuzla in Bosnia-Herzegovina plus Kostolac and Nikola Tesla in Serbia). A similar SO_2 distribution for October 2019, as shown upper left, is found for the TROPOMI-COBRA product, however related to less noise in the signal (lower left and right)

Сл. 6.6. Сателитска мјерења SO_2 добијена са TROPOMI. Средње вриједности SO_2 у Добсоновим јединицама (ДУ) за Балкан приказане су за октобар (горе лијево) и новембар 2019 (горе десно) за оперативни производ TROPOMI. Црвени кругови наглашавају двије вруће тачке SO_2 на Балкану које су истражене у овој студији (које садрже емисије SO_2 из електрана Угљевик, Какањ и Тузла у Босни и Херцеговини, плус Костолац и Никола Тесла у Србији). Слична дистрибуција SO_2 за октобар 2019, као што је приказано у горњем лијевом углу, налази се за производ TROPOMI-COBRA, али се односи на мање шума у сигналу (доље лијево и десно)

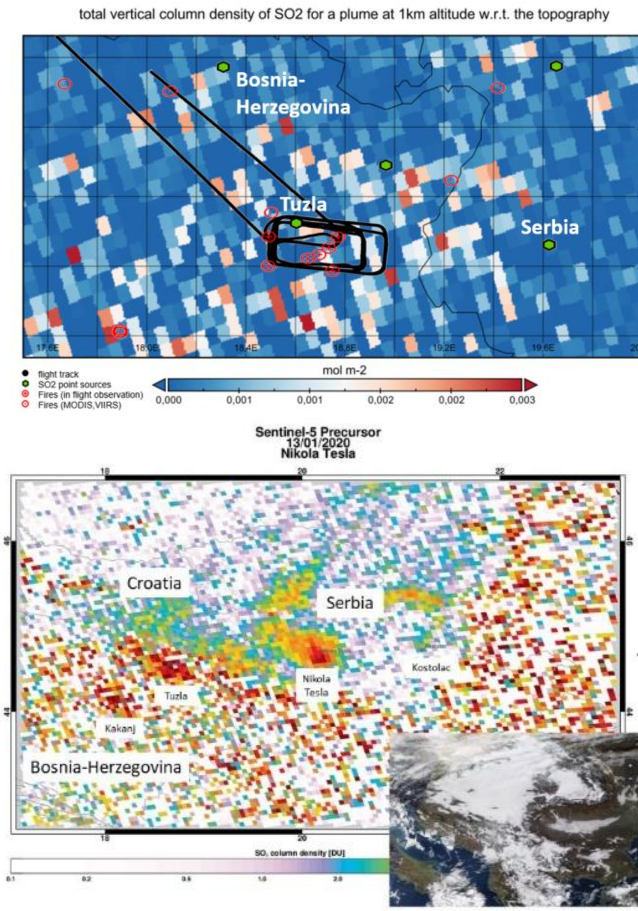


Fig. 6.7. Satellite measurements of SO_2 obtained with the TROPOMI-COBRA retrieval (color-coded unit in mol m^{-2}) including the Falcon flight track (in black) from 2 November 2020 investigating the SO_2 emissions from the Tuzla power plant (top image). In the bottom image, SO_2 obtained from the operational TROPOMI retrieval (color-coded unit in DU) is shown for 13 January 2020 where an extended, triangular snow cover was present over Serbia, as indicated in the lower right satellite cloud image

Сл. 6.7. Сателитска мјерења SO_2 добијена TROPOMI-COBRA узорковања (бојом означена јединица у mol m^{-2}) укључујући стазу лета Falcon (у црној боји) од 2. новембра 2020. која истражује емисије SO_2 из електране Тузла (горња слика). На доњој слици, SO_2 добијен из оперативног узорковања TROPOMI (бојом означена јединица у DU) приказан је за 13. јануар 2020. гдје је над Србијом био присутан проширени троугласти сњежни покривач, као што је приказано на доњем десном сателитском снимку облака

However, the second flight to the Nikola Tesla power plants in Serbia (November 7th, 2020) was less affected by noise in the SO_2 retrieval and was found to be suitable for a validation. To select reliable TROPOMI-SO_2 pixels from the Serbian flight, the SO_2 data was combined with additional TROPOMI products (i.a. QA and CF), as mentioned in Section 6.2. Reliable SO_2 pixels were defined when QA≥50 and CF≤0.10. Presently the TROPOMI/S5P pixel size is 3.6 x 5.6 km^2. For the selected validation area, the 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. 6.8**)**.

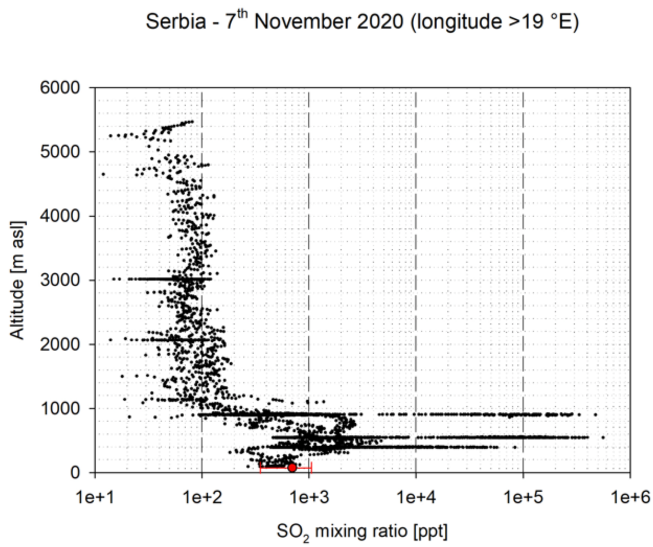


Fig. 6.8. Vertical SO_2 profile from the DLR Falcon-20 flight over Serbia on 7th November 2020 (black dots). Superimposed is the daily mean value and standard deviation (red dot with bar) of the ground-based SO_2 measurements in Obrenovac (76 m asl), located near the Nikola Tesla power plants, on 7th November 2020. The SO_2 mixing ratios are given in parts per trillion (ppt)

Сл. 6.8. Вертикални профил SO_2 са лета DLR Falcon-20 flight изнад Србије 7. новембра 2020 (црне тачке). Приказани су слојеви (layers) са вриједностима средње дневне вриједности и стандардне девијације (црвена тачка са цртицом) приземних мјерења SO_2 у Обреновцу (76 м.н.в.), који се налази у близини електране Никола Тесла, 7. новембра 2020. Односи мијешања SO_2 су дати у дијелова на трилион (ppt)

From this figure 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 ~150-300 m), which was also confirmed by HYSPLIT and MECO(n) (>90%).

Therefore, the airborne measurements were vertically interpolated between these layers and then integrated over the inversion layer to receive VCD values comparable with satellite measurements. Both the airborne measurements and the ground-based measurements from the nearby site Obrenovac and HYSPLIT indicate that the contribution to the VCD from the layer below ~400 m was less important on this day (note the logarithmic scale).

6.4. 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. 6.9. (left).

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/NTB power plants are advected with the wind to the northwest and are distinguishable from each other. Unfortunately, one of the two airborne SO_2 instruments (CI-ITMS) had problems to measure the unexpectedly high SO_2 mixing ratios inflight (as confirmed later by laboratory test). Such time sequences were replaced by measurements from the second SO_2 instrument onboard (Thermo Scientific 43i).

In Fig. 6.9. (right), several TROPOMI/S5P products have been added to the airborne measurements. Noticeable is the agreement of the location of the highest SO_2 values for the airborne and spaceborne TROPOMI 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 only the westernmost part of the flight (including the NTB, but not NTA plume) is suitable for a validation. This part of the flight was also flown at three different vertical levels, as mentioned in Section 6.3.

Therefore, 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.

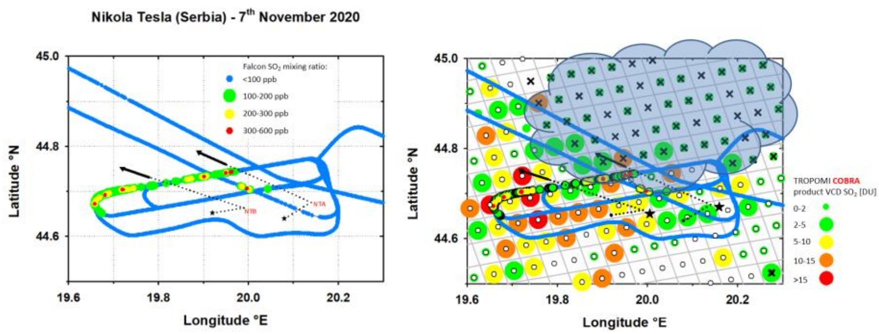


Fig. 6.9. Left: SO_2 mixing ratios (in ppb) along the DLR Falcon-20 flight track in vicinity of the power plants Nikola Tesla A (NTA in red) and B (NTB also in red) on 7th November 2020. The dotted lines with a star at the end indicate the measured wind direction at the height of the power plant chimneys (~150-300 m): 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 southeast. Right: Underlaid below the flight track is the corresponding TROPOMI-SO_2 COBRA product in DU (also color-coded). 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

Сл. 6.9. Лијево: Односи мијешања SO_2 (у ppb) дуж лета DLR Falcon-20 у близини електрана Никола Тесла А (NTA црвено) и Б (NTA такође црвено) 7. новембра 2020. Испрекидане линије са звјездицом на крају показују измјерени смјер вјетра на висини димњака електране (~150-300 m): сјевероисточни ка источни. Испрекидане линије са стрелицом на крају означавају смјер вјетра мјерен на нивоима лета Фалкон низ вјетар од електрана (~540-900 m); претежно са југоистока. Десно: испод стазе за летење је одговарајући TROPOMI-SO_2 COBRA производ у DU (такође означен бојом). Поред тога, фракција облака (CF) је суперпонирана као црни крстови (CF≥0,20) и као бијели кругови (CF<0,20), а схематски је приказан положај остатка облака из јутарње магле

6.5. Discussion

The final step of this first attempt of a validation of spaceborne SO_2 measurements with airborne SO_2 measurements over the Western Balkan region also includes the intercomparison with model simulations, as introduced in Section 6.2. In Fig.

6.10. results for the flight to Serbia on November 7th, 2020, is presented for the HYSPLIT, WRF-Chem and MECO(n) models. In the westernmost part of the flight track, where reliable TROPOMI and Falcon measurements are available, the models indicate a SO_2 vertical column density (VCD) in the range of 2.45-11.0 DU.

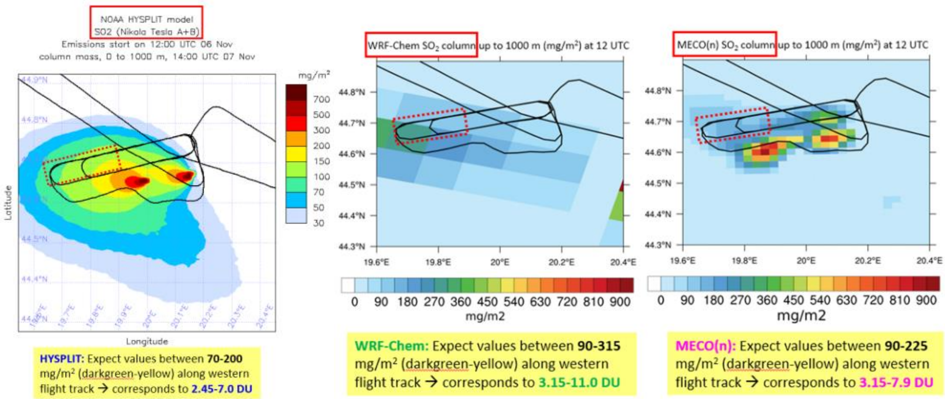


Fig. 6.10. SO_2 model simulations for the flight to the Nikola Tesla power plants in Serbia on November 7th, 2020, (left: HYSPLIT, center: WRF-Chem, right: MECO(n)). The red dotted box indicates the area of interest downwind of the power plants.

Сл. 6.10. Симулације модела SO_2 за лет до електрана Никола Тесла у Србији 7. новембра 2020. (лијево: HYSPLIT, средина: WRF-Chem, десно: MECO(n)). Црвена тачкаста кутија означава област интересовања низ вјетар од електрана.

Next the SO_2-results from the Falcon vs. TROPOMI measurements and the model simulations are combined in Fig. 6.11. (top) to receive an overall picture. Both VCD-SO_2 values obtained from airborne (Falcon-20, y-axis) and spaceborne (TROPOMI) measurements (COBRA retrieval, x-axis) are shown on the basis of the selected 7 pixels (Fig. 6.9), indicating a clear positive correlation (r=0.70). However, the number of data points is unfortunately rather limited, since only 7 reliable TROPOMI pixels (QA≥50 and CF≤0.10) with coincidental Falcon measurements at 3 vertical levels are available (needed for the calculation of representative VCDs).

In Fig. 6.11. (bottom) the SO_2 mean±std values of all 7 selected data points (Falcon/TROPOMI-COBRA) in Fig. 6.11. (top) were determined in addition to the values for the TROPOMI operational product. For TROPOMI, the value for the COBRA product is slightly lower (~10%) than for the operational product (due to less noise and lower biases in COBRA). In addition, the determined airborne

Falcon-SO_2 values are slightly lower (also ~10%) than the obtained COBRA values, though the Averaging Kernels (AK) have been applied to the airborne data set.

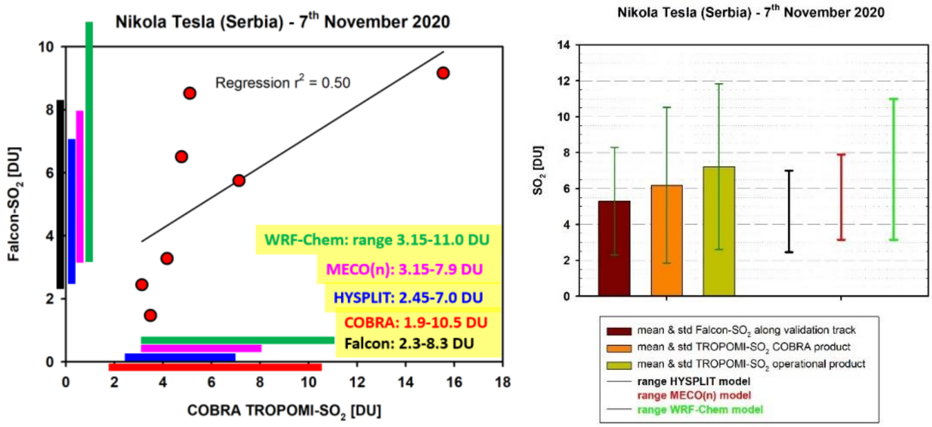


Fig. 6.11. Validation of the TROPOMI-SO_2 retrieval based on the COBRA algorithm with vertical column density (VCD) estimates in DU from airborne SO2-measurements obtained from the DLR Falcon-20 in vicinity of the Nikola Tesla B power plant on 7th November 2020 (top). For model simulations the ranges obtained from Fig. 6.10. are given. For the TROPOMI and Falcon-20 measurements the range of the mean values ± standard deviation of the 7 selected TROPOMI pixels (top figure) is given in the second figure (bottom) and shown as ranges (columns) in the top figure. Averaging kernels (AK) have been applied to the Falcon-SO_2 data set

Сл. 6.11. Валидација узорковања TROPOMI-SO_2 заснованог на COBRA алгоритму са проценама густине вертикалних стубова (VCD) у DU из ваздушних мјерења SO_2 добијених са DLR Falcon-20 у близини електране Никола Тесла Б 7. новембра 2020. (горе). За симулације модела дати су опсези добијени са Сл. 6.10. За TROPOMI и Falcon-20 мјерења опсег средњих вриједности ± стандардна девијација 7 изабраних TROPOMI пиксела (горња слика) је дат на другој слици (доље) и приказан као опсези (колоне) на горњој слици. Усредњавајућа језгра (АК) су примијењена на Falcon-SO_2 скупу података

To quantitatively compare satellite measurements with airborne measurements and model results, the AKs have to be calculated and applied (Theys et al. 2021a). The reason for this step is that the sensitivity of the instrument varies with height, due to the interaction of e.g. spectroscopy, optical characteristics (e.g. Rayleigh scattering), radiative transfer and spectral response with atmospheric properties. Furthermore, various interference factors attenuate the signal strength, as the thermal and chemical composition of the atmosphere (e.g. interferences from O_3 absorption in the UV), aerosols and the surface albedo (Krotkov et al. 2008). The AK describes the profile sensitivity (low values = low sensitivity, e.g. for SO_2 at ground level) and has to be applied (=multiplied) to the airborne measurements (and model results) to mimic the altered signal that the satelliteborne instrument observes. For the relevant pressure levels (900-950 hPa = 500-900 m), where the inversion layer with the elevated SO_2 mixing ratios was located (Fig. 6.5), the AK values of the most relevant TROPOMI pixels (certainly not influenced by clouds: pixel number 4-8), vary between 1.0-1.3 (Fig. 6.12).

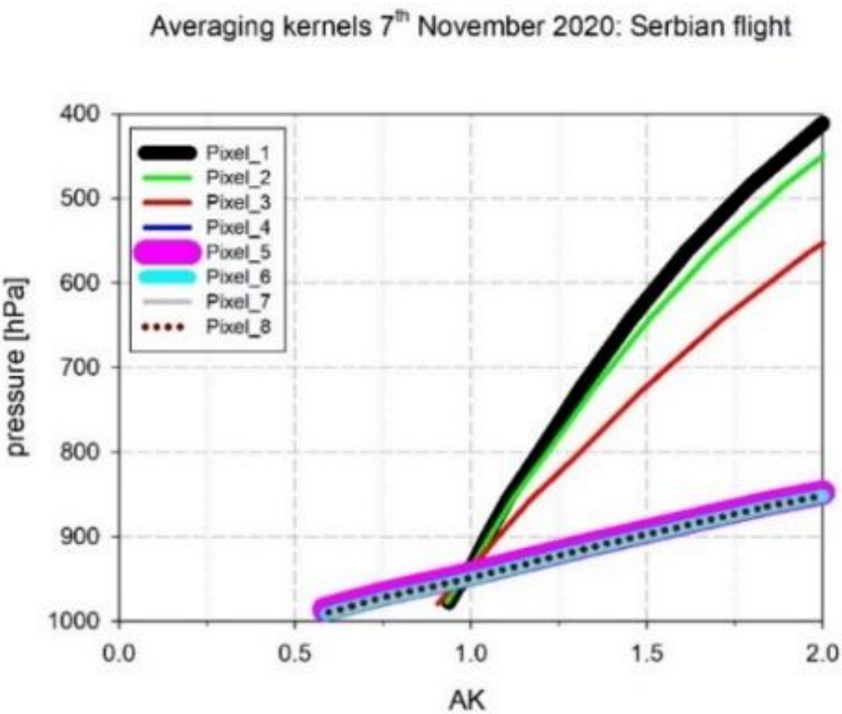


Fig. 6.12. For eight selected TROPOMI pixels from the Falcon flight to Serbia on November 7th, 2020, the vertical distribution (with pressure) of the Averaging Kernels (AK) is shown.

Сл. 6.12. За осам одабраних TROPOMI пиксела са лета Falcon за Србију 7. новембра 2020. приказана је вертикална дистрибуција (са притиском) просјечних језгара (АК).

By applying (=multiplying) these AK values to the airborne Falcon-SO_2 measurements, SO_2-VCDs in a similar range as obtained from satellite measurements were achieved (on average only 10% difference to COBRA, Fig.

6.11). No striking differences between model results – airborne measurements – satellite measurements were observed meaning that the first validation attempt looks promising (used source strength of NTB 83.500 tons SO_2/yr in the models seems to agree with measurements). The presented data set is though very sparse due to the lack of more reliable TROPOMI pixels. This would require measurements in a different season when noisy TROPOMI signals are less pronounced (not winter) and when an extended area, where the power plants are located, is cloudfree to allow for a more detailed probing.

6.6. Conclusion and outlook

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 attempt to conduct a satellite validation (here TROPOMI-SO_2 instrument on Sentinel-5 Precursor) with airborne SO_2 measurements for the power plants in this region to verify the quality of satellite measurements monitoring the pollution situation in the Western Balkans.

We found that:

- a satellite validation is difficult in winter due to the low sun position and the frequent, widespread occurrence of smoke clouds causing noise in the TROPOMI-SO_2 signal (less pronounced in the COBRA retrieval compared to the operational retrieval and less on days with extended snow cover),
- however, a satellite validation with airborne in situ measurements is partly simplified due to the presence of pronounced inversion layers, facilitating the perception of the vertical extension of emitted SO_2 power plant plumes, an essential parameter needed to estimate the Vertical Column Density (VCD) of SO_2 from airborne measurements for the validation with TROPOMI measurements,
- for the Falcon flight to Serbia on 7th November 2020, a coarse validation of the SO_2 plume from the Nikola Tesla B power plant was feasible, despite the availability of only few TROPOMI data points associated to a number of uncertainties, and despite the time difference of 2-3 h between the satellite overpass and the airborne measurements,
- for the Falcon flight to Bosnia-Herzegovina on 2nd November 2020, a validation of the SO_2 plume from the Tuzla power plant was not feasible, because of the presence of too noisy TROPOMI-SO_2 signals (caused by smoke clouds from coal heating and other local fire acivities).

We learned that:

- in the Western Balkan region satellite instruments measuring SO_2 from space (here TROPOMI investigated) have considerable difficulties to capture the pollution situation in the winter season compared to the summer season,
- satellite measurements require cloudfree meteorological sceneries, which are rare in winter and frequently disrupted by man-made smoke clouds,
- satellite validation with airborne measurements is generally a huge challenge in the Western Balkan region: during the METHANE-To-Go-Europe field campaign described here the conditions were in addition hampered by the COVID-19 pandemic, since no stop-overs for the aircraft were possible in this region and the duration of the airborne measurements in the region was very short compared to the total flight time, which prohibited a more complete validation with satellite measurements,
- flight permission to conduct flights with a German research aircraft at very low altitude in vicinity of power plants is very difficult to receive in Western Balkan countries: in Serbia the METHANE-To-Go-Europe team finally succeeded through the support by local scientists; in Bosnia-Herzegovina the flight near the Tuzla power plant was performed as several so-called “missed approaches” to the Tuzla airport,
- flight planning was complicated by the unreliability of the meteorological wind forecasts between flight plan submission (1-2 days in advance of the flight) and the performance of the flights: HYSPLIT-SO_2 forecasts helpful,
- finally, the foreseen airborne instrument for SO_2 (CI-ITMS) was not capable of measuring the extremely high SO_2 mixing ratios (up to several hundred ppb = nmol mol^{-1}) observed ~20-40 km downwind of the investigated power plants, fortunately measurements from a less precise SO_2 backup instrument onboard were available.

Outlook:

- More airborne SO_2 measurements of this kind are needed in the Western Balkan region to improve satellite algorithms for SO_2,
- a new measurement technique for airborne probing of point sources (as power plants) has been set up by the METHANE-To-Go-Europe team and successfully tested in 2022 in vicinity of coal mines in Poland, consisting of a helicopter sonde (HELiPOD) flown in a wire below a helicopter (local helicopter companies can be hired, which simplifies the flight permission process considerable, furthermore flights closer to the targets are possible compared to aircraft). Next measurements on sulphur dioxide and methane emissions from oil and gas activities are planned in Oman.

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Емисије сумпор-диоксида (SO_2) из термоелектрана на угаљ у Србији и Босни и Херцеговини: први покушаји валидације резултата сателитског in situ мјерења у ваздуху помоћу TROPOMI

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Сажетак

Регион западног Балкана познат је по алармантним количинама сумпор-диоксида (SO_2) емитованог у ваздух из термоелектрана на угаљ. Иако је посљедњих година донесен велики број прописа у области заштите животне средине који се односе на рјешавање овог проблема (нпр. о системима за одсумпоравање, о изградњи модерних електрана), вриједности концентрација SO_2 и даље су много веће од прописаних.

За праћење растућег загађења у региону западног Балкана, дјелимично узрокованог великом потражњом за енергијом извана (западна Европа), потребне су разноврсне анализе. Неколико од десет највећих загађивача SO_2 у Европи налази се у Босни и Херцеговини и Србији.

У овом поглављу представљен је рад о првим in situ мјерењима сумпор-диоксида у овом региону спроведеним у ваздуху помоћу њемачког истраживачког авиона. Сарађивали су научници из Босне и Херцеговине и Србије. За мјерења, у касну јесен и почетком зиме 2020. године, изабране су двије термоелектране на угаљ са највећим емисијама наведеног загађивача: „Тузла“ у Босни и Херцеговини и „Никола Тесла“ у Србији. Мјерења су углавном вршена у граничном слоју (испод ~1 km надморске висине). Мјерењем низ вјетар од електрана установљене су изузетно високе вриједности SO_2, које прелазе 100 јединица на милијарду (ppb = nmol mol-1). Мјерење је вршено на удаљености ~20-40 km од извора. Облаци дима са садржајем SO_2 из електрана задржавали су се у добро дефинисаним инверзионим слојевима између ~500-1000 m надморске висине. Резултати оваквих проучавања могу се користити за валидацију синхроних мјерења SO_2 у свемиру помоћу TROPOspheric Monitoring Instrument (TROPOMI) onboard the Sentinel-5P satellite.

Након разилажења облака који су често у одређеном периоду покривали дијелове посматраних територија у хладнијим мјесецима, могла су се извршити мјерења изнад Термоелектране „Никола Тесла" помоћу TROPOMI – SO_2. Урађена је и валидација, јер је мјерење извршено кад није било облачно вријеме, док су била добро дефинисана вертикална проширења сондираног облака дима са SO_2 (потребно за процјену густине вертикалне колоне (VCD), мјерено сателитом). Иначе, ова мјерења у ваздуху, праћена симулацијама модела, могу се користити за одређивање јачине емисије SO_2 из термоелектрана и затим за упоређивање са извјештајима издатим од оператера електране. Овим радом указано је на умјерену усклађеност између мјерења у ваздуху, резултата модела, инвентара емисија и сателитских мјерења за Термоелектрану „Никола Тесла".

Кључне ријечи: Емисије сумпор-диоксида, термоелектране на угаљ, мјерења у ваздуху, сателитска валидација, Балкан