First airborne in situ SO₂ observations of two coal-fired power plants in Serbia and Bosnia-Herzegovina: Potential for top-down emission estimate and satellite validation

<u>Theresa Klausner</u>¹, Heidi Huntrieser¹, Heinfried Aufmhoff¹, Robert Baumann¹, Alina Fiehn¹, Klaus-Dirk Gottschaldt¹, Pascal Hedelt², Predrag Ilić⁵, Patrick Jöckel¹, Sanja Mrazovac Kurilić³, Diego Loyola², Ismail Makroum¹, Mariano Mertens¹, Zorica Podraščanin⁴, Nicolas Theys⁶ and Anke Roiger¹,

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¹Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
 ²Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Methodik der Fernerkundung, Oberpfaffenhofen, Germany
 ³University Union-Nikola Tesla, Faculty of Ecology and Environmental Protection, Belgrade, Serbia
 ⁴University of Novi Sad, Department of Physics, Faculty of Sciences, Novi Sad, Serbia
 ⁵Institute for Protection and Ecology of the Republic of Srpska, Banja Luka, Bosnia-Herzegovina
 ⁶Royal Belgian Institute for Space Aeronomy (BIRA-IASB), Atmospheric trace gases, Brussels, Belgium



Feel free to contact me!

Knowledge for Tomorrow

Outline

- 1) Global and regional sulfur dioxide (SO₂) emissions
- SO₂ observations and modeling activities in the vicinity of two coal-fired power plants in Serbia and Bosnia-Herzegovina
 - 2.1) Airborne research campaign METHANE-To-Go-Europe
 - 2.2) Sentinel-5 Precursor (S5P) with **TROPOMI** instrument
 - 2.3) Ground-based measurement network
 - 2.4) Global/regional nested chemistry climate model MECO(n)
 - 2.5) Regional online coupled **meteorology-atmospheric chemistry model** WRF-Chem
- 3) First **results**
 - 3.1) Serbia mission flight3.2) Bosnia mission flight
- 4) Summary and Outlook













1) Global and regional sulfur dioxide (SO₂) emissions





1) Global SO₂ emissions

- Globally ~2/3rd of SO₂ are directly emitted by anthropogenic activities, mainly by combustion of sulfurrich fossil fuels; ~1/3rd is indirectly emitted (sulfur containing gases) by natural sources and oxidized to SO₂ (Theys et al., 2015)
- Shutdown of older power plants, switching from coal to natural gas and/or installation of flue gas desulfurization systems led to severe reductions in western Europe, throughout northern America and Russia (Klimont et al., 2013)
- SO₂ emissions increased in Europe in the Balkan region, as well as in India and eastern China







1) Regional SO₂ emissions in the Balkan

- SO₂ emission hotspot in Serbia and Bosnia-Herzegovina was recently also observed from space by TROPOMI and OMI (Fioletov et al., 2020)
- The two coal-fired power plants Nikola Tesla (Serbia) and Tuzla (Bosnia), with no or incomplete SO₂ removal, belong to the ten strongest SO₂ emitters in Europe (Chronic Coal Pollution)
- According to the World Health Organization, both countries are among the most polluted ones in Europe
- Tuzla and surrounding cities are also among the ten most polluted cities in Europe based on PM2.5 (www.NZZ.ch)

TROPOMI and OMI mean SO₂ (April 2018 to March 2019) smoothed by oversampling/pixel averaging with R=30 km. Black dots indicate SO₂ emissions sources. (Fioletov et al., 2020)



Top 10 largest SO₂ emitters among power plants in 2016 (Chronic Coal Pollution)





TROPOMI = TROPOspheric Monitoring Instrument; OMI = Ozone Monitoring Instrument

2) SO₂ observations and modeling activities in the vicinity of two coal-fired power plants in Serbia and Bosnia-Herzegovina



2.1) Airborne research campaign METHANE-To-Go-Europe

• DLR Falcon 20 aircraft performed 13 flights from October 22nd to November 7th, 2020









- In situ instrumentation:
 - isotopically on-line calibrated Chemical Ionization Ion Trap Mass
 Spectrometer (CI-ITMS) for SO₂
 - complemented by greenhouse gas (CO₂, CH₄), aerosol number concentration, and other short-lived pollutant (CO, NO, NO_v) measurements
- Two flights, on November 2nd and 7th, focused on characterizing the pollution plumes downwind of Tuzla (Bosnia) and Nikola Tesla (Serbia)



2.1) Airborne research campaign METHANE-To-Go-Europe

- **Challenges** during flights to the Balkan region:
 - \circ No stopover to refuel due to Covid-19 \rightarrow limited measurement time
 - \circ Low-flight permit difficult in Balkan \rightarrow performance of "low approach" at nearby airports
 - Target region has to be **cloud free** for satellite validation
 - Changing wind forecasts hamper SO₂ pollution plume forecast by HYSPLIT
 - Extreme high pollutant volume mixing ratios (VMR)
 - HYSPLIT forecast: SO₂ (mean VMR) from selected point sources









15 UTC

2.2) Sentinel-5 Precursor (S5P) with TROPOMI instrument

- ESA satellite Sentinel-5P launched in October 2017
 -multichannel spectrometer TROPOMI (UV-VIS-NIR, SWIR)
 -measuring Vertical Column Density (VCD) of O₃, NO₂, HCHO, SO₂, CO₂, CH₄, aerosols and clouds
- Specifications

 polar orbit, 14-15 Orbits / day
 equator passing: 13:30 h (local)
 swath: 2600 km wide → daily global coverage
 pixel resolution: 5.5 x 3.6 km² (at the beginning: 7 x 3.5 km²)
- SO₂ Retrieval
 - -band 3 (310-405 nm)
 - -besides TROPOMI operational product also COBRA product which improves low SO₂ VCDs (< \sim 0.25 DU)
 - -VCD via air mass factor (AMF), knowledge on the SO₂ vertical profile needed



TROPOMI mean SO₂ (April 2018 to March 2019) smoothed by oversampling techniques with R=30 km. (Fioletov et al., 2020)



0.48

0.8

4.8 DU

0.24



2.3) Ground-based measurement network

					Bosnia-Herzegovina
Country Bosnia- Herzgewonia	Location	Lat °N	Lon °E	Measured species	
5	Caparde (Zvornik)	44.3986	19.0089	SO ₂	Ground-based stations
	Bijeljina	44.7568	19.2185	SO ₂	
	Ugljevik	44.6840	18.9696	NO, NO ₂ , NOx, PM10	Turla
	Skver Tuzla	44.5408	18.6733	SO ₂ , NO, CO, O ₃ , PM2.5	
	BKC Tuzla	44.5319	18.6547	SO ₂ , NO, CO, O ₃ , PM2.5	Google Earth
	Bukinje Tuzla	44.5236	18.6003	SO ₂ , NO, CO, O ₃ , PM2.5	10 km
	Lukavac	44.5333	18.5349	SO ₂ , NO, CO, O ₃ , PM2.5	Serbia
	Živinice	44.4539	18.6483	SO ₂ , NO, CO, O ₃ , PM2.5	
Serbia					
	Obrenovac	44.6595	20.2045	SO ₂ , PM10, NO ₂ , CO	Nikola Tesla
	Kostolac	44.7175	21.1734	SO ₂ , NO ₂	Nikola Tesla B _{Nikola} tesla A
					Google Earth



2.4) Global/regional nested chemistry climate model MECO(n)

- On-line coupling of the global CCM EMAC with regional CCM COSMO-CLM/MESSy → regional refinement (e.g. Hofmann et al., 2012; Kerkweg and Jöckel, 2012a and 2012b; Mertens et al., 2016)
 - EMAC
 - nudging (operational ECMWF) horizontal resolution ~280 km, 90 vertical levels time step 720 s
 - 3 COSMO-CLM/MESSy nests

nudging (optional) horizontal resolution 50 / 12 / 2 km, 50 vertical levels time step 240 / 60 / 30 s

- Simulation of various anthropogenic SO₂ emissions
 - P-SO2: modified point source emissions from E-PRTR (2016)
 - E-SO2: EDGAR v5.0 gridded inventory (2015)
 - O-SO2: OMI-HTAP v2.2 gridded inventory (2010)



CCM = chemistry-climate model; MECO(n) = MESSy-field ECHAM and COSMO models nested n times; EMAC = ECHAM/MESSy Atmospheric Chemistry; COSMO-CLM/MESSy =COSMO model in CLimate Mode / Modulet Earth Submodel System; E-PRTR=European Pollutant Release and Transfer Register; EDGAR=Emissions Database for Global Atmospheric Research; OMI-HTAP=OMI-based and HTAP emission inventory





2.5) Regional online coupled meteorology-atmospheric chemistry model WRF-Chem

- High model resolution of 10 x 10 km and 151 × 151 grid points
- Center of domain: 45.25°N and 19.85°E
- NOx, SOx, PM2.5, PM10 and NH₃ emissions from EMEP (10x10 km) for all anthropogenic SNAP sectors
- Used as forecast and analysis simulation



EMEP = European Monitoring and Evaluation Programme, SNAP = selected nomenclature for reporting of air pollutan DLR.de • Chart 13 > EGU 21-5912 > Klausner et al. • vPICO presentation > April 28, 2021

3) First results





DLR

3) Effect of low sun position and smoke aerosols on TROPOMI products



November to January: very noisy (ground-based smog due to usage of low quality coal) \rightarrow hampers validation

3.1) Falcon flight downwind of Nikola Tesla (Serbia)



3.1) Falcon flight downwind of Nikola Tesla (Serbia)

• Detailed vertical profile indicates that SO_2 is mainly located in boundary layer (BL) \rightarrow important knowledge for VCD calculation • First comparison with WRF-Chem indicates more easterly wind (-25°) and lower SO₂ mixing ratios





3.1) Operational TROPOMI and COBRA products around Nikola Tesla (Serbia)

- Operational and COBRA products are different → challenging for validation with airborne measurements
- Flight track was designed to allow for satellite evaluation in space and time; due to fog the flight was delayed by two hours and took place from ~13:30 to 14:30 UTC



*COBRA is mostly meant to improve the low SO_2 VCDs (< ~0.25DU) and therefore not performing the retrievals for the alternative windows (325-335 nm and 360-390nm) as in the operational algorithm



3.1) Combination of Falcon flight & TROPOMI around Nikola Tesla (Serbia)



Potential steps for validation of emission plume (left chimney):

- Estimate VCD-SO₂ from Falcon
- Combine with simulated SO₂ to determine total VCD-SO₂ in designated area indicated by "
- Compare with VCD-SO₂ from TROPOMI

3.2) Falcon flight downwind of Tuzla (Bosnia)

- Flight pattern on November 2nd, 2020
 - low approach at Tuzla airport (I)
 - "box" at different heights without (2.5, 1.2 km; II) and with (1.1, 0.9, 0.8, 0.6 km; III) **SO₂ enhancements of up to 100 ppb**





3.2) Falcon flight downwind of Tuzla (Bosnia)

- Mass balance for rectangle (in red) to calculate instantaneous SO₂ flux [kg s⁻¹]
- Airborne measurements grouped into bins of 300 x 100 m (horiz. x vert.), interpolated between flight legs, lowest flight leg valid down to ground

$$F = \iiint \overrightarrow{U}(\theta, z) \sin(\alpha) \cdot (C(\theta, z) - C_{\text{bg}}) L \, d\theta \, dz,$$

ppb to g m⁻³ by ideal gas law; integrated in horizontal and vertical; L=difference between two points on regular grid of 300x100 m, U=wind speed, α =wind angle

Interpolated measurements along red line: from north to east, south and west



 Preliminary flux ~3.8 kg s⁻¹, which is a factor ~2 higher than average emissions from EDGAR, OMI HTAP and E-PRTR of 1.8 kg s⁻¹



3.2) Ground-based measurements at Tuzla (Bosnia)



- "Cluster" detects SO₂ mixing ratios at similar magnitude as observed aboard Falcon
- Lukavac latest in time, but wind direction changes during the day

3.2) MECO(n) simulations of Tuzla (Bosnia)



Clean inflow
Uniform ver

Uniform vertical $P-SO_2$ distribution in boundary layer \rightarrow consistent with mass balance assumption

 Ongoing effort to minimize model bias in temperature, relative humidity and wind direction





3.2) COBRA product around Tuzla (Bosnia)

- Both, COBRA and the operational product, are **noisy** (lots of smaller fires which influence the sensitivity due to enhanced amount of aerosol)
- Useful validation needs to be further examined



COBRA (orbit at 11:32 UTC)

4) Summary and Outlook



4) Summary and Outlook



- Airborne in situ SO₂ measurements were for the first time successfully conducted downwind of two major coal-fired power plants in Serbia and Bosnia-Herzegovina
- Two TROPOMI underpass validation flights were conducted and revealed important lessons learned for future flights
- Validation of **TROPOMI-SO₂** with Falcon-SO₂ looks promising for the flight in Serbia
- Mass Balance estimate of Falcon-SO₂ looks promising for the flight in Bosnia
- Improvements on Falcon-SO₂ data quality of extreme high SO₂ mixing ratios
- Ongoing refinements on mass balance estimation, MECO(n) and WRF-Chem model simulations
- Comparison of SO₂ Vertical Column Densities over designated areas using TROPOMI and combined Falcon-MECO(n)-SO₂ data
- Future TROMOPI-SO₂ validation flights envisaged within the framework of METHANE-To-Go





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Additional slides





Cloud Situation in the morning over Croatia, Bosnia & Serbia – 7 November 2020



Flight to Nikola Tesla A (left) & B (right) – 7 November 2020 at 13:23 UTC (lat 44.89°N, lon 19.76°E)



Flight to Nikola Tesla A (right) & B (left) – 7 November 2020

Ν



