Study of $SO_2$ pollution in the Middle East using MERRA-2, CAMS data assimilation products, and high-resolution WRF-Chem simulations

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Key Points:

- Anthropogenic emissions and their vertical distribution are the critical factors for simulating air-pollution
- Fine-resolution WRF-Chem simulations with the most accurate $SO_2$ "top-down" emissions agree well with observations
- In Riyadh and Jeddah, the European $SO_2$ air-quality limit (125 $\mu g/m^3$) is exceeded in about 75 days per year

Key Words: OMPS, OMI, Anthropogenic Emissions, Air-quality, Arabian Peninsula, GEOS-5, OMI-HTAP

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Abstract

Oil recovery, power generation, water desalination, gas flaring, and traffic are the main contributors to \( SO_2 \) emissions in the Middle East (ME). Satellite observations suggest that the traditional emission inventories do not account for multiple \( SO_2 \) emission sources in the ME. This study aims to evaluate the most frequently used \( SO_2 \) emissions datasets over the ME by comparing high-resolution regional model simulations and meteorology/chemistry assimilation products, MERRA-2 and CAMS, with satellite and available ground-based air-quality observations.

Here, we employ the WRF-Chem-3.7.1 regional meteorology-chemistry model and conduct simulations for the period 2015-2016 with 10 km grid spacing using HTAP-2.2 emission datasets and the new OMI-HTAP data, which is based on the combination of the near-surface \( SO_2 \) emissions taken from the HTAP-2.2 inventory with strong (>30 kt/year) \( SO_2 \) point sources obtained from the satellite Ozone Monitoring Instrument (OMI) observations.

We find that conventional emission inventories (EDGAR-4.2, MACCity, and HTAP-2.2) have uncertainties in the location and magnitude of \( SO_2 \) sources in the ME and significantly underestimate \( SO_2 \) emissions in the Arabian Gulf. The WRF-Chem, run in conjunction with the new OMI-HTAP emissions, improves comparisons between the satellite and ground-based \( SO_2 \) observations. Our simulations show that \( SO_2 \) surface concentrations in Jeddah and Riyadh frequently exceed European air-quality limits.

The ME generates about 10% of global anthropogenic \( SO_2 \) emissions, on par with India. Therefore, the development of effective emission controls and improvement of air-quality monitoring in the ME are urgently needed.

1 Introduction

Atmospheric pollutants, both of natural and anthropogenic origin, have a considerable negative impact on human health and can cause premature mortality (Lelieveld, Evans, et al., 2015; Lelieveld et al., 2019). According to the World Health Organization (WHO), outdoor air pollution caused 4.2 million premature deaths worldwide in 2016 (WHO, 2018).

Rapid economic and population growth, urbanization, industrialization, excessive traffic emissions and burning of fossil fuels are the main causes of dramatically deteriorating air-quality in developing countries (Janssens-Maenhout et al., 2015). On-going climate change will only exacerbate this problem (Leung & Gustafson Jr, 2005). Along with particulate matter (PM), sulphur dioxide (\( SO_2 \)) is one of the most common pollutants. \( SO_2 \) is a toxic gas produced by burning sulfur-containing fossil fuels, and it is known to cause inflammation of human airways. In order to protect human health, the WHO have issued air-quality guidelines (WHO, 2006) recommending that daily average \( SO_2 \) surface concentrations should not exceed 20 \( \mu g/m^3 \). This limit is quite restrictive relative to most national air-quality requirements. The European Union Air Quality Directive (EUEA, 2008) limits \( SO_2 \) daily averaged surface concentration at 125 \( \mu g/m^3 \) and a number of exceedances of this limit during the year should not be more than three. According to the air quality standards issued by the Presidency of Meteorology and Environment (PME) of the Kingdom of Saudi Arabia (KSA), daily averaged \( SO_2 \) surface concentrations should not exceed 365 \( \mu g/m^3 \) more than one time per year. KSA PME sets annual average limit to 80 \( \mu g/m^3 \) (PME, 2012).

Not only is \( SO_2 \) a harmful and toxic gas, it is also photochemically converted in the atmosphere into sulfate aerosol, which contributes to PM mass. Particulate sulfate also has significant adverse effects on human health (Lelieveld, Evans, et al., 2015; Lelieveld...
et al., 2019) and ecology, and impacts the Earth’s radiation balance (Charlson et al., 1992; C. C. Chuang et al., 1997; Myhre et al., 2013; Ramanathan et al., 2005).

Ground-based air-quality monitoring networks over the ME are sparse. However, high levels of \( \text{SO}_2 \) can be detected from space using satellite based instruments. In addition, modern data assimilation products that provide 3D atmospheric chemistry and aerosol fields can be employed to assess air-quality in the ME. We use here the Modern-Era Retrospective Analysis for Research and Applications (MERRA-2) (Randles et al., 2017; Buchard et al., 2017) developed at National Aeronautic and Space Administration (NASA), and the Copernicus Atmosphere Monitoring Service (CAMS) operational analysis (CAMS-OA) (Flemming et al., 2015; Inness et al., 2015) developed at the European Centre for Medium-range Weather Forecast (ECMWF). The MERRA-2 (https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2) and CAMS-OA (https://atmosphere.copernicus.eu/) global products have lower spatial resolution in comparison with regional models, and therefore could smooth concentration extremes. Although the aerosol total column loading is corrected by assimilating the aerosol optical depth (AOD), gaseous concentrations are generally not assimilated by either CAMS-OA or MERRA-2.

To calculate the spatial-temporal distribution of atmospheric pollutants, emissions of \( \text{SO}_2 \) and other species must be provided to the free running or data assimilating models. To quantitatively evaluate air-quality accurately, emissions must be correctly characterised and represented. However, conventional emissions inventories, such as the Emissions Database for Global Atmospheric Research version 4.2 (EDGAR-4.2) (Janssens-Maenhout et al., 2013), Hemispheric Transport of Air Pollution version 2.2 (HTAP-2.2) (Janssens-Maenhout et al., 2015), and Monitoring Atmospheric Composition and Climate CityZen (MACCity) (Granier et al., 2011), are based on countries emissions reports. These reports are often incomplete or outdated, particularly in developing countries, creating major uncertainty in the data. This is especially true in the ME (McLinden et al., 2016).

Therefore, a comparison of model outputs with reanalysis data and their thorough test against available observation is essential. C. Li, McLinden, et al. (2017) also pointed out that substantial uncertainties exist in the estimated emissions, especially for regions experiencing rapid changes in the economy such as China and India. For instance, a comparison of different anthropogenic emission inventories using WRF-Chem over South-east and South Asia has been performed in Amnuaylojaroen et al. (2014) and Sharma et al. (2017). Amnuaylojaroen et al. (2014) showed that the uncertainties of carbon monoxide surface mixing ratios in simulations with different anthropogenic emission inventories reach 30%. Sharma et al. (2017) showed that the modeled ozone mixing ratios during noontime are sensitive to the choice of an emission inventory.

Advances in satellite measurements have yielded new data and techniques that help to evaluate and improve conventional inventories (“bottom-up” approach). In particular, McLinden et al. (2016); Fioletov et al. (2016) developed a novel ”top-down” method for mapping the major \( \text{SO}_2 \) emission sources using \( \text{SO}_2 \) column loadings for the period 2005-2016 from the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006; C. Li et al., 2013). They released a catalog of \( \text{SO}_2 \) point emissions, where they identified more than 500 point sources (some of which are not present in the conventional EDGAR-4.2 and HTAP-2.2 emission datasets), with annual \( \text{SO}_2 \) emission rates ranging from about 30 kt/yr to more than 4000 kt/yr. For example, 14 unaccounted \( \text{SO}_2 \) sources located in the ME were detected, 12 of which are related to oil and gas exploration and refining. F. Liu et al. (2018) combined the point sources identified in Fioletov et al. (2016) with \( \text{SO}_2 \) emissions from the HTAP-2.2 inventory to develop the OMI-HTAP dataset. They deployed this dataset in the Goddard Earth Observing System, version-5 (GEOS-5) global atmospheric model (Molod et al., 2015) and tested the GEOS-5 output with the dense \( \text{SO}_2 \) air-quality observations measured over Europe and the US.

In this study we focus on the ME, which is one of the most polluted areas in the world. Because of extremely high levels of natural particulate pollution driven by dust
aerosols (Tsiouri et al., 2015; Kalenderski & Stenchikov, 2016; Jish Prakash et al., 2016; Parajuli et al., 2019), the region experiences extreme pollution episodes, compounded by a strong contribution of anthropogenic aerosols and gases (Lelieveld, Beirle, et al., 2015; Lelieveld et al., 2009; Ahmed, 1990; Barkley et al., 2017; Karagulian et al., 2015; Ukhov et al., 2020). ME emits about 10% of the total global anthropogenic SO$_2$ (Klimont et al., 2013). These emissions could be involved in the monsoon circulation and have a global effect on atmospheric composition (Lelieveld et al., 2018). SO$_2$ conventional emissions inventories have a high level of uncertainty over the ME and air-quality observations are sparse. To our best knowledge, there is no thorough inter-comparison of the emission inventories over the ME so far.

Thus, this study aims to address the following scientific questions:

1. What is the impact of SO$_2$ emissions on air-quality over the ME region and how well SO$_2$ pollution is depicted by assimilation products or free-running models?

2. How do conventional anthropogenic emission datasets (HTAP-2.2, EDGAR-4.2, and MACCity) compare with the newly developed SO$_2$ emission dataset based on the "top-down" approach for the ME region?

3. How do SO$_2$ column loadings and surface concentrations from MERRA-2, CAMS-OA, GEOS-5, and the WRF-Chem model compare with satellite observations and in situ measurements over the ME?

To answer these questions, we calculated SO$_2$ distribution for the period 2015-2016 using the WRF-Chem-3.7.1 model with 10 km resolution. We evaluated the output from the WRF-Chem runs with HTAP-2.2 and OMI-HTAP emissions datasets, GEOS-5 model output, and MERRA-2, CAMS-OA products against the satellite retrievals of SO$_2$ column loadings obtained from the Ozone Mapping Profiler Suite (OMPS) (C. Li, Krotkov, et al., 2017; Zhang et al., 2017) and against in situ observations of SO$_2$ surface concentrations conducted by the Saudi Authorities for Industrial Cities and Technical Zones (MODON) in major cities of Saudi Arabia. Our comparisons show that using WRF-Chem with the OMI-HTAP emissions dataset allows to overcome some deficiencies inherent in the currently available assimilation products, and our approach can capture air pollution patterns at finer spatial resolutions.

The rest of the paper is organized as follows: we describe the satellite retrievals of SO$_2$ column loadings and the assimilation products used in this study and we describe the WRF-Chem model setup in Section 2. In Section 3, we evaluate the SO$_2$ emission datasets used in this study and we compare the SO$_2$ column loadings and surface concentrations obtained from the models with the satellite observations and in situ measurements. We present our conclusions in Section 4.

2 Data, model setup, and data assimilation products

Below, we discuss satellite and ground-based observations, assimilation products, and WRF-Chem model setup.

2.1 SO$_2$ satellite observations

SO$_2$ column loadings are the most widely used observation products. Long-term datasets are currently available from measurements conducted by NASA Earth Observing System (EOS) Aura Ozone Monitoring Instrument (OMI: 2004-current) (Levelt et al., 2006; C. Li et al., 2013) and NASA-NOAA Suomi National Polar-orbiting Partnership (Suomi-NPP) Ozone Mapping and Profiling Suite Nadir Mapper (OMPS-NM: 2011-current) (C. Li, Krotkov, et al., 2017; Zhang et al., 2017).
We use NASA OMPS total column SO$_2$ (OMPS-NPP-NMSO2-PCA) dataset (C. Li, Krotkov, et al., 2017; Zhang et al., 2017). The dataset is processed using principal component analysis (PCA) SO$_2$ retrieval algorithm, which is consistent with the OMI operational product (OMS02) (C. Li et al., 2013). The OMPS pixel size is 20×20 km$^2$, but we use here the gridded (0.5°×0.5°) monthly average SO$_2$ columns obtained assuming fixed planetary boundary Layer (PBL) SO$_2$ profile shape. Although due to its lower than OMI spatial resolution OMPS can detect fewer small point SO$_2$ emitters, for large point sources the OMPS-derived "top-down" SO$_2$ emissions are consistent with those from the OMI SO$_2$ catalogue (Fioletov et al., 2016; Zhang et al., 2017).

2.2 CAMS

The Copernicus Atmosphere Monitoring Service has been conducting near real-time analysis of SO$_2$ since July 2012 until present within the Monitoring Atmospheric Composition and Climate (MACC) project (urlhttps://atmosphere.copernicus.eu/). CAMS-OA product has a resolution of 0.8°×0.8° before 21 June 2016 and 0.4°×0.4° after with 60 vertical levels and a top level at 0.1 hPa. CAMS-OA calculates the atmospheric composition using an extended version of the Carbon Bond chemical mechanism 5 (CB05) (Yarwood et al., 2005) that has been implemented in the ECMWF Integrated Forecast System (IFS) (Flemming et al., 2015). CB05 comprises 54 chemical species and 126 chemical reactions complementing the already integrated modules for greenhouse gases and aerosols, including dust, sea salt, sulphate, black carbon and organic matter. CAMS-OA uses anthropogenic emissions from the MACCity inventory (Granier et al., 2011) on a 0.5°×0.5° grid built over the period 1960-2010 (see Fig. 1a). MACCity emissions are based on the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) inventory but has improved seasonal variability (Lamarque et al., 2013).

The generation of sulfate within CAMS-OA is simulated independently of the atmospheric chemistry calculated by CB05. For this purpose, CAMS-OA assumes an independent SO$_2$ tracer from CB05, driven by the same SO$_2$ and dimethyl sulfide (DMS) emissions, and converts them into sulfate aerosols. SO$_2$ oxidation is parameterized using a prescribed latitude-dependent e-folding timescale ranging from 3 days at the equator to 8 days at the poles (Yarwood et al., 2005). CAMS-OA assimilates MODIS optical depth to constrain aerosols and also only significant stratospheric SO$_2$ loadings generated by volcanic eruptions. Thus, tropospheric SO$_2$ remains largely unconstrained.

2.3 MERRA-2 and GEOS-5

The Modern-Era Retrospective analysis for Research and Applications, version 2 (MERRA-2) (Randles et al., 2017; Buchard et al., 2017) assimilates both meteorological and aerosols/chemistry observations. To simulate atmospheric processes MERRA-2 utilizes the GEOS-5 model (Molod et al., 2015). Both GEOS-5 and MERRA-2 use the Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model (Chin et al., 2002) to simulate dust, sea salt, black and organic carbon, SO$_2$ and sulfate. Hydroxide (OH) for SO$_2$ oxidation is calculated interactively. MERRA-2 assimilates AOD at 550 nm obtained via a neural network retrieval trained on AERONET data. The MERRA-2 aerosol analysis corrects aerosol loadings but does not assimilate any SO$_2$ observations. Thus, SO$_2$ concentrations are completely unconstrained (Randles et al., 2017). The output fields are available on a 0.625°×0.5° latitude-longitude grid and 72 terrain-following hybrid σ – p model layers with the top layer at 0.01 hPa.

MERRA-2 uses EDGAR-4.2 (Janssens-Maenhout et al., 2013) emissions available globally on a 0.1°×0.1° grid for 2008. It comprises emissions of gaseous (SO$_2$, NO$_x$, CO, non-methane volatile organic compounds, NH$_3$), and particulate (PM$_{10}$, PM$_{2.5}$, black and organic carbon) air pollutants. This inventory is based on annual national-level industrial fuel consumption data, agricultural statistical records, empirical information on
technological standards influencing the amount of emissions, and emission reduction measures applied. Spatial source mapping uses a set of proxy location data of industrial, agricultural, transport and urban objects and population density. This approach potentially could cause uncertainty in location and magnitude of emission sources. E.g., we detected in EDGAR-4.2 one “hot-spot” of unknown origin located in the middle of the desert west of Riyadh. It does not appear in the latest emission inventories. An updated version EDGAR-4.3.1 for 2010 (Crippa et al., 2016) was recently released.

2.4 WRF-Chem

To calculate fine-resolution \( \text{SO}_2 \) fields, we use the Weather Research and Forecasting (WRF) community model (Skamarock et al., 2005) coupled with chemistry (WRF-Chem) (Grell et al., 2005). The WRF-Chem model is extensively used for prediction and simulation of weather, dust storms and aerosol interactions with atmosphere and radiation (Cahill et al., 2017; Jish Prakash et al., 2016; Kalenderski & Stenchikov, 2016; Anisimov et al., 2017; Osipov & Stenchikov, 2018; Parajuli et al., 2019). For air-quality research the WRF-Chem model has been used in many regions of the globe: East Asia (Wang et al., 2010), the US (Kim et al., 2006; M.-T. Chuang et al., 2011), Europe (Forkel et al., 2012; Ritter et al., 2013), South America (Archer-Nicholls et al., 2015) and ME (Ukhov et al., 2020).

In this study we employ WRF-Chem-3.7.1 and configure it over the ME. Simulations are carried out for the whole period of 2015-2016, for each month separately, starting from the last week of the preceding month. This week is considered to be a spin-up and is excluded from further analysis. The simulation domain (see Fig. 1) with 450×450 grid points is centered at 28°N, 42°E. We use a 10×10 km² Mercator projection horizontal grid and a 50-level vertical grid with enhanced resolution closer to the ground comprising 11 model levels within the near-surface 1-km-layer. The model top boundary is set at 50 hPa.

To improve the representation of the meteorological fields, we apply spectral nudging (Miguez-Macho et al., 2004) above the PBL (>5.0 km) to horizontal wind components \((u \text{ and } v)\) toward MERRA-2 wind fields. The nudging coefficient for \(u\) and \(v\) is set at 0.0001 s\(^{-1}\). We nudged waves with wavelengths larger than 450 km. This allows to keep the large-scale motions close to “observations”, and let the model freely develop small-scale features.

The aerosol/chemistry initial conditions and boundary conditions (IC&BC) are calculated using MERRA-2 output using the newly developed Merra2BC interpolation utility; see Appendix A1. To be consistent with aerosol/chemistry IC&BC, we also define the meteorological IC&BC using MERRA-2 output (see Appendix A1).

The Regional Atmospheric Chemistry Mechanism (RACM) (Stockwell et al., 1997) containing 77 chemical species and 237 chemical reactions, including 23 photochemical reactions, is used for the modeling of the atmospheric chemistry. It is embedded into WRF-Chem using the Kinetic PreProcessor (KPP) (Damian et al., 2002). The role of KPP is to integrate the system of stiff nonlinear ordinary differential equations, which represents the specified set of chemical reactions. An online computation of the photolysis rates for the 23 photochemical reactions of the RACM gas-phase chemical mechanism is implemented according to Madronich (1987). The set of physical parameterizations used in WRF-Chem runs is presented in Table 1. More details on the physical parameterizations can be found at [http://www2.mmm.ucar.edu/wrf/users/phys_references.html](http://www2.mmm.ucar.edu/wrf/users/phys_references.html).

Similar to MERRA-2, the GOCART aerosol and chemistry module is used to calculate \( \text{SO}_2 \) to \( \text{SO}_4 \) oxidation (Chin et al., 2002) by the hydroxide radical \( \text{OH} \) whose abundance is simulated by RACM.
Table 1. Parameterizations used in WRF-Chem calculations

| Parameterization       | Scheme                      | Namelist option               |
|------------------------|-----------------------------|-------------------------------|
| Surface Layer          | Revised MM5                 | sf_sfclay_physics=1           |
| Land Surface           | Unified Noah                | sf_surface_physics=2           |
| Boundary Layer         | Yonsei University (YSU)     | bl_pbl_physics=1               |
| Cumulus                | Grell 3D Ensemble           | cu_physics=5                  |
| Micro Physics          | WRF single moment           | mp_physics=4                  |
| SW Radiative Transfer  | RRTMG                       | ra_sw_physics=4               |
| LW Radiative Transfer  | RRTMG                       | ra_lw_physics=4               |
| Aerosol/Chemistry      | GOCART/RACM                 | chem_opt=301                  |
| Photolysis             | Madronich                   | phot_opt=1                    |
| Dust emissions         | GOCART                      | dust_opt=1                    |

To evaluate the dependence of $SO_2$ concentrations on emissions, we ran WRF-Chem with two of the latest emission datasets: HTAP-2.2 (Janssens-Maenhout et al., 2015) and the new OMI-HTAP, developed in F. Liu et al. (2018).

### 2.4.1 HTAP-2.2 Emissions

HTAP-2.2 is the most recent gridded emission inventory (Janssens-Maenhout et al., 2015) built for 2008 and 2010 on a $0.1^\circ \times 0.1^\circ$ global grid. It includes anthropogenic emissions of $PM_{10}$, $PM_{2.5}$, $NO_x$, $CO$, non-methane volatile organic compounds (NMVOCs), $SO_2$, $BC$, $NH_3$ and OC.

HTAP-2.2 presents emissions as sector-specific grid maps. The sectors for all substances are defined based on the emission category: international and domestic air traffic, international shipping, industry, transportation and residential. HTAP-2.2 emissions are based on the latest official regional information consistent with air pollutant grid maps from the US Environmental Protection Agency and Environment Canada for North America, European Monitoring and Evaluation Programme (EMEP) (Visschedijk et al., 2009) for Europe, Model Inter-Comparison Study (MIX) (M. Li et al., 2015) for Asia. For most countries of the ME and North Africa, HTAP-2.2 emissions are identical to those defined in the newly released EDGAR-4.3.1 dataset for 2010 (Crippa et al., 2016). The "hot-spot" of unknown origin (see Sec. 2.3) is not present in EDGAR-4.3.1 and in HTAP-2.2 emissions datasets (see Fig. 1).

### 2.4.2 OMI-HTAP Emissions

Fioletov et al. (2016) used OMI observations for the period 2005-2016 to develop a catalog of strong (>30 kt/year) $SO_2$ point emissions of industrial origin. They found that over the ME there is a significant amount of important $SO_2$ sources unaccounted for in the recent HTAP-2.2 emissions inventory. The uncertainties are especially high over the Arabian Gulf, as HTAP-2.2 has a tendency of moving sources from over the Gulf to the nearby coastal areas.

F. Liu et al. (2018) combined $SO_2$ emissions related to the residential and transportation sectors from the HTAP-2.2 inventory with the point emissions catalogue of Fioletov et al. (2016). Namely, in designated areas associated with significant emissions, F. Liu et al. (2018) substituted the industrial emissions in HTAP-2.2 with the emissions from the point sources catalogue. They refer to this $SO_2$ emission dataset as OMI-HTAP. F. Liu et al. (2018) conducted simulations using the global GEOS-5 model (Molod et al., 2015) to test OMI-HTAP dataset. GEOS-5 is the same atmospheric model as in MERRA-2.
but instead of assimilating of meteorological and AOD observations, F. Liu et al. (2018) used a GEOS-5 replay mode when the model meteorological fields are nudged to the MERRA-2 reanalysis fields. They showed that in the US and Europe this new dataset improves comparisons of $SO_2$ surface concentrations with air-quality observations.

Here we apply the OMI-HTAP emissions dataset for 2015-2016 within the WRF-Chem regional model configured for the ME region, where Fioletov et al. (2016); McLinden et al. (2016) found significant uncertainties in the conventional emission inventories. We have to modify OMI-HTAP slightly as we found that the location of the point source associated with the Jeddah power plant was shifted in the catalogue Fioletov et al. (2016). Thus we corrected this error as we know the precise location of this plant.

Original OMI-HTAP (F. Liu et al., 2018) is provided without shipping and aviation emissions. Since shipping emissions are important over the Red Sea and Arabian Gulf we supplemented OMI-HTAP with shipping emissions from HTAP-2.2. The aviation emissions are not accounted for as they are relatively small. Natural sulfur emissions associated with volcanoes are negligible in the ME relative to anthropogenic inputs and thus we do not account for them. Dimethyl sulfide (DMS) emissions are parameterized within the GOCART (Chin et al., 2002). Emissions of all other constituents ($PM$, black and organic carbon, etc.) are taken without any modification from the HTAP-2.2 inventory.

$SO_2$ emissions reported in Fioletov et al. (2016) are annual mean since the catalogue does not provide information on their monthly variability. F. Liu et al. (2018) assigned some plausible seasonal variability to OMI-HTAP, but here we use the OMI-HTAP dataset in the annual mean format.

### Table 2. Inventory of all products/runs and corresponding emission datasets with description of vertical distribution of $SO_2$ emissions

| Product/Run | Emission Inventory for $SO_2$ | Vertical distribution of $SO_2$ emissions |
|-------------|-------------------------------|------------------------------------------|
| CAMS-OA     | MACCity                       | All emissions: lowest model layer         |
| (0.4°×0.4°) | (0.5°×0.5°) for 2010          |                                          |
| MERRA-2     | EDGAR-4.2                     | Traffic+residential+ships: lowest model layer |
| (0.625°×0.5°) | (0.1°×0.1°) for 2008       | Industrial emissions: 100-500 m         |
| GEOS-5      | OMI-HTAP                      | Surface emissions: lowest model layer    |
| (0.625°×0.5°) | (0.1°×0.1°) for 2014       | Elevated emissions: 100-500 m           |
| WRF-Chem    | HTAP-2.2                      | Traffic+residential+ships: 0-1000 m     |
| (0.1°×0.1°) | (0.1°×0.1°) for 2010        | Industrial emissions: 120-1000 m        |
| WRF-Chem    | OMI-HTAP                      | Surface emissions: 0-1000 m             |
| (0.1°×0.1°) | (0.1°×0.1°) for 2015, 2016   | Elevated emissions: 120-1000 m         |
3.1 Comparison of $SO_2$ emission datasets

Figure 1 compares the conventional $SO_2$ emission datasets: EDGAR-4.2 for 2008, MACCity for 2010, HTAP-2.2 for 2010 with the OMI-HTAP dataset for 2016. All emission inventories were conservatively interpolated on the WRF-Chem $10 \times 10 \text{ km}^2$ grid. In Figure 1d the point sources of the OMI-HTAP dataset are shown by red and yellow circles of different magnitude depending on their emission rate. The yellow circles correspond to the surface point sources, and red circles to the elevated point sources.

All four emission maps share the common spatial pattern but differ in magnitude and specific positions of the point and distributed sources. Total $SO_2$ emissions from the entire domain in MACCity, Edgar-4.2, HTAP-2.2, and OMI-HTAP are, respectively, 12.5,
12.3, 15.3, and 14.8 Mt/year. Thus, the ME accounts for over 10% of global anthropogenic emissions of SO$_2$ (Klimont et al., 2013). The MACCity and EDGAR-4.2 emissions are close in total amount but not in their spatial distribution. The MACCity emission inventory has coarser spatial resolution than the other datasets.

As in the ME domain, the HTAP-2.2 mainly adopts emissions from the EDGAR-4.3.1; therefore Fig. 1b and Fig. 1c reflect the evolution of the EDGAR inventory, from the version 4.2 to 4.3.1. HTAP-2.2 shows a substantial increase of SO$_2$ emissions in comparison to EDGAR-4.2 over the west and east coast of the Arabian Peninsula.

Total emissions in the OMI-HTAP and HTAP-2.2 are close and exceed the MACCity and EDGAR-4.2 total emissions by about 20%. OMI-HTAP accounts for emission sources in the Arabian Gulf, which are not present in HTAP-2.2, but HTAP-2.2 tends to overestimate emissions over land in the eastern Arabian coastal areas in comparison with OMI-HTAP. In both OMI-HTAP and HTAP-2.2, the significant emission sources are located on the Red Sea coast of the Arabian Peninsula.

To better characterize the spatial distribution of emissions in the different datasets, we define ten spatial sub-regions, as shown in Fig. 1 and calculate annual SO$_2$ emissions in each sub-region for all emissions datasets (see Fig. 2). For HTAP-2.2, we split the total emissions into industrial and traffic+residential+shipping emissions. Following F. Liu et al. (2018) total OMI-HTAP emissions are split into elevated that are energy emissions and surface that are non-energy emissions, which comprise industry, residential, and transportation sources.

![Figure 2](image_url)

**Figure 2.** Annual SO$_2$ emissions in Mt/year from HTAP-2.2, OMI-HTAP, MACCity, and EDGAR-4.2 integrated over the sub-regions (see Fig. 1). The HTAP-2.2 emissions are split into emissions from traffic+residential+ships and industrial sectors. The OMI-HTAP emissions are split to elevated and surface emissions following F. Liu et al. (2018).

HTAP-2.2 emissions are larger compared with MACCity and EDGAR-4.2 in the sub-regions of southeastern Europe, Syria, Iraq, Iran, and Turkey. It is not the case in the central Arabian Peninsula, which may be due to the improved characterization of the emission sources in this region. We have to mention that the latest period accounted for in all emission datasets (except OMI-HTAP) is 2010, but emissions over the eastern US and Europe have been reducing during the last decade, and the recent war in Iraq and Syria also suppressed industrial activities (Krotkov et al., 2016). At the same time,
the economic development and population growth in Saudi Arabia and Iran drive the emissions up in the Red Sea region and over Iran citelelieveld2015abrupt. Thus, actual emissions in 2015-2016 may by slightly different than those assumed in the standard emission inventories.

Over the Arabian Gulf, OMI-HTAP emissions differ significantly from other datasets because the "top-down" approach helped to identify unaccounted sources associated with gas flaring on oil platforms. Consistently, MACCity, EDGAR-4.2 and HTAP-2.2 report unrealistically low emissions over the Arabian Gulf. For almost all other sub-regions, the OMI-HTAP dataset shows lower emissions than in HTAP-2.2. On a national level, the contribution of residential and traffic sources to the total emissions in HTAP-2.2 is relatively small. However, within the cities, traffic and residential emissions could contribute substantially to total air pollution.

3.2 Vertical distribution of $SO_2$ emissions

Emissions can be distributed vertically, either using a fixed profile (Benedictow et al., 2009; Schaap et al., 2005; Visschedijk & van der Gon, 2005; Visschedijk et al., 2007), or a plume-rise model (Guevara et al., 2014; Houyoux et al., 2000; Byun & Schere, 2006). Plume-rise models calculate the rise of a plume using buoyancy, exit momentum and stack height. The Briggs plume rise algorithm is one of the most commonly used (G. A. Briggs, 1969; G. Briggs, 1975, 1984). It can predict plume heights up to 2000 m (Houyoux et al., 2000). However, this estimate contains significant uncertainties. According to Gordon et al. (2018), the Briggs algorithm may underestimate plume heights by up to 50%. In other cases the plume height is consistently overestimated by the Briggs algorithm (VDI, 1985).

F. Liu et al. (2018) in their global GEOS-5 simulations with the OMI-HTAP emission inventory $SO_2$ from energy-related sources (elevated emissions) emitted in the elevated 100-500 m layer with a constant mixing ratio. The non-energy (industrial, residential, and transportation) sources (surface emissions) were placed in the lowest model layer (see Tab. 2). MERRA-2 that uses EDGAR-4.2 emission inventory releases all $SO_2$ industrial emissions in the elevated 100-500 m layer and traffic, residential, and shipping emissions - in the lowest model layer (Buchard et al., 2014). CAMS-OA places all emissions in the lowest model layer to be mixed up within the PBL. The global models use this convention uniformly all over the globe. However, in desert regions, like the ME, PBL and mixing processes are quite different than in the mid-latitudes (Bieser et al., 2011). The height of the unstable desert PBL can reach 6 km and strong turbulence mixes pollutants vertically more effectively than in the mid-latitudes. For example, Z. Liu et al. (2008) showed that during storm events in the Saharan and Arabian deserts, dust particles have been carried aloft to a maximum altitude of 6.6 km.

The catalogue presented by Fioletov et al. (2016) does not provide information on $SO_2$ point source emission heights, but we know that in Saudi Arabia the height of the stacks in power and desalination plants, or petrochemical facilities, is typically between 35 and 150 m (Nayebare et al., 2016). Therefore, in our simulations in the ME we place elevated OMI-HTAP $SO_2$ emissions in the 120-1000 m layer, and surface OMI-HTAP $SO_2$ emissions in the 0-1000 m layer with a constant mass mixing ratio. A similar assumption we made in the WRF-Chem simulation with HTAP-2.2 emissions, where industrial emissions were released in the elevated 120-1000 m layer, and traffic, residential, and shipping emissions were placed in the lower 1000 m layer (see Tab. 2). We apply this assumption uniformly over the entire simulation model domain, as currently the WRF-Chem model does not allow to do it selectively. Buchard et al. (2014) stated that changing the vertical distribution of the emissions did not affect their $SO_2$ fields, but we found that this assumption is only true for total $SO_2$ loadings and fails for surface concentrations. We will discuss these issues in more detail below.
3.3 Evaluation of $SO_2$ column loadings

Figure 3. Annually averaged for 2015-2016 $SO_2$ column loadings (DU) presented on the 10x10 km$^2$ WRF-Chem grid from a) CAMS-OA with MACCity emissions, b) WRF-Chem run with HTAP-2.2 emissions, c) MERRA-2 with EDGAR-4.2 emissions, d) WRF-Chem run with OMI-HTAP emissions, e) GEOS-5 for 2014 with OMI-HTAP emissions, f) OMPS observations.

To evaluate the emissions datasets, here we compare simulated annual averages for 2015-2016 of $SO_2$ column loadings with the corresponding OMPS observations (Fig. 3). We choose to analyze the annual means, as this eliminates high-frequency processes that might not be adequately described in the models, and is consistent with the annual mean emissions dataset we use in this study. We do not use OMI observations for comparison, as they have already been employed to construct the catalogue sources and cannot be treated as independent. In addition, OMPS instrument retrievals are better suited for the model evaluation as the OMPS has lower $SO_2$ retrieval noise than OMI and contiguous spatial coverage.

In the comparison we include outputs from GEOS-5, MERRA-2, CAMS-OA and from two WRF-Chem runs conducted with HTAP-2.2 and OMI-HTAP emission datasets. For comparison purposes $SO_2$ loading fields from OMPS, GEOS-5, MERRA-2 and CAMS-OA are conservatively interpolated on the WRF-Chem 10x10 km$^2$ grid to retain fine spatial features developed in the WRF-Chem runs. Loadings are presented in Dobson units (1 DU = 2.687e+20 molecules m$^{-2}$). GEOS-5 run is for 2014 and conducted by F. Liu et al. (2018) with OMI-HTAP $SO_2$ emissions.

The loadings in Fig. 3 reflect the total column amounts of $SO_2$ in the atmosphere, which are defined by emissions, dry and wet deposition, and $SO_2$ conversion to sulfate through photochemical and wet-phase in-cloud oxidation. We tested the sensitivity of
loadings to the vertical distribution of the emissions, and found it to be quite weak consistently with Buchard et al. (2014). Thus, considering the OMPS observations as ground truth, the loading is an ideal characteristic for testing \( \text{SO}_2 \) emissions.

![Figure 4](image-url)

**Figure 4.** Annually averaged for 2015-2016 and over the sub-domains (see Fig. 1) vertical profiles of \( \text{SO}_2 \) mixing ratios in \( \mu g/kg \) from the GEOS-5 (for 2014) with OMI-HTAP emissions, WRF-Chem runs with HTAP-2.2 and OMI-HTAP emissions, as well as from MERRA-2 with EDGAR-4.2 and CAMS-OA with MACCity emissions: a) Red sea and Western Arabian coast, b) Iraq and Syria, c) Iran, d) central Arabian Peninsula, e) Arabian Gulf, f) Eastern Arabian coast.

Both assimilation products and the free model runs in Fig. 3 show similar main spatial features with high (~0.5-1 DU) \( \text{SO}_2 \) column loadings along the west and east coast of the Arabian Peninsula and eastern Iraq (~0.1-0.3 DU). The unknown ”hot-spot” in EDGAR-4.2 mentioned in Sec. 2.3 produces in MERRA-2 \( \text{SO}_2 \) column loadings about 0.7 DU, which is higher than in Riyadh (see Fig. 3c). All simulated fields, except CAMS-OA, overestimate loadings in southern Europe and the eastern Mediterranean. CAMS-OA consistently overestimates \( \text{SO}_2 \) over the southern Red Sea coast because it uses overestimated in this region MACCity emissions (Fig. 1a). All products underestimate \( \text{SO}_2 \) over the Arabian Gulf except the WRF-Chem and GEOS-5 runs with OMI-HTAP emissions, which show higher than observed loadings of 0.5-1.0 DU. This is consistent with the findings of Fioletov et al. (2016) and McLinden et al. (2016).

Loading is a vertically integrated characteristic, which, as we found, is not sensitive to the detailed vertical profile of emissions. However, transport, chemical transformations and especially near-surface concentrations of \( \text{SO}_2 \) depend on \( \text{SO}_2 \) vertical distribution. Figure 4 shows the annual mean vertical profiles of \( \text{SO}_2 \) mixing ratios obtained from WRF-Chem simulations, CAMS-OA, MERRA-2 and GEOS-5 outputs averaged over the spatial sub-regions (see Fig. 1). CAMS-OA and GEOS-5 exhibit monotonous pro-
files of \( SO_2 \) mixing ratios with a maximum at the surface layer, but MERRA-2 and WRF-Chem runs in most cases show an elevated maximum. Therefore, e.g., in the WRF-Chem run with OMI-HTAP emissions, the \( SO_2 \) content in the PBL over the Arabian Gulf is the highest, but GEOS-5 shows larger surface concentrations.

To further the evaluation of the simulated loadings and associated emissions datasets, we show in Fig. 5 the differences between CAMS-OA, MERRA-2, GEOS-5, and WRF-Chem loadings with OMPS observations. We calculated the differences on the OMPS grid to avoid the side effects depicted in Fig. 5f, where the difference of the WRF-Chem and OMPS loadings is presented on the original WRF-Chem grid. The RMS error (RMSE) in Fig. 5f is bigger than in Fig. 5d because the coarse-resolution OMPS observations do not resolve the fine-scale spatial features developed in the WRF-Chem run. The mean BIASes and RMS errors for all panels in Fig. 5 are calculated for the entire domain and presented in Table 3. The WRF-Chem run with OMI-HTAP has the smallest RMSE and mean BIAS.

The \( SO_2 \) column loadings obtained from the WRF-Chem run with the OMI-HTAP emissions are in good agreement with the observations by OMPS, both in spatial distribution and magnitude. The WRF-Chem run with HTAP-2.2 emissions overestimates the \( SO_2 \) loadings for Jeddah vicinity, eastern part of the Saudi Arabia, Iraq and Syria compared to the OMPS.

Figure 5. Annual mean differences of \( SO_2 \) column loadings with respect to OMPS observations (DU) presented on the OMPS 0.5° × 0.5° grid for a) CAMS-OA with MACCity emissions, b) WRF-Chem run with HTAP-2.2 emissions, c) MERRA-2 with EDGAR-4.2 emissions, d) WRF-Chem run with OMI-HTAP emissions, e) GEOS-5 run with OMI-HTAP emissions for 2014, f) WRF-Chem run with OMI-HTAP emissions (presented on 10×10 km\(^2\) WRF-Chem grid). RMSE and Mean Bias calculated for the entire domain are shown on the panels and in Table 3.
Table 3. RMSE and Mean BIAS (DU) relative to OMPS calculated on the OMPS grid except for the last row, where the WRF-Chem grid was used.

|                | RMSE (DU) | Mean BIAS (DU) |
|----------------|-----------|----------------|
| CAMS-OA. MACCity | 0.090 | -0.027         |
| MERRA-2. EDGAR-4.2 | 0.088 | -0.030         |
| GEOS-5. OMI-HTAP | 0.092 | 0.022          |
| WRF-Chem. HTAP-2.2 | 0.122 | 0.026          |
| WRF-Chem. OMI-HTAP | 0.083 | 0.013          |
| WRF-Chem. OMI-HTAP | 0.103 | 0.008          |

Figure 6 shows the RMSE and Mean BIAS of the simulated SO$_2$ loadings with respect to the OMPS for each sub-region (see Fig. 1). Overall, the WRF-Chem run with OMI-HTAP emissions shows improvements in RMSE and mean BIAS almost in all sub-regions compared with all other products. Based on these results, we conclude that the SO$_2$ loadings from the WRF-Chem run with OMI-HTAP emissions compare better with the OMPS observations than MERRA-2, CAMS-OA and the WRF-Chem run with HTAP-2.2. As expected, MACCity and EDGAR-4.2 tend to underestimate emissions as loadings are negatively biased, but both HTAP-2.2 and OMI-HTAP are positively biased relative to OMPS and, presumably, slightly overestimate emissions.

3.4 Evaluation of SO$_2$ surface concentrations

SO$_2$ column loadings characterize the total content of SO$_2$ in the atmosphere but regional air-quality is determined by the surface concentration of SO$_2$ where it most af-
ffects humans and environmental systems. This characteristic can not be easily retrieved
from satellite observations but is available from model simulations, assimilation prod-
ucts, and field observations at specific sites. To find out which of the products produces
more accurate surface \( \text{SO}_2 \) concentrations, we use in situ observations conducted by air-
quality stations that continuously measured \( \text{SO}_2 \) surface concentrations. These measure-
ments were performed during 2015-2016 by MODON using stationary Air Quality Moni-
toring Systems (AQMS) installed in the major cities of Saudi Arabia: Riyadh, Jeddah,
Dammam. The measurements were conducted following a strict protocol and instruments
were re-calibrated on a quarterly basis. Detailed information on the deployed instruments
is presented in Appendix A2.

Figure 7 shows daily mean \( \text{SO}_2 \) surface concentrations from the WRF-Chem runs
with HTAP-2.2 and OMI-HTAP emissions, MERRA-2, CAMS-OA for period 2015-2016
sampled at three AQMS locations, and compared with the daily averaged in situ mea-
surements. The dash-dotted line corresponds to the WHO air-quality guideline for the
daily averaged \( \text{SO}_2 \) surface concentration, i.e. 20 \( \mu g/m^3 \). The dashed line corresponds
to the EU air-quality limit for the daily averaged \( \text{SO}_2 \) surface concentration, i.e. 125 \( \mu g/m^3 \).
The plots on the right of each panel in Fig. 7 show the corresponding annual averaged
values. The KSA PME air-quality limit for annual averaged \( \text{SO}_2 \) surface concentration,
i.e. 80 \( \mu g/m^3 \) is shown by the solid blue line. In all cities the WHO guideline limit is
exceeded. In Jeddah, the European \( \text{SO}_2 \) air-quality limit (125 \( \mu g/m^3 \)) is exceeded through-
out the whole summer. WRF-Chem simulations with OMI-HTAP emissions fit the av-
eraged observed \( \text{SO}_2 \) concentration quite well. However, because we only use the annual
mean emissions and do not account for the seasonal power consumption variability due
to air-conditioning, the simulations underestimate the \( \text{SO}_2 \) concentration in Summer.
WRF-Chem run with HTAP-2.2 emissions severely overestimates concentrations in Jed-
dah therefore we removed this run from the Jeddah’s daily plots. MERRA-2 underes-
timates pollution in all cities. CAMS-OA and GEOS-5 concentrations compare quite rea-
sonably with AQMS observations, but due to the lower spatial resolution the peak concen-
trations may be underestimated in comparison to the fine resolution WRF-Chem runs.
Annual KSA PME limit 80 \( \mu g/m^3 \) is not exceeded anywhere except for Jeddah in the
WRF-Chem run with HTAP-2.2 emissions. This is probably because a big emission source
in HTAP-2.2 is shifted to be too close to the location of AQMS station in Jeddah. Thus,
we conclude that the WRF-Chem run with OMI-HTAP provides \( \text{SO}_2 \) surface concen-
trations that compare well with the AQMS measurements and we will use WRF-Chem
with OMI-HTAP for our further air-quality estimates.

Figure 8 shows the geographic distributions of \( \text{SO}_2 \) surface concentrations fields
obtained from CAMS-OA, MERRA-2, WRF-Chem and averaged over 2015-2016, sur-
face concentrations from GEOS-5 averaged over 2014. CAMS-OA, MERRA-2 and GEOS-
5 fields were conservatively interpolated on the \( 10 \times 10 \text{ km}^2 \) WRF-Chem grid. The at-
mospheric lifetime of \( \text{SO}_2 \) in the troposphere is typically about one week but in dry en-
vironments it could be 2-3 times longer; thus, in the ME \( \text{SO}_2 \) could be transported over
large distances. The peaks of \( \text{SO}_2 \) surface concentrations occur in the vicinity of strong
\( \text{SO}_2 \) sources; see Fig. 1. Annual average \( \text{SO}_2 \) surface concentrations within these areas
exceed 100 \( \mu g/m^3 \). Despite their low spatial resolution, this effect is more pronounced
in the CAMS-OA and GEOS-5 fields, because in these models, significant emissions are
released in the surface layer (see Fig. 4). The highest \( \text{SO}_2 \) surface concentrations are found
over the western and eastern coasts of Saudi Arabia, where the major \( \text{SO}_2 \) emissions sources
are located. Eastern Iraq, the Arabian Gulf and Mediterranean countries also suffer from
high \( \text{SO}_2 \) surface concentrations. Down the stream, \( \text{SO}_2 \) is oxidized to produce sulfate
aerosols that are the subject of a separate study. The geographic distributions of sul-
fate concentration obtained from WRF-Chem simulations with OMI-HTAP dataset are
discussed in Ukhov et al. (2020).
Figure 7. Left: Daily averaged $SO_2$ surface concentrations ($\mu g/m^3$) measured by AQMS and sampled from WRF-Chem, MERRA-2, CAMS-OA at the locations of AQMS in Jeddah, Riyadh, and Dammam for a) 2015 and b) 2016 (GEOS-5 is not shown because it is for 2014). Time series of the WRF-Chem run with HTAP-2.2 for Jeddah exceed the plot limits and therefore not shown. The dot-dashed and dashed lines correspond to WHO guideline (20 $\mu g/m^3$) and EU limit (125 $\mu g/m^3$), respectively. Right: Corresponding annual mean $SO_2$ surface concentrations ($\mu g/m^3$). Solid line correspond to KSA PME annual average limit (80 $\mu g/m^3$).

Figure 9 shows the 2015-2016 annual mean $SO_2$ surface concentrations for the sub-regions, see Fig. 1. We see more differences between the products in surface concentrations than in the column loadings. This could be related to the different model configurations, as we treat the concentrations in the lower model layer as a surface concentration, but the models have different vertical grids. The differences in the $SO_2$ vertical distribution (see Fig. 4) also translate to the differences in the surface concentrations. However, the averaging over the sub-domains reduce the differences associated with the varying spatial resolution. Consistent with previous findings, CAMS-OA shows highest concentrations for the eastern Mediterranean coast and the western Arabian coast sub-regions. MERRA-2 concentrations are the lowest. The two WRF-Chem runs are similar except for the Arabian Gulf and the eastern Arabian coast. GEOS-5 concentrations significantly exceed those in the WRF-Chem run with OMI-HTAP emissions in all regions except for
Figure 8. Annually averaged 2015-2016 \( SO_2 \) surface concentrations (µg/m³) from a) CAMS-OA with MACCity emissions b) WRF-Chem run with HTAP-2.2 emissions, c) MERRA-2 with EDGAR-4.2 emissions, d) WRF-Chem run with OMI-HTAP emissions, e) GEOS-5 run for 2014 with OMI-HTAP emissions, f) map showing major cities of the ME. Egypt and the central Arabian Peninsula. This difference is explained by different vertical distribution of \( SO_2 \) emissions (see Fig. 4).

Figure 10 shows the annual mean \( SO_2 \) concentrations and 90th percentile of daily concentrations for the major cities of the ME. Concentrations were sampled from the WRF-Chem run with OMI-HTAP emissions. The number of days during 2015-2016 when the daily \( SO_2 \) surface concentrations exceeded the European Union air-quality limit (125 µg/m³) was also calculated. Saudi cities are among the most polluted in the ME. In Jeddah and Riyadh, the 90th percentile of daily mean \( SO_2 \) concentrations exceeded 125 µg/m³. The number of days when the daily averaged \( SO_2 \) surface concentration exceeded the European Union air-quality limit in Jeddah and Riyadh during 2015-2016 period is 154 and 150, respectively. The NEOM region located in the northwest of the Kingdom (see Fig. 8f) is relatively clean although it is affected by trans-boundary transport from North Africa and the eastern Mediterranean.

4 Conclusions

In this study we test how well the assimilation products (MERRA-2, CAMS-OA) and the free-running models (GEOS-5, WRF-Chem) in combination with emission databases (MACCity, EDGAR-4.2, HTAP-2.2, OMI-HTAP) reproduce \( SO_2 \) column loadings and surface concentrations in the ME on a regional basis during the period 2015-2016. We use satellite observations and \textit{in situ} ground-based measurements of \( SO_2 \) surface con-
Figure 9. Annual averaged for 2015-2016 $SO_2$ surface concentrations ($\mu g/m^3$) over the sub-regions (see Fig. 1) calculated for CAMS-OA with MACCity emissions, MERRA-2 with EDGAR-4.2 emissions, WRF-Chem run with HTAP-2.2 emissions, WRF-Chem run with OMI-HTAP emissions and GEOS-5 run for 2014 with OMI-HTAP emissions. Numbers indicate the magnitude of the corresponding columns.

Figure 10. Annually averaged for 2015-2016 $SO_2$ surface concentrations (solid bars) and 90th percentiles ($\mu g/m^3$) (hatched bars) calculated using daily mean $SO_2$ concentrations for the major cities obtained from WRF-Chem run with OMI-HTAP emission dataset. Numbers over the solid bars correspond to the number of days during 2015-2016 when daily averaged $SO_2$ surface concentrations exceeded the European Union air-quality limit (125 $\mu g/m^3$) for $SO_2$. For the city concentration, we took the maximum concentration from 3x3 cells covering the city.

The $SO_2$ column loadings show a strong dependence on the emissions datasets used, both in the assimilation products and free model runs, and therefore they help to evaluate emissions, when used in combination with the OMPS observations.

We specifically test and improve the newly developed $SO_2$ emission dataset (OMI-HTAP) based on the combination of the HTAP-2.2 $SO_2$ emissions from residential and traffic sources with the catalogue of the significant (>30 kt/year) $SO_2$ point sources observed by the OMI instrument (“top-down” approach), and use it along with the conventional HTAP-2.2 emissions dataset in the WRF-Chem simulations.

The OMI-HTAP and the conventional emissions inventories such as MACCity, EDGAR-4.2, HTAP-2.2, which are based on the “bottom-up” approach, show that the differences between the domain-integrated annual emissions of $SO_2$ range from 12.3 to 15.3 Mt/year.
All datasets underestimate emission rates over the Arabian Gulf except for OMI-HTAP. For example, though OMI-HTAP and HTAP-2.2 agree well on the location and strength of most of the $SO_2$ sources, the latter underestimates $SO_2$ emissions over the Gulf by $\sim 0.55 \text{ Mt/year}$ relative to OMI-HTAP.

The location and strength of some of the $SO_2$ sources are also inconsistent among the datasets. For example, the MACCity exaggerates emissions over the southern Red Sea coast. Overall, based on the test with OMPS loadings, MACCity and EDGAR-4.2 underestimate emissions over the ME, whereas HTAP-2.2 and OMI-HTAP are positively biased.

Along with $SO_2$ column loadings, we evaluated the $SO_2$ surface concentrations and their effect on air-quality in the major ME cities. The air-quality observations were made available from \textit{in situ} measurements conducted by the AQMS installed in Jeddah, Riyadh and Dammam. The simulated surface concentrations appear to be sensitive not only to the magnitude of emissions, but to their vertical distribution and model spatial resolution, thus making it more difficult to simulate air-quality than column loadings. For example, the information on the exact location of the significant $SO_2$ source and its emission height in the OMI-HTAP dataset becomes of particular importance when we compare with AQMS observations taken within the vicinity of large emission sources. Due to limitations of the method that was used to construct the point source catalogue (Fioletov et al., 2016), the coordinates of the point sources can not be exactly determined. Since we know the exact location of the Jeddah power plant, we corrected the coordinates in the developed dataset, which led to better agreement of our simulations with the observations.

Thus, the main findings of this study are as follows:

1. ME countries are prolific $SO_2$ emitters, and the ME is responsible for more than 10% of global anthropogenic $SO_2$ emissions.
2. The older datasets, MACCity and EDGAR-4.2, underestimate emissions across the ME. The new HTAP-2.2 and OMI-HTAP overestimate emissions over the ME, but the OMI-HTAP allows to reduce biases and RMSE in $SO_2$ loadings with respect to the OMPS observations.
3. All products reproduce $SO_2$ loadings relatively well, but WRF-Chem with OMI-HTAP emissions exhibits the smallest RMSE and bias.
4. $SO_2$ surface concentrations are less consistent between products than loadings, as they depend not only on the emissions, but they are also sensitive to the model resolution and the vertical distribution of $SO_2$ emissions.
5. The vertical distribution of $SO_2$ emissions should not be spatially uniformly prescribed in the models over the Globe, but has to be skillfully parameterized to account for specific meteorological conditions.
6. Higher spatial resolution in WRF-Chem allows to compare the simulated concentrations with air-quality measurements more reliably in comparison with the coarse resolution global models that smooth concentration extremes, and therefore improves model calibration and emissions evaluation.
7. CAMS-OA overestimates $SO_2$ surface concentrations, while MERRA-2 tends to underestimate them. WRF-Chem run with OMI-HTAP emissions compares better with \textit{in situ} air-quality measurements than with other products.
8. Populated coastal areas exhibit the largest $SO_2$ pollution. In Jeddah and Riyadh, the European $SO_2$ air-quality limit is exceeded around 75 days per year.

Developing a denser air-quality monitoring network and measurements of vertical distribution of air pollutants is urgently needed to improve air-quality modeling in the ME. The $SO_2$ retrievals used in this work do not allow to extract information on the monthly variability of sources, but adding the seasonality to the emissions dataset is a work in progress.
progress. The developed framework presented here can be used to evaluate the effect of other air pollutants like NO\textsubscript{x} and O\textsubscript{3}. The results of this study could serve as the basis for a regional air-quality forecast system that interactively calculates high-resolution atmospheric chemistry and aerosol processes driven by anthropogenic emissions. This system could be especially valuable for the prediction of extreme pollution events, and could also improve our understanding of the impact of anthropogenic pollution on air-quality and human health in the ME.

**Code and data availability**

1. The MERRA-2 reanalysis is available at [https://disc.gsfc.nasa.gov/daac-bin/FTPSubset2.pl](https://disc.gsfc.nasa.gov/daac-bin/FTPSubset2.pl)
2. The CAMS-OA is available at [http://apps.ecmwf.int/datasets/data/cams-nrealtime](http://apps.ecmwf.int/datasets/data/cams-nrealtime)
3. The monthly OMPS level 3 retrievals on 0.5°×0.5° grid are available at [https://avdc.gsfc.nasa.gov/pub/data/satellite/Suomi_NPP/L3/NMSO2m/NMSO2_0.5x0.5_monthly](https://avdc.gsfc.nasa.gov/pub/data/satellite/Suomi_NPP/L3/NMSO2m/NMSO2_0.5x0.5_monthly)
4. The Merra2BC interpolator is available at [https://doi.org/10.5281/zenodo.3695911](https://doi.org/10.5281/zenodo.3695911)

**Anthropogenic emission inventories**

1. EDGAR-4.2 is available at [http://edgar.jrc.ec.europa.eu/overview.php?v=42](http://edgar.jrc.ec.europa.eu/overview.php?v=42)
2. MACCity is available at [http://accent.aero.jussieu.fr/MACC_metadata.php](http://accent.aero.jussieu.fr/MACC_metadata.php)
3. HTAP-2.2 is available at [http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123](http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123)
4. OMI-HTAP is available at [https://avdc.gsfc.nasa.gov/pub/data/project/OMI_HTAP_emis](https://avdc.gsfc.nasa.gov/pub/data/project/OMI_HTAP_emis)

**Appendix A**

**A1 Merra2BC interpolator**

*Merra2BC* interpolator (available at [https://github.com/saneku/Merra2BC](https://github.com/saneku/Merra2BC)) creates time-varying chemical boundary conditions based on MERRA-2 reanalysis for a WRF-Chem simulation by interpolating chemical species mixing ratios defined on the MERRA-2 grid to the WRF-Chem grid for initial conditions and boundary conditions. In the case of initial conditions, interpolated values are written to each node of the WRF-Chem grid. In the case of boundary conditions, only boundary nodes are affected.

*Merra2BC* utility is written on Python. The utility requires additional modules which need to be installed in Python environment: NetCDF4 - interface to work with netCDF files and SciPy’s interpolation package.

The full MERRA-2 reanalysis dataset including aerosol fields is publicly available online. Depending on the requirements, all or one of the following aerosol and gaseous collections needs to be downloaded: \texttt{inst3\_3d\_aer\_Nv} - gaseous and aerosol mixing ratios, (kg/kg) and \texttt{inst3\_3d\_chem\_Nv} - Carbon monoxide and Ozone mixing ratios, (kg/kg). In addition to downloaded mixing ratios, pressure thickness \texttt{DELP} and surface pressure \texttt{PS} fields also need to be downloaded. Spatial coverage of the MERRA-2 files should include the area of the WRF-Chem simulation domain. The time span of the downloaded files should match with the start and duration of the WRF-Chem simulation. For more information regarding MERRA-2 files specification please refer to Bosilovich et al. (2016).
A11 Mapping chemical species between MERRA-2 and WRF-Chem

Merra2BC input file config.py contains multiplication factors to convert MERRA-2 mixing ratios of gases given in kg/kg into ppmv. Aerosols are converted from kg/kg to ug/kg. In the case of using the GOCART aerosol module in the WRF-Chem simulation, all MERRA-2 aerosols and gases are matched with those from WRF-Chem. To convert MERRA-2 aerosol mixing ratios given in kg/kg into ug/kg, multiply by a factor of $10^6$. In case of gases, multiply MERRA-2 mixing ratios by a ratio of molar masses $M_{\text{air}}/M_{\text{gas}}$ multiplied by $10^6$ to convert kg/kg into ppmv, where $M_{\text{gas}}$ and $M_{\text{air}}$ are the corresponding molar masses. If another aerosol module is chosen in WRF-Chem, then different multiplication factors should be used.

A12 Typical workflow

Below are the steps describing how to work with the Merra2BC utility:

1. Run real.exe, which will produce the initial wrfinput.d01 and boundary conditions wrfbdy.d01 files required by WRF-Chem simulation;
2. Download required MERRA-2 collection files;
3. Download the Merra2BC code from https://github.com/saneku/Merra2BC;
4. Edit config.py file which contains:
   (a) mapping of chemical species and aerosols between MERRA-2 and WRF-Chem;
   (b) paths to wrfinput.d01, wrfbdy.d01, met_em_* files;
   (c) path to the downloaded MERRA-2 collection files;
5. Program real.exe sets default boundary and initial conditions for some chemical species. Merra2BC adds interpolated values to the existing ones and it may cause incorrect concentration values. To avoid this, run “zero_fields.py” script, which will zero the required fields;
6. Run script “main.py”, which will perform the interpolation; as a result, files wrfinput.d01, wrfbdy.d01 will be updated by the interpolated from MERRA-2 values;
7. Modify WRF-Chem namelist.input file at section &chem: set have_bc_chem = .true. to activate updated boundary conditions from MERRA-2 and, if it is needed, chem_in_opt = 1 to activate updated initial conditions;
8. Run wrf.exe program.

For the usage of the Merra2BC interpolator the following python modules need to be installed:

- netcdf4: https://github.com/Unidata/netcdf4-python
- scipy: https://github.com/scipy/scipy

A13 Meteorological Boundary and Initial Conditions

To be consistent with BC&IC for chemical species and aerosols, we utilized the same procedure to build meteorological BC&IC from MERRA-2 reanalysis for all required meteorological parameters. In particular, the following 3D parameters were processed: pressure (Pa), geopotential height (m), temperature (K), meridional and zonal wind components (m/s), relative humidity (%); 2D parameters: surface pressure (Pa), sea level pressure (Pa), meridional and zonal wind components at 10 meters height (m/s), temperature at 2 meters height (K), relative humidity at 2 meters height (%), skin temperature (K), ice mask (0/1), terrain height (m), land/sea mask (1/0), soil temperature at 0-10 (cm), 10-40 (cm), 40-100 (cm) and 100-200 (cm); soil moisture at 0-10 (cm), 10-40 (cm), 40-100 (cm) and 100-200 (cm); snow depth (m); snow water equivalent (kg/m²).
A2 MODON measurements

The "AF22M" by Environnement-S.A (see Fig. A1) is a continuous ambient air-quality monitoring analyzer, based on the ultraviolet fluorescence principle, which is the standard method for the measurement of SO₂ concentrations. The measurements are conducted at regular intervals and the collected data is transmitted in real time to servers at MODON for processing and storage. To provide confidence in the operational status of each AQMS, a comprehensive physical audit is conducted by Ricardo-AEA Ltd, (United Kingdom) on a quarterly basis. An external view of the AQMS installed in Jeddah is presented in Fig A2.

![AF22M Analyzer](image_url)

**Figure A1.** The analyzer "AF22M" by Environnement-S.A for continuous detection of ambient SO₂ concentration.

**Author contribution** A. Ukhov wrote the manuscript and constructed IC&BC for aerosol and chemistry species based on MERRA-2 reanalysis, and took part in planning the calculations. S. Mostamandi performed the calculations, constructed meteorological BC&IC based on MERRA-2 reanalysis and took part in the discussions. G. Stenchikov planned the calculations, led the discussion, and reviewed and improved the manuscript. Y. Alshehri collected, filtered and validated SO₂ observational data and wrote the section on the SO₂ measurement procedures. N. Krotkov, J. Flemming, C. Li, V. Fioletov, C. McLimden, A. da Silva and A. Anisimov participated in the discussion, advised on the products and emission datasets, and helped to formulate the research program, and reviewed the manuscript.

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

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a) Total $SO_2$ emission rate 12.5 Mt/year

b) Total $SO_2$ emission rate 15.3 Mt/year

c) Total $SO_2$ emission rate 12.3 Mt/year

d) Total $SO_2$ emission rate 14.8 Mt/year

$SO_2$ emission rate, $kg/m^2/year^{-1}$
Figure 5.
SO₂ column loadings differences, DU

a) CAMS. MACC-MCity - OMPS
RMSE = 0.090
BIAS = -0.027

b) WRF-Chem. HTAP-2.2 - OMPS
RMSE = 0.122
BIAS = 0.026

c) MERRA-2. EDGAR-4.2 - OMPS
RMSE = 0.088
BIAS = -0.030

d) WRF-Chem. OMI-HTAP - OMPS
RMSE = 0.083
BIAS = 0.013

e) GEOS-5. OMI-HTAP - OMPS, 2014
RMSE = 0.092
BIAS = 0.022

f) WRF-Chem. OMI-HTAP - OMPS
RMSE = 0.103
BIAS = 0.008
