Analysis of ambient SO_2 concentrations in Dimitrovgrad, Bulgaria, 2023:

An investigation of the contribution to SO_2 concentrations due to emissions from the Martisa 3 coal fired power plant

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Summary

This study investigated the contribution of emissions from the Maritsa 3 coal-fired power plant (CFPP) to ambient air concentrations of sulphur dioxide (SO_2) during 2023 in the adjacent city of Dimitrovgrad, in southeastern Bulgaria.

Maritsa 3 CFPP operated sporadically through 2023 and considerable differences were seen in ambient air SO_2 concentrations between times when Martisa 3 was operating and those times when it was not operating, with higher concentrations and far greater variability in levels during operating times.

This investigation provides strong evidence that the operation of the Martisa 3 CFPP is likely responsible for increased SO_2 concentrations in Dimitrovgrad

Key findings included:

- The average SO_2 concentration during operating times (26.9 μg/m^3) was 1.8 times higher than when not operating (15.3 μg/m^3).
- The daily average SO_2 concentration frequently exceeded 40 μg/m^3, above the World Health Organisation (WHO) 24-hour average guideline value.
- The highest SO_2 concentrations occurred far more frequently when Martisa 3 was operating (and in the 24 hours after operating). Operating times accounted for 90% of the 50 highest hourly SO_2 concentrations, and 83% of the 100 highest values.
- Analysis indicated that differences in SO_2 concentrations between operating and non-operating times were not principally driven by differences in wind patterns between these times, but were likely to have resulted from differences in SO_2 emissions.
- Analysis indicated an appreciable SO_2 source to the east of the monitoring station, in the direction of the Maritsa 3 complex, a pattern most clearly seen during times when the CFPP was operating.
- SO_2 concentration patterns suggest that higher levels seen during parts of winter were associated with Martisa 3 operating and were not solely due to other additional local sources during winter months, such as domestic coal combustion.

Introduction

The Maritsa 3 coal-fired power plant (CFPP) is located on the eastern edge of Dimitrovgrad, in southeastern Bulgaria (Figure 1). In addition, a cluster of CFPPs is located to the north-east of Dimitrovgrad, at between 30 to 50 Km. These include the Brikel, AES, Contour Global and Maritsa East 2 CFPPs (Figure 2).
Electricity generated output data reported on the ENTSO-e transparency platform indicate that the Maritsa 3 CFPP operated sporadically through 2023, with the plant operating approximately one third of the year.

Fossil fuels, particularly coal and oil, can contain the element sulphur. When these fuels are burned the air pollutant sulphur dioxide (SO_2) is produced. SO_2 is a toxic gas with adverse impacts for both human health and the environment. Sulphur emissions also contribute to the formation of $PM_{2.5}$ air pollution.

The most important sources for SO_2 emissions globally are coal power stations, smelters, oil and gas industry sites and volcanoes. The European Environment Agency provides sector emission estimates for Agriculture, Energy supply, Manufacturing and extractive industries, Residential, commercial and industrial, Transport, and Waste. Energy supply is consistently the sector responsible for the largest contribution to SO_2 emissions in Bulgaria. In 2021 energy supply generated 72% of Bulgaria's atmospheric emissions of SO_2 (EEA, 2024a).

This analysis was carried out to investigate the contribution of SO_2 emissions from the Maritsa 3 CFPP to ambient air concentrations of sulphur dioxide (SO_2) in the vicinity of Dimitrovgrad

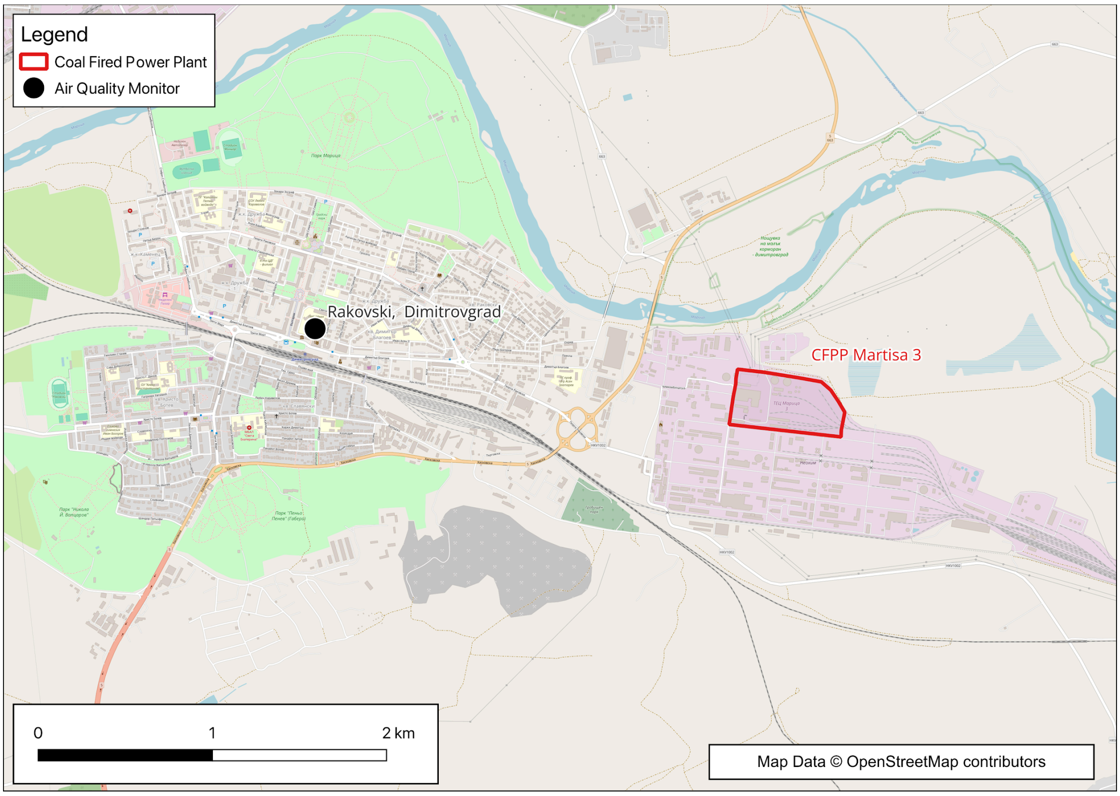


Figure 1. Locations of the Maritsa 3 coal fired power plant and Dimitrovgrad air quality monitoring station. Map data © OpenStreetMap contributors (2015), and available under the Open Database License from https://www.openstreetmap.org

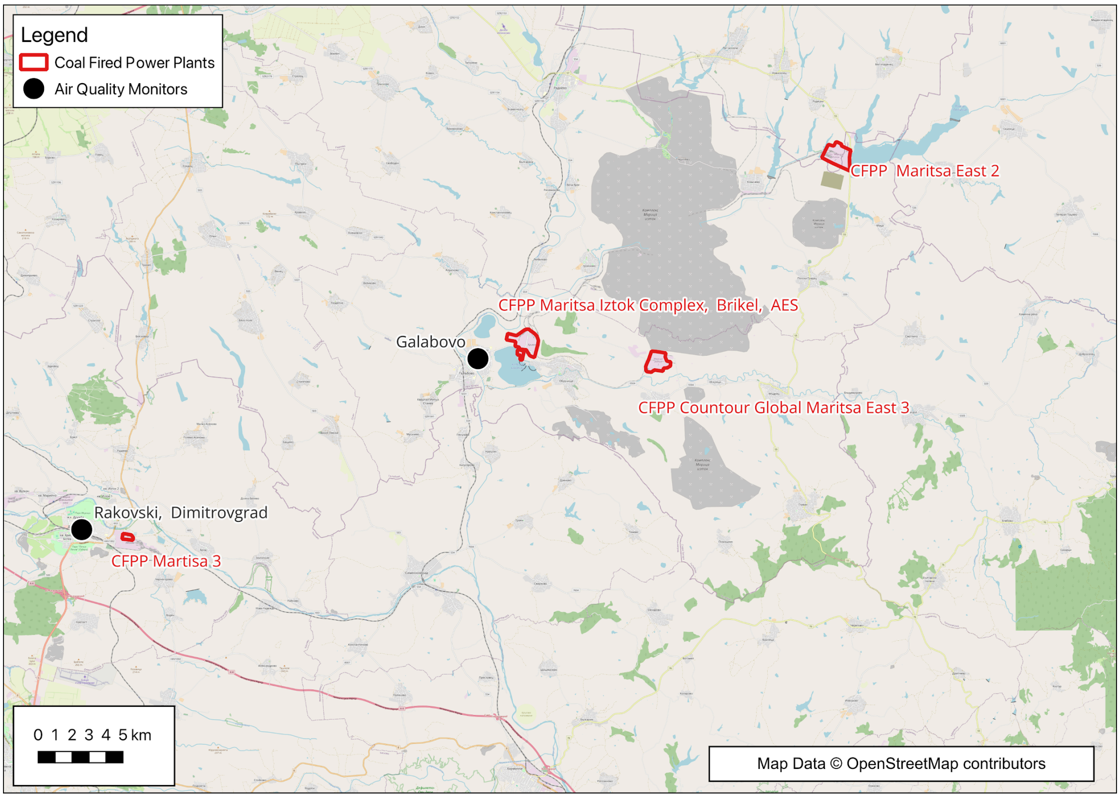


Figure 2. Locations of industrial complexes that include coal fired power plants, and locations of air quality monitoring stations in the study area. Map data © OpenStreetMap contributors (2015), and available under the Open Database License from https://www.openstreetmap.org

Data and methods

Local Air Quality Data

Ambient concentrations of SO_2 are measured by the Rakovski air quality monitoring station in central Dimitrovgrad (Figure 1). The Martisa 3 CFPP is located 2.5 Km to the east of the Rakovski monitor (in the direction 100-110°).
Data from this monitoring station are made available to the public by the Bulgarian Environmental Executive Agency (EEA 2024b). This analysis used reported SO_2 data for the whole year of 2023 (26.01.23 to 31.12.23). These data are also available via the European Environment Agency Air Quality Download service (EEA 2024c) which confirms that they have been verified and validated according to the agency's quality assurance process.

Local Meteorological Data

In this analysis, pollution data (SO_2) was processed in combination with wind data to investigate patterns of pollution movement and dispersion. SO_2 concentrations, at times when wind was and was not travelling from the direction of a potential pollutant source, were compared to test the hypothesis that the source was contributing to local air pollution. The data were analysed using the Openair R software package (Carslaw and Ropkins, 2012).

The Rakovski monitoring station provides meteorological data (wind speed and direction), in addition to ambient SO_2 concentrations. Analysis of the meteorological data from this station, identified that wind speed records do not include any data at wind speeds below 4.5 m/s (Figure 3, c) and that the observations have a significantly skewed distribution of wind speeds when compared with the two modelled wind datasets (Figure 4). It is possible that instrument malfunction has prevented the recording of winds below 4.5 m/s, or that nearby buildings affect wind speed at the site. This reduces confidence in the reliability of these meteorological observations. Analysis of SO_2 observations from the Rakovski monitor did not identify any similar concerns with the pollutant concentration data.

The modelled meteorological datasets are likely to provide a reasonable representation of wind speeds typical at the regional scale. Regional scale wind patterns, rather than very-local scale patterns are most relevant to this study which investigates air pollution dispersion over distances of many kilometres.

It was, therefore, considered that meteorological data from the Rakovski monitoring station were not suitable for this analysis.

This report makes use of modelled meteorological data in order to provide greater confidence in the analysis results for SO_2 dispersion. The modelled wind speed and wind direction data were downloaded from Visualcrossing.com (Visual Crossings, 2023a). Visual Crossing combines observations from many sources to construct global historical weather databases. The sources of data include raw observations at a variety of temporal resolutions, commonly hourly observations from meteorological monitoring stations at airports. Global coverage is achieved by interpolation based on the proximity of weather stations to the location. A complete list of data sources is provided at Visualcrossings.com (Visual Crossings, 2020).

Visual Crossings historical weather data is a composite product made by merging weather data from nearby meteorological monitoring stations, remote sensing data from satellite and radar, as well as modelled forecast data. Input data are weighted when the data are merged, according to the distance to the observation point and the quality of the data. Surface observations are weighted ahead of remote and modelled data, and official monitoring stations are weighted ahead of lower quality data sources (Visual Crossings, 2022, 2023b).

This analysis includes use of two Visual Crossings datasets constructed using:

1) only remotely sensed and forecast data; referred to as 'regional modelled wind data'
2) local surface observations, remotely sensed and forecast data; referred to as 'modelled wind data including local observations'

To provide a greater understanding, we choose to analyse data using both datasets rather than only using the dataset that includes local observations. This is because there are few local surface observation stations in the study region. Those that exist are a significant distance from Dimitrovgrad and may not be representative of local weather conditions.

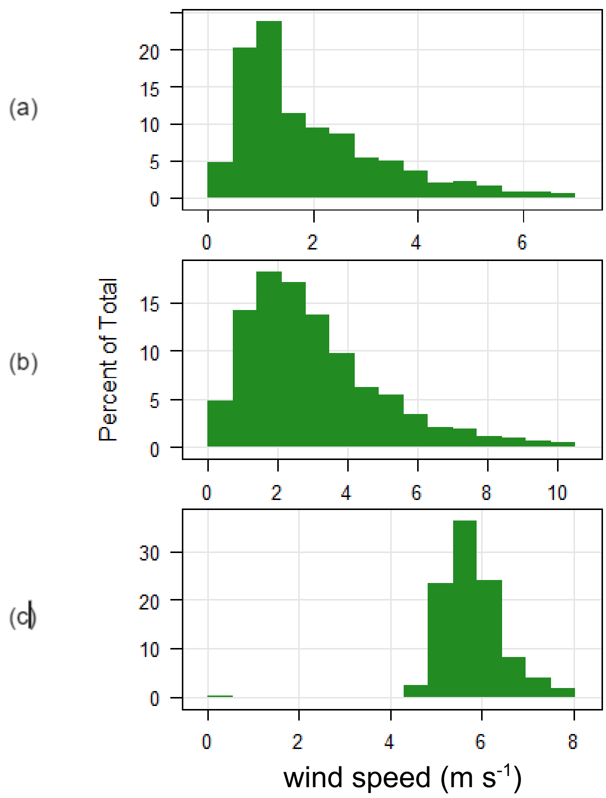
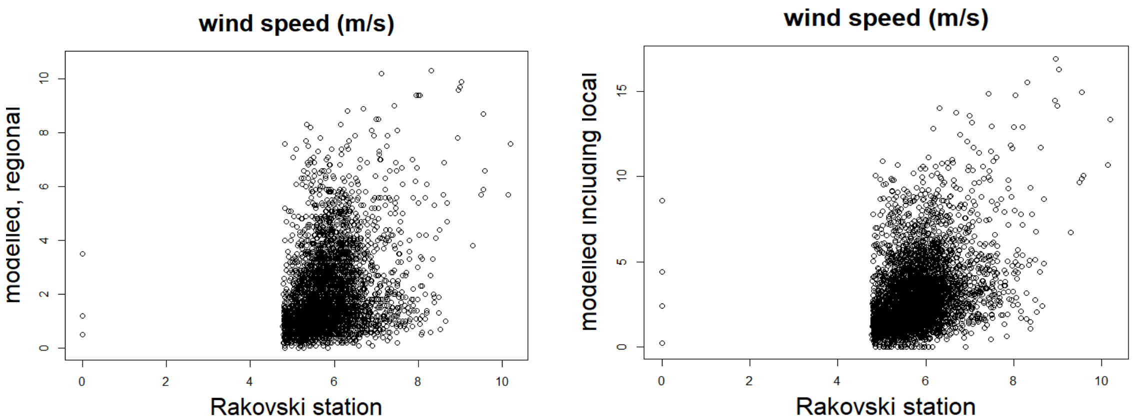


Figure 3. Wind speed frequency counts for 2023 for (a) regional modelled wind dataset, (b) modelled wind data including local observations, and (c) data from the Rakovski station



(a) (b)

Figure 4. Wind speed reported by the Rakovski station (x-axes) compared to data for the same time point from the modelled dataset (y-axes): (a) regional modelled wind dataset, (b) modelled wind data including local observations. Neither comparison identifies a strong positive correlation between wind speed observations and modelled datasets, and both identify that the Rakovski station does not record wind speeds below 4.5 m/s.

Modelled wind datasets

The distribution of wind speeds and directions for the two modelled wind datasets are shown in Figure 5. The dataset which incorporates local observations shows generally higher wind speeds. Both datasets indicate northwest as a prevailing wind direction, though the dataset incorporating local observations has a larger proportion of westerly and easterly winds.

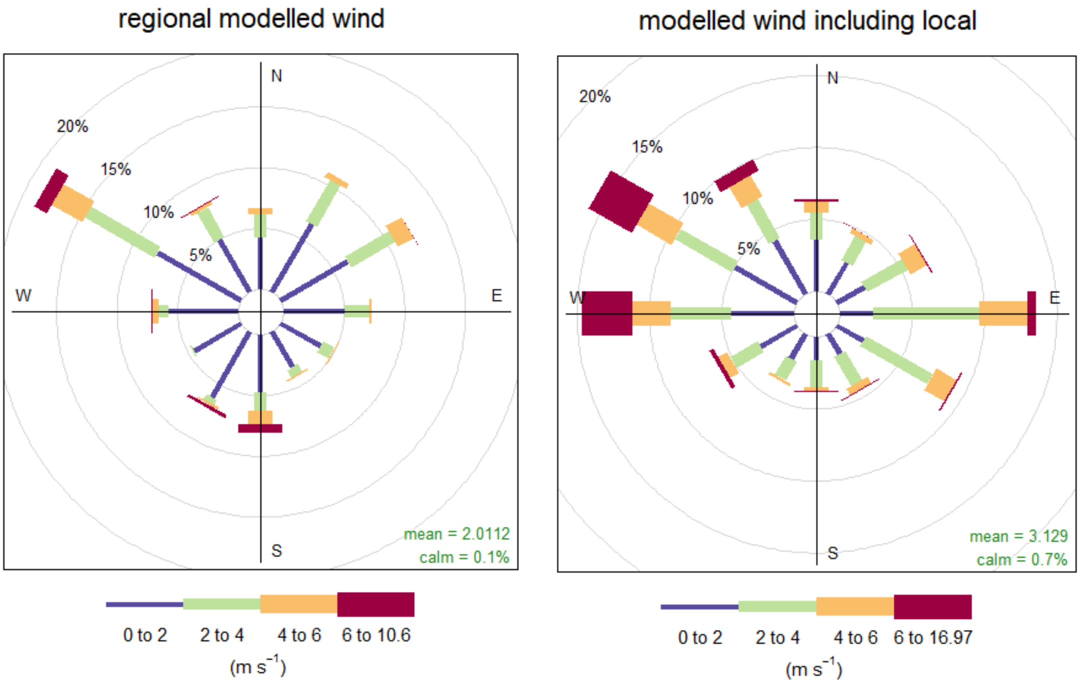


Figure 5. Wind speed frequency counts by wind direction for the regional modelled data and the modelled data including local observations during 2023

SO_2 ambient air concentrations

Concentrations of SO_2 in ambient air are regulated in the EU. There is a legal requirement to meet 24-hour and 1-hour limit values that were first introduced in 2008. The World Health Organisation (WHO) also promotes a stricter 24-hour health based guideline, last updated in 2021 (Table 1). The EU is currently working to adopt new, stricter limit values for SO_2, it is expected that there will not be a legal requirement for these to be met until 2030 (European Council 2024).

Table 1. 24-hour average and 1 hour average SO_2 concentration limits included in EU standards and WHO air quality guidelines.

Average Period	EU	WHO Guidelines
24-hour average	125 µg/m^{3*}	40 µg/m^{3***}
1-hour average	350 µg/m^{3**}	-

*3 permitted exceedances each year. **24 permitted exceedances each year. ***99th percentile (i.e. 3–4 permitted exceedances per year). Source: European Council 2008, WHO 2021.

Analysis of SO_2 data from the Rakovski monitoring station reveals that in 2023 the average SO_2 concentration was 19 µg/m^3, though there were considerable variations throughout the year (Figure 6). On many occasions the daily average SO_2 concentration exceeded 40 µg/m^3, reaching values of around 60 µg/m^3, occasionally higher. In comparison, the World Health Organisation (WHO) sets a guideline value for SO_2 of 40 µg/m^3 for a 24-hour average (WHO 2021). The EU standard sets a maximum of 125 µg/m^3 for a 24-hour average, with 3 exceedances allowed per year (European Council 2008). Data from the Rakovski station did not exceed this EU standard. The EU also sets a maximum hourly average of 350 µg/m^3, with 24 exceedances allowed per year. The highest reported hourly average concentration was 644 µg/m^3, though the EU limit of 350 µg/m^3 was not exceeded more than 24 times in the year.

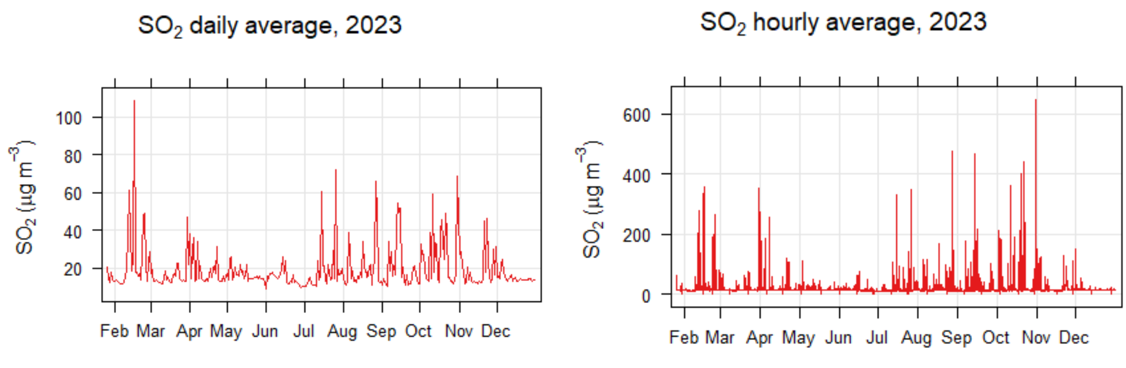


Figure 6. Daily average, and hourly average SO_2 concentrations (µg/m^3) reported by the Rakovski monitor for 2023

SO_2 concentration and Maritsa 3 CFPP operations

The Maritsa 3 CFPP did not operate continuously during 2023, meaning that it is possible to compare SO_2 concentrations observed at the air quality monitoring station at time that Maritsa 3 was operating and when it was not operating. The concentrations observed are controlled by factors including emissions from the CFPP, other local emission sources and meteorology.

The distribution of wind speeds and directions between operating/not operating times were very similar for both modelled wind datasets (Figure 7). This suggests that any differences observed in SO_2 concentration between operating/not operating times were not principally driven by wind patterns, and were likely to have resulted from differences in SO_2 emissions.

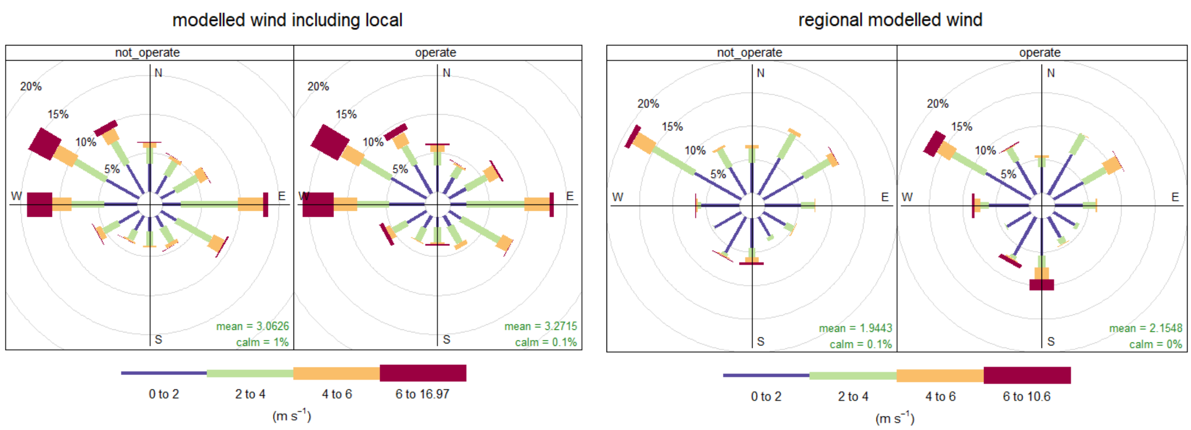


Figure 7. Comparison of wind speed frequency counts by wind direction between times when Martisa 3 was operating and times when it was not operating, for both modelled datasets during 2023

Considerable differences were seen in SO_2 concentrations between times when Martisa 3 was operating and those times when it was not operating in 2023. Higher levels and far greater variability in concentrations were seen during times that the CFFP was in operation (Figure 8 and 9). This included;

- The average SO_2 concentration over the times Martisa 3 was operating in 2023 was 26.9 μg/m^3, 1.8 times higher than when not operating (15.3 μg/m^3).
- The highest hourly average SO_2 concentration during operating times was 644 μg/m^3, almost double the highest equivalent concentration when not in operation (360 μg/m^3)
- Of the times when the hourly average SO_2 concentrations exceeded 200 μg/m^3, 91% occurred when Martisa 3 was operating and a further 6% occurred during the 24 hour period after Maritsa 3 ceased operating and when it might be expected that SO_2 from the CFPP may still influence the monitoring station)
- Of the times when the hourly average SO_2 concentrations exceeded 100 μg/m^3, 81% occurred when Martisa 3 was operating and a further 8% of such exceedances occurred during the following 24 hour period.
- Of the 50 highest hourly observations, 90% were recorded during hours when Martisa 3 was operating, a further 4% occurred within 24 hours of Martisa 3 ceasing operations.
- Of the 100 highest hourly observations, 83% were during hours when Martisa 3 was operating, a further 8% occurred within 24 hours of Martisa 3 ceasing operations.
- During Martisa 3 operating hours, there was a considerably higher variation in SO_2 concentrations over time compared to non-operating times. This is consistent with

significant quantities of SO_2 being emitted from a nearby major source with either variable emission rates or dispersion conditions. A measure of variability in concentration, the standard deviation, was 4 times higher when operating (42.0 µg/m^3 compared to 10.5 µg/m^3).

- SO_2 concentrations averaged by hour-of-the-day and by month-of-the-year were consistently higher when Martisa 3 was operating compared to other times (Figure 9).

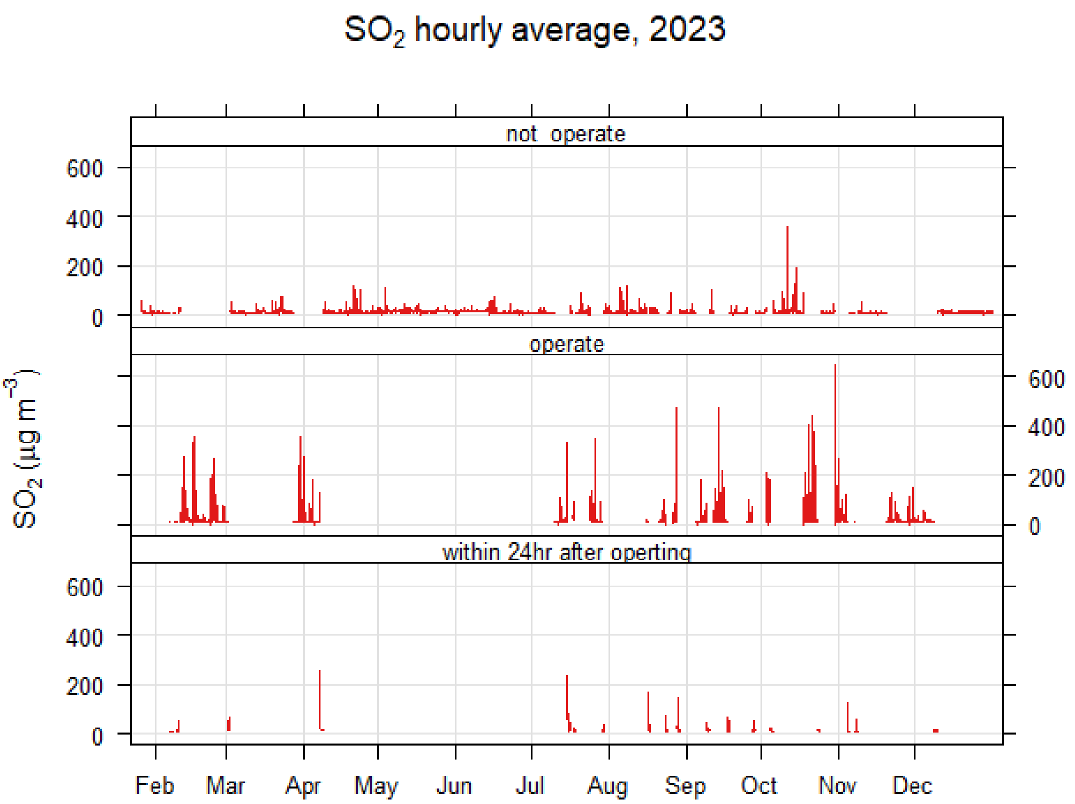


Figure 8. Hourly average SO_2 concentrations (µg/m^3) reported by the Rakovski monitor for 2023, split into times when the Martista 3 CFPP was not operating (top), was operating (middle) and during the 24 hour period after operating to account for SO_2 dispersal (bottom)

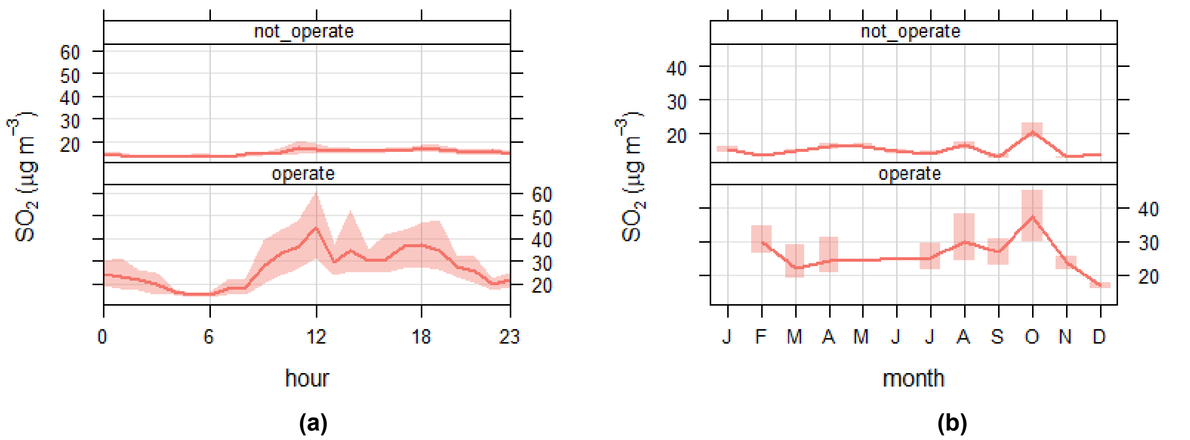


Figure 9. Average SO_2 concentrations (µg/m^3) for (a) the hourly diurnal cycle, and (b) the monthly seasonal cycle during 2023, separated by times when Martisa 3 was operating (bottom) and was not operating (top). The solid line indicates the average value, with the shaded areas above and below the line providing a measure of the variability of the SO_2 concentrations (the 95% confidence interval of the average, which is the range in which 95% of concentration values were found)

Many of the times when high hourly average SO_2 concentrations coincided with operation of Martisa 3 were during winter (Figure 8). It is possible that other local sources of SO_2 increase during winter, such as domestic coal combustion, and this could contribute to the elevated hourly average SO_2 concentrations. This factor, however, does not appear to account for the pattern observed, given that higher SO_2 levels were also seen at times when Martisa 3 operated in summer, and also that lower SO_2 levels were observed during times in winter when Maritsa 3 was not operating. The data suggest that the effect is associated with Martisa 3 operating, not due to other additional local sources during winter months.

SO_2 concentration and meteorology

Analysis of SO_2 concentrations recorded when different wind conditions occurred can give an indication about the location of major SO_2 emission sources relative to a monitoring station. For example, where high SO_2 concentrations routinely coincide with the direction of the wind coming from the west, this can indicate that a major source is located to the west of the monitoring location.

Throughout 2023, relatively high concentrations of SO_2 at the Rakovski monitor coincided, at different times, with nearly all wind directions. A similar pattern was seen using either of the modelled wind datasets (Figure 10). Averaged across the year, there was not a strong connection between higher SO_2 concentrations and a specific wind direction, though higher concentrations were moderately more frequent with winds from an arc between northeast and southeast.

In Figure 10, the inner black segments indicate the frequency that wind movement was from each direction across the whole year; The larger a segment is, indicates that the wind travelled more frequently **from** that direction. Differences in the relative sizes of these segments between the two graphics reflect differences in the two modelled wind datasets used in the analysis, as discussed above.

The colours for each wind direction indicate the frequency with which different concentrations of SO_2 occurred when the wind was travelling from that direction; a larger proportion of a certain colour indicates that the corresponding concentration range occurred more frequently during the times when the wind was travelling from that direction. As the graphics show, for the large majority of the time SO_2 concentrations were below 20 $\mu g/m^3$, with the notably higher concentrations occurring infrequently. Concentrations over 20 $\mu g/m^3$ occurred most often when the wind direction was between north-northeasterly and south-southeasterly.

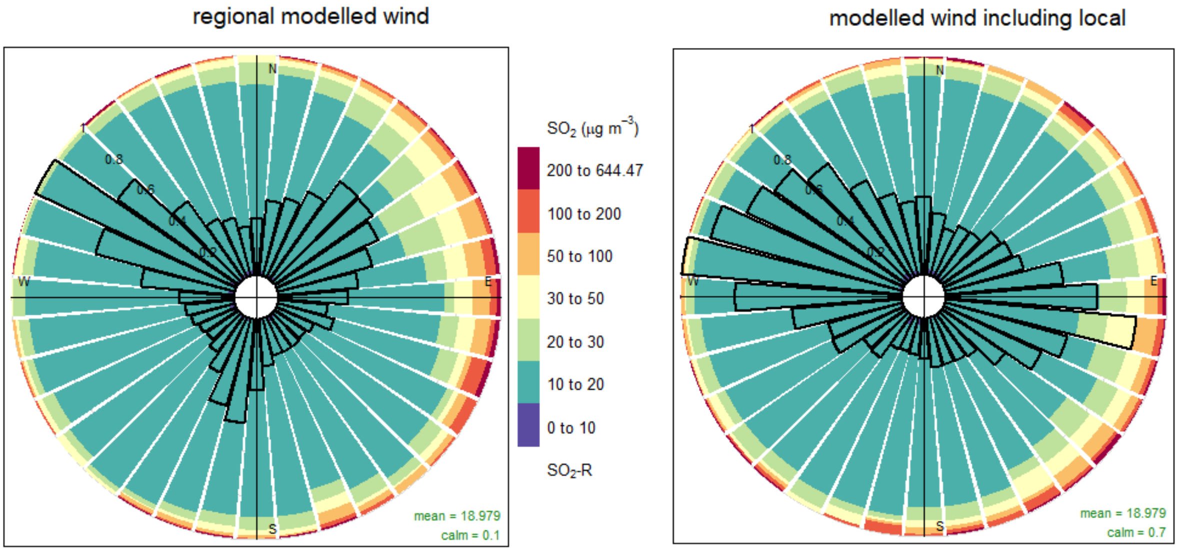


Figure 10. Frequency of SO_2 concentration according to wind direction, using both modelled wind datasets during 2023. Black outlines show wind direction frequency. Colour segments show the frequency of different hourly-average SO_2 concentrations observed for each wind direction sector.

Comparing times when Martisa 3 was operating against times when it was not operating, however, shows notable differences between these two conditions (Figure 11). Higher SO_2 concentrations occurred far more frequently during operating times (and in the 24 hours after operating), consistent with the information above.

The data indicate a notable SO_2 source to the east, in the direction of the Maritsa 3 complex, which is located at 100-110° from the Rakovski monitor. Higher SO_2 concentrations did also coincide with wind coming from other directions during times when Maritsa 3 was operating - especially when that analysis uses modelled wind data that includes local observations, which might be expected to provide a better representation of true wind patterns in the local vicinity.

It should be remembered, however, that the 'local' wind observations were made a significant distance from Dimitrovgrad and may not be representative of weather conditions in the immediate area. The modelled meteorological data with local observations includes many more hours of winds from directions away from the Maritsa 3 complex relative to the modelled meteorological data without local observations (Figure 5), meaning that it is not surprising this meteorological dataset causes high SO_2 concentrations to be distributed across more wind sectors.

The combined effects of wind speed and frequency of wind direction create significant complexities in the interpretation of air pollutant concentrations. For example, some of the high SO_2 concentrations occurring with wind directions away from the Maritsa 3 complex also coincide with low wind speeds. At low wind speed SO_2 dispersion is limited, leading to higher concentrations regardless of wind direction.

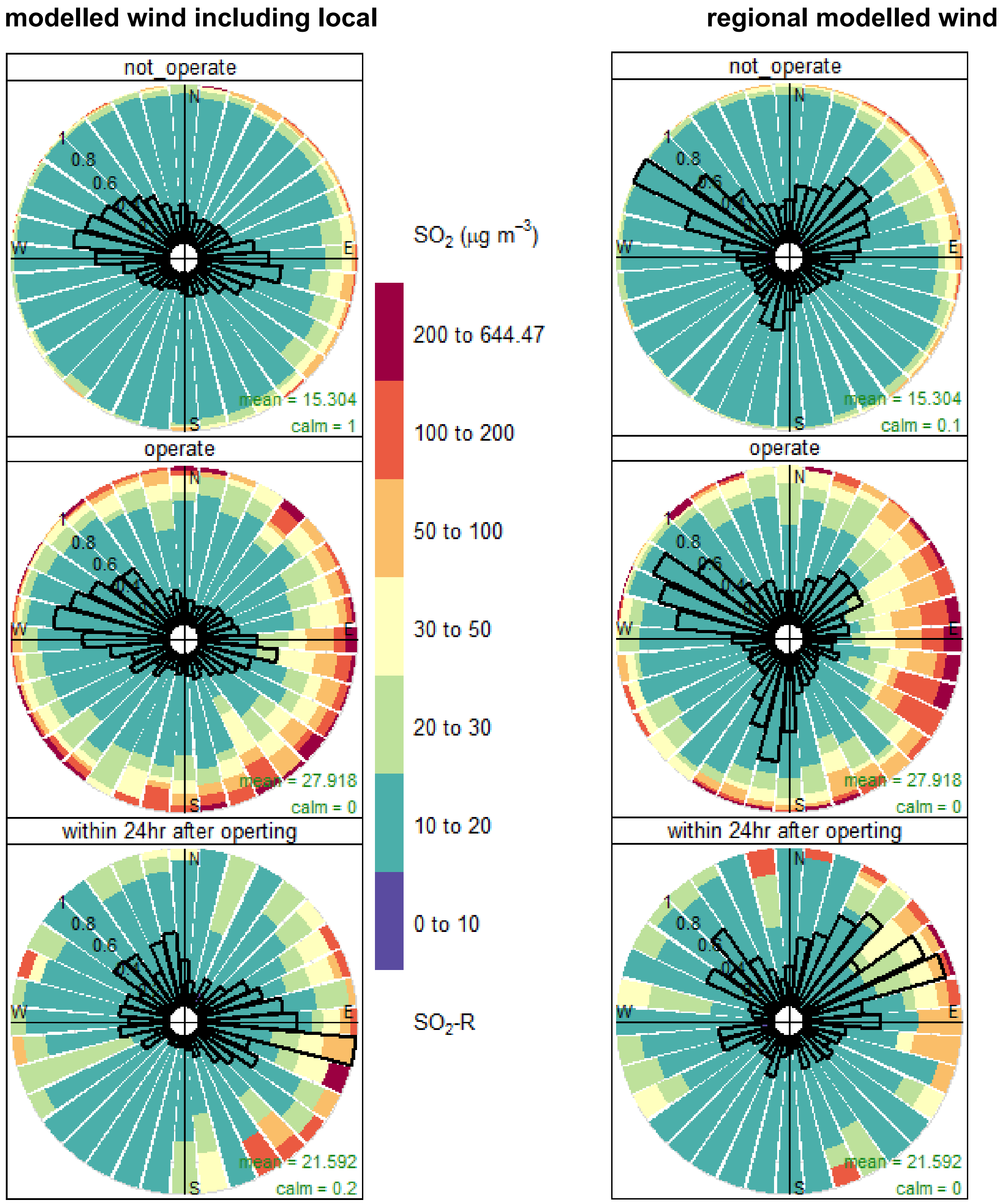


Figure 11. Frequency of SO_2 concentration according to wind direction, using both modelled wind datasets during 2023 and split according to the times when Marisa 3 CFPP was not operating (top), was operating (middle), and during the 24 hour period after operating (bottom). Black outlines show wind direction frequency in each case. Colour segments show the frequency of different hourly-average SO_2 concentrations observed for each wind direction sector.

An apparent relationship between SO_2 concentration, wind direction and wind speed (rather than frequency of wind direction) adds further evidence that emissions from the direction of the Maritsa 3 complex are an important air pollution source in the area. The highest SO_2 concentrations in both modelled meteorological datasets appear to coincide with easterly wind directions and with wind speeds between 0-10 m/s (Figure 12). When the observations are differentiated according to the Maritsa 3 CFPP operating times and standby times these clusters of relatively high SO2 concentrations were more clearly associated with times when the CFPP was known to be operating. It is also apparent during the 24-hour periods following operational periods, but the association is much weaker during non-operating periods (Figure 13).

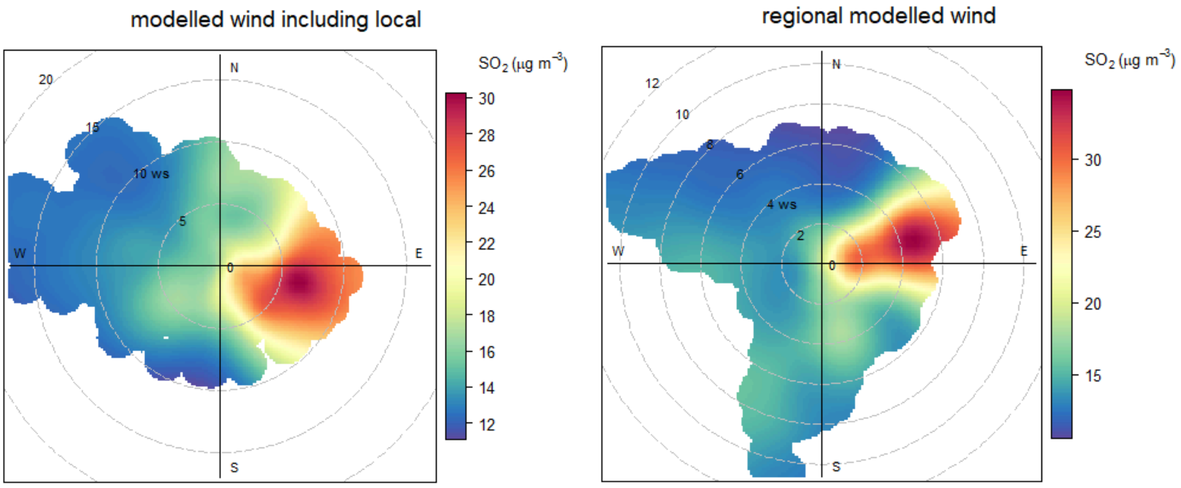


Figure 12. SO_2 concentrations in relation to wind speed (m/s) and wind direction data from both modelled datasets during 2023

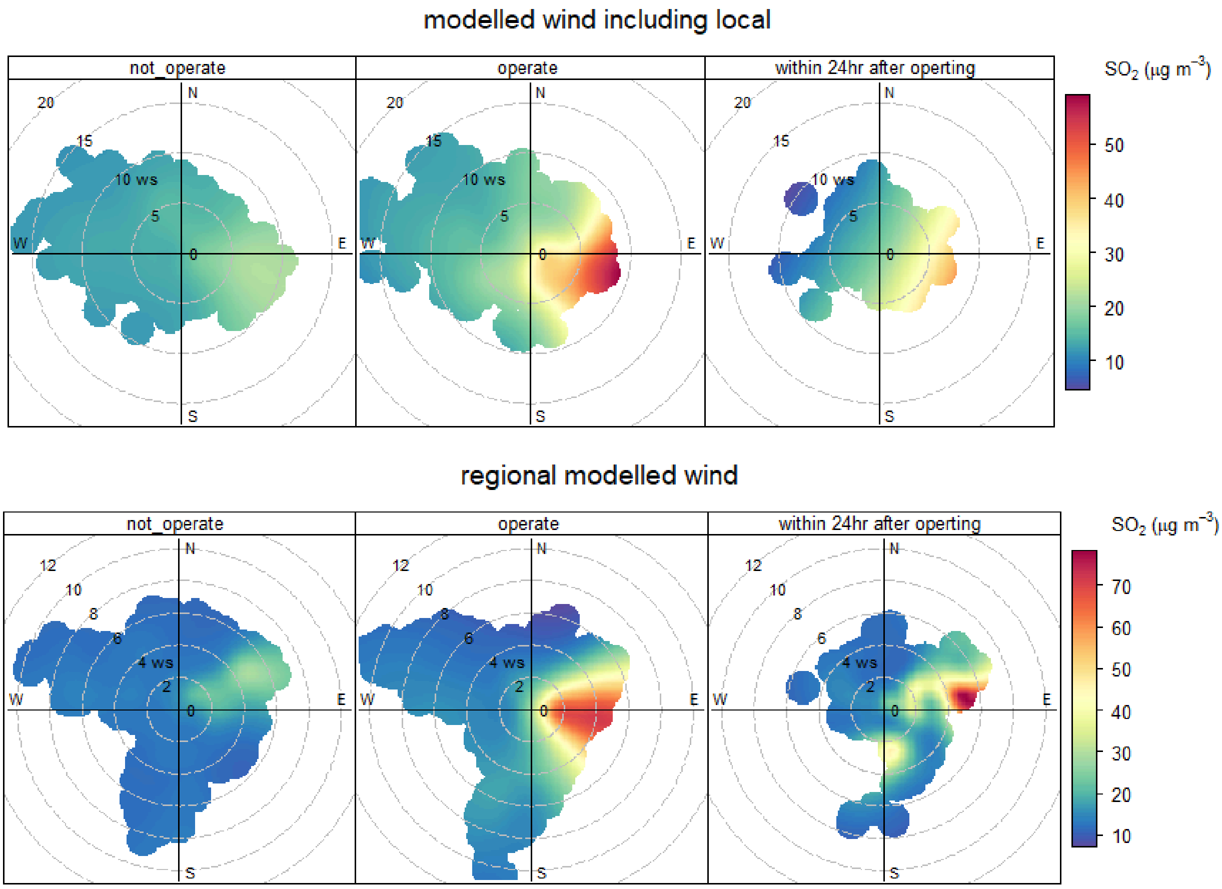


Figure 13. SO_2 concentrations in relation to wind speed (m/s) and wind direction data from both modelled datasets during 2023, split into times when the Martista 3 CFPP was not operating (left), was operating (middle), and during the 24 hour period after operating to account for SO_2 dispersal (right)

To assess the contribution to the annual SO_2 burden at the air quality monitoring station we weight SO_2 observations by the frequency of occasions when the wind originated from each direction and speed (Figure 14). This provides an indication of the wind directions and speeds that dominate the overall mean concentrations and hence the most important wind directions and speeds for exposure to SO_2 air pollution. The weighted mean concentration of SO_2 shows that annual mean concentrations appear to be driven by lower wind speeds and that the wind speed-direction pairing that contributed most to the annual mean is from the east, in the approximate direction of Maritsa 3 CFPP.

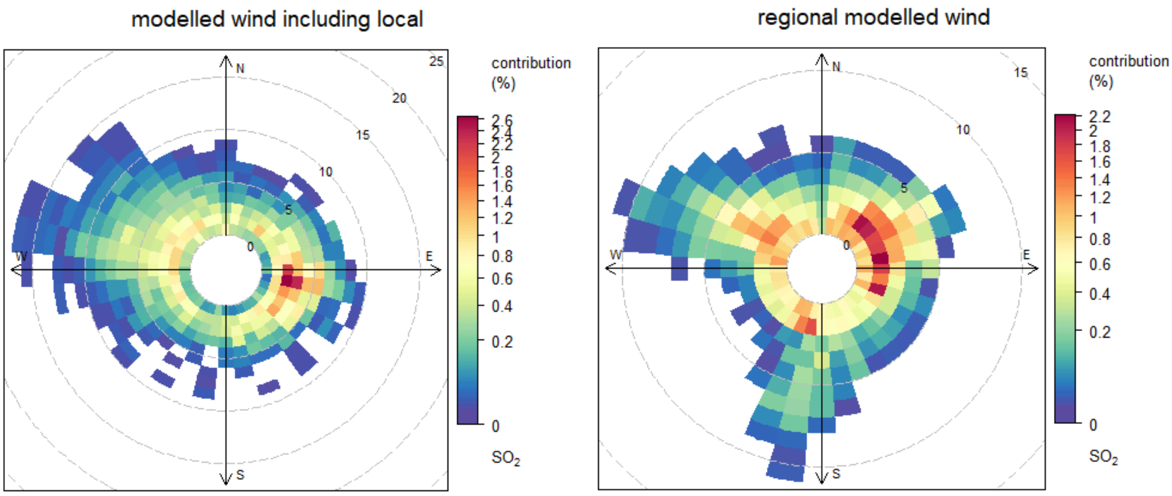


Figure 14. The contribution of SO_2 concentrations with respect to wind speed (m/s) and wind direction data from both modelled datasets during 2023

Conclusions

The Martisa 3 CFPP is located 2.5 Km to the east of central Dimitrovgrad where there is an air quality monitoring station. Hourly ambient concentrations of SO_2 measured by the air quality monitoring station during 2023 were analysed in conjunction with modelled meteorological data and data describing the operation of the Martisa 3 CFPP.

In 2023 the average SO_2 concentration was 19 µg/m^3. The daily average SO_2 concentration exceeded 40 µg/m^3 on many occasions, and hence exceeded the WHO's guideline value. The highest reported hourly average concentration was 644 µg/m^3.

SO_2 concentrations differed significantly when Martisa 3 was operating compared to times when it was not operating. Higher levels and far greater variability in concentrations were seen during times that the CFFP was in operation.

Combined concentration data, and wind direction and frequency data indicate that higher SO_2 concentrations recorded at the Rakovski monitoring station were moderately more frequent with winds originating along an arc between northeast and southeast. This arc broadly coincides with the upwind location of the Maritsa 3 complex.

In short, combined concentration, wind direction and wind speed data during 2023 indicate an appreciable SO_2 source to the east, in the direction of the Maritsa 3 complex.

The greatest contribution to the annual SO_2 burden at the air quality monitoring station is from winds aligned with the approximate upwind location of the Maritsa 3 complex.

The data presented here provide strong evidence that the operation of the Martisa 3 CFPP is likely responsible for increased SO_2 concentrations in Dimitrovgrad

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