SOME OBSERVATIONS ON MEASUREMENTS OF BACKGROUND CONCENTRATIONS OF ATMOSPHERIC BLACK CARBON AND SULPHUR DIOXIDE IN THE CENTRAL MEDITERRANEAN REGION

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ABSTRACT

Previous studies reported that the two main contributors to air pollution in the central Mediterranean region are ship emissions and emissions originating from the volcano on Mount Etna in Sicily. The objective of the work presented here is to investigate a possible link between the said pollution sources and concentration measurements obtained during a 4-month monitoring campaign which took place in 2021. Data was collected at the Giordan Lighthouse monitoring station, on the island of Gozo, forming part of the Maltese archipelago, in the central Mediterranean, which is ideally located to study regional atmospheric pollution. Furthermore, a relationship between black carbon in atmospheric aerosol and sulphur dioxide was sought. From February to May 2021, real-time measurements of black carbon and sulphur dioxide concentrations were obtained using an aethalometer, a multi-angle absorption photometer and a sulphur dioxide analyser. Meteorological parameters, e.g., wind speed and direction, were also measured concurrently. It was observed that during episodes linked to peak concentrations of the said pollutants, the prevailing wind was from the east–south sector, which diverges significantly from the direction of the two primary sources relative to the monitoring site, implying that there exists the possibility that the emission source/s associated with the data used in this current research differs from the ones previously documented. The outcome of this study necessitates further investigation of the air pollution scenario in the central Mediterranean.

Keywords: central Mediterranean, black carbon, sulphur dioxide, air pollution meteorology.

1 INTRODUCTION

Atmospheric aerosols play a significant role in atmospheric physical and chemical processes, the biosphere, climate change and human health. In recent years, the subject has been gaining substantial attention due to the impact of atmospheric aerosols on the energy balance of the earth (and hence climate change). Furthermore, airborne particles have a notable influence on human health. According to the European Environment Agency, in Europe, in 2020, long-term exposure to concentrations of fine particulate matter ($PM_{2.5}$) resulted in approximately 238,000 premature deaths [1].

A primary atmospheric aerosol component of note is black carbon, which is emitted directly into the atmosphere during combustion of fossil fuel or biomass burning. Black carbon plays an important role in the earth's climate due to its strong ability to absorb incoming solar and outgoing terrestrial radiation, which alters the earth's radiation budget, leading to a warming effect. Consequently, aerosol black carbon is emerging as a crucial anthropogenic forcing agent in climate change, second only to carbon dioxide [2].

As a constituent of $PM_{2.5}$, black carbon affects human health. Inhalation of these particles causes short-term symptoms such as asthma and bronchitis, as well as more grievous long-term symptoms including chronic irritation and inflammation of the respiratory track, which can even lead to cancer and other diseases.

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Society is encountering significant challenges in connection with climate change and atmospheric pollution. Apart from black carbon, an atmospheric pollutant that oxidizes in the atmosphere to form climate-influencing aerosols is sulphur dioxide (SO_2), which is a key player in the Earth's radiative balance since it is a precursor to the formation of sulphate aerosol [3]. The latter is dependent on the sulphur content of fuels. Furthermore, similar to black carbon, SO_2 is a primary pollutant that has a significant impact on human health.

Certain meteorological parameters such as precipitation, wind speed and direction have a considerable influence on the atmospheric residence time of aerosol particles. Wet and dry deposition serve as the primary means of black carbon removal from the atmosphere. Depending on local meteorological conditions, the atmospheric residence time of black carbon aerosol particles typically ranges from days to weeks. Dry deposition, albeit at a slow pace, can eliminate larger particles within a few days and the sub-micron fraction over several weeks. On the other hand, the mechanisms underlying wet deposition remain inadequately understood, rendering it one of the most uncertain processes in current models. Nevertheless, previous studies have shown that wet deposition emerges as the predominant process for rapidly mitigating the impacts of black carbon on climate, human health, and ecosystems. However, the scavenging of black carbon is contingent upon its source, i.e., whether it originates from biomass burning, or fossil fuel combustion [4]. Variations in wind speed and direction, also exert a significant influence on both the temporal and spatial fluctuations in black carbon concentrations. Increased wind speeds contribute to a reduction in concentrations of black carbon close to ground [5]. The direction of the wind determines the flow pattern and distribution of black carbon across the area. Knowledge of wind direction serves as indicator for determining the relevant source of pollution.

Relevant removal processes have received increasing attention in recent years as they play a crucial role in determining the atmospheric residence time of sulphur compounds. Thus, understanding these processes is vital for accurately assessing the environmental impact of both natural and anthropogenic emissions of sulphur compounds. As in the case of black carbon, wet and dry deposition are the two removal processes applicable to atmospheric sulphur compounds. Wet deposition involves the incorporation of sulphur compounds into cloud droplets (rainout) and their subsequent removal through falling precipitation (washout). On the other hand, the dry deposition process involves the direct collection of gaseous and particulate species on land or water surfaces. It has been observed that roughly half of the SO_2 emitted into the atmosphere is removed by dry deposition. The remaining portion undergoes oxidation to (eventually) form sulphates, which are then eliminated through precipitation. The atmospheric residence time for sulphur is approximately 5 days [6].

Meteorological parameters such as wind speed has direct bearing on the variations of SO_2 atmospheric concentration. It was determined that high wind speeds lead to a reduction in SO_2 concentration primarily due to the dilution effect [7]. As is the case of black carbon, wind direction has been empirically shown to be crucial in determining emission sources.

2 MONITORING STATION

Giordan Lighthouse, the monitoring station referred to in this study, is located on the northern coast of the small island of Gozo, forming part of the Maltese archipelago, in the Malta–Sicily channel, in the central Mediterranean (refer to Fig. 1). The exact location of the monitoring station is 36°04'24" North, 14°13'09" East, 167 m above sea level. This ambient air monitoring site lies on a hill top, in an area that is relatively unpolluted and unaffected by pollution from the main island of Malta. Due to its opportune location, greenhouse and reactive trace gases, and atmospheric aerosol concentrations are being recorded continuously, in real-time.

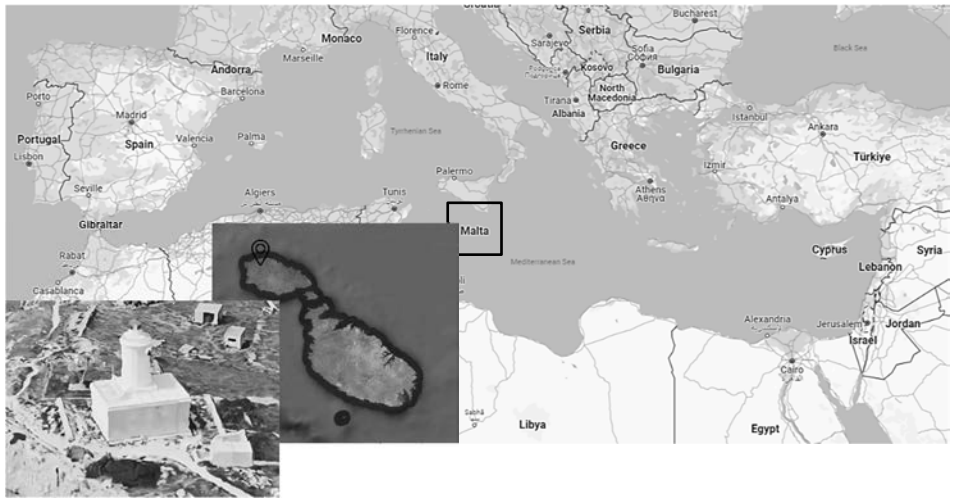

Figure 1: A map of the Mediterranean region, showing the location of the monitoring station located at Giordan Lighthouse.

In the region, one of the main sources of black carbon is diesel engines, used in (sea) transport. Ship emissions are continuously increasing due to the increase in global-scale trade contributing to regional, e.g., coastal, and global air pollution. Ship emissions primarily depend on the engine's size and the type of fuel used. Vessels equipped with diesel engines running on heavy fuel oil release gases such as SO_2, as well as black carbon in the form of particles. Giordan Lighthouse is ideally located on a main (and densely trafficked) shipping lane in the Mediterranean, which extends from the Suez Canal to the Strait of Gibraltar. For these reasons, black carbon and SO_2 are amongst the pollutants whose concentration is being measured at the monitoring station. Some of the collected data were analysed as part of the work being presented here.

The other important source of pollution in the region, is the volcano on Mount Etna, near the city of Catania on the eastern coast of nearby Sicily. Due to its close proximity, Etna eruptions have an influence on the Maltese islands. This is another reason the monitoring station was set up at Giordan Lighthouse.

3 INSTRUMENTATION

The monitoring station is equipped with various ambient air monitoring equipment for measuring concentrations of several gases and aerosols. At present, Giordan Lighthouse is equipped with monitors for greenhouse gases (specifically, carbon monoxide, carbon dioxide, methane and atmospheric water content), reactive gases (specifically, SO_2, nitrogen oxides and ozone) and aerosols. The aerosol monitoring system consists of an aethalometer, multi-angle absorption photometer (MAAP), optical particle counter (OPC) and a scanning mobility particle sizer (SMPS) in conjunct with a differential mobility analyser (DMA). In addition to this equipment, the station also has installed two low-volume air samplers. Furthermore, various meteorological instruments and sensors, which measure wind speed and direction, as well as ambient temperature and relative humidity are installed on the roof top of the lighthouse.

The monitors are fed through two sample inlets, namely a PM_{10} aerosol inlet and an inverted U-shape polytetrafluoroethylene (PTFE) tube. The latter feeds the gas monitors. These two inlets are mounted on the roof top of Giordan Lighthouse. The gas and aerosol monitors are installed in an air-conditioned room immediately below the roof top of the building. The instrumentation room is kept at a temperature of about 25°C.

3.1 Black carbon instrumentation

Mass and number concentration of atmospheric aerosol particles in the range from 10 nm to 10 μm are measured using the four instruments mentioned previously, i.e., aethalometer, MAAP, OPC and the SMPS-DMA. The volume flow rate for the entire aerosol sampling and measurement system is maintained at 16.7 l min^{-1}. The system, whose schematic is shown in Fig. 2, is located in the room immediately below the roof top where the PM_{10} inlet and a water trap are installed. The flow line is split; one branch leads to the OPC and the other to a PM_1 cyclone, which limits the size of particles to under 1 μm. Following the cyclone is an isokinetic splitter that leads to three Nafion driers, each connected to the aethalometer, MAAP and SMPS-DMA, separately. Note that the Nafion driers ensure that the relative humidity is maintained at 40% or less in accordance with the World Meteorological Organization – Global Atmosphere Watch standards, given that the monitoring station at Giordan Lighthouse is a recognised Global Atmospheric Watch Station.

In this study, and as indicative in Fig. 2, two instruments were employed to measure the concentration of black carbon present in atmospheric aerosol particles, as detailed hereunder.

Black carbon mass concentration in atmospheric aerosol particles can be measured using different techniques. The optical attenuation method is one such technique, and is extensively

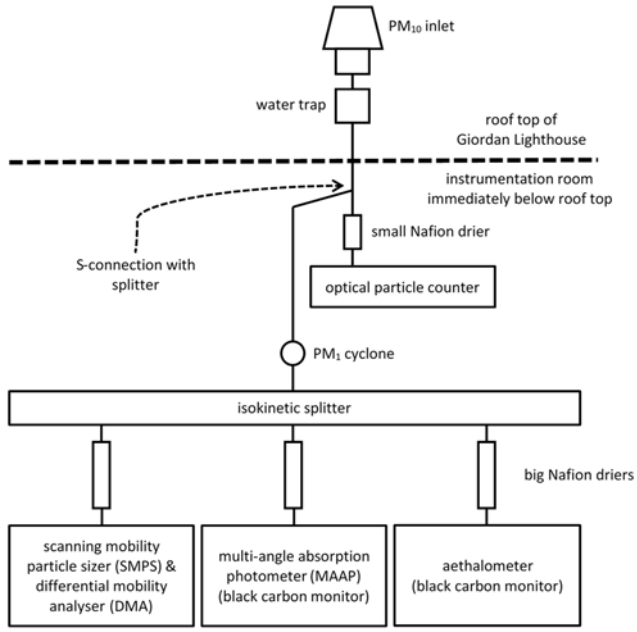

Figure 2: Schematic diagram of the instrumentation setup at Giordan Lighthouse for measuring the concentration of aerosols in ambient air.

employed for real-time measurements. It offers a non-contaminating and non-destructive means of measurement. One of the most widely used instruments, which is based on this technique, is the aethalometer [8]. The aethalometer used in this study is the Magee Scientific Model AE33. Featuring high time resolution and high sensitivity, this instrument measures light absorption at seven different wavelengths, namely 370, 470, 520, 590, 660, 880 and 950 nm, by collecting aerosol particles on a quartz fibre filter [2]. In this current study, data relating to the 880 nm wavelength was used. It is worth noting that the AE33 aethalometer incorporates the dual-spot algorithm. The latter provides significant advantages. For instance, the algorithm allows for compensation with regards to the spot-loading effect and also gives values for the 'loading compensation' parameter in real-time. The net result is that the instrument/technique provides thorough knowledge of the optical properties of the sampled aerosol [9]. In this technique, two measurements are obtained simultaneously, from two sample spots with different rates of accumulation of aerosol mass, for a given single air stream. These two measurements are then combined mathematically to eliminate nonlinearities and provide the compensated particle light absorption and black carbon mass concentration [10].

Another filter-based instrument that was used in this study is the MAAP. Specifically, the Thermo Scientific Model 5012 instrument was used on this occasion. The MAAP measures atmospheric black carbon mass concentration at a single nominal wavelength of 637 nm [11]. The technique used for measuring aerosol black carbon mass concentration comprises the determination of the absorption of an aerosol sample collected on a glass fibre filter. The distinctive aspect of measuring the absorption coefficient using multi-angle absorption photometry is that the signals scattered to angles of 130° and 165° are measured, additional to the typical transmission measurement [12]. Furthermore, the MAAP uses a radiative transfer model, where the single scattering albedo and the optical depth of the aerosol loaded filter coincide with the measured transmitted (0°) and reflected (130° and 165°) signals. As a result, the absorption coefficient is determined, and the black carbon mass concentration is computed [13].

3.2 Gas sampling and measurement system: Sulphur dioxide analyser

The greenhouse and reactive gas, i.e., trace gas monitors are connected to a glass manifold, which is located inside the room immediately below the roof top of the lighthouse, where the instruments are installed. The said manifold is fed via the (inverted) air inlet at the roof top, and is equipped with a water trap. A blower is used so as to compensate for the pressure drop and thus concentrations are measured at ambient atmospheric pressure. The sampling and measurement setup for the trace gases, as it pertains to SO_2, is shown in Fig. 3.

In this study, the SO_2 concentration was measured using a Thermo Scientific Ambient Sulphur Dioxide Analyser Model 43i, which operates on the basis of the fact that SO_2 molecules absorb ultraviolet (UV) light, becoming excited at one wavelength, and subsequently emitting UV light at a different wavelength as these molecules decay to a lower energy state [14]. The associated pulsed fluorescence technology is able to measure SO_2 concentrations down to 0.05 parts by billion (ppb), with a time resolution of 300 s (as averaging time) [15].

Ambient air is drawn through the PTFE tubing and into the analyser through a 47 mm × 5 μm PTFE filter, to remove airborne dust particles [16]. The air sample passes through a hydrocarbon 'kicker', which eliminates hydrocarbons from the sample by forcing the hydrocarbon molecules to permeate through the tube wall. The 'clean' air sample then flows

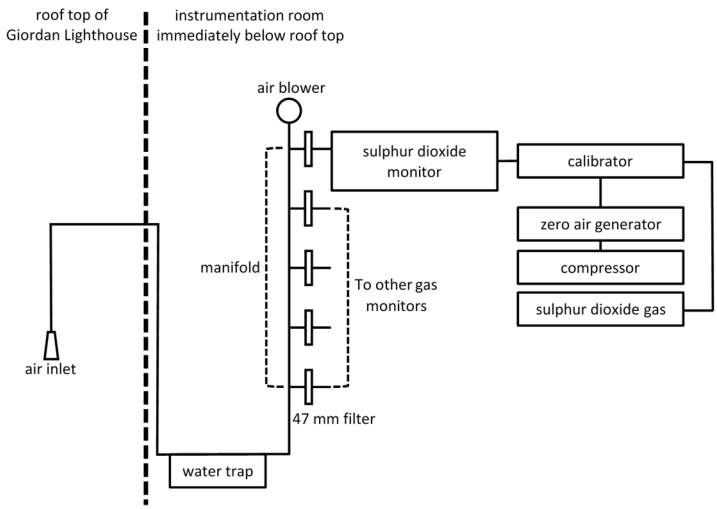

Figure 3: Schematic diagram of the instrumentation setup at Giordan Lighthouse for measuring the concentration of trace gases (SO_2, in particular) in ambient air.

into the fluorescence chamber, where pulsating (at 10 Hz) UV light excites the SO_2 molecules. It is worth noting that the condensing lens focuses the pulsating UV light into four selective mirrors that reflect only the wavelengths capable of exciting SO_2 molecules. Once these excited SO_2 molecules decay to lower energy levels, UV light is re-emitted in proportion to the SO_2 concentration present in the chamber (and sampled air). The reflective band pass filter limits the light reaching the photomultiplier tube to the SO_2 fluorescence wavelengths [14].

4 DATA COLLECTION, PROCESSING AND ANALYSIS

The black carbon and SO_2 concentration data reported here were collected during a 4-month period, i.e., February to May 2021, using the aethalometer, MAAP and SO_2 analyser as detailed previously. The sample flow rate was set to 5.0, 8.0 and 0.5 l min^{-1}, respectively. Concentrations were recorded every minute and then hourly averages were calculated accordingly, during the processing stage.

Fig. 4 shows the time series of black carbon and SO_2 concentrations measured at Giordan Lighthouse for the mentioned time period. The black carbon concentration ranged from a minimum of zero to a maximum of 1.64 μg m^{-3} (mean value of 0.25 ± 0.19 μg m^{-3}; the latter being the relevant standard deviation) and from 0.01 to 1.56 μg m^{-3} (mean value of 0.27 ± 0.19 μg m^{-3}; the latter being the relevant standard deviation) for the MAAP and aethalometer, respectively. The SO_2 concentration ranged from 0.10 ppb to 1.16 ppb (mean value of 0.23 ± 0.10 ppb; the latter being the relevant standard deviation). In most cases, from these plots, one notices that high concentration levels of black carbon were not associated with high concentration levels of SO_2.

Fig. 5 depicts sections of the time series shown in Fig. 4, but with better abscissa/timeline resolution, and combining data from all three parameters, so as to highlight the relevant peaks in black carbon (from both instruments) and SO_2 concentrations. Good correlation between concentration of the two pollutants is not obvious. In other words, peaks in black carbon

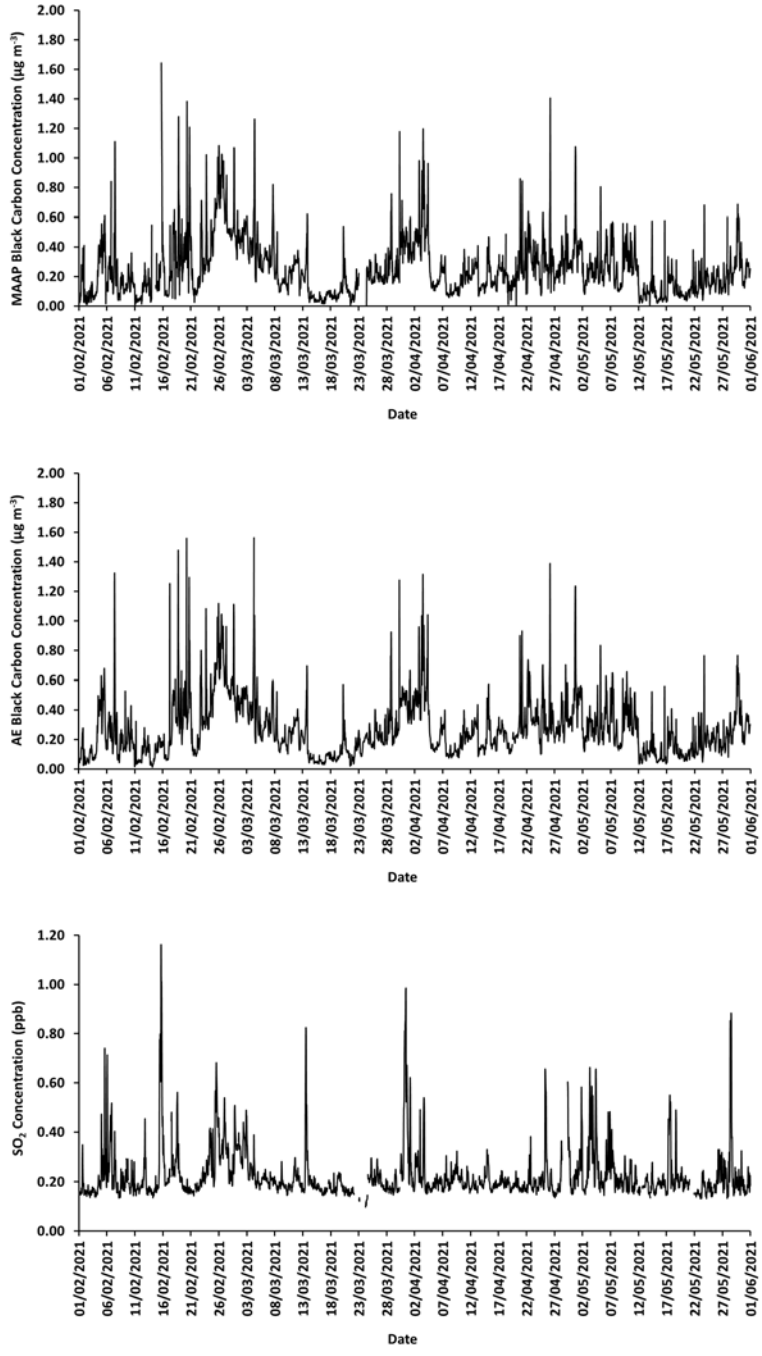

Figure 4: Time series (February to May 2021) for black carbon mass concentrations measured with the MAAP and aethalometer (AE), as well as for SO_2 concentration. All measurements were obtained at the monitoring station located at Giordan Lighthouse.

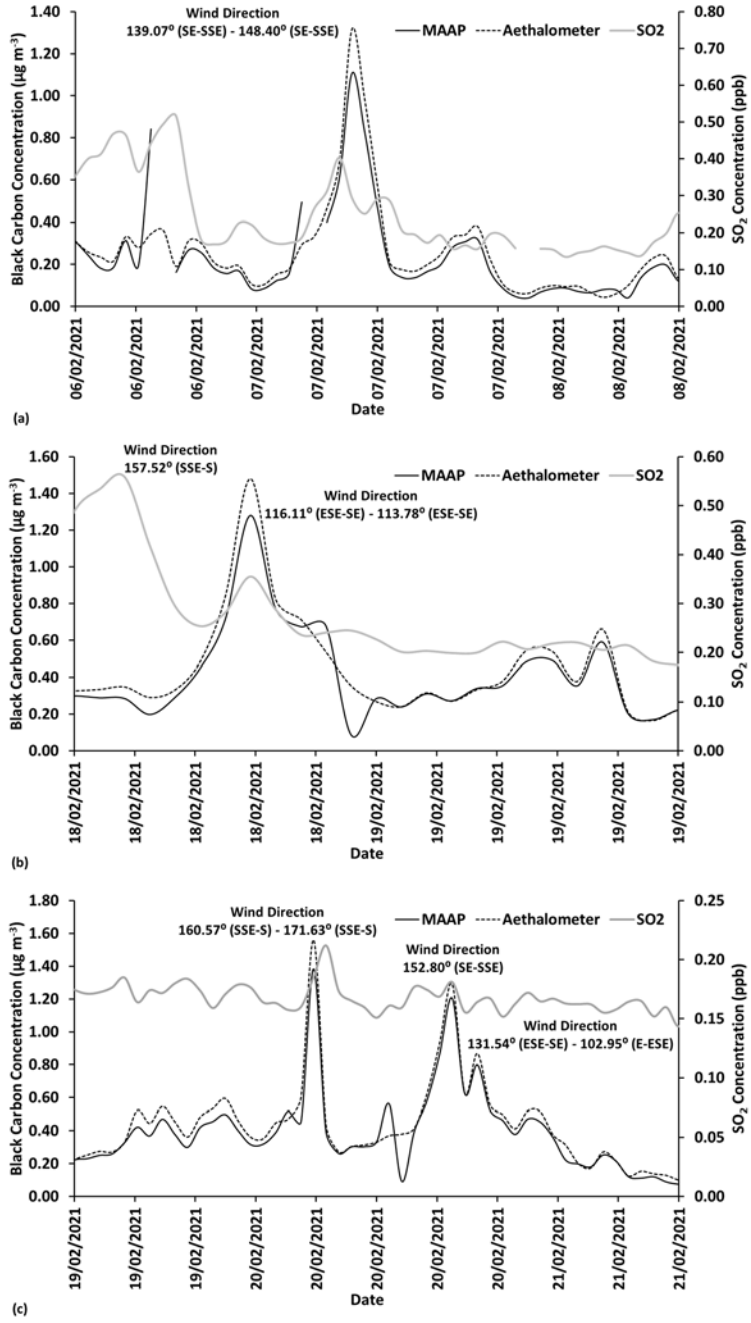

Figure 5: Peaks in black carbon concentrations obtained from the MAAP and aethalometer in comparison with the time series for the SO_2 concentration as measured at the monitoring station located at Giordan Lighthouse. The wind direction associated with peaks is indicated.

concentration were not accompanied by maxima in SO_2 concentration, which was the case in earlier investigations (refer to [17] and [18]). Furthermore, the findings in this study fail to agree with previous studies by Ziola et al. [12], where concentrations of all trace gases under study, including SO_2, correlated very strongly with the concentration of black carbon. Indeed, this fact highlights the need for further, and more detailed, investigation.

The data analysis resulting from the current study, indicates a moderate correlation between black carbon and SO_2 concentrations (with a slope of approximately 0.6 and a correlation coefficient which is just under 0.7). This is possibly due to the intricate nature of the relevant emission sources and the partial prevalence of particular sources during certain measuring periods. Nevertheless, such hypotheses require rigorous testing.

Another critical factor that influences the outcome is meteorology, especially wind direction and speed. In fact, wind is considered a significant factor that influences local-scale weather conditions and affects black carbon concentration due to variability in advection [19]. From Fig. 5, one can further observe that the prevailing wind, for the peaks shown ((a), (b) and (c)), was from the east–south sector. This was also the case for other identified peaks that are not shown here, except for one specific SO_2 peak, where the wind was from the south–south-west sector. Another important observation is that while the MAAP and aethalometer compared quite well (with a slope of approximately 1.0 and a correlation coefficient which is equal to 0.96), the latter measures consistently higher than the MAAP, even if marginally. According to an inter-comparison study carried out by the Leibniz Institute for Tropospheric Research and the European Centre for Aerosol Calibration and Characterization, it was found that the aethalometer, used in the current study, typically gives up to 35% higher values than the MAAP, and that such differences could be due to varying sensitivities to the type of aerosols present [20].

As highlighted earlier on, the two relevant primary sources of atmospheric pollution identified in the Malta–Sicily Channel are ship emissions and the volcano on Mount Etna. From a recent study [21], using data obtained at the monitoring station located at Giordan Lighthouse, for pollution concentration to correlate with ship emissions, the prevailing wind has to be from the west–north-east sector. However, apportioning black carbon in atmospheric aerosols to specific ship plumes poses challenges. Factors such as overlapping of several (ship) plumes, fluctuating wind speed and direction, and intense sea traffic make it difficult to distinguish which ship is associated to which plume. Complicating matters further, an elevated mesoscale background concentration and noise often overshadow the relatively lower concentrations found in ship plumes, which makes it even more complex to identify ship-related pollution episodes or peaks [22].

The effect of the Etna volcanic ash plumes or clouds, on the Maltese islands, was investigated using data obtained from the monitoring station at Giordan Lighthouse [23]. Once again, wind turns out to be an important factor in the dispersal and dilution of volcanic plumes, as both its direction and speed regulate the dispersion of volcanic ash. For Etna's volcanic cloud to hover over the Maltese islands, the geostrophic wind must blow from the north-east towards the islands. However, the prevailing wind direction on the islands is from the north-west. This means that the percentage occurrence of wind speed and direction favourable to steering Etna's volcanic clouds toward the islands is only 13% [23]. Nevertheless, more research is necessary to better understand the effects of the volcano on Mount Etna on the Maltese islands, and more generally, to better understand volcanic ash dispersal.

For the data used in the current study, the prevailing wind was from the east–south sector, which is way off from the direction of the two sources discussed earlier. Hence, there exists the possibility that the main pollution source pertaining to the data used in the current study,

is neither ship nor volcanic emissions from Mount Etna but some other activity and thus further study is required. As regards peaks of black carbon and SO_2 concentrations, and how they relate to each other, identifying the location of the original source/s through back trajectory calculations is necessary and decisive.

5 CONCLUSIONS

Continuous measurements of black carbon and SO_2 concentrations were collected at Giordan Lighthouse monitoring station, using an aethalometer, MAAP and an SO_2 analyser over a 4-month period, i.e., February to May 2021. Research efforts were focused primarily on the investigation of peaks in black carbon and sulphur dioxide concentrations; checking for coincidences. Furthermore, a preliminary attempt was made to identify the location of the emission source/s of the said pollutants.

A moderate correlation between black carbon and SO_2 concentrations was noted. Nevertheless, from the time series of concentrations for the considered 4-month period, it seems that relatively high concentrations of black carbon were not accompanied by high concentrations of SO_2.

Certain meteorological parameters, in particular wind speed and direction, affect black carbon and SO_2 concentrations. During the monitoring period under consideration, the prevailing wind associated with the observed concentration peaks was from the east–south sector, except for one specific case, when the wind was from the south–south-west sector. This is in disagreement with previous studies. In the latter, it was found that the two main sources of atmospheric pollution (atmospheric black carbon and SO_2, in particular) in the central Mediterranean, are ship and volcanic (referring to Mount Etna) emissions, which are located in the west–north-east sector and the north–east sector, respectively, with respect to the monitoring station. Thus one can conclude that there exists the possibility that the emission source pertaining to the data used in this current research, is different from the ones previously reported. For this reason, further studies are required mainly to identify the associated source/s. One possible approach would be back trajectory modelling and source apportionment through chemical analysis using specific markers (if possible).

As a side note, it was also observed that the correlation between the two instruments measuring black carbon concentration, i.e., the aethalometer and MAAP, was strong (as expected). This can be observed from Fig. 5, since peaks from the MAAP coincided quite well with the peaks obtained from the aethalometer. Furthermore, it can be noticed that the latter measures consistently higher than the MAAP, even if marginally, which agrees with previous studies.

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