ANALYSIS AND OBSERVATIONS CONCERNING CONCENTRATIONS OF GREENHOUSE GASES MEASURED OVER AN 11-YEAR PERIOD IN THE CENTRAL MEDITERRANEAN REGION

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ABSTRACT
Measurements of concentrations of various greenhouse gases were collected over a time period of 11 years (2012–2023). The data were collected at Giordan Lighthouse monitoring station on the island of Gozo, forming part of the Maltese archipelago, in the central Mediterranean region. The associated data analysis and observations are presented. It turns out that the methane concentration is increasing at a relatively substantial rate of 9.6 parts per billion by volume (ppbv) per year. Similarly, carbon dioxide is showing a mean increase of 2.15 parts per million by volume per year. Similar trends have been reported in the eastern Mediterranean region. A minimal increase is observed in carbon monoxide concentration and atmospheric water content, at a rate of 0.32 ppbv per year and 0.02% per year, respectively. Both methane and carbon dioxide show maxima in winter and minima during the summer months. Carbon monoxide also shows a maximum in winter but a minimum during summer and autumn. However, carbon monoxide exhibits a secondary maximum in August. In contrast, atmospheric water content presents a clear maximum in summer and a minimum in winter as expected for the Mediterranean climate. With respect to wind direction, methane, carbon monoxide and carbon dioxide clearly show maxima when the prevailing wind is from the north sector and secondary broad maxima when the wind is blowing from the south-east sector. Greenhouse gases exhibit a clear diurnal variability with a maximum in the early morning and a minimum in the afternoon. Both the diurnal maximum and minimum of carbon dioxide precede that of carbon monoxide.
Keywords: central Mediterranean, greenhouse gases, methane, carbon monoxide, carbon dioxide, water vapour.

1 INTRODUCTION

Global warming and climate change have become a hot topic during the last decades. Anthropogenic activities since the industrial revolution have gradually contributed to the increasing concentration of atmospheric greenhouse gases (GHGs). Evidence shows that elevated concentrations of GHG emissions are significantly responsible for global warming and climate change [1].

Climate change has a pronounced effect on sensitive regions like the Mediterranean basin. Giorgi [2] considers the Mediterranean region as a climate change hotspot due to its responsiveness to climatological variability. Modelling studies indicate that the Mediterranean region is likely to warm up (referring to surface temperature) at a rate of 20% more than the global annual mean [3]. Another analysis has shown that the eastern Mediterranean and Middle East region is warming almost two times faster than the global average and more rapidly than most other inhabited parts of the world [4]. Most noticeably this impact has increased during the last 10 years as it was the warmest decade since 1880 [5]. The rise in the average temperature is triggering extreme weather events such as heatwaves, droughts, dust storms, and torrential rain [4].

Atmospheric carbon dioxide (CO_2) and methane (CH_4) are the most important anthropogenic GHGs as they play a significant role in the radiation balance of the earth. The

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primary sources of CO_2 emissions are those associated with the burning of fossil fuels, cement production, and land use changes due to deforestation [6]. Land and oceans are major sinks of CO_2, and remove about half of the anthropogenic emissions. On a global scale, evidence shows that during the decade 2013–2022, land CO_2 sinks accounted for 31%, while ocean CO_2 sinks accounted for 26% of the total CO_2 anthropogenic emissions [7]. During the year 2023, the global atmospheric CO_2 concentration reached 419.3 parts per million by volume (ppmv), which is approximately 51% more than that during pre-industrial times [7]. The atmospheric residence time of CO_2 varies from 5 to 200 years due to the different rates of uptake by different removal processes [8].

CH_4 is the second most important contributor to global warming after CO_2. Although CH_4 is far less abundant in the atmosphere than CO_2, and has a mean lifetime of approximately one order of magnitude less than that of CO_2 [5], it has the potential to trap more heat per molecule than CO_2. CH_4 has a global warming potential that is 86 times stronger per unit mass on a 20-year timescale, and 28 times more powerful on a 100-year time scale, than CO_2 [9]. The atmospheric lifetime of CH_4 molecules is about 12 years. It is worth noting that, CH_4 emissions can be classified into thermogenic, biogenic and pyrogenic [10]. In 2019, atmospheric CH_4 concentration reached 1866 parts per billion by volume (ppbv), which is an increase of 156% over that in the year 1750 [11].

Due to the impact of GHGs on the climate and air quality, GHGs are continuously monitored around the globe using various techniques such as ground-based instruments, airborne and satellite measurements. In this paper, we present an analysis of a data set concerning GHGs, that spans 11 years. The said data set, which is longest of its kind in the region, consists of ground-level observations of atmospheric carbon monoxide (CO), CO_2, CH_4, and water content (H_2O) concentrations, as well as relevant meteorological parameters, e.g. wind speed and direction. Gas concentration measurements were made using a cavity ring-down spectrometer, specifically Model G2401 by Picarro, installed at the monitoring station located at Giordan Lighthouse, on the island of Gozo, forming part of the Maltese archipelago, in the central Mediterranean. The monitoring station forms part of the Global Atmospheric Watch network of the World Meteorological Office.

2 METHODOLOGY: MONITORING SITE, INSTRUMENTATION AND DATA PROCESSING

The atmospheric monitoring station at Giordan Lighthouse is located on the northwest side of the island of Gozo; approximately 36°40′N, 14°30′E, and 167 m above sea level. It is representative of the atmosphere in the region in that it is considered background on the local scale, since there are no significant anthropogenic activities occurring in the vicinity except for some farming activity in the surrounding fields. The monitoring station is close to the coastline and faces the sea within the sector 260°–60°. During summer, the area is dominated by high-pressure zones leading to strong solar radiation, high temperature and generally low wind speeds [12]. In Summer, the general weather conditions are relatively homogenous for the whole Mediterranean basin and are characterized by north-westerly winds and sea-breeze circulations driven by land-sea thermal contrast [13].

The monitoring station started its operations in 1997 and is equipped to measure, trace gas and aerosol concentrations, as well as meteorological parameters. A detailed description of the installed instruments and acquired initial funding are given by Nolle et al. [14] and Azzopardi et al. [15].

At Giordan Lighthouse monitoring station data is recorded at one-minute intervals, and is flagged daily according to the established EBAS standard procedure. Thorough quality control and quality assurance procedures are adhered to as per the requirements of EBAS. It is worth noting that, EBAS is a database hosting observation data of atmospheric chemical composition and physical properties (https://ebas-data.nilu.no/), developed and operated by the Norwegian Institute for Air Research. On an annual basis, the data are aggregated to hourly values and then uploaded to the EBAS database in Greenwich Mean Time (GMT) format. Indeed, the data set used in the current study is available for download from the EBAS database. However, the time series has been corrected to GMT+1 to correspond to the Malta time zone.

The measurements of CO, CO_2, CH_4 and H_2O concentrations started on 9 October 2012 and lasted until 31 July 2023. Measurements were recorded with a cavity ring-down spectrometer (Model G2401 by Picarro). This is the longest data set of GHG measurements in the central Mediterranean. However, it should be noted that there are two major gaps in the data set. The first major data gap corresponds to the period from 4 April 2016 to 21 April 2017. The second data gap occurred from 10 November 2021 to 25 July 2022. On both occasions, the instrument developed a major fault and had to be returned to the original manufacturer in California, USA for repairs, a general check and calibration. On 31 July 2023, the monitoring station was completely shut down due to the start of major works on the only country road leading to Giordan Lighthouse, making the site inaccessible. By the time of writing this paper, the measuring facility is still not operational. It is hoped that operations are resumed by the end of summer 2024.

A span calibration check of the Picarro analyser is done twice a year for the CO, CO_2 and CH_4 channels. The certified span gas mixture used during the checks has specific concentrations of the relevant gases, as follows: CO at 200 ppbv, CO_2 at 300 ppmv and CH_4 at 1.5 ppmv, and comes in a 10-litre cylinder from Air Liquide, Germany.

The data for the 11-year long observational period was prepared into a Microsoft Excel file and then imported into International Business Machines (IBM) Corporation Statistical Package for the Social Sciences (SPSS) software. The built-in date and time wizard function of IBM SPSS was used to create new variables for year, month and day of the year from the date variable. Furthermore, the recode built-in function was used to transform the wind direction data into 36 bins of 10-degree intervals. The recode function was also used to assign the correct season to each day of the year. Data averages according to year, month, wind direction and season were all accomplished through IBM SPSS. The results, i.e., data processing outputs of the IBM SPSS were then imported into a new Microsoft Excel file for further analysis and plotting of graphs.

3 RESULTS OF THE DATA ANALYSIS

In the following subsections, the obtained results are discussed in detail. First, a discussion of the trends is given followed by details concerning seasonal and diurnal variability. The influence of wind direction on trace gas concentrations is also discussed.

3.1 Trends analysis

The analysis of the GHG data reveals a clear annual trend for CO_2 and CH_4 concentrations, as demonstrated in Fig. 1 and Fig. 2, respectively.

An overall increase in concentration is observed for both CO_2 and CH_4 with annual mean concentrations ranging from 386 ppmv and 1851 ppbv in 2012 to 441 ppmv and 1963 ppbv in 2023, respectively. Similar observations have been reported by Gialesakis et al. [5] for the eastern Mediterranean, from measurements at Finokalia, on the island of Crete. The authors reported a long-term increase in concentration for both CO_2 and CH_4 with yearly

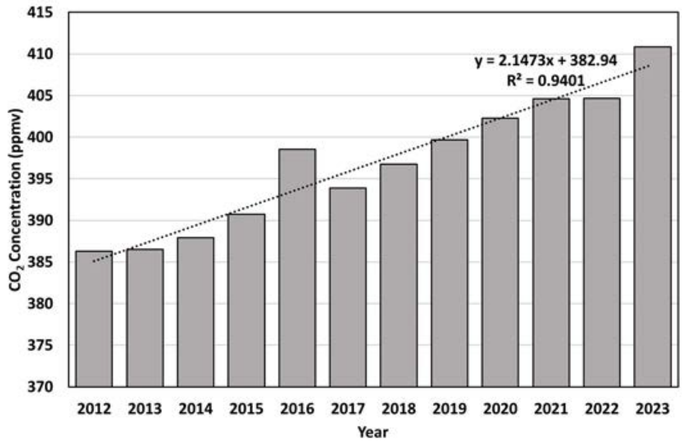

Figure 1: Time series of the annual average carbon dioxide concentration for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

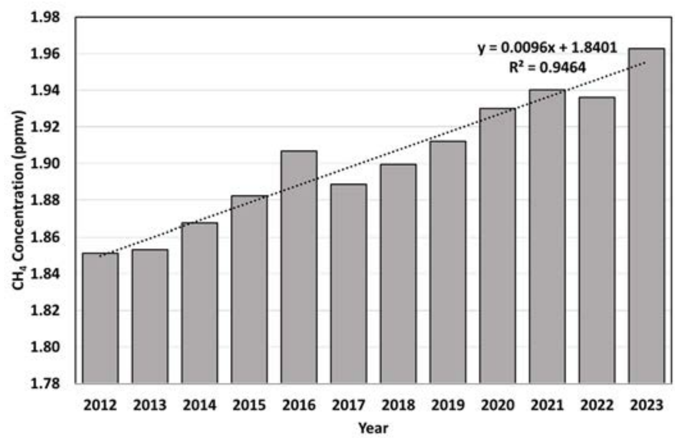

Figure 2: Time series of the annual average methane concentration for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

average concentrations ranging from 375 ppmv and 1862 ppbv in 2002 to 417 ppmv and 1978 ppbv in 2021, respectively. Evidently, the mean concentration for CH_4 is higher in the eastern Mediterranean as compared to the central Mediterranean. However, the CO_2 yearly mean concentrations are much higher in the central Mediterranean. This can be attributed to anthropogenic emissions originating from marine traffic in the said region. The Strait of Sicily is the busiest shipping route in the Mediterranean as it provides a vital link between Asia, Europe and North America [16]. Ship emissions are a major source of anthropogenic pollution in the Malta–Sicily channel. From a geographical point of view, the monitoring station at Giordan Lighthouse is closer to the international shipping lane than the station at Finokalia, which is located on the northeast of Crete. Therefore, atmospheric concentration of CO_2 originating from ship emissions is more pronounced at Giordan Lighthouse monitoring station than at the observatory of Finokalia.

The undertaken trend analysis also reveals that the CH_4 concentration is increasing at a relatively substantial rate of 9.6 ppbv per year. Similarly, the CO_2 concentration is exhibiting a mean increase of 2.15 ppmv per year. This is comparable with what has been reported for the eastern Mediterranean where the CH_4 concentration is increasing at a mean rate of 7.5 ppbv per year, while that of CO_2 is increasing at a rate of 2.4 ppmv per year [5]. These values are in line with the northern hemisphere mean growth rate observed over the same period at Mauna Loa (Hawaii), which is a reference site of the Earth System Research Laboratories, National Oceanic and Atmospheric Administration. At the latter reference site, the mean growth rate for CH_4 and CO_2 concentrations was 10.34 ppbv per year and 2.44 ppmv per year, respectively (https://www.esrl.noaa.gov/gmd/ccgg/trends/gr.html). A 10-year CO_2 concentration measurement campaign, from May 1992 to March 2002, on the island of Lampedusa (approximately 155 km southeast of Gozo) also shows an average increase of 1.7 ppmv per year [17]. Hence, one can conclude that in-situ measurements indicate that the CO_2 concentration in the central Mediterranean has been continuously on the increase during the last three decades.

3.2 Seasonal variability

The monthly mean concentrations of CO, CO_2, CH_4 and H_2O were calculated from the hourly values. Fig. 3 shows the monthly average concentration for each month of the year in the case of CO and CO_2. Consequently, the plots in Fig. 3 give the seasonal cycle for the two atmospheric trace gases. The CO_2 monthly mean concentration ranges from 387 ppmv in August to 403 ppmv in March, while that of CO ranges from 100 ppbv in October to 132 ppbv in March. The percentage monthly amplitude for CO and CO_2 is 28.0% and 4.1%, respectively. Note that the percentage monthly amplitude is calculated as the difference between the average monthly maximum and minimum divided by the annual mean. Therefore, the seasonal variability of the CO concentration is more pronounced than that of CO_2. Both the CO and CO_2 concentrations show minima in summer/autumn and maxima in spring. However, the CO concentration exhibits a secondary maximum in August, which can be attributed to the biomass burning from the nearby fields. The atmospheric CO_2 concentration is influenced by vegetation activity through both photosynthesis and respiration processes [18] contributing to seasonal variations.

The seasonal cycles for CH_4 and H_2O concentrations are shown in Fig. 4. It is evident that, CH_4 and H_2O concentrations have a strong seasonal inverse correlation behaviour. CH_4 monthly mean concentrations range from 1871 ppbv in July to 1924 ppbv in March. On the other hand, H_2O monthly average values range from 1.1% in February to 2.5% in August. The percentage annual amplitude for both CH_4 and H_2O concentrations is 2.8% and 83.3%, respectively. H_2O and CH_4 concentrations have a pronounced seasonal variability.

3.3 Diurnal variability

The diurnal variations for CO and CO_2 concentrations are presented in Fig. 5. The CO concentration exhibits a maximum at 08:00 and a secondary maximum at 19:00, and two minima, one at 03:00 and another at 15:00. The CO_2 concentration shows two maxima, one

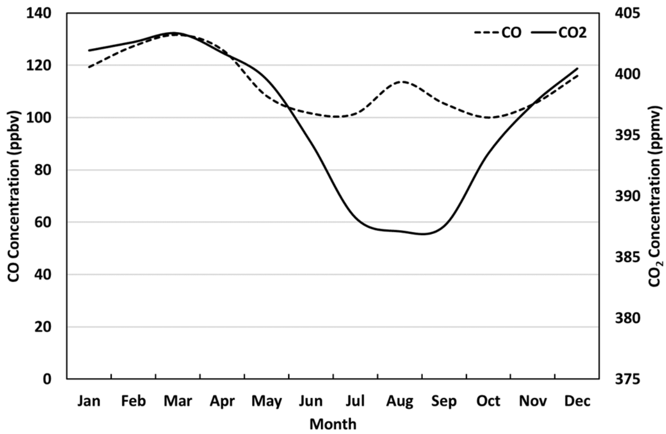

Figure 3: Average monthly (and seasonal) variability in carbon monoxide and carbon dioxide concentrations for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

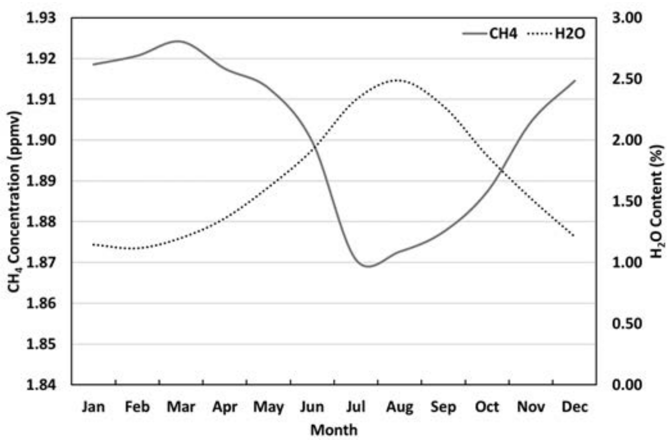

Figure 4: Average monthly (and seasonal) variability in methane concentration and atmospheric water content for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

at 04:00 and another at 20:00, with one minimum at 13:00. Between midnight and 06:00, CO and CO_2 concentrations exhibit inverse correlation. The diurnal variability of the CO_2 concentration can be attributed to several factors including atmospheric boundary layer height dynamics, proximity of the observation/monitoring site to anthropogenic emissions and their variability, the abundance of vegetation, proximity to the sea, altitude of the monitoring site, altitude of the sampling height, season, climate and meteorological variations [19]. During daytime, both CO and CO_2 concentrations correlate reasonably well.

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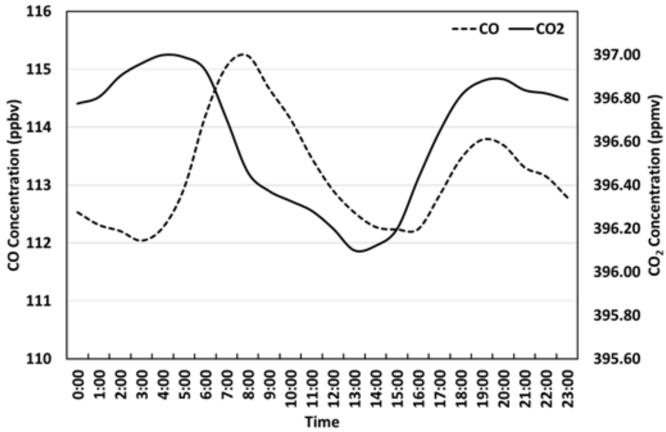

Figure 5: Average diurnal variability in carbon monoxide and carbon dioxide concentrations for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

The maximum and minimum of the CO_2 concentration precede those of the CO concentration. This variability, that appears to follow solar illumination, is probably due to (daytime) photosynthesis.

Fig. 6 demonstrates the diurnal variability for CH_4 and H_2O concentrations. Both compounds show broad nocturnal maxima due to the absence of sunlight.

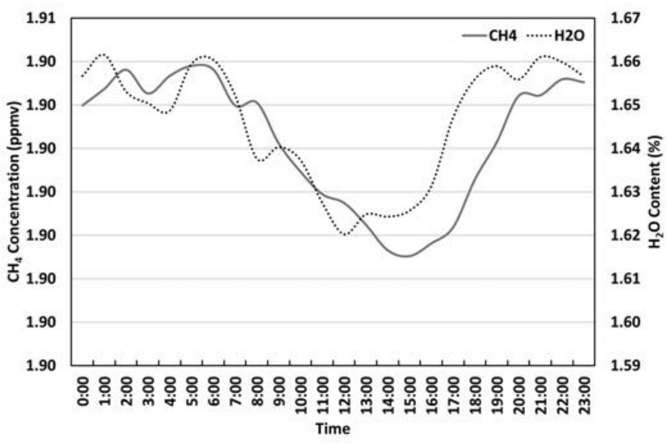

Figure 6: Average diurnal variability in methane concentration and atmospheric water content for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

However, the H_2O concentration reaches a minimum at noon when solar illumination is at its maximum, while the CH_4 concentration attains a minimum at 15:00. The percentage diurnal amplitudes, after normalisation, for CO, CO_2, CH_4 and H_2O concentrations are 2.8%, 0.2%, 0.2% and 2.5%, respectively, for the considered 11-year period.

3.4 Influence of wind direction

The concentrations of the four GHGs that were considered in the study, as a function of wind direction, are presented in Figs 7 and 8. The CO, CO_2 and CH_4 concentrations show maxima when the prevailing wind is from the north sector and secondary broad maxima when the

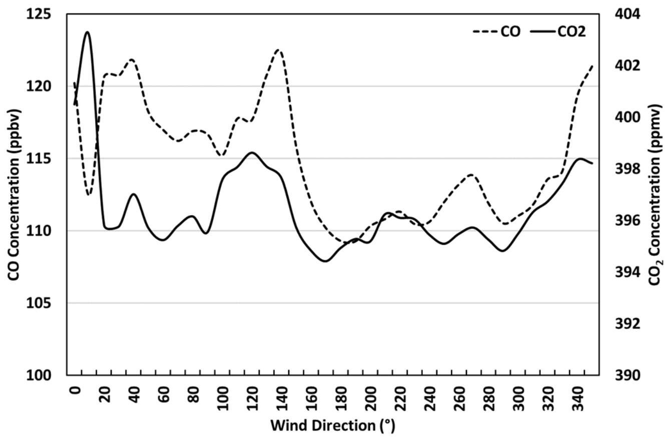

Figure 7: Average carbon monoxide and carbon dioxide concentrations as a function of wind direction for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

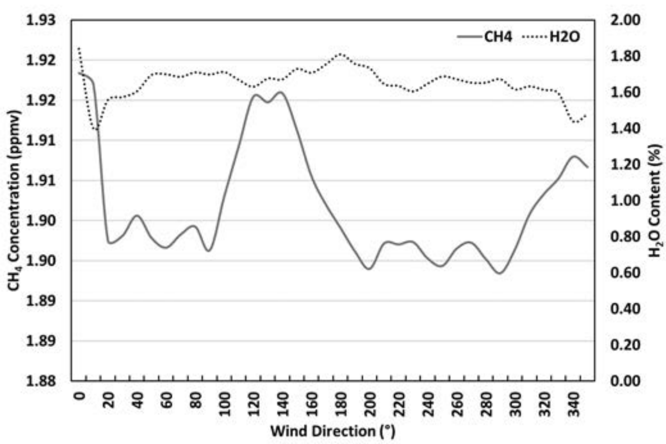

Figure 8: Average methane concentration and atmospheric water content as a function of wind direction for the time period 2012–2023, measured at the monitoring station located at Giordan Lighthouse, Gozo, Malta.

wind is blowing from the south-east sector. A modelling study of CO emissions indicated that countries surrounding the Mediterranean contribute relatively little to CO pollution in the region. In this respect, the strongest influences in the Mediterranean are from western Europe, notably France, Germany and northern Italy [20]. Consequently, the Mediterranean basin is a receptor of CO long-range transport from Europe and mostly originating from burning of fossil fuels. Therefore, the elevated GHG concentrations when the prevailing wind is from the north sector cannot be only attributed to shipping emissions crossing the Strait of Sicily given that the long-range transport of pollutants from mainland Europe is substantial. Conversely, when the wind is blowing from the south-east sector, the CO concentration is more pronounced than that of CO_2, which is a clear indication of the anthropogenic influences originating from the main island of Malta. Meanwhile, the H_2O concentration does not seem to be a function of the prevailing wind as atmospheric water vapour content is largely independent of anthropogenic activities.

4 CONCLUSIONS

Analysis and observations concerning concentrations of GHGs measured over an 11-year period in the central Mediterranean, are presented. The relevant data were collected at Giordan Lighthouse atmospheric monitoring station, located on the island of Gozo, forming part of the Maltese archipelago, in the central Mediterranean. These data consist of the longest records of ground level GHG concentrations for the central Mediterranean, in recent times. Annual, seasonal, diurnal and wind direction variabilities of the said GHG concentrations were investigated. Trend analysis reveals that concentrations of CH_4 and CO_2 are increasing at a relatively substantial rate of 9.6 ppbv per year and 2.15 ppmv per year, respectively. This is comparable with what was reported for the eastern Mediterranean by Gialesakis et al. [5] and is quite similar to what was determined for Mauna Loa (Hawaii). CO_2 and CH_4 are the most important GHGs due to their role in anthropogenic global warming and climate change. Both CO_2 and CH_4 concentrations have a substantial annual mean rate of increase compared that for CO and H_2O concentrations. The latter two are increasing at a minimal rate of 0.32 ppbv per year and 0.02% per year, respectively. The four GHGs exhibit seasonal variability. Monthly averages for CO_2 concentrations vary from 387 ppmv in August to 403 ppmv in March. While monthly mean concentrations of CH_4 fluctuate from 1871 ppbv in July to 1924 ppbv in March. However, the percentage monthly amplitude analysis reveals that both H_2O and CO concentrations have more pronounced seasonal variability compared to concentrations of CO_2 and CH_4. Seasonal variations in CO_2 concentration reveal its dependence on the photosynthesis by plants with maximum CO_2 concentration in autumn and winter, and a minimum in summer. Similarly, concentration of CH_4 attains its maximum in winter and the minimum in summer. As regards seasonal variability CH_4 and H_2O concentrations exhibit an inverse correlation but show a good correlation in the diurnal variability. Both CO and CO_2 concentrations exhibit a diurnal variability. During night time, there is an inverse correlation between CO and CO_2 concentrations. It is worth noting that the CO_2 gas that is trapped in the atmospheric boundary layer achieves higher concentration as the mixing height descends during night time. During daytime, CO and CO_2 concentrations show a good correlation. Both the diurnal maximum and minimum of CO_2 concentration precede that of CO. This phenomenon is due to plant photosynthesis which is high throughout the day due to solar illuminance. Concentrations of CO, CO_2 and CH_4 show a strong correlation with the prevailing wind direction. Concentration maxima are associated with the north sector due to the influence of ship emissions that traverse the Strait of Sicily and the transport of long-range pollution from continental Europe. These three GHGs also show secondary broader maxima when the wind is blowing from the south-east sector, which is evidence of the anthropogenic emissions from the main island of Malta.

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