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A different perspective on air pollution measurements

Hava kirliliği ölçümlerine farklı bir bakış

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A Different Perspective on Air Pollution Measurements

Highlights

- *A mobile equipment is designed and manufactured for measuring air pollution.*
- *SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$ and VOC parameters are measured and analysed.*
- *Monthly average concentrations of pollutants maps are prepared for Karabük city.*
- *O_3 and VOC parameters show a different trend as compared to other pollutants.*
- *The range of $PM_{2.5}$ concentration is between 127.24 μg/m^3 to 295.2 μg/m^3 in Winter.*

Graphical Abstract

The pollutant measurements in Karabük province are taken at fifty points with 8 portable intermittent equipments to obtain hourly, and monthly averages. These measurements are then analysed and mapped to comment on air pollution due to multi-pollution sources in Karabük Province

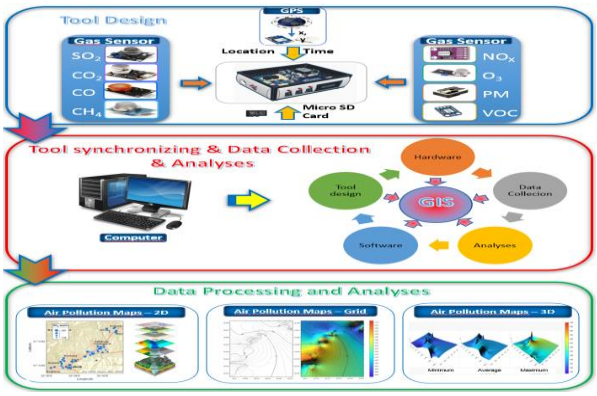

Figure. Graphical Abstract

Aim

This study aims to determine the air pollution of SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$, and VOC parameters in the Karabük province. For this purpose, a new equipment has been designed and used for pollution mapping

Design & Methodology

The data, collected by designed tools, is analysed statistically, and monthly time series are obtained to comment on ambient air quality by many programs including MATLAB for analyses, SURFER for mapping, EXCELL for databases and ARCGIS for mapping.

Originality

The number of stations are not enough to obtain air quality maps in Karabük province. To be able to make them, a portable air pollution measurement equipment is designed and manufactured for collecting air pollution data at any point of the Karabük province.

Findings

The relations with high correlations and R-squared values are good representations of the emissions originating from the same sources. However, throughout the day, the concentration is changing considerably and correlations between parameters are very low between some hours. It means Karabük is under many emission sources' influence.

Conclusion

For isolated measurement points which are far away from the roads, industries and households, the correlated regression relations between parameters are obtained. During the winter months the air pollution increases considerably. However, O_3 and VOC parameters show a different trend as compared to other pollutants.

Declaration of Ethical Standards

The author(s) of this article declare that the materials and methods used in this study do not require ethical committee permission and/or legal-special permission.

A Different Perspective on Air Pollution Measurements

Araştırma Makalesi / Research Article

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ABSTRACT

This study aims to determine the air pollution in Karabük province. For this purpose, a new equipment has been designed. The equipment can measure the SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$, and VOC pollution alongside with many atmospheric parameters. The measurement period has been decided to be one year starting from June 2021. The measurement period was one year, starting from June 2021. The measurements were taken at fifty points with 8 portable intermittent equipment. Then hourly and monthly averages were calculated. The calculation of the averages depends on many statistical analyses. The mean (geometric, harmonic, root, interquartile, Winsorized), median, midrange, Skewness, and Kurtosis analyses were done to obtain correct daily, and monthly averages. These analyses are necessary to comment on the intermittent measurement averages. The analyses of the collected data showed that the concentrations are changing considerably through the measurement period. The highest concentration was observed for the SO_2, CO, NO_X, and $PM_{2.5}$ with respective values of 186.4, 170, 204.9, and 265 µg/m^3. All these values are dangerous for human health. Elevation, temperatures, atmospheric pressure, and wind are sensitive parameters for atmospheric pollution. In Karabük province, most of the measurement points are affected by multi-pollution sources. The scatter diagrams also support this fact. During winter months, the pollution increases instantly. However, O_3 and VOC parameters show different trends as compared to other pollutants. The concentration of these two parameters, namely O_3 and VOC, increases during spring months. The O_3 and VOC increase by 78.1%, and 43.2%, respectively due to photochemical reactions in the atmosphere in spring.

Keywords: A new equipment, air pollution, statistical analyses, kriging, gis mapping.

Hava Kirliliği Ölçümlerine Farklı Bir Bakış

ÖZ

Bu çalışmanın amacı Karabük ilindeki hava kirliliğini tespit etmektir. Bu amaçla tarafımızdan yeni bir hava kirliliği ölçüm cihazı tasarlanmış ve üretilmiştir. Ölçüm cihazı SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2,5}$ ve VOC kirletici parametreleri ile birlikte birçok atmosferik parametreyi de aynı zamanlı olarak ölçebilmektedir. Ölçümler Haziran 2021' den başlayarak bir yıllık süre için yapılmıştır. Ölçüm noktalarının sayısı 50 olarak belirlenmiş ve 8 portatif ekipman ile anlık olarak ölçülmüştür. Bu ölçüm değerleri kullanılarak saatlik ve aylık ortalama değerler hesaplanmıştır. Ortalamanın hesaplanması için birçok istatistiksel analiz yapılmış ve doğru ortalama değer istatistisel analizler ile belirlenmiştir. Ortalama (geometrik, harmonik, kök, çeyrekler arası, Winsorized metodu), medyan, orta aralık, çarpıklık ve basıklık analizleri yapılarak en doğru günlük ve aylık ortalama değerler, ölçümlerdeki uç değerlerin veri setlerinden çıkarılması ile hesaplanmıştır. Bu analizler aralıklı ölçüm değerlerinin ortalamasını bulmak için oldukça önemlidir. Veri analizlerine göre konsantrasyonlar, ölçüm süresi boyunca önemli ölçüde değişmektedir. En yüksek konsantrasyon, sırasıyla 186,4, 170, 204,9 ve 265 µg/m^3 değerleriyle SO_2, CO, NO_X ve $PM_{2,5}$ için gözlenmiştir. Bütün bu değerler standardların üzerindedir ve insan sağlığı için tehlikelidir. Yükseklik, sıcaklıklar, atmosferik basınç ve rüzgar, atmosfer kirliliği için hassas parametrelerdir. Karabük ilinde ölçüm noktalarının çoğu çoklu kirlilik kaynaklarından etkilenmektedir. Dağılım diyagramları da bu gerçeği desteklemektedir. Kış aylarında kirlilik önemli ölçüde artmaktadır. Ancak O_3 ve VOC parametreleri diğer kirleticilere göre farklı bir eğilim göstermektedir. Bu iki parametrenin konsantrasyonu bahar mevsiminde sırasıyla %78,1 ve %43,2 oranında artmaktadır. Atmosferdeki sıcaklık artışına bağlı olarak oluşan fotokimyasal reaksiyonlar sonunda bu parametrelerin konsantrasyonlarının arttığı görülmektedir.

Anahtar Kelimeler: Yeni ölçüm modülü, hava kirliliği, istatistiksel analizler, kriging, cbs haritalama.

1. INTRODUCTION

Fuel consumption for energy production is responsible for 80% of atmospheric pollution [1]. Pollutants released into the atmosphere are accumulated there and in the long term they produce chemical, physical, and biological effects on the environment [2]. These effects are important not only for today but also for future generations. The countries in the world are trying to minimize all these effects through International Agreements, which are ratified by countries to diminish the damages caused by pollutants released into the atmosphere [3]. However, the solution to this problem requires legal monitoring on a global scale and take remedies according to the results. If the air pollution problems are not solved internationally and legally, national studies will not be enough to stop this threat [4].

The measured parameters are, often, limited to SO_2 and PM in many air pollution measurement stations in Turkey. Additionally, CO, NO_2, NO_X, and O_3 are also measured in some special stations. Continuous

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measurement of these parameters is important to find out the temporal variation of the pollutants at one location. This is called "pollution monitoring". Pollution monitoring is a difficult process because of expensive measurement systems, calibration, and maintenance of the equipments [5]. Distribution and residence time of the pollutants in the atmosphere, physical and chemical interaction of these pollutants with other atmospheric components are all important parameters to be considered for air pollution research. Clean atmosphere can be achieved by continuous monitoring of the pollutants and keeping them under control [6]. In order to keep the pollutants under control, emission control systems should be used, and the ambient air quality should also be controlled by making measurement on real time basis.

The number of measured parameters is very important for establishing the air quality measurement network. In most cases, the measurements are not revealing the exact result for the districts, provinces, or regions where pollution measurements are made. It is a common statement that the measurement stations only represent the pollution where they are located [7; 8].

Increasing the number of measurement stations is not a solution for monitoring the ambient air quality in a district. The measurement stations require energy, a budget to keep the station running, maintenance expenses and calibration of the equipments [9]. Besides, it is very costly to run a a station. Instead of an expensive station, a simple, mobile, microcontroller, and multi-purpose measuring devices can be preferred for the measurement of many pollutants at the same time as a supplementary to support stationary measurement stations [10]. It is cheaper and increases the correctness of any local study [11]. There are some examples of this type of tool. For example, Cao T. and Thompson, J.E. "developed a portable device that measures PM parameter on time dependent basis. The instrument is programmed via an Arduino microcontroller and a PM sensor [12]. Also, CO_2, CO, and NO_2 parameters were measured with a microcontroller. According to the measured concentrations of these parameters, the traffic is tried to be controlled by GIS techniques. The microcontroller is again an Arduino based instrument [13]. These type of equipments can also be used in mining processes. The measurement of some pollutant parameters including temperature and humidity are used for the prediction of mine air quality, and the results are used to alert the miners in case of emergency conditions [14]. The validation of the sensors is one of the important issues for these types of tools. The sensor sensitivity is also very important. There could be some errors due to environmental factors. The device is a low-cost equipment, and some errors can be present as compared to the advanced measurement equipments. However, the errors are acceptable due to limitations of lower than 10 $\mu g/m^3$ [15].

Aim of this study and the steps taken to conduct the study: The main aim of this study is to produce pollution maps for a region that cannot be obtained by regular air pollution measurement station data. In this study, the atmospheric pollution maps are produced with ArcView Programs. To obtain these maps, the study is divided into 5 steps (Figure 1): The first step which is "Designing measurement tools" is the starting point of this study. An Arduino-based microcontroller processor for measuring SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$, and VOC is designed and produced. It measures not only pollutant parameters but also meteorological parameters including temperature, humidity, and pressure. In the production of the devices, a cheap, open-source Arduino microcontroller card is preferred. It has a wide usage area due to its software, and other hardware additions are possible. The sensor, module, and other parts in the devices have been selected so that they will be compatible with the Arduino microcontroller. Arduino microcontroller needs Arduino software to operate the sensors and modules connected to it, and also to exchange data with sensors. The necessary codes and programs were compiled by us and uploaded to the Arduino microcontroller with a USB cable in the computer environment. Eight measurement tools are produced in our laboratory one is stationary, and the others are mobile equipment. Mobile equipment measures the pollutant concentrations at the pre-determined points.

The second step in the study is the preparation of the "Air Pollution Database" to process the collected data. This step is divided into data and analysis parts. Data is collected from the measurement points according to coordinates, calendar, and clock. Analyses parts contain figures and statistical analyses. The main aim of this section is to determine the corrected daily and monthly averages.

The third step in the study is "Collecting pollution data" which is the most difficult part of this study. The measurements for 50 points lasted for one year. The measurements are discontinuous except for one point. To take the averages of each station, at least 21 hrs measurements on different days are made. Each measurement in any station lasted for one hr.

The fourth step in the study is "Comparison of measurements with the data of the local measurement station". The data obtained from the study is not the same as official data. According to the comparison of SO_2, CO, NO_X, O_3, and $PM_{2.5}$ concentrations as the official data, monthly trends are found to be very similar. These five parameters are not measured at all stations in Karabük provinces. Therefore, CO, NO_X, and O_3 values obtained at the local governmental stations are not reflecting the Karabük province.

The fifth step in the study is "Preparing maps" and it is the last step, and the measured data are used after obtaining monthly averages for the measurement points. The number of measurement points is tried to be increased to obtain the quality of the maps. GIS queries are used in order to prepare the maps [16].

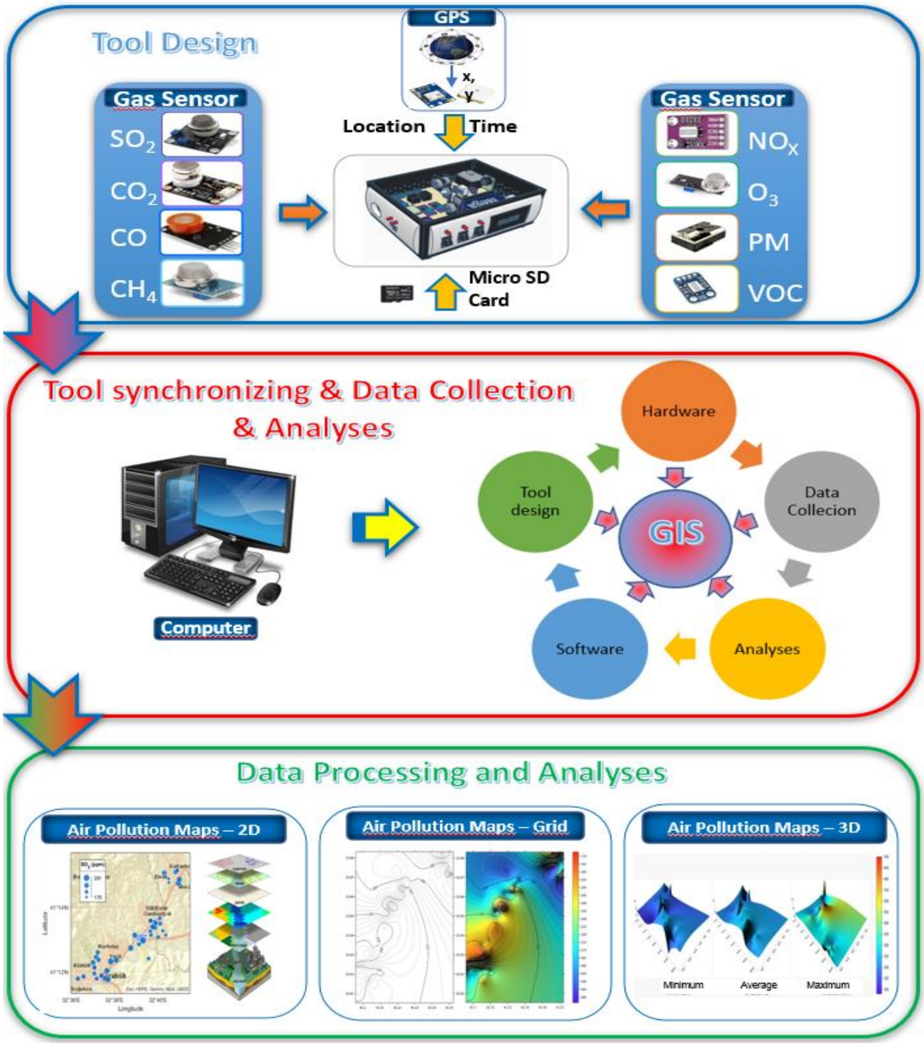


Figure 1. Methodological approaches of the study

2. MATERIAL AND METHOD

The coverage area of this study is the Karabük Province. However, this methodology can be applied to any place where air pollution is a big problem. The study concentrates on SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$, and VOC parameters. If the emission sources are not multi-sourced, then the correlation relations between emission parameters are also determined. The data is analysed statistically, monthly time series are obtained to comment on ambient air quality. Also, GIS techniques and many programs including MATLAB, SURFER, EXCELL and ARCGIS were used.

2.1. Tool Design and Data Collection

2.1.1. Tool Design

The tools are Arduino based, and they can measure 8 parameters including SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$ and VOC. PM sensor is an optical sensor. Besides these parameters, the air temperature, pressure, and humidity are also measured. Every 10 seconds, it records each parameter on the SD card. During the transportation, the coordinates are also recorded by GPRS connection. The coordinates are very important for producing maps. The measurement results are also sent to a touch control screen on the equipment. The instantaneous measurements can be seen on the screen. The units of ppm, µg/m^3, or mg/m^3 can be selected for the concentration. The sensors are very sensitive even to small concentrations of parameters. The basic programming language for the control Arduino card is C++. The air flows through a propeller passing over the sensors, and then air exits out through a second propeller. The speed is automated due to the wind speed in the atmosphere.

The isokinetic structure is very important for trustable pollution measurements. The electronic design and structural establishments of the tool are considered carefully not to cause any trouble with the measurements. The cable and other electronical components are hidden in the air-flowing duct. The measurement tools can be charged and reused for approximately four hours after charging. The stability of the equipment is also important. Each piece of equipment is calibrated for correct measurements. The measurements are made on different points at the same time after the necessary adjustments of the equipment are done.

2.1.2. Databases

The databases are formed due to the daily collection of data. The databases have been separated into two groups. The first group is the data parts including hour (in the hour: minutes: second units), day, month, year, coordinates, parameters (original units of sensors – ppm or $\mu g/m^3$), humidity, atmospheric temperature and pressure. In this part, only the original collected data is kept. The measurement points in the Karabük province are separated according to the coordinates. During the study, the measurement points are not changed. The 24-hours measurements have been settled for one point every month. Safe measurements are the main consideration for the selection of these measurement points.

2.2. Analyses

The second group is the analyses containing: regression analysis, time series analyses, and statistical analyses. In this section, an automated graphical analysis besides statistical analyses including regression analyses between parameters, and parametric mean (geometric, harmonic, root square, trim, interquartile, midrange, Winsorized, tri), variance, standard deviation, interquartile range, range, mean difference, deviation, Skewness, and Kurtosis are studied. With these analyses, the uncertainty level of the collected data is considerably decreased, and the error is minimized.

2.2.1. Statistical Methods

Statistical analyses are used to obtain corrected averages. They are very helpful to determine the daily and monthly averages of the measurements. There is no strict rule to obtain the corrected data. But statistical analyses are always the best way to determine it. Figure 2 summarizes the general statistical approaches.

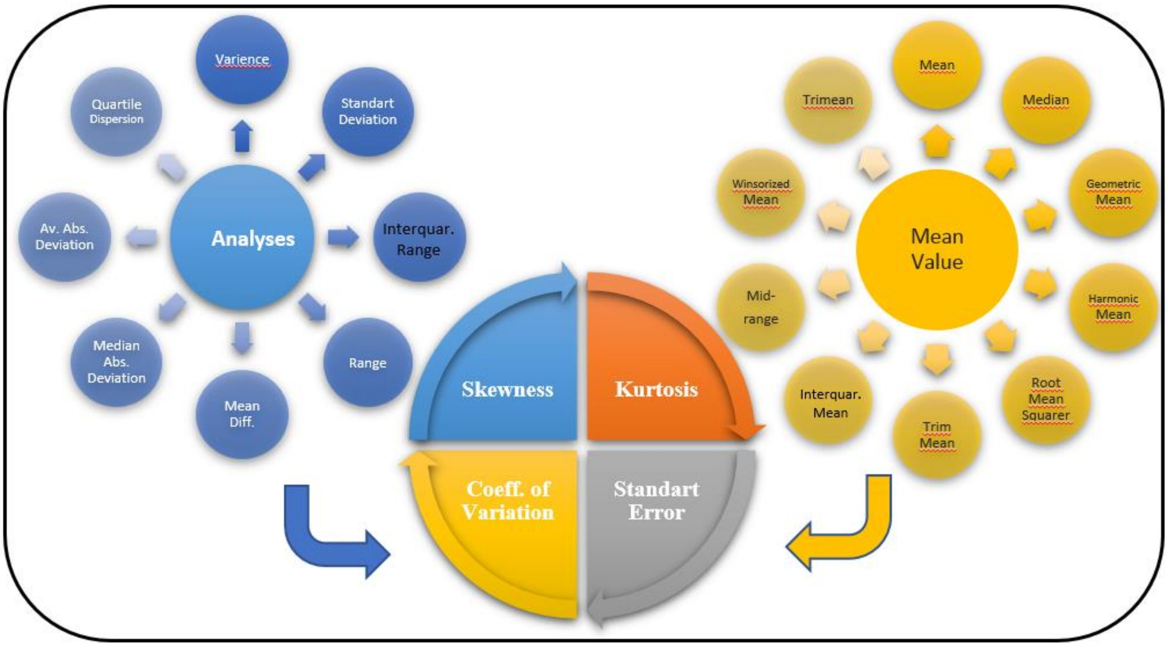


Figure 2. Statistical analyses

The mean values of the data sets were calculated by using several approaches to determine the most accurate results [17]. The mean values of the series are calculated by the median, geometric mean, harmonic mean, midrange, and root mean square. To reduce the effect of outlier values, trim mean, and interquartile mean were also studied. In order to support the results and to comment on the averages which are less affected by outliers, Winsorized mean and TriMean analyses are also applied to the dataset. Many researchers are using these methods to analyse datasets statistically [18; 19; 20].

The calculation of the mean value has a little meaning. The dispersion of the values according to the mean, and averages are also important to comment on the data set certainty. The mean value with the variance and the standard deviation are giving the general characteristic of the data set. The calculations for mean difference, median absolute deviation, average absolute deviation, quartile dispersion, and relative mean difference are also important to comment on the distribution of the data sets. The expectation is always to obtain a normal distribution data set [21; 22; 23]. The calculated mean values are checked for reliability using standard error and coefficient of variation. Variance and standard deviation are insufficient to explain the series distribution and symmetry. Coefficients of skewness and kurtosis are used to support the distribution character of the data set [24].

Arithmetic mean and median show the center of the dataset. They are important statistical parameters to comment on the pollution parameters. They are used to compare the results with EPA air quality standards [25], and National Air Quality Standards [26]. Moreover, geometric mean to investigate the existence of variation in measurement results, harmonic mean to calculate the average of ratios, and midrange to determine the fluctuation from the median are also necessary to evaluate and comment on intermittent measurements. The spread between numbers of the data set, and the spread of the data to the mean are determined by variance and standard deviation.

Interquartile range, mean difference, median absolute deviation, average absolute deviation, quartile dispersion, relative mean difference, standard error, and coefficient of variation are also calculated to assist the evaluation of the pollution parameters. The statistical methods are a very useful tool to identify the best central values of air pollution measurements rather than continuous measurements. To support the standart statistical measurements, Root Mean Square, Trim Mean, Interquartile Mean, Winsorized Mean, Skewness, and Kurtosis are also used.

Root Mean Square is a statistical criterion for determining the size of changes in positive and negative fluctuations (such as -5 to +5). It is obtained by Eq. (1):

$$X_{RMS} = \sqrt{\frac{X_1^2 + X_2^2 + \cdots + X_n^2}{n}} \tag{1}$$

Trim Mean is useful when there are too many outliers or when the distribution is extremely skewed. It is calculated by using Eq. (2). The average value without considering a specified percentage of the largest, and smallest values in the data set is determined (such as 10%, 20%):

$$\overline{X}_{TM} = \frac{\sum_{k=p+1}^{n-p} X_k}{n - 2p} \tag{2}$$

Interquartile Mean is calculated by averaging the middle 50% of the data in the data set. It is calculated with Eq. (3). It is very useful for commenting on the outliers with the other statistical calculations.

$$\overline{X}_{IQM} = \frac{2}{n} \sum_{k=\frac{n}{4}+1}^{\frac{3}{4}n} X_k \tag{3}$$

Winsorized mean is calculated by averaging the smallest and largest values in the data set after replacing them with the closest values Eq. (4).

$$\overline{X}_{Win} = \frac{X_n \ldots X_{n+1} + X_{n+2} \ldots X_n}{n} \tag{4}$$

Trimean represents the general trend of the dataset. It is defined as the median and the weighted average of the upper and lower quartiles. It is calculated by Eq. (5):

$$\overline{X}_{Tri} = \frac{1}{2}(Q_2 + \frac{Q_1 + Q_3}{2}) \tag{5}$$

Skewness refers to a distortion or asymmetry in a data set that deviates from the normal distribution. It gives information about the direction of outliers. A positive value indicates that many outliers are located on the right side of the distribution. If it is negative, then the outliers are located on the left side of the distribution (Eq. (6)):

$$SK = \frac{\sum_k^n (x - \overline{x})^3}{(n-1) * s^3} \tag{6}$$

The Kurtosis coefficient is zero in the normal distribution. The positive kurtosis coefficient indicates a pointed distribution, and the negative kurtosis coefficient indicates a flattened distribution. If the distribution is like the normal distribution, it takes a value in the range of (-1, +1) (Eq. (7)):

$$KU = \frac{\sum_k^n (X_k - \overline{X})^4}{\sum_k^n (X - \overline{X})^2} \tag{7}$$

2.2.2. Functional Analyses

A scatterplot matrix is a useful way to investigate the relationship between variables [27; 28]. To make a two-dimensional scatterplot matrix, each variable must be inserted both horizontally, and vertically [29]. In this study, scatterplot matrixes for pollutant parameters were studied, and the relationship between the parameters has been investigated. If there is no relation between the parameters, the emission sources will be multi-sources, and the area survey must be done to comment on the effect of each pollutant source. In the Karabük province, the emission sources are limited to industrial, households (residential), and road transportation. The agricultural effects can be observed, but their effect is not large.

Functional relationships between the pollutants (SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$ and VOC) are determined by using linear, polynomial (2^{nd}, and 3^{rd} order) and logarithmic regression models [30; 31].

2.2.3. Uncertainty Analyses

The uncertainty percentage is calculated by using the equations Eqs. 8 and 9. The (sample size -1) degrees of freedom value is obtained by the 95% of confidence interval on student t-table [32].

$$\text{Std. Error of Mean (SEM)} = \frac{\text{Standard Deviation}}{\sqrt{\text{Sample size}}} \quad (8)$$

$$\text{Uncertainty (\%)} = \frac{2 * \text{SEM} * t_{0.05,df}}{\bar{X}} * 100 \quad (9)$$

2.3 Data Processing

2.3.1 Kriging Interpolation Method

The most basic problem in air pollution studies is the limitation of the number of measurement stations. When a pollution map is tried to be produced, it is not possible to obtain an accurate map due to this limitation. The obtained data which are gathered from enough measurement points are necessary to figure the pollution on a map. The interpolation between the measured points gives the results of the points which are not measured. The method of this interpolation is Kriging [33; 34]. Kriging is a term used in geostatistics to describe estimation or interpolation. It is the process of finding the optimum values of unknown spatial data at any point by using data from points close to this location [35]. The most important feature that sets this method apart from others is that the weights are calculated with the least amount of estimation error possible [36]. The Kriging interpolation method is commonly used in air pollution mapping studies [37]. The general equation is given in Eq. (10):

$$N_P = \sum_{k=1}^{n} P_k * N_k \quad (10)$$

2.3.2. Geographic Information Systems

Geographical information systems (GIS) are a digital platform for storing, viewing, analysing, and creating maps using data associated with their locations. It enables any work on the planet to be performed in a digital environment [38]. The relationships between the data are determined, and evaluation of the data is provided due to the maps produced, which utilize spatial data. In recent years, GIS has become increasingly common for estimating air pollution [39].

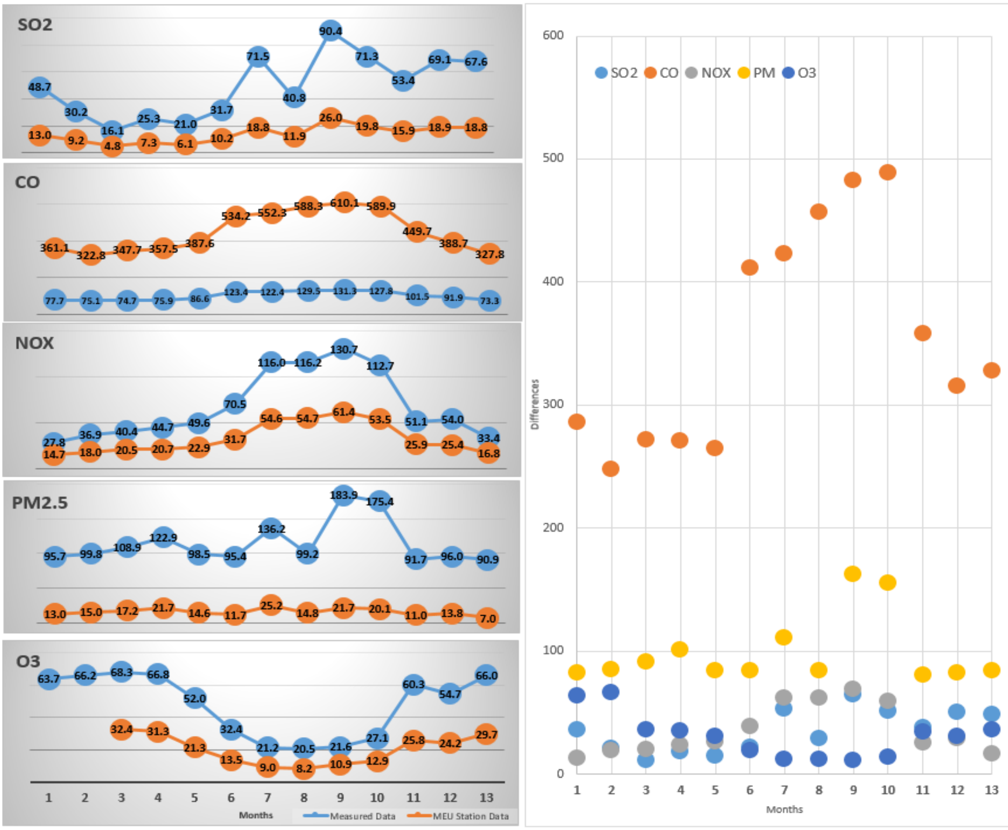
The maps are prepared by using collected data. As the number of measurement points increases, the less uncertain maps are prepared. For that reason, each month the number of measurement points is tried to be increased. The maps are prepared by using GIS, SURFER, and MATLAB programmes. The coordinate range is approximately 600 km^2 area. The data is processed by kriging methodology, and the map is prepared by Surfer, and GIS programs. It is also possible to compare how big the pollution problem is in the measurement points using MATLAB programs.

3. RESULTS AND DISCUSSION

The main aim of this study is to produce pollution maps for the parameters SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$, and VOC. The measurements have been done for one year period. Monthly measurements have been increased after producing measurement tools number. In the beginning, the pollution measurements were done only for 3 points, and the necessary data couldn't be collected for producing maps. The period of this study extended from 2021 June to 2022 June. The number of pollution measurement points during this time increased by more than 50 points per month. The pollution maps are produced by using these measurements.

3.1. General

The steps of studies are categorized as designing measurement tools, preparing pollution databases, collecting pollution data, comparison of measurements with local measurement station data, and preparing maps. After setting the database, the air pollution measurement station of the Ministry of Environment, Urbanization and Climate Change (MEU) data is also collected from the webpage of MEU. The collected data is first checked with the station data. This check is very important for commenting on data harmonization. Some errors have been found in the station data, and these errors are due to the measurement elevation of pollution parameters. The data of MEU is not at the ground level. The data is collected 10 m above the ground. Therefore, there will be some errors between these two measurements. In the Karabük Province, there are 5 measurement stations, one is in the Safranbolu district, and 4 stations are in the Karabük province. The parameters are limited to SO_2, PM, CO, O_3 and NO_X. With the data obtained in these stations, the pollution maps cannot be prepared.



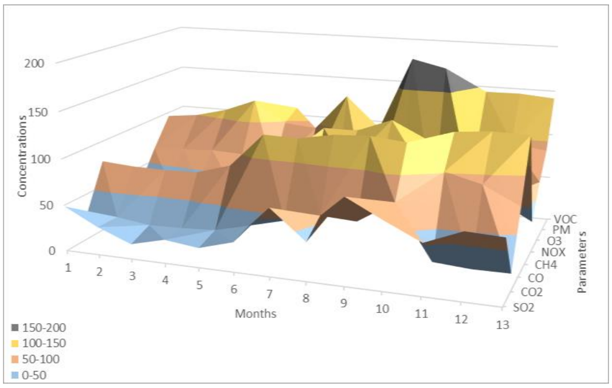
Units: (μg/m^3)

Figure 3. Comparison of measured data and the scatter diagram

When the data are compared, the largest differences are observed for CO (Figure 3). As can be seen in the figure the difference reaches 488 μg/m^3 in October. The second largest difference is seen in the O_3 parameter. The other parameters SO_2, NO_X, and $PM_{2.5}$ are in the range of 67.8-73.7%, 47.1-55.1%, and 81.5-92.3%, respectively. These differences are expected due to the difference of the elevation of sampling points. The MEU measurements are done at 10 m above the ground. The air is taken into the measurement tool by a pipe flow. The second important aspect is the monthly data can only be compared due to the confidence of instantaneous measurements at the stations. The instantaneous data measurements are not shared by the ministry because they are confidential. Therefore, the small instant uncertainties are increased through comparisons of monthly averages. These comparisons cannot be accepted owing to the gathered errors of instant data. Another important thing is the measured data in this study is not continuous due to the charging problem. During the charging of the tools, the measurements are stopped, and these conditions also create an important error. Under these circumstances, these types of differences can be expected, and the quality of the measured data can be increased by the comparison of produced equipment measurements rather than the comparisons of the MEU air pollution measurements at the stations. Although there are big differences in some pollution parameters, the trend of air pollution parameters in both are certainly very similar to each other.

3.2. Discussion

The measured data analyses are the main considerations for this study. As one can observe in Figure 4, the monthly averages of the data for the Karabük provinces are changing considerably throughout the study period.



Unit: (μg/m^3); VOC unit is ppm

Figure 4. Monthly averages of the pollution parameters in the Karabük Province

The highest concentration was observed in the $PM_{2.5}$, VOC, and CH_4 with respective values of 184 μg/m^3, 145 ppm, and 136 μg/m^3. The highest changes were observed in NO_X, $PM_{2.5}$, and CH_4. The respective changes between minimum, and maximum values are 102.9, 93.0, and 85.6 µg/m^3, respectively. Besides, the concentration of SO_2, NO_X and CO are also very close to the critical values. The daily consideration of the parameters could be very important according to the air quality criteria.

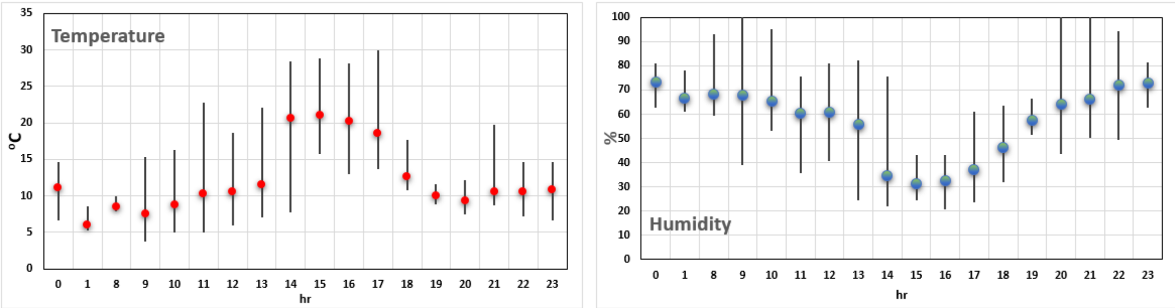
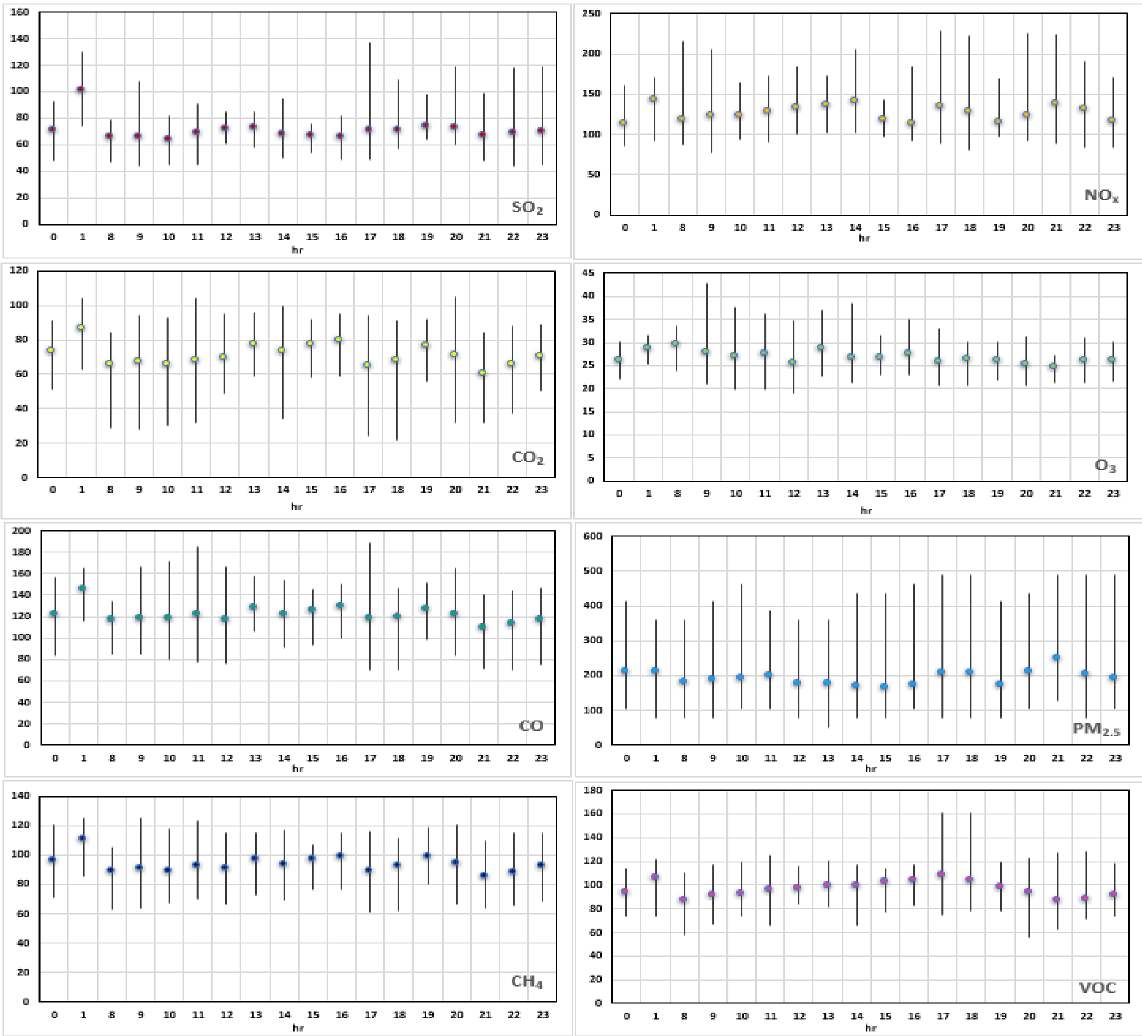


Figure 5. Daily temperature (°C), and humidity (%) changes in Karabük Province

The temperature and humidity of the atmosphere are also important parameters for air pollution. In a day, the temperature is changing from 4 °C to 30 °C, and the humidity is changing from 20% to 99%. These changes are also affecting pollution throughout the days (Figure 5). The pollution parameters show different trends in a day (Figure 6).



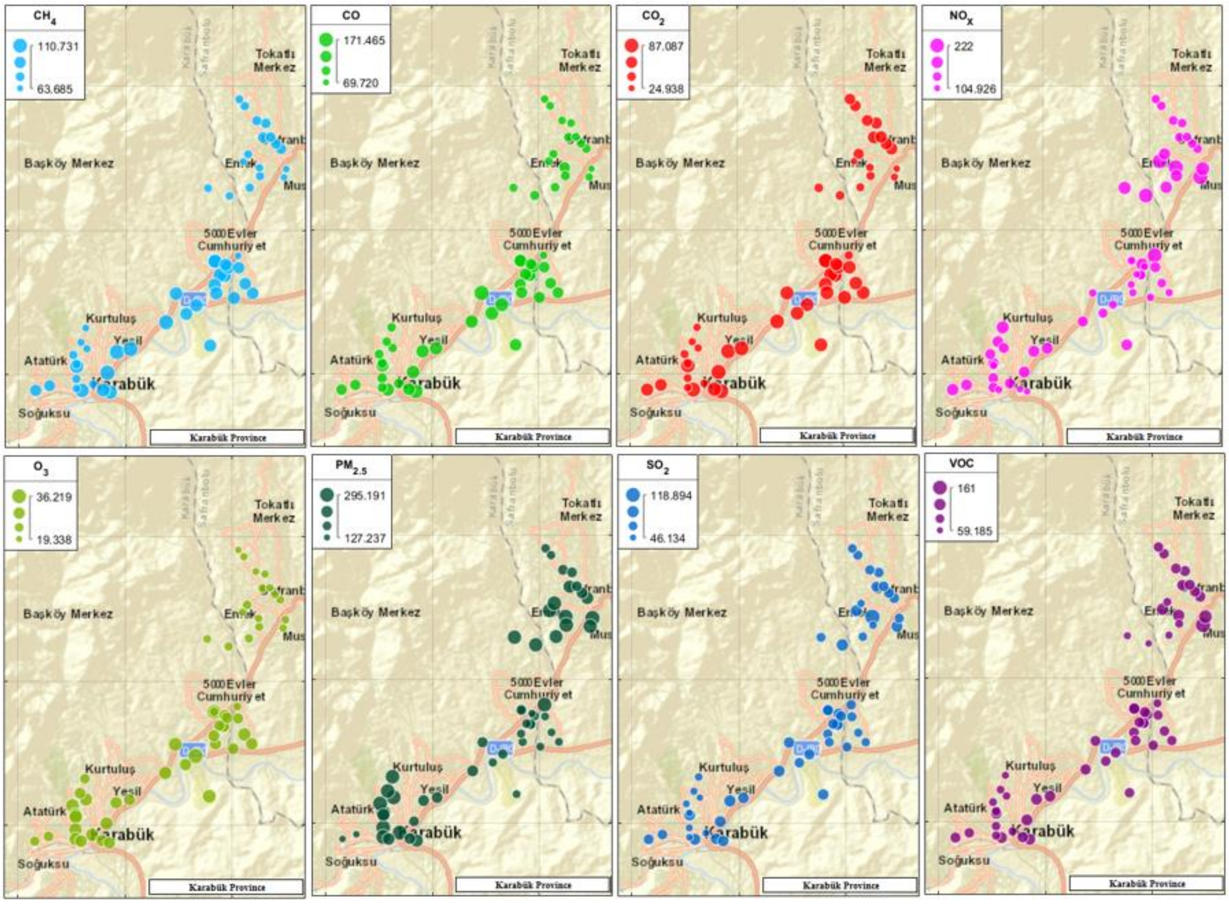
Unit: (µg/m^3); VOC unit is ppm

Figure 6. Daily and hourly concentrations of pollution parameters

The concentrations of SO_2, VOC, and CO_2 are very low in the morning hours. However, concentrations of NO_X, VOC and CH_4 are very high in the afternoon. The NO_X and O_3 trends are opposite as compared to each other. The daily concentrations of pollution parameters are fluctuating throughout the entire day. The main reasons for this fluctuation are not only atmospheric conditions but also emission sources.

The hourly measurements are showing the general characteristics of the parameters. Throughout the day, the concentration is also changing considerably. The minimum concentrations of parameters are determined usually around 1 o'clock. Through hr 1, and 8, the concentrations of parameters are not changing too much. For that reason, these measurements were eliminated in Figure 6. According to figure 6, the lowest measurement values of SO_2, CO_2, CO, CH_4, NO_X, O_3, and $PM_{2.5}$ parameters through the night are 43.6, 22.0, 69.7, 61.0, 77.5, 18.8, and 51.6 µg/m^3, and that of VOC is 55.0 ppm. On the other side, the highest measurement values of SO_2, CO_2, CO, CH_4, NO_X, O_3, and $PM_{2.5}$ parameters are 137.1, 105.0, 189.5, 125.0, 228.5, 42.8, and 489.8 µg/m^3, and that of VOC is 161 ppm. VOC is given in unit ppm for comparison purposes with other pollution parameters.

The average pollution values of each parameter are given in Figure 7. The measurements are done in around 50 points in Karabük Province. Each pollutant is figured due to the values of measurement points. The circle radius is scaled due to the pollutants' quantity. As the pollutant is increasing, the radius of the circle is also increasing.

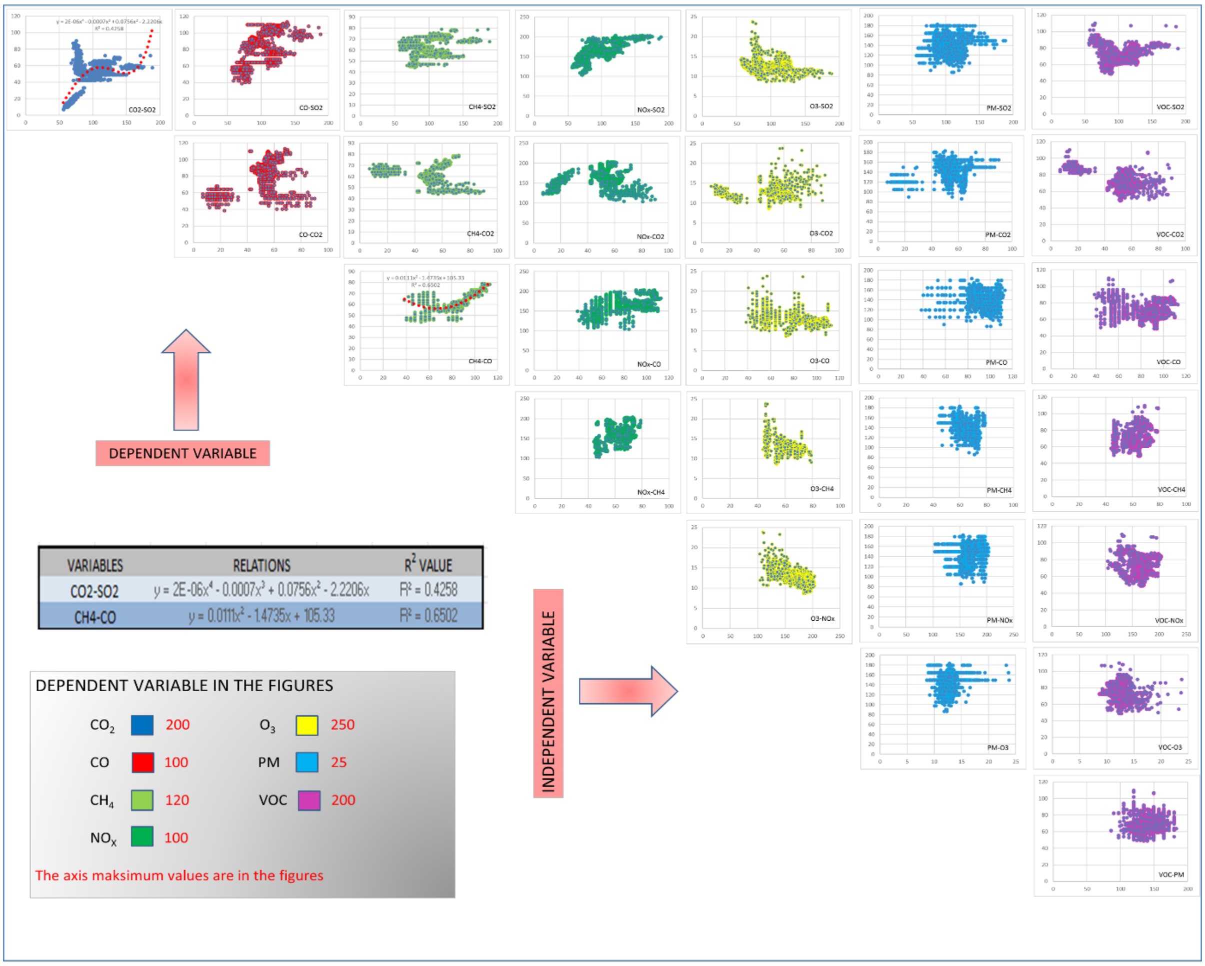


Unit: (µg/m3); VOC unit is ppm

Figure 7. Average pollution values of measurement points in the Karabük Province

The concentrations of all parameters are in a range changing from minimum to highest value. For example, the CH_4 concentration is changing between 63.66 µg/m^3 to 110.73 µg/m^3. The highest range is occurring in the PM concentrations with a minimum value of 127.24 µg/m3 to a maximum value of 295.2 µg/m^3. The smallest range is determined for the O_3 concentrations. The range is only between 19.3 µg/m^3 to 36.2 µg/m^3. The relations between the pollution parameters are one of the important indicators to comment on the pollution sources. If the pollution originates from more than one source, then the relations will not be strict, and the figures will not show exact relations between pollutants. When Figure 8 is observed, it can be easily concluded that the emissions are not originated from one source.

The highest relations are observed with a 0.65 R-squared value between CH_4, and very low CO. In the Karabük province, the industry is the main emission source. The annual percentage of industrial emissions is changing between 50% and 60%. The industrial emission sources are followed by household fuel combustion emissions with values changing between 25% and 35%, and it is followed by road transportation with values in the range of a minimum of 10% to a maximum of 25%. The regression analyses between pollutants with high correlations are also determined in this study. The relations with high correlations and R-squared values are good representations of the emissions originating from the same sources (Figure 9). Another important conclusion is that, if the points are very close to big industries, as seen in Figure 9, there was a higher correlation between parameters. The trend can be linear, 2nd order, 3rd order, or logarithmic. The highest R-squared value determines these relations. Deciding on emission sources is the basic step to control them However, some figures show no relations. It means the pollution parameter analyses on the measurement points are so uncertain that this place is surveyed for the emission sources. For that reason, the points in this study are selected on the roads, close to the residential areas or inside the industries. To reflect the real situation, from the city center of the Karabük province to Safranbolu district, 50 points are determined and studied, and a general conclusion about the emission sources can be reached according to the results shown in Figure 8.

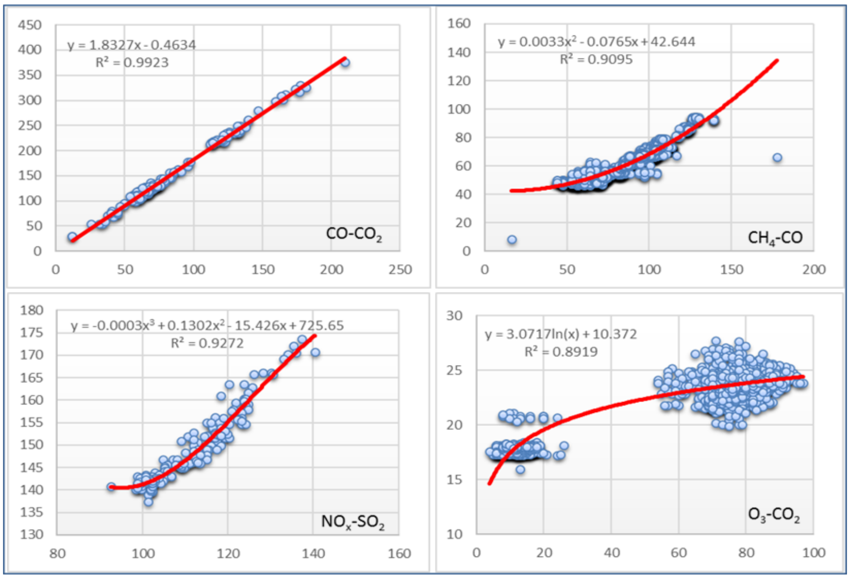


Units: (µg/m^3); VOC unit is ppm

Figure 8. Relations between the pollution parameters (Q-Q Plot)

Figures 8 and 9 show the relations where the parameters are correlated, and if the high R-squared values are obtained, the emission sources of parameters will be the same. The measurement points are affected by many sources such as households, transportation, and industry.

The situation is different in the Safranbolu district. This point is far from the road and industrial plants. The parameter relations are very high as compared to the other points. For this point, depending on the atmospheric conditions, the concentrations can be higher or smaller. It is certain that when the wind speed is high the concentration is decreasing, and the relations between the pollution parameters are not observed as seen in Figure 9. The higher correlated concentration values are observed under stable atmospheric conditions.



Units: (µg/m^3)

Figure 9. Highly correlated concentrations on "point 48" in the Safranbolu district

The 0.9923 R-squared values for CO-CO_2 figure show a big correlation between these two parameters. The relation is linear. CO and CO_2 pollution originate from the same source. The second-order relation between CH_4-CO is also stronger with a value of 0.91 R^2. It represents that the combustion is inefficient, and the result is a high rate of CH_4 and CO emission. This type of burning occurs in solid fuels such as lignite, coke, asphaltite, and other types of coals. The NO_X and SO_2 relations with 0.9272 R^2 value also show a high-rate combustion of solid fuels. The O_3 and CO_2 have some probabilities due to the 0.8919 R^2 value. The high concentration of CO_2 in the atmosphere has a great effect on ground O_3 level. The relations are logarithmic.

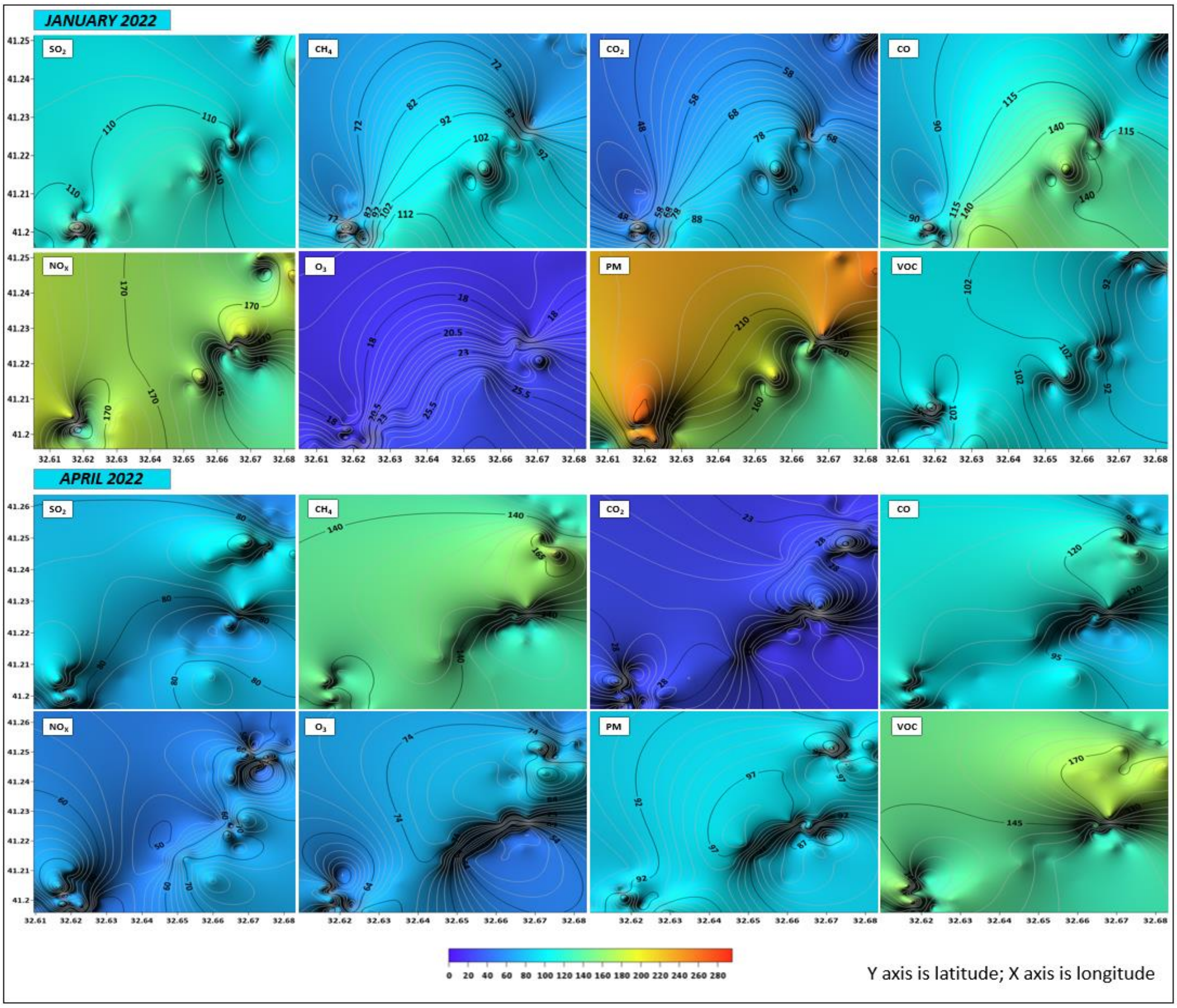
The daily and monthly averages of the pollution parameters are given in Table 1. The data and the calculated averages are analysed by applying statistical analyses. First, the range is considered, then the obtained mean and the averages show the general characteristics of the measurements. If the calculated values contain some outliers, then the values out of the standard deviations according to the skewness are deleted to obtain a normal distribution.

Table 1. The analyses of the pollutant concentrations

	Average Monthly Pollutant Values							
	SO_2	CO_2	CO	CH_4	NO_x	O_3	$PM_{2.5}$	VOC
Minimum Value	86.08	38.08	69.86	62.18	97.29	15.78	112.59	83.14
Maximum Value	186.4	92.67	170	117.41	204.9	28.1	265.02	119.06
Mean	108.16	62.52	114.7	83.45	165.11	20.49	202.42	98.4
Median	107.17	59.98	110.03	80.02	168.02	18.73	219.74	100.96
Geometric Mean	107.78	61.04	111.97	82.06	164.4	20.13	198.37	98.22
Harmonic Mean	107.41	59.59	109.32	80.74	163.66	19.8	193.91	98.04
Root Mean Square	108.54	64	117.41	84.86	165.77	20.86	206.01	98.57
Trim Mean (10%)	108	62.19	114.09	82.84	165.81	20.32	203.66	98.5
Interquartile Mean	107.3	61.53	112.88	81.42	167.77	19.46	212.64	99.76
Midrange	136.24	65.38	119.93	89.79	151.1	21.94	188.81	101.1
Winsorized Mean	108.13	62.24	114.19	83.03	165.41	20.45	202.2	98.27
TriMean	107.58	61.36	112.56	81.5	167.49	19.71	210.4	99.48
Variance	82.87	186.99	629.28	237.93	219.15	15.53	1467.76	33.89
Standard Deviation	9.1	13.67	25.09	15.42	14.8	3.94	38.31	5.82
Interquartile Range	12.33	24.58	45.1	28.22	16.43	7.23	63.4	8.35
Range	100.33	54.59	100.14	55.23	107.61	12.32	152.43	35.92
Mean Difference	10.23	15.67	28.75	17.51	16.23	4.34	42.25	6.35
Median Abs. Deviation	5.85	12.06	22.13	13.11	7.96	2.02	18.61	2.24
Average Abs. Deviation	7.26	11.94	21.9	13.53	11.17	3.31	31.4	4.57
Quartile Dispersion	0.06	0.2	0.2	0.17	0.05	0.17	0.16	0.04
Relative Mean Diff.	0.09	0.25	0.25	0.21	0.1	0.21	0.21	0.06
Standard Error	0.11	0.16	0.3	0.18	0.18	0.05	0.45	0.07
Coef. of Variation	0.08	0.22	0.22	0.18	0.09	0.19	0.19	0.06
Skewness	0.54	0.26	0.26	0.41	-0.83	0.62	-0.66	-0.49
Kurtosis	4.13	1.84	1.84	1.83	3.21	1.83	2.02	2.71
SEM Range	2.20-3.22	0.74-2.73	2.43-5.0	2.14-3.07	1.57-4.08	0.60-1.99	0.73-8. 25	1.40-2.62
Uncert. Range (%)	8.25-11.57	12.0-16.86	9.86-16.86	6.19-14.19	8.96-10.39	11.41-12.28	3.18-17.05	5.78-7.31

Unit: (µg/m^3); VOC unit is ppm

In Table 1, such as SO_2, and the VOC values do not show a normal distribution due to kurtosis values, which are not between -1, and +1. Therefore, outliers are present in the measurements. The mean (geometric, harmonic, root, interquartile, Winsorized), median, midrange, and averages also support the Skewness, and Kurtosis values. These values show the sides of the outliers. The difference between interquartile, and mean values show how the averages are affected by the outliers. The determined outliers are omitted for the calculation of daily and monthly data. The uncertainty values of the CO_2 and CO are very high as compared to the other parameters with a value of 16.9%. This quantity is acceptable due to IPCC Good Practice Guidance [40]. The maps are produced by GIS techniques, the pollution data are listed according to the coordinates on the map, and then by using the Kriging method, the data are processed on the map of the province. The contours are obtained by using SURFER programs. The winter and the spring maps are given in Figure 10. The pollution is very high in the winter season, especially for PM parameters. It is the effect of households burning fossil fuels for heating purposes. In the winter season, coal production is very high in Karabük. Although natural gas is used for heating purposes, the number of households using coal is not low. In 2022, the cost of natural gas is also effective for the preference of the stoves used in homes. In spring season, the pollution is lower as compared to the winter season as expected.



Unit: (µg/m^3); VOC unit is ppm

Figure 10. Pollution maps obtained by GIS techniques and Kriging methods

In the maps, some points are very noticeable which are seen as the pointed hill. These points are exposed to pollution from highly dense transport roads, and combustion of apartments or houses. Due to the continuous emissions, these points are always measured higher in pollutant concentration. According to Figure 10, all parameters except CH_4, O_3 and VOC measured in January are higher in concentration than that measured in April. The decrease in concentration of these parameters is due to decrease on road transportation afterdense snowy days in January 2022. This result can be expected generally. However, the differences between the parameters are not very large when the maps are considered. The maximum differences are determined for $PM_{2.5}$ and NO_X parameters with the respective value of 102.94, and 98.74 µg/m^3. It can be concluded that the emission from the households in the winter season changes the pollutant concentrations by not more than 50%. The industries are very effective in the Karabük Province due to continuous emissions. The calculations are done by using average values. The detailed figures are also given in Appendix A.

The topography of the region is also very effective for concentration distribution. Higher concentrations are generally measured at higher elevations. Regions close to the industries, high-traffic roads, and centers of the province are determined to be highly contaminated with pollution. According to average values after omitting outliers, the higher concentration is determined for $PM_{2.5}$ as 265 µg/m^3. The average concentration is around 200 µg/m^3 for the winter season, which means the $PM_{2.5}$ concentration values at some points are very low as compared to other points. If the maps are observed, the south of the city at a lower elevation has been determined to be cleaner than the north-east region, where the wind speed is more than 3 m/s. This wind generally transfers the pollution to the north-east. This flow line is in an increasing elevation, and it ends at the center of the Safranbolu district.

The O_3 and VOC parameters show a different trend as compared to other pollutants. The concentration of these two parameters increases in the spring season. The O_3 concentration increases due to increasing rate of photochemical reactions in the atmosphere when the conditions are right. Another important factor to cause increase in the ozone concentration is the presence of VOCs in the atmosphere. VOC mostly originates from inefficient burning of fuels in road transportation.

4. CONCLUSION

The air pollution is a big problem in industrial and populated big cities. Although a high percentage of air pollution originates from the industry, other sources can also be very effective in the creation of pollution. In order to understand the effect of all sources on pollution, the measurements of atmospheric pollution must be carried out in the entire city. The pollution measurement stations installed in the city are very important for the control of air quality. In these measurement stations measurement of many parameters (pollutants) can be done on continuous basis. In the Karabuk province, there are 5 main stations. There is only one station in the Safranbolu district. These stations are not enough to make the pollution map or air quality map of the Karabük province. In order to be able to make the air pollution map of the Karabük province, a portable air pollution measurement equipment is designed and manufactured for measuring air pollution at any point of the Karabük province. It is an Arduino-based control system. It can measure SO_2, CO_2, CO, CH_4, NO_X, O_3, $PM_{2.5}$ and VOC parameters. The measuring tool is a mobile system that is connected to satellites by using GPRS to find out the coordinates of the location where measurement is made. It is possible to measure the concentration of the air pollution parameters at any point in the region of interest without any other supplementary equipment needed. The measured values are recorded on the SD card. The meteorological parameters such as atmospheric temperatures (ºC), humidity (%) and pressure (Pa) are also measured automatically. If it is needed, an anemometer can also be connected to the equipment to measure the wind speed. By using these data collected, detailed pollution maps can be prepared for any region. The effects of winds and atmospheric temperatures on pollution distribution are also considered and studied. The northeast winds with a speed of more than 3 m/s are very effective in the transportation of pollutants to the Safranbolu district. During these transportations, the ground-level concentrations of pollutants in Safranbolu are always very high.

Elevation, temperatures, atmospheric pressure, and wind are sensitive parameters for the distribution of atmospheric pollution. In the Karabük province, most of the measurement points are affected by different pollution sources. This can be seen clearly from the scatter diagrams.

For isolated measurement points which are far away from the roads, industries and households, the correlated regression relations between parameters are obtained. This can be expressed with a high R-squared value. All types of relations including linear, polynomial (2nd, and 3rd order) and logarithmic can be obtained between the parameters. The daily and monthly average concentrations of pollutants are used for the analyses and mappings. The averages are determined by mean (geometric, harmonic, root, interquartile, Winsorized), median, midrange, Skewness and Kurtosis analyses. These statistical analyses give an acceptable average value after omitting many outliers in the data sets.

During the winter months the air pollution increases considerably. However, O_3 and VOC parameters show a different trend as compared to other pollutants. The concentration of these two parameters increases in the spring season. The O_3 concentration increases due to the increasing rate of photochemical reactions in the atmosphere. VOC, which originates mainly from inefficient burning of fuels from road transportation, causes increase of ozone concentration by photochemical reactions under high atmospheric temperatures of sunlight. This increase can be easily seen in the spring maps.

The range of concentrations of all parameters are found to be as follows: For example, CH_4 concentration changes between 63.66 µg/m^3 to 110.73 µg/m^3. The range of $PM_{2.5}$ concentration is between 127.24 µg/m^3 to 295.2 µg/m^3. The smallest range is determined for the O_3 concentrations. The range is only between 19.3 µg/m^3 to 36.2 µg/m^3.

Nomenclature		$\overline{X}_{Win}$	Winsorized mean
b	Intercept	$\overline{Y}$	Average independent variable
c	Constant (real number)	Y_k	Dependent variable
$\overline{\ln X}$	Logarithmic independent variable average	**Abbreviations**	
$\overline{\ln Y}$	Logarithmic dependent variable average	AAD	Average absolute deviation
M	Midrange	CH_4	Methane
m	Slope	CO	Carbon monoxide
n	Observation numbers	CO_2	Carbon dioxide
N_k	Height value of points	CV	Coefficient of variation
N_p	Desired height value of point P	GIS	Geographic information systems
p	Median value	GPS	Global positioning system
P_k	Weight value of point P	GPRS	General packet radio service
Q_1	Lower quarter	KU	Kurtosis
Q_2	Middle half	LCD	Liquid crystal display
Q_3	Upper quarter	MAD	Median absolute deviation
R	Range	MD	Mean difference
R^2	R squared value	MEU	Ministry of Environment, and Urbanization
s	Standard deviation	NO_X	Nitrogen oxides
s^2	Varience	O_3	Ozone
U_k	Observed value	PM	Particulate matter
V_k	Fitted value	QD	Quartile dispersion
$\overline{X}$	Average dependent variable	RMD	Relative mean difference
$\overline{X}_G$	Geometric mean	SD card	Secure digital memory card
$\overline{X}_H$	Harmonic mean	SE	Standard error
$\overline{X}_{IQM}$	Interquartile mean	SK	Skewness
X_k	Observed value (independent variable)	SO_2	Sulfur dioxide
$\overline{X}_M$	Mean value	SSE	Sum of squares of the residual error
X_{max}	Maximum value	SST	Treatment sum of squares
X_{min}	Minimum value	VOC	Volatile organic compounds
X_{RMS}	Root mean square	TVOC	Total volatile organic compounds
$\overline{X}_{TM}$	Trim mean	2D	Two dimensional
$\overline{X}_{Tri}$	Trimean	3D	Tri dimensional

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DECLARATION OF ETHICAL STANDARDS

The authors of this article declare that the materials and methods used in this study do not require ethical committee permission, and/or legal-special permission.

AUTHORS' CONTRIBUTIONS

Ali CAN: Preparing Pollution Databases, Collecting Pollution Data, Analyses, Writing, Editing.

Hasan ÖZSOY: Help in Preparing Pollution Databases, Collecting Pollution Data, Analyses, Writing.

CONFLICT OF INTEREST

There is no conflict of interest in this study.

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