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# Original Research Spatial-Temporal Variations of Sulphur Dioxide Concentration, Source, and Probability Assessment Using a GIS-Based Geostatistical Approach

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# Abstract

Ground-level sulphur dioxide is one of the air pollutants of high concern as a typical indicator of urban air quality. To inform decisions regarding, for instance, the protection of public health from elevated SO<sub>2</sub> levels in the city of Balikesir, an understanding of spatial-temporal variance of SO<sub>2</sub> patterns is necessary. Therefore, the aim of this study is to locate sample points, characterize distribution patterns, perform the probability map, and map SO<sub>2</sub> distributions by means of spatial information sciences. In this work, the data were compiled from 48 sampling sites using passive sampling on 10-17 March 2010 (in winter) and on 13-20 August 2010 (in summer). The estimations of SO<sub>2</sub> levels at unsampled locations were carried out with the inverse distance weighted method. Finally, locations exceeding the Turkish Air Quality Standard threshold value were determined in the Balikesir by use of geostatistical algorithms (Indicator kriging). The capability of the methods to predict air quality data in an area with multiple land-use types and pollution sources were then discussed. The results of the passive sampling study show that the winter and summer average concentrations are 32.79  $\mu g/m^3$  and 28.27  $\mu g/m^3$  for SO<sub>2</sub>, respectively. It is expected that where industrial activity is not excessively important, traffic and domestic heating systems are the main source of SO<sub>2</sub> precursors. Moreover, using Indicator Kriging, results show that there are multiple hotspots for SO<sub>2</sub> concentrations and they are strongly correlated to the locations of industrial plants, traffic, and domestic heating systems in Balikesir.

Keywords: sulfur dioxide, GIS, inverse distance weighted, spatial analysis, Balikesir

#### Introduction

Air pollution in an urban atmosphere has been known to cause adverse effects on human health and the environment. In many countries, cities are facing increasing urban air pollution and its negative effects. Rapidly increasing population, unplanned urbanization, industrialization, and motor vehicle use are major contributors to urban air pollution in developed countries. In order to protect the public health, governments need to set control strategies.  $SO_2$  is one of the major urban air pollutants. The main sources of  $SO_2$  emissions are fossil fuel combustion, smelting, and the manufacture of sulphuric acid. Coal burning is the largest anthropogenic source of  $SO_2$ , accounting for about 50% of annual global emissions. It causes serious air pollution problems that can be exasperated in urban areas depending on meteorological conditions, topographical characteristics, city planning, and design and human activities.  $SO_2$  is the main air quality indicator and the most monitored pollutant is urban air.  $SO_2$  has an adverse effect on the human respiratory system and the environment because of its contributions to the acidification of the

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ecosystems in soil and water quality. The acid deposition can impact aquatic ecosystems in rivers and lakes and cause damage to forests, crops, and other vegetation. SO<sub>2</sub> emissions also contribute, as a secondary particulate pollutant, to the formation of particulate matter in the atmosphere, an important air pollutant in terms of its adverse impact on human health. Globally, the SO<sub>2</sub> level shows a downward trend in urban atmospheres. Emissions of SO<sub>2</sub> have decreased by 76% between 1990 and 2009 in European Environment Agency member countries [1]. Fig. 1 shows past emission trends of SO2 in the EEA-32 and EU-27 group of countries. Strong progress in reducing SO<sub>2</sub> emissions has been made by many countries; EEA-32 emissions of SO<sub>2</sub> have decreased by 74% between 1990 and 2008. Within the EEA-32 group of countries, all have reported lower emissions in 2008 compared to 1990 except Malta (+4%), Turkey (+28%), and Iceland (+262%) (Fig. 1) [2]. SO<sub>2</sub> concentrations are still an air pollution problem in urban areas that have had uncontrolled urbanization and industrialization. Many Turkish cities have been affected negatively by SO<sub>2</sub> pollution originating from the use of low-quality fossil fuels, rapid urbanization, and industrialization-associated energy consumption and production systems [3, 4]. Air quality statistics in Turkey indicate that annual SO<sub>2</sub> levels exceed the limit values of WHO, EU, and EPA standards in some cities where coal is used for domestic heating and industrial activities [5].

There are many proposed solutions for air pollution reduction in cities. Monitoring studies are considered essential for improving air quality and protecting human health and the environment. An air pollution map of the city is a powerful tool for management of spatial and temporal air quality data. GIS has gained importance in the field of air pollution mapping.

Many air pollution studies have engaged spatial interpolation methods to produce maps of air pollution concentrations. Interpolation is necessary when the data does not cover the domain of interest completely to predict the value-of-attribute at unsampled sites, by using the known measurements made at locations within the same area. The interpolation techniques based primarily on distanceweighting methods [6, 7] and kriging [8, 9] have been employed. These methods differ only in their choice of sample weights [10].

Most air pollution monitoring networks typically do not resolve the actual spatial variability of air pollution levels due to monitor placement unfairness. Therefore, kriging is problematic when used on small geographic scales that have an insufficient number of monitors [11], because if there are a small number of spatial observations, kriging can smooth the spatial pattern of pollution levels by not capturing the spatial complexity of the pollutant. IDW is a popular form due to its assigning more weight to nearby points than to distant points [11, 12].

Air pollution monitoring in large areas, such as an urban one, is performed by operating a number of monitoring stations located in several sites. Passive sampling is one of the most widely used methods for the monitoring and mapping of air pollution.

In this study, the evaluation of the temporal and spatial variances of the air quality of Balikesir based on SO<sub>2</sub> concentrations was carried out using passive sampling distributed in urban and rural areas. To make informed decisions regarding, for instance, the protection of public health from elevated SO<sub>2</sub> levels in Balikesir, data on spatial-temporal variances of SO<sub>2</sub> patterns are necessary. Therefore, the aim of this study is to locate sample points, characterize distribution patterns, perform the probability map, and map distributions of SO<sub>2</sub> by means of spatial information sciences, thus providing useful information for decision-makers.

#### Methods

#### The Study Area

The population of Balikesir, situated at the Marmara Region but including land within the border of the Aegean Region according to the population census in 2009 is about

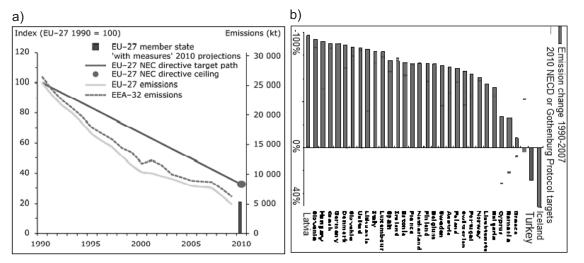


Fig. 1. (a) Emission trends of sulphur oxides (EEA member countries, EU-27 Member States), (b) Change in emissions of  $SO_2$  compared with the 2010 NECD and Gothenburg protocol targets (EEA member countries).

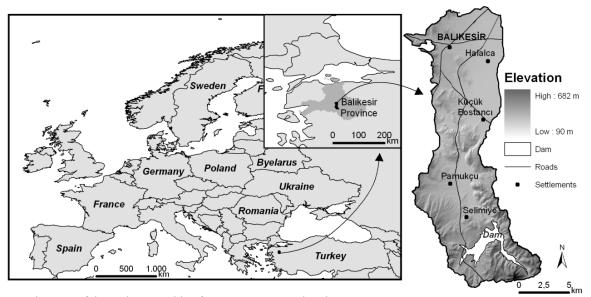


Fig. 2. Location map of the study area, with reference to Europe and Turkey.

259,000 (Fig. 2). Balikesir is affected by air pollution in winter months, which is especially caused by heating systems. According to the SO<sub>2</sub> and PM values in the winter period of 2002-03 and 2005-06, the level of the air pollution in the city center was placed into the category of 1st Group Polluted Cities. The city center of Balikesir was placed among the most polluted cities in Turkey for a number of years according to the conventional SO<sub>2</sub> and PM measurements. The concentrations of SO2 decreased in the period of 1996-2006. SO<sub>2</sub> mean is 78.4  $\mu$ g/m<sup>3</sup>, and the standard deviation is 95.71  $\mu$ g/m<sup>3</sup>. It is clear from the results of the statistics related to SO<sub>2</sub> compiled for 10 years in the period of 1996-2006 that the  $SO_2$  trend is on the decrease [13]. Besides heating systems, industrial and traffic sources, the topographic structure, and city layout, negative meteorological conditions also contribute to the general perspective of air pollution in the city center. The geographical structure of the city center is in the shape of a bowl, the decrease in the dominant winds in winter months, high air pressure, the

decrease in the temperature of air and the frequently occurring foggy days are cause to increase the effect of the pollution.

The statistical data showing the long-term distribution of meteorological parameters consisting of daily average values obtained from a study conducted for 26 years (1980-2006) by Tecer (2008) [13] are summarized in the following paragraphs. When the local meteorological conditions are assessed from the data obtained from the meteorology station in the city center for each parameter, the findings are as follows;

The average winds blow with a speed of 2.73 m/sn while the velocity of 45.3% of the average winds in 24 hours is lower than 2.1 m/s, and only12.4% of the average wind speed was higher than 5.7 m/s. The windrose diagram and wind class frequency distribution were prepared using WRPLOT software and depicted in Fig. 3. The average temperatures were 14.57°C, while the lowest temperature was -9.2°C, and the highest temperature was 32.6°C.

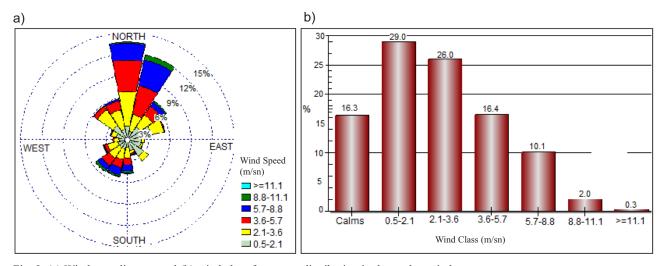


Fig. 3. (a) Wind rose diagram and (b) wind class frequency distribution in the study period.

The average relative humidity of 70.5% has a standard deviation of 21.6%. The 25% of relative humidity, which was measured as 27.3% as minimum and 99.7% as maximum, was higher than 80.3%. These values show that the relative humidity, in the city is high. The statistical data in relation to the rain, cloud, humidity and pressure parameters show that there are stable weather conditions in the city center that prevent the dispersion of the pollutants into the city-wide atmosphere, in which a homogenous mixture is not obtained [13].

#### Data and Method

#### Data

The data were compiled from 48 passive sampling stations geographically distributed throughout the study area, which provided information about the SO<sub>2</sub>, spatial coordinates, and collection time. The period of temporal data collection was from March 10 to March 17, 2010, and 13-20 August for winter and summer, respectively (Table 1). To demonstrate the seasonal change of SO<sub>2</sub> levels, a winter campaign during the heating season in the Balikesir urban area, and summer campaign were organized on a one-week sampling period. Sample locations were selected according to several pollution sources in the urban areas, including: high traffic density regions, residential areas (representing coal consumption for heating), industrial areas, and rural areas. The modified analyst type samplers were chosen to measure SO<sub>2</sub> concentrations in Balikesir Atmosphere [14, 15]. Diffusion tubes were mounted to select points such as street lights, trees, and traffic lights at 2-3.5 m above the ground. During the sampling campaign, passive samplers were used in exposed and sheltered conditions. The samplers were analyzed in the Anadolu University Environmental Engineering Laboratories using ion chromatography. Table 1 shows the description of sampling sites including site codes, number, typology, and exposure time.

Other data used in the study is an adigital elevation model (DEM) created from the scanned 1:25,000 scale georeferenced topographic maps in order to draw the study area boundary. It was determined from watershed by using ArcHydro analysis because watershed is a key factor in showing  $SO_2$  variability and to minimize the impact of topography. The study area includes the city itself and the surrounding area.

#### Spatial Interpolation Methods

After obtaining all ground-level  $SO_2$  measurements, the spatial distribution of this pollutant in the city and around the city was analyzed for winter and summer periods. Later, the  $SO_2$  level at other locations where direct measurements were not carried out was predicted. Because of the fact that the factors that determine the values of environmental variables are numerous, largely unknown in detail, and interact

Sampling code	Site description	Sampling periods			
	She description	Winter	Summer		
MWAY	Rural Motorway, 4 sites	8 days 8 days			
URB	Urban-Residential, 3 sites	8 days	8 days		
URB+TR.	Urban, Traffic, 2 sites	8 days	8 days		
IND	Industry, 2 sites	8 days	8 days		
SUBURB	Suburban, 4 sites	8 days	8 days		
RUR	Rural, 3 sites	8 days	8 days		

Table 1. Details of the sampling sites.

with a complexity that we cannot unravel, we can regard their outcomes as random.

We selected Inverse distance weighting (IDW) to predict  $SO_2$  air concentrations. This method is widely used to interpolate the climatic data to create spatial models [16]. Interpolation weights in IDW are computed as a function of the distance between observed sample sites and the site at which the prediction has to be made [17, 18]. The power parameter in IDW interpolation controls the significance of surrounding points. Hence a higher power of point two was set for creating air quality models as well as to study the errors. Estimation was done using winter, summer, and annual mean.

In the decision-making process care must be taken in using a map of predicted SO<sub>2</sub> for identifying unsafe city areas because it is necessary to understand the uncertainty of the predictions. Probability maps show the degree that the interpolated values exceed a specified variable's threshold. A number of air quality studies have used the geostatistical capabilities of GIS to compute the probability that air quality target thresholds are exceeded locally [19-21]. In the second step we used the Indicator Kriging (IK) technique to calculate the conditional probability of the occurrence of SO<sub>2</sub> concentrations, to show locations that exceeded the critical threshold SO<sub>2</sub> value in the city area because IK makes better predictions than traditional kriging [20, 22]. In the study we accepted 20 µg/m<sup>3</sup>, the Turkish limit value which is to be met by 1 January 2014, as an annual critical threshold. Interpolated values show prediction probabilities (ranging from 0 to 1, i.e. least probable to most probable) of the annual limit of 20 µg/m3 being exceeded throughout the year.

The mean prediction error (0.05) and the mean standardized prediction error (0.07) are close to zero. This shows that the predictions are unbiased and the model is accurate. Also, because the root mean-square prediction error (0.53) is close to the average kriging standard error (0.46), it can be accepted that the prediction errors are correctly assessed in the model.

To determine local autocorrelation in the  $SO_2$  data, ArcGIS 9.3 software (ESRI Corp, Redlands, CA) was used to implement the geostatistical interpolation method.

Sampling code	Site description	SO <sub>2</sub> Concent	Ratio	
	She description	Winter	Summer	Winter/Summer
MWAY	Rural Motorway, 4 sites	40.29	29.44	1.37
URB	Urban-Residential, 3 sites	30.49	18.86	1.62
URB+TR.	Urban, Traffic, 2 sites	43.87	38.48	1.14
IND	Industry, 2 sites	41.73	42.85	0.97
SUBURB	Suburban, 4 sites	25.39	25.68	0.99
RUR	Rural, 3sites	26.93	26.52	1.02
	Total means	32.79	28.27	2.48

Table 2. SO<sub>2</sub> concentrations averaged over the sampling period (8 days).

## **Results and Discussion**

# Temporal Variation of SO<sub>2</sub> Concentrations

SO<sub>2</sub> concentrations of different sampling sites in winter and summer seasons are presented in Table 2. One-week sampling concentrations of SO<sub>2</sub> ranged from 25.39 µg/m<sup>3</sup> to 40.29 µg/m<sup>3</sup> in winter and from 18.86 µg/m<sup>3</sup> to 42.85 µg/m<sup>3</sup> in summer. As shown in Table 2, the mean annual SO<sub>2</sub> concentrations at all the sites were 30.53 µg/m<sup>3</sup>, while mean winter concentrations were higher than the mean in summer (i.e 32.79 µg/m<sup>3</sup> and 28.27 µg/m<sup>3</sup>). In the study period, annual mean of SO<sub>2</sub> concentrations were under the Turkish Air Quality Limit (60 µg/m<sup>3</sup>), but exceeded the European Union limit (20 µg/m<sup>3</sup>) on long-term average at most sites. During winter seasons, motorway, residential areas, urban traffic, and industry sites had higher levels of SO<sub>2</sub> than the suburban and rural sites. These high values might be due to residential heating, urban traffic, and industrial emission.

# Spatial Variation of SO<sub>2</sub> Concentrations

The IDW technique was used to obtain spatial distribution of SO<sub>2</sub> concentrations over Balikesir. Fig. 4 illustrates the spatial distributions of SO<sub>2</sub> in the area. Many "hot spots" with levels above 40  $\mu$ g/m<sup>3</sup> were measured both in the center of the city (where there is dense traffic and building) and in industrial areas on the southern part of the city.

The north-south trend in air quality can be attributed to an  $SO_2$  difference between the urban and the suburban. Less traffic and prevailing wind direction are contributing factors to the relatively low values in the northern part of the area. The north has the least contaminated neighborhoods of Balikesir city, coinciding with the most extensive blue areas of the city. As can be seen in Fig. 4, the city center is the most contaminated place due to the significant emissions of road vehicles, and also high-density building. The high concentration of industrial areas leads to high levels of pollution in the southern part of the study area. Also,

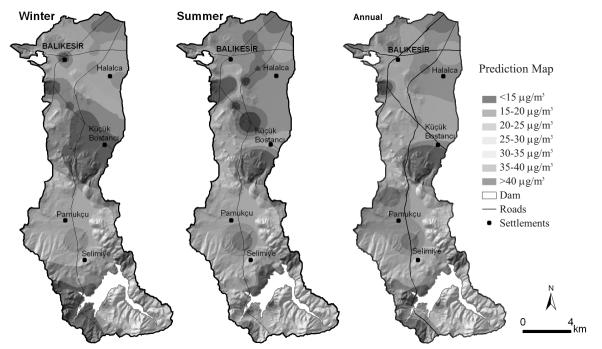


Fig. 4. Interpolated SO<sub>2</sub> ( $\mu$ g/m<sup>3</sup>) distribution in Balikesir.

le 3. Cross tabl	e of aspect and winter $SO_2$ disturbance on the study area.
Classification	Aspect

Classification SO <sub>2</sub> [µg/m <sup>3</sup> ]	Aspect						Total			
	Flat	Ν	NE	Е	SE	S	SW	W	NW	Total
20.0-21.0	0.3	0.0	1.3	4.9	16.7	48.0	25.8	2.9	0.1	100
21.0-25.0	0.0	3.2	11.8	30.0	30.9	15.7	5.1	2.2	1.0	100
25.0-30.0	1.2	9.2	13.2	23.8	28.0	13.8	5.5	1.8	3.4	100
30.0-35.0	5.3	8.0	19.4	31.2	19.0	7.0	4.6	1.5	4.0	100
35.0-37.7	0.5	4.3	16.1	37.4	18.6	12.1	6.1	3.0	2.0	100

population and building density are higher in the southern part. Another sensitive area is the southeast, where there are major roadways and heavy traffic activity. Also of concern is the military airport situated east of the city area.

It is clear that the SO<sub>2</sub> concentrations are higher in winter than in summer. In the industrial, residential, and motorway sites SO<sub>2</sub> levels have exceeded the long-term annual European Union limit of  $20 \ \mu g/m^3$ . This result indicates that SO<sub>2</sub> pollution in Balikesir is mainly a problem and product of the winter heating season, due to the combustion of highsulphur contain coal for residential and industrial activities. The motorway sites (14 sites) were situated just near the Bursa-Balikesir-Izmir Motorway (D565) from the northsouth direction in the study area. This motorway passes through the city center. So, at these sites, high density of traffic and residential activities might cause higher SO<sub>2</sub> concentrations.

Table 3 shows relationships between aspect and disturbance of  $SO_2$ . In the study area, the highest levels of  $SO_2$  are on the E, SE, and NE aspects, respectively. A topographic effect on SO<sub>2</sub> disturbance during the winter seasons is the north. In the winter season, dominant wind directions are NNE, NNW, and N, respectively [23]. Because of these dominant wind directions, the areas with the highest SO<sub>2</sub> concentrations moved from the north and the northwest of Balikesir city to the southwest of the city. The pattern in Fig. 5 shows that the SO<sub>2</sub> concentration was not only caused by industrial, residential, and traffic emission sources, but also may be influenced by meteorological factors, especially wind speed and direction. The analysis of the relationships between SO<sub>2</sub> concentrations and local meteorological factors indicated that low temperature and wind speed might result in higher SO<sub>2</sub> levels in winter than in summer.

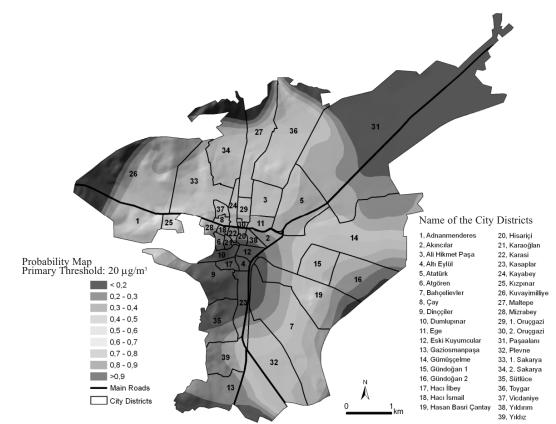


Fig. 5. Probability that the SO<sub>2</sub> value exceeded 20 µg/m<sup>3</sup> in winter in Balikesir, Turkey.

Tabl

# Probability Mapping of SO<sub>2</sub> Concentrations

Geostatistics allows one to predict the probability that a known critical event produces harm to air quality in excess of a given concentration threshold. This evaluation, developed in the form of maps, can be considered as a map of spatial risk, useful for all local air quality management efforts. The hazardous probability that SO<sub>2</sub> concentrations exceed the Turkish Air Quality Standard of 20  $\mu$ g/m<sup>3</sup> at any of the unsampled sites was determined by IK. Fig. 5 shows the probability map for sites where SO<sub>2</sub> would exceed this critical value in winter.

The probability map shows that there are multiple hotspots of hazard probability in the study area. The hotspots are located in the central and northeastern parts of the study area in the vicinity of the city center and high-traffic system. In these red lines, considerable improvements in air quality are needed to prevent exceeding the air-qualitystandard target level in the future. In the northwestern and southeastern regions of the city, predominately suburban background, the predicted probability of exceeding the target value is lower.

## Conclusion

The aim of our paper was to locate sample points, characterize distribution patterns, perform the probability map, and map distributions of  $SO_2$  in Balikesir urban and rural areas. To do so, the spatial and temporal assessment of air pollution was carried out using passive sampling at selected sites and the GIS approach.

Our first contribution is to show that the map of air pollution made by the IDW interpolation technique is a powerful tool for the determination of high concentrations of and locations affected by air pollution sources. Therefore, the technique is one of the essential methods to analyze and monitor air pollution in large areas like cities. The results indicate that residential heating, industry, and traffic are responsible for pollution in the area. Higher SO<sub>2</sub> concentrations in winter are pointed out by sites dominated by coal combustion for heating systems. These results are similar to the finding reported by previous studies [3, 15], which found that the SO<sub>2</sub> levels are higher in winter due to coal combustion. Industrial and traffic emission sources contributed to the annual SO2 concentrations at relatively higher levels. In addition, the ambient SO<sub>2</sub> concentration may be influenced by biogenic sources as well as from longrange transport and from adjacent areas [24, 25].

Furthermore, the mapping of probability of target threshold exceedances and associated city districts provided additional insight to improve air quality in the decisionmaking process by using IK. The use of  $SO_2$  prediction and probability mapping makes it easier to identify areas where air quality is a problem. The Balikesir urban area is characterized by more than one hot-spot site, these being mainly influenced by road-traffic and domestic heating emissions. The highest  $SO_2$  values were found near streets with high traffic volume. The resulting data of this study shows that the passive sampling method and spatial analysis using GIS are well suited for the assessment of air pollution in a region of complex terrain like an urban area. It is expected that the results of the study will be useful for improving air quality, protecting human health, and preparing effective clean air strategies. However, it should be considered that the number of points used in the IDW procedure is important and affects the resulting map [26]. Finally, this study's general methodology could certainly be extended to other atmospheric pollutants and to other environmental variables.

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