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Urban scale air quality analysis due to coal-based residential heating

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Abstract

The environmental effects of air pollutants released to the atmosphere from coal-based residential heating should be regarded as one of the primary environmental concerns in cities. Unfortunately, in Turkey, hundreds of medical cases still occur due to gas poisoning from coal-based conventional stoves used for heating purposes. This study attempts to investigate the effects of coal-based residential heating on CO and SO_2 air quality in a city of the south Marmara Region located between Europe and Asia. A total of 138 chimneys were sampled in the heating season that falls from October 1st through March 31st in the city. Ambient air pollutants released from those chimneys were analyzed to evaluate the background air quality variations in the city. The mean of CO concentrations was approximately 11,000 mg/m^3, with variations from nearly 9500 to 12,500 mg/m^3, while the mean of SO_2 concentrations was roughly 173 mg/Nm^3 ranging from 108 to 240 mg/Nm^3 in the sampled chimneys. The AERMOD predicted the maximum daily mean CO concentration for the model was 41.5 µg/m^3 on February 29th at midnight for the downtown area and exceeded the official limits. The predicted highest periodic SO_2 concentration was 45.1 µg/m^3 on February 29th at midnight in the heating season. The highest periodic SO_2 concentration was observed in the old settlements of the downtown, where the most coals were utilized for residential heating with antiquated systems. It is confirmed that the AERMOD results are valid by using meteorological and air pollution data for the modeling study.

Keywords Coal · Residential heating · Carbon monoxide · Sulfur dioxide · AERMOD

Introduction

Coal has been utilized for residential heating in parallel with industrial developments. Today, coal and its derivatives are mainly used to meet the heating demands and energy needs in the world. According to the WHO, residential heating is an essential need for people, and approximately 3 billion people still use solid fuels, such as coal and wood for residential heating and cooking purposes worldwide (WHO 2015). Despite the increasing use of natural gas and electricity for heating purposes, the utilization of coals for residential heating is considered to be a traditional and mutual practice in many places in the world (Kerimray et al. 2017). This situation is highly related to the state of the development and acquisition of domestic raw material reserves. Coal and other types of raw materials, such as municipal wastes, forestry residuals, and agricultural wastes, may be used for heating purposes. The large proportion of families in some countries revert to residential heating practices by using low-moisture solid wastes, such as wood scraps and discarded or surplus furniture, due to economic turndown or living difficulties in their countries (Saffari et al. 2013; WHO 2015). From a general point of view, coal consumption for residential heating seems to continue for a while in developing and undeveloped countries. This situation might be changed by improving the economic conditions of those countries and the use of alternative fuel types everywhere and by the long-term strategies in reducing and prohibiting coal utilization for home or space heating purposes.

Coal utilization may release elements and compounds, such as arsenic, sulfur, mercury, and lead, that might be mainly harmful to human well-being. The short- and long-term exposures to coal smoke may affect human health because smoke consists of approximately 28 different pollutants, including 14 pollutants classified as carcinogenic compounds (Smith et al. 2014; Loomis et al. 2013). Zhang and Smith (2007) corroborated that a significant correlation

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was found between coal smoke exposure and lung cancer in many epidemiological evaluations (Liu et al. 1991). Lung cancer apprehension, severe respiratory illnesses, lung function reductions, immune system impairments, and poisonous coal endemics in mainly CO poisoning might be highly related to coal smoke exposure in indoor and ambient conditions. Coal combustion products, for example, carbonaceous and sulfurous pollutants, directly harm humans (Zhang and Smith 2007; Zhang et al. 2008), and they might be serious threats for our habitats and ecosystem in the form of several environmental concerns, such as acid rains, haze, and the impairment of visibility. Additionally, coal consumption may have an indirect impact on climate change (Borm 1997; Finkelman et al. 2002; Finkelman 2004; Zhang et al. 2008).

Coal or coke as a coal-derived fuel has been utilized as a priority choice in Germany in the 1960s, and a similar situation exists in France, Denmark, and Canada. After two decades, the use of coal or its derivatives for residential heating was declined in Canada, Norway, and Sweden due to the increasing use of oil or natural gas nationwide (Schipper et al. 1985). In the Netherlands, coal was used as a major source for heating from the 1950s through the 1960s; however, the use of coal ended in the mid-1970s (Dzioubinski and Chipman 1999). In the USA, coals were used for residential heating by 55% of homes in the mainland, and the coal utilization rate has dramatically decreased since the 1940s. The rate was less than 1% in the USA in the 1980s (USCB 2011). In China, the use of coals for residential heating may contribute to 7% of national SO_2 emissions (WHO 2015), and they are currently considered primary ambient emission sources, such as fine particles ($PM_{2.5}$), carbon monoxide (CO), carbon dioxide (CO_2), and sulfur dioxide (SO_2) (Li et al. 2017; Tian et al. 2018). Some European countries where coal burning for heating predominantly exists may be over the average global emissions. Thus, residential coal utilization may be responsible for 4% of SO_2 and 1% of NO_x emissions worldwide (WHO 2015). Coal utilization has been mostly replaced with natural gas in North America and most part of Europe and will also be banned entirely in the capital of China by 2020 due to the emergence of various fuel alternatives and the deleterious effects of coal-burning on air quality (Sickles and Shadwick 2015; Kerimray et al. 2017; Li et al. 2017). Nonetheless, in many countries, a high proportion of residences and homes use coal-based heating systems, thereby increasing demand for coal consumption (Kerimray et al. 2017).

In Turkey, several medical cases occur due to carbon monoxide poisoning from old-fashioned stoves used for heating purposes. Unfortunately, many cases resulted in death. In line with previous studies, Metin et al. (2011) have concluded that poisoning from coal-based heating cases have been occurred mostly in Marmara Region. In 2016, there were a total of 831 cases of flue gas poisoning, and 141 of these cases resulted in deaths (Akgun 2017). A total of 129 cases have been reported in the Marmara Region where this study was conducted, and 19 of these cases have resulted in death (Akgun 2017). According to the statistics, February and March were determined as the months with the highest incidence, especially in heating seasons. Accordingly, the ambient concentrations of CO and SO_2 pollutants must be analyzed in heating seasons when heating needs intensively emerge in Balikesir's downtown.

This study attempts to investigate the effect of coal-based residential heating on ambient CO and SO_2 air quality in Balikesir. The study consists of two stages. The first one includes the sampling of coal-based chimneys, and for this reason, a total of 138 chimneys were sampled to determine emitted CO and SO_2 levels. The second part of the study covers the air quality modeling of those air pollutants; therefore, the AERMOD (American Meteorological Society/Environmental Protection Agency Regulatory Model) air quality dispersion modeling was performed to determine whether those chimneys might affect the ambient air quality levels of the city, especially in a cold season.

Materials and methods

Study area

The study was conducted in the north-western part of Turkey. Balikesir, a mid-sized city, has more agricultural and livestock activities than industrial applications. Figure 1 illustrates that Balikesir Province is located between 39.20° and 40.30° north parallels and 26.30°–28.30° east meridians. The city has a total population of approximately 1.2 million people, where nearly 350,000 people live in the downtown of the city (TUIK 2017). The general climatic characteristics of Balikesir Province, including mostly coastal areas, are considered the Mediterranean climate that means that the summers are hot and dry and that the winters are more likely warm and rainy. The lowest temperature and the highest amount of precipitation occur in winter, whereas the highest temperature and the lowest precipitation occur in summer (Table 1). The continental climate effect increases toward inner zones, from the west to the east and from the north to the south, of the city; hence, winters are colder in the inner zones. Moreover, due to its unique topographical form, the city has partly settled in the north, north-west, and south-west parts of the mountainous elevations; therefore, the city has almost a semi-bowl-shaped elevation. Dominant wind directions are formed over natural corridors formed in the northern directions of the city center. The downtown is also located in the inner zone, as is shown by the support of the digital image of Google Earth in Fig. 1.

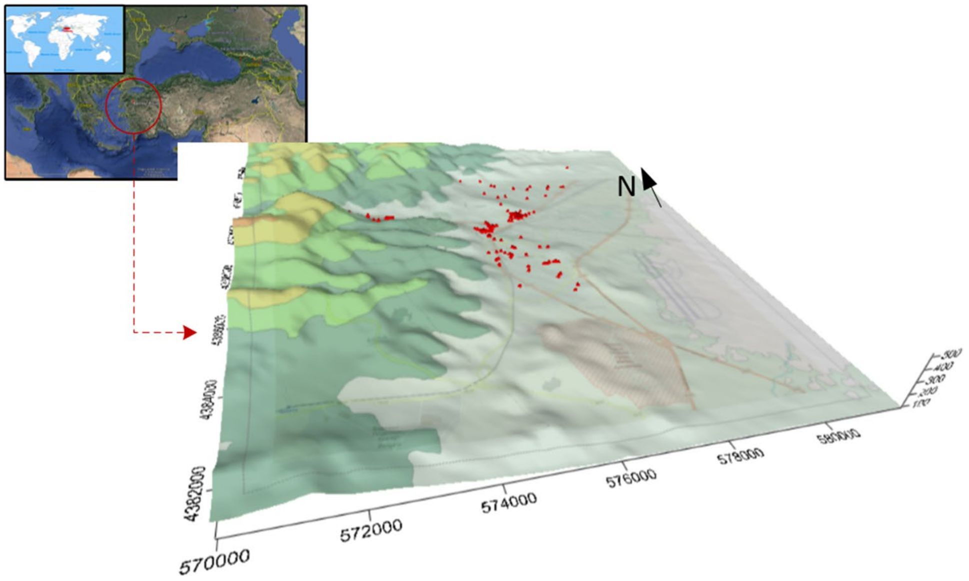


Fig. 1 The study area in different zones and aerial view of sampled chimneys as pointed in red over the downtown

In 2016, the total amount of the coals used for residential heating was approximately 68,000 tons in a portion of 65% delivered by domestic sources and the rest provided by imported coal. Conversely, the total amount of the coals used for industrial purposes was approximately 32,000 tons that could be considered as almost half the amount of coals consumed for residential heating (CAAP 2018). In this manner, it may be identified that residential heating is the major source of air pollution for Balikesir City.

Approximately 72% of the total natural gas was consumed for residential heating purposes, while the rest of the fuel was used for industrial processes (CAAP 2018). Generally, the frequent use of coal for residential heating and industrial processes, including low calorie, high content of sulfur, and other toxic compounds (Jingchao et al. 2018; Li et al. 2018a, b; Zhao and Luo 2018), and improper burning techniques or insufficient combustion performance, may lead to air pollution at any processing stage (Van der Lans et al. 1998).

Meteorological background of the study area

Meteorological occurrences, such as wind speed and direction, temperature, solar radiation, and precipitation, are the factors that may significantly affect ambient pollutant concentrations. For instance, winds play an essential role in the transportation, dispersion, and dilution of air pollutants. Wind speed and direction data provide reliable information on the transport of pollutants from a source to a receptor. Wind data are also used to assess the relationships between pollutant sources and the local air quality monitoring station (AQMS). Air temperature and solar radiation affect the chemical reactions in the atmosphere, while precipitation allows pollutants to be removed or reduced from the atmosphere, such as particulate matter. The change in meteorological parameters appears to have an impact on the atmospheric concentrations and dispersions of ambient pollutants. For instance, in most places where coal is used for domestic heating, SO_2 pollution decreases with increasing air temperature, and, on the contrary, SO_2 pollution increases with decreasing temperatures. This negative correlation indicates that SO_2 pollution is released by the combustion of fuels used for residential or space heating purposes. The dispersion of air pollutants must be correlated with meteorological parameters to see the effect of heating. Table 1 presents the historical, long-term, and meteorological changes from 1938 through 2017, including the major meteorological parameters (TSMS 2018).

According to the long-term meteorological data presented in Table 1, the mean of precipitation was much higher in winters than in other seasons. The month of August is the driest month with a mean precipitation of 6.1 kg/m^2, while the precipitation reaches its peak with a mean of 94.9 kg/m^2 in December. In terms of temperature, the month of July is the warmest month with a mean of 24.8 °C, and the month of January is considered to be the coldest month with a mean of 4.8 °C.

The Clean Air Act Plan for Balikesir stated that the significant sources of air pollution are residential heating processes, particularly in the winter seasons (CAAP 2018). Furthermore, air pollution levels in Balikesir tend to increase due to bowl-shaped topographical conditions and the reduction of wind speeds and the number of blows in the heating seasons.

Sampling sites and procedures

The measurement of air pollutants was made in the chimneys of apartment complexes in which coals were mainly

Table 1 An overview of the meteorological parameters for Balikesir Province (1938–2017)

Meteorology	January	February	March	April	May	June	July	August	September	October	November	December	Annual
Max. temp., °C	23.5	25.2	30.7	35.2	38.5	42.5	43.2	43.7	40.3	36.4	29.9	26.1	20.4
Mean temp., °C	4.8	5.9	8.2	12.9	17.8	22.4	24.8	24.6	20.7	15.7	10.5	6.6	14.6
Min. temp., °C	−18.6	−18.8	−8.0	−4.0	0.6	4.0	9.1	6.0	4.0	−2.3	−7.9	−12.9	9.0
Max. mean Temp.,m°C	8.8	10.5	13.6	19.3	24.5	29.2	31.2	31.2	27.7	22.0	15.9	10.6	43.7
Min. mean Temp., °C	1.3	1.9	3.3	6.9	11.0	15.0	17.7	17.9	14.1	10.2	6.0	3.1	-18.8
Mean solar Rad., hr	2.9	3.6	4.8	6.3	8.7	10.7	11.6	10.9	8.6	6.2	4.2	2.5	81.0
Mean rainy days	14.4	11.7	11.5	9.3	7.6	4.5	1.6	1.4	3.4	6.7	9.4	13.7	95.2
Mean Precip.,mm	85.0	68.9	60.8	50.1	41.2	24.8	8.3	6.1	21.9	45.6	75.4	94.9	583.0
Mean wind speed m sn^{-1}	1.90	2.16	2.18	1.82	1.65	2.09	2.86	2.93	2.29	1.90	1.51	1.79	2.09
Prevailing wind dir	NNE	NNE	SSW	SSW	SW	NNE	NNE	NNE	NNE	NNE	NNE	NNE	NNE

utilized for residential heating and daily usage purposes. The apartment floors range from 4 to 7 based on topography. In Balikesir, the majority of apartments have four floors by regulation. There are two reasons for this legal limitation, namely, protection from earthquakes and the presence of a military airbase located close to the downtown (Fig. 1). The height of buildings is approximately 12 m, including roof sections, and the height of chimneys is mainly 1.5 m from the roof base.

The sampling period was from February through March 2016 in a heating season of the city. A total of 138 chimneys were sampled, and the ambient air pollutants, carbon monoxides (CO), and sulfur dioxides (SO_2) were mainly measured to analyze the local air quality variations. The other parameters related to the coal-burning process, such as oxygen levels in percentage, were measured during the study. Moreover, other parameters related to heating boiler performance, such as combustion efficiency, were also determined in the sampling procedure to identify if there was a correlation between those parameters and the measured air pollutants. Figure 1 depicts the sampled chimneys of the apartments with their actual coordinate points.

As illustrated in Fig. 1, the sampling points are in red dots, and they are mainly located in the center of the downtown area. There are a few apartment complexes rarely scattered farther north and north-west of the downtown.

The measurements were conducted using a portable gas analyzer (Madur, GA-21 Plus, Poland) with an accuracy level for O_2 (± 0.01%), CO (± 5 ppm), and SO_2 (± 5 ppm). The gaseous parameters were measured using electrochemical cells, while the other physical parameters, such as ambient and stack temperatures, were measured using specific resistive electrochemical sensors and standard Ni-CrNi thermocouples. The analyzer could detect the measured toxic gas concentrations in ppm and mg/Nm^3 after the readings were adjusted on the basis of reference oxygen percent as stated in the "National Regulation on Air pollution Control Caused by Heating" (RCAPCH 2005). CO and SO_2 measurements were corrected for the 8% oxygen content according to the relevant national air quality regulation. The other parameters that were directly related to coal-burning processes, such as oxygen levels and combustion efficiency in percentage, were also measured to determine their effects on the air pollutants during the study.

All the combustion of the heating boilers at the sampling site were utilized by using coals. The main parameters consistently measured in all chimneys were gas temperature, combustion efficiency, O_2 (as in %), CO, and SO_2 during the study. The regular heating season officially includes months from October through March at the study area (RAQAM 2008). According to official statistics, approximately 60% of the residence and nearly 16% of the industrial site use natural gas in the downtown (CAAP 2018).

Dispersion of local air pollutants

Air quality dispersion models have been used for more than 30 years (Venkatram 1979; Fox 1984; Weil et al. 1992; Stein and Wyngaard 2001; Irwin 2014), and they are a useful method to predict local or regional air pollution levels that might exist in the ambient under distinct scenarios, such as meteorological conditions, topographic properties, and divergent sources (USEPA 1998; Kesarkar et al. 2007; Stein et al. 2007; Seangkiatiyuth et al. 2011). Dispersion models are considered an alternative method when pollutants from different sources may not be technically feasible to measure specific points or places (USEPA 2009; O'Shaughnessy and Altmaier 2011). The models have also provided reliable results in time-based periodic epidemiological studies (Zou et al. 2010).

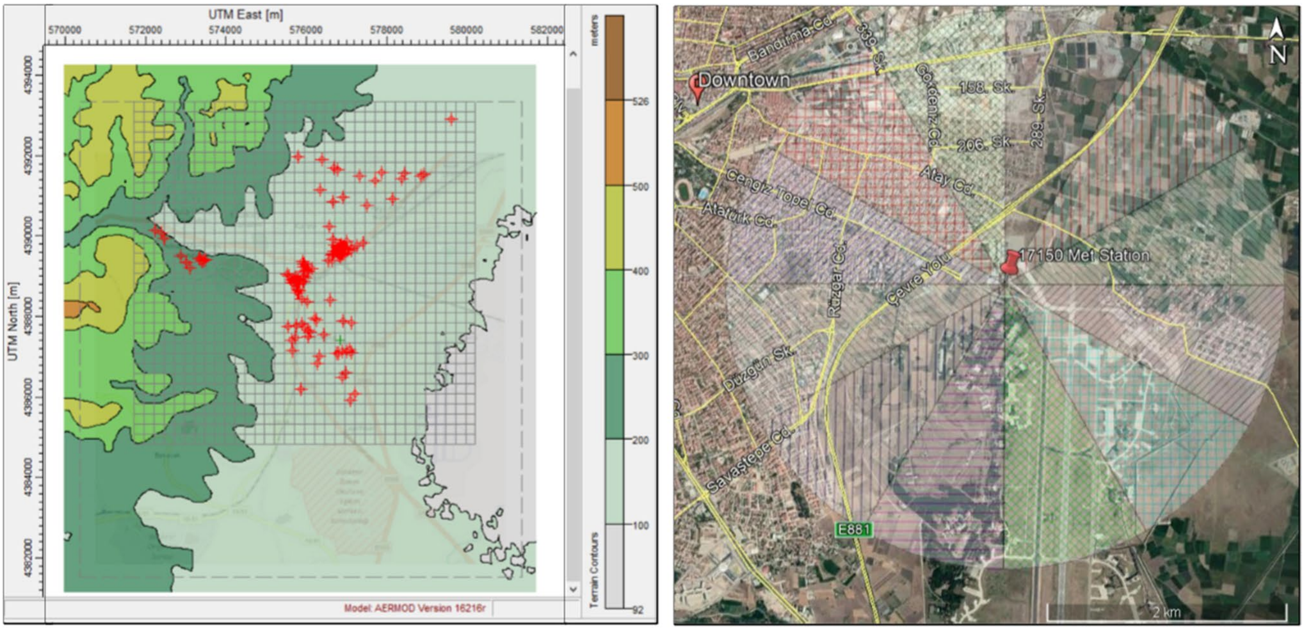
The AERMOD is a steady-state plume model, and its algorithm contains three separate components: AERMIC (dispersion model), AERMAP (terrain preprocessor), and AERMET (meteorological data preprocessor) (USEPA 2003). The USEPA has initially presented the AERMOD as a new dispersion model in April 2000. According to previous studies by Tartakovsky et al. (2016) and ADMGO (2016), the AERMOD was highly efficient for the dispersion modeling of air pollutants up to a roughly 50-km diameter of the pollutant source.

In the modeling design, the AERMOD modeling algorithm was employed to estimate the concentrations of CO and SO_2 airborne pollutants. The ground-level concentrations have been predicted by constructing a total of 1228 ground-level uniform spaced receptors by covering the study area on a Cartesian grid system. Moreover, all sampled chimneys have been individually pointed in the model, and the modeling layer including topographical properties of the study area, the sampled chimneys and old settlements area of the city are illustrated in Fig. 2a.

Before starting the modeling stage, the representative meteorological and terrain data are necessary for the modeling study. The other components of the AERMOD, such as AERMET, were employed using local meteorological hourly data. All the meteorological data were obtained from the Provincial Agency of Meteorology with a ratio of missing data, especially for wind speed, which was approximately 24% in the modeling study.

All the necessary meteorological data such as hourly wind speed, direction and frequency, temperature, precipitation, pressure, cloudiness, and surface characteristics for determining boundary layers that were analyzed in AERMET, and then the met data was compiled for the AERMOD (USEPA 2004).

According to the implementation guide of USEPA for the AERMOD, the surface characteristics are required to determine the boundary layer and three major surface characteristics, including surface roughness length (z_o), albedo (r), and Bowen ratio (B_o) that must be defined by using the AERMET. The surface roughness length is related to any



(a) (b)

Fig. 2 **a** Topographical layer of the study area with sampled chimneys in red mark and old settlements area in blue dashed circle. **b** Lad use distribution of the study area obstacles within a dimension when the mean horizontal wind speed is zero. The surface roughness length is considered to be an important factor in determining the stability of the boundary layer. The albedo is used for the reflectivity of solar radiation and is defined as the fraction of the total amount of solar radiation reflected by the surface layer back to the atmosphere. The Bowen ratio indicates the availability of surface moisture on the layer and is the ratio of the sensible heat to latent heat fluxes (USEPA 2019). The recommended domain is limited to a 10 km × 10 km region for the AERMOD application in terms of representative surface characteristics. The effects of the Bowen ratio and albedo parameters on meteorological measurements and plume dispersions are different from surface roughness. The Bowen ratio and albedo are used to express the power of convective turbulence during unstable conditions by determining how much of the incoming radiation is converted to sensible heat fluxes (USEPA 2019). The representative surface characteristic values have been determined on the basis of the local land cover observations. The city has approximately 30% forestry; 35% pastureland and meadows; and 15% horticultural fields including olive, vegetable, and orchid gardens for this modeling study (CAAP 2018). In the modeling process, the study area has been divided into 12 individual regions by setting 30° angles. The albedo ranged from 0.21 to 0.33, the surface roughness parameter ranged from 0.04 to1.0, and the Bowen ratio ranged from 0.75 to 4.75. Figure 2b presents the land use and topographical characteristics of the study area.

The horizontal datum, modeling domain, that includes all specified receptor and source locations and the terrain data for the study area are necessary, and they are processed in the AERMAP to reflect surface characteristics, including the base elevations of receptors and sources, discrete grids for all receptors, sensitive points on the grids that might be influenced by a dispersion pollutant, and surface roughness for the study area (USEPA 2018). In this study, Cartesian grids with uniform grid spaces were used in the modeling setup because the polar grids were employed for the more specific source that must specify its position (USEPA 2018).

In the meteorological data process, the local representative wind data play an essential role, and it must be well defined for the modeling procedure. Figure 3 exhibits the wind data and the prevailing wind directions for the modeling period. Northern winds were dominant in the study area when the wind data were analyzed on the basis of the wind direction, wind speed, and wind blowing frequency.

All the meteorological data of the representative months of February and March were comprehensively arranged and prepared as two separate databases, namely, surface and upper meteorological data for the modeling process (Fig. 4). The surface meteorological data were obtained from the local air-base station (WMO IDWMO ID#17,150), and the upper (radiosonde) data were provided by the Provincial Agency of Meteorology. There are currently eight meteorological bases that collect radiosonde measurements in Turkey, and the upper data were provided from the nearest center located in Istanbul, Kartal. The AERMET processor transformed the two different databases (surface and upper) into ".sfc" and ".pfl" files. The files were ready to compile in the AERMOD modeling processor (Version 9.4.0, Lakes Environmental Software 2017, Ontario, Canada).

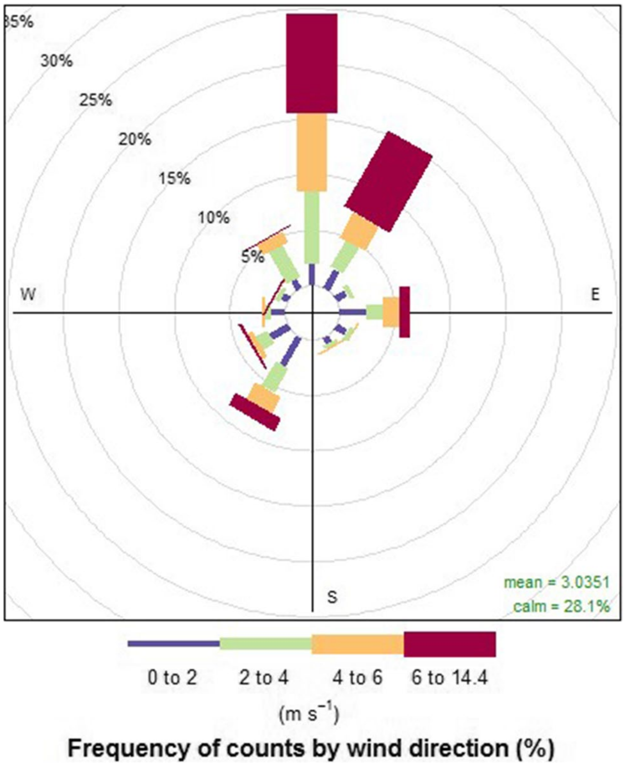


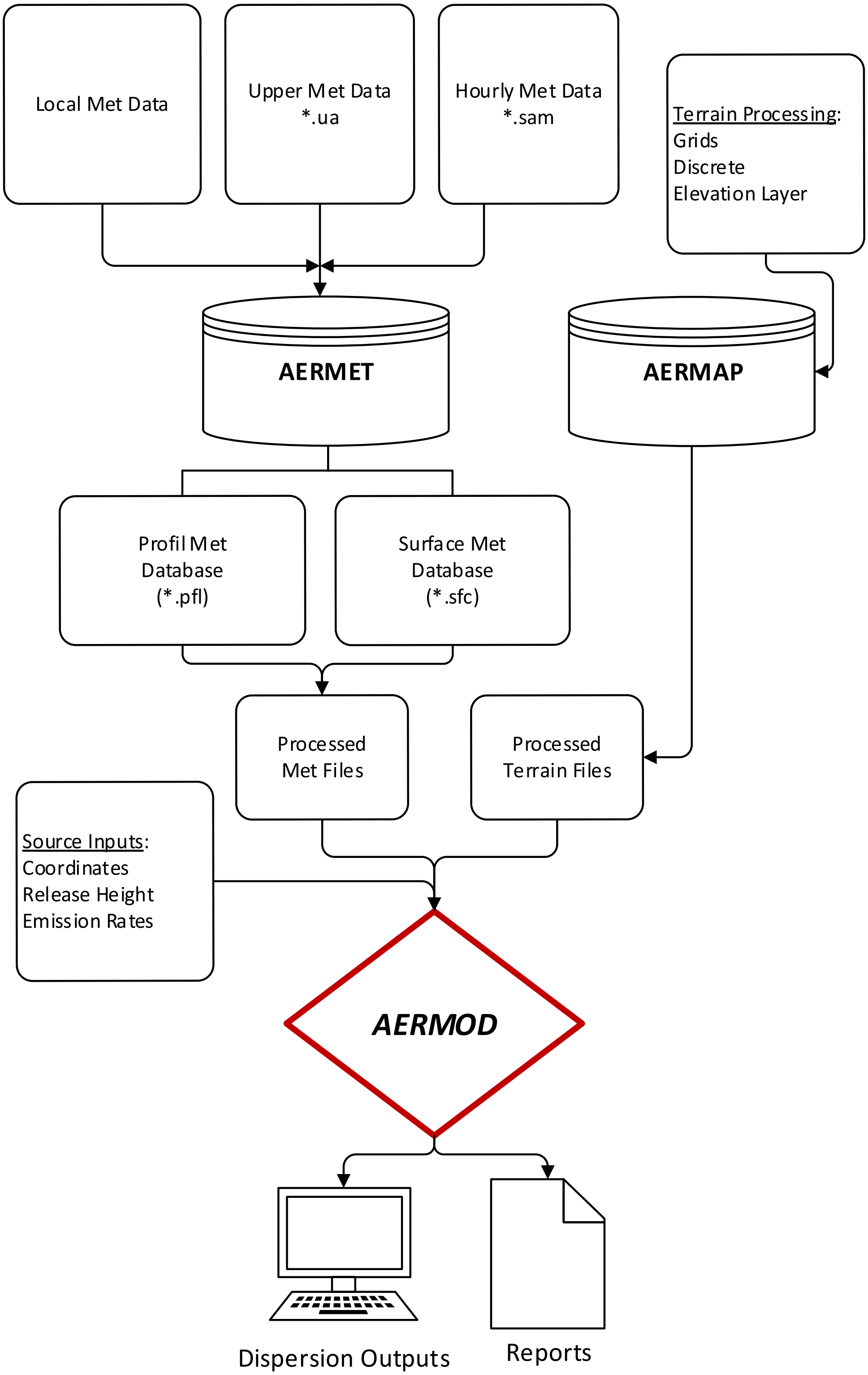
Fig. 3 Windrose diagram for the modeling period (February–March)

The AERMOD was run to characterize the dispersion of local air pollutants in the downtown area due to higher residential heating processes. The model outputs include the predicted highest ground-level pollutant concentrations for this study.

Validation of AERMOD results

The validation of the AERMOD model prediction is a critical process to verify all the predictions for the study. The measured parameters and the AERMOD-predicted parameters should be used together in a proper statistical analysis (Chang and Hanna 2004; Kumar et al. 2006; Abril et al. 2016). There might be four primary uncertainty sources that could be identified for typical air quality models. Basic uncertainties may occur due to the construction of model algorithms, representative model input data, the accuracy of monitoring data, and

Fig. 4 The AERMOD modeling steps



incomplete meteorological data (Dresser and Huizer 2011). In the multiple model performance analysis, for our case, one parameter was chosen from the meteorological side, and one data set was selected from the pollutants. The wind speed, which is considered to be one of the most essential meteorological parameters used in air quality modeling studies and SO_2 concentrations, was used to conduct the performance evaluation of the AERMOD. The AERMOD was employed for the local air quality monitoring station (AQMS) in the downtown. Figures 7 and 8 exhibit the location of the

AQMS. Additionally, the measured ambient SO_2 concentrations from the AQMS and the modeled SO_2 ground-level concentration, specifically for the AQMS, were used in a validation process, and the CO concentrations were not evaluated due to limited measured data in the downtown. The measured and predicted values of wind and SO_2 concentrations were examined by the residual analysis of bias, and the analysis results are presented in Table 2.

In the performance analysis, the calculation process of model validation parameters was thoroughly discussed by Hanna et al. (1991), Hanna (1993), Chang and Hanna (2004), and Hanna and Chang (2012). The comparing parameters, U_{me} and U_{mo}, refer to the measured wind speed and the modeled wind speed values generated in the AERMOD, respectively. The validation parameters including fractional bias (FB) measures systematic biases between measured and predicted values where the positive FB indicates an under-prediction, the negative FB refers to an over-prediction by the used model (Chang 2002; Chang and Hanna 2004; Hanna and Chang 2012). The normalized mean square error (NMSE) implies the overall error of the standardized values between the observed and modeled data. The factor of two (FAC2) is described as the proportion of estimation within a factor of 2 of the observed parameters (Hanna and Chang 2012; Irwin 2014). Also, the normalized absolute difference (NAD) for threshold-base and refers the fractional area for errors (Hanna and Chang 2012).

According to Chang and Hanna (2012) the study results asserted that the AERMOD had better performance on the dispersion of concentrations over a region. Furthermore, "a good model" must meet at least one factor within their acceptance intervals. Therefore, the acceptance intervals of these control parameters are given in Table 3. The data used in the validation process have been obtained after removing irrelevant and missing data in order to calculate validation parameters including FB, NMSE, FAC2, and NAD. The number of data (N) used in validation analysis for both wind speed and SO_2 are given in Table 2.

According to the validation results, as presented in Table 3, the FB, NMSE, FAC2, and NAD satisfied the acceptance criteria for the wind analysis, which indicated that the wind data used in the model were reliable. Furthermore, the performance of the AERMOD was also satisfied with SO_2 concentrations for the FB, NMSE, FAC2, and NAD. In this case, the FB is expected to be close to 0.0 for a perfect model and measures only the systematic bias of the model (Chang and Hanna 2004). According to Chang and Hanna (2004), Langner and Klemm (2011), and Dresser and Huizer (2011), the negative FB refers to be overestimated by the AERMOD for the wind predictions.

Table 3 Model validation parameters with acceptable ranges

Validation Parameters	Acceptance Criterias	Model Performances			
		Wind	Acceptable	SO_2	Acceptable
FB	≤ 0.67	−0.03	✓	0.64	✓
NMSE	≤ 6	0.14	✓	1.88	✓
FAC2	≥ 0.3	1.03	✓	0.52	✓
NAD	≤ 0.5	−0.02	✓	0.32	✓

FB fractional bias, *NMSE* normalized mean square error, *FAC2* fraction of predictions within a factor of two of observations, *NAD* normalized absolute difference

In addition to the validation process, the results of model validation also include a good agreement with the FAC2, which has been defined as the most robust measure because it has not been impacted by the extreme (max. or min.) outlier by Chang (2002). During the study period, only ambient SO_2 has been continuously measured by the AQMS in the downtown. Hence, the FAC2 for SO_2 has been calculated as 0.52 by meaning that the SO_2 levels were satisfied by the AERMOD.

The comparison of observed and modeled SO_2 levels for modeling period of February and March are illustrated in Fig. 5. AERMOD has predicted ground level SO_2 generally underestimated at the low levels comparing the ambient SO_2 level from the AQMS. This result has also been confirmed with the calculated positive FB value. However, similar fluctuations were observed in both measured and modeled SO_2 levels. Previously, Zou et al. (2010) and Gibson et al (2013) concluded that AERMOD results showed a good agreement between modeled and measured SO_2 levels for a long-term time (monthly or annual) period.

Table 2 Results of residual analysis for AERMOD validation

Parameters	N	Means		Std.dev		Model validation parameters			
		Me	Mo	σ_{Me}	σ_{Mo}	FB	NMSE	FAC2	NAD
Wind, m/sn	601	3.9	4.1	2.4	2.3	-0.03	0.14	1.03	-0.02
$SO_{2,}$ μg/m^3	691	4.8	3.52	3.5	6.02	0.64	1.88	0.52	0.32

N number of data (obtained after irrelevant and missing data removed), *Me* measured means, *Mo* modeled means, *FB* fractional bias, *NMSE* normalized mean square error, *FAC2* The fraction of predictions within a factor of two of observations, *NAD* normalized absolute means

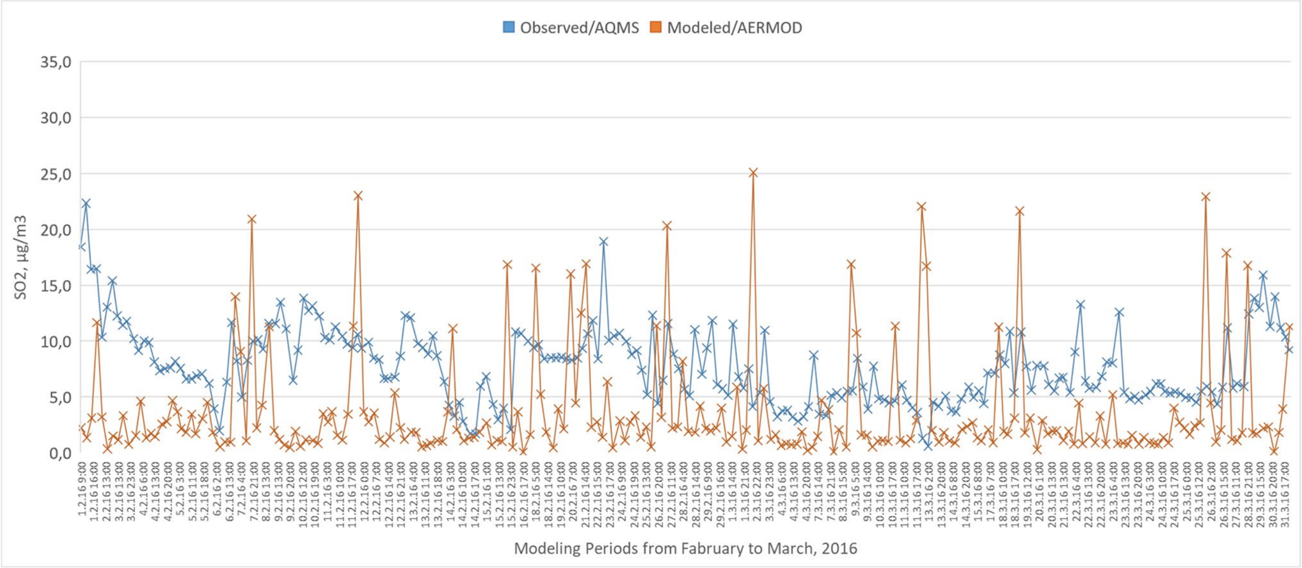


Fig.5 Comparison of measured SO_2 by local AQMS and modeled SO_2 by AERMOD

Study results

Results of gas sampling in chimneys

Multiple chimneys were sampled on daily-basis from February to March. Figure 6 illustrates the daily means of those sampling results. The measured CO concentrations at the chimneys exceeded national limits (10,000 mg/m^3). However, the measured SO_2 concentrations at the chimneys were below the national limits (2,000 mg/m^3). The measured CO concentrations at chimneys have had increasing trends in the middle of the weekdays, especially on Sundays, when people were spending more time at homes. The measured SO_2 concentrations did not show a significant trend during the weekdays, but it has showed similar trends as measured CO levels on Sundays. Both measured gas concentrations had lower rates on Saturdays when the local people were leaving homes for shopping or any other social activities caused by lessening heating requirements at their homes. The temporal variations of the measured CO and SO_2 concentrations at chimneys are given as daily means in Fig. 6.

A correlation analysis using statistical software (IBM-SPSS 2016 Version 20 USA) was performed to identify whether any statistical relations with released CO and SO_2 concentrations have boilers combustion efficiencies. A correlation analysis was also performed to determine whether a combustion efficiency might affect the release of CO and SO_2 gases. As presented in Table 4, the combustion efficiency for this study had wide range from 26.6 to 98.6%. This wide range might also affect

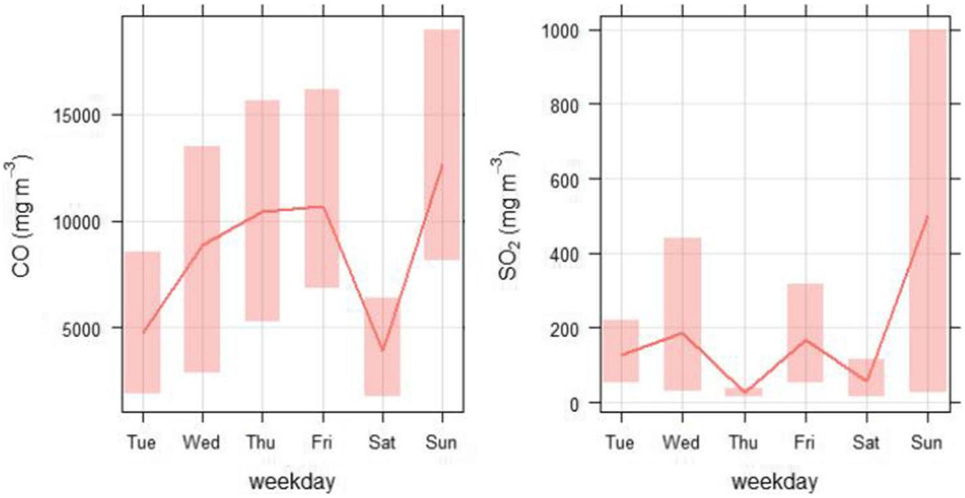


Fig. 6 Temporal variations of measured CO and SO_2 at the chimneys

Table 4 Measured parameters for the all sampled chimneys

Sampled chimneys	Gas exit temp (°C)	CO mg/Nm^3	Emission rate (g/s)	SO$_2$ mg/Nm^3	Emission rate (g/s)	Degree of sooty	O$_2$ %	Combustion efficiency (%)	Sampled chimneys	Gas exit temp (°C)	CO\ mg/Nm^3	Emission rate (g/s)	SO$_2$ mg/Nm^3	Emission rate (g/s)	Degree of sooty	O$_2$ %	Combustion efficiency (%)
1	117.9	3072	8.9	1139	1.45	1	17.3	66.2	70	63.2	2310	5.8	20	0.02	1	19.1	73.8
2	99.8	13,684	38.0	366	0.44	2	19.1	40.1	71	62.1	2393	6.0	17	0.02	4	18.8	77.7
3	117.2	2175	6.3	1873	2.38	6	16.6	73.1	72	70.4	25,115	64.3	28	0.03	2	19.6	55.8
4	136.3	5449	16.6	1598	2.13	1	17.9	52.5	73	29.6	14,651	33.0	82	0.08	2	20.5	80.3
5	20.2	10,480	22.9	31	0.03	2	19.7	88.1	74	41.4	11,523	27.0	19	0.02	1	18.7	88.3
6	70	10,999	28.1	122	0.14	2	18.4	75.5	75	34.6	9792	22.4	53	0.05	1	20.3	69.5
7	62.2	18,366	45.9	127	0.14	4	19.7	84.5	76	36.5	10,167	23.5	35	0.04	1	19.2	87.8
8	136.4	1562	4.8	152	0.20	1	17.1	77.9	77	43.6	28,880	68.1	12	0.01	2	17.8	96.3
9	217.6	23	0.1	140	0.22	2	12.6	76.2	78	31.1	35,645	80.8	15	0.01	5	18.3	95.6
10	125.2	8947	26.6	18	0.02	1	18.9	28.6	79	71.4	9230	23.7	21	0.02	1	17.9	74.8
11	36.8	7558	17.5	17	0.02	1	18.8	84.6	80	64.8	9397	23.7	161	0.18	2	19.7	58.1
12	80.3	2136	5.6	870	1.00	6	17.9	74.6	81	118	2811	8.2	62	0.08	4	17	70.4
13	60.4	3901	9.7	188	0.20	2	16.4	87.4	82	201.4	515	1.8	17	0.03	2	15.3	61.4
14	96.1	2055	5.7	92	0.11	3	18.9	50.1	83	56.1	4919	12.1	17	0.02	2	17.4	87.5
15	200.5	812	2.9	527	0.81	4	16.4	50.2	84	41.7	20,889	49.0	133	0.14	1	19.9	76.6
16	59.1	9172	22.7	23	0.02	1	19.4	59.5	85	71.5	3715	9.5	27	0.03	2	19	68.7
17	76.5	2278	5.9	15	0.02	5	18.5	66.7	86	28.7	15,544	35.0	25	0.02	2	19.5	92.6
18	59.9	7274	18.0	23	0.02	4	19.3	63.6	87	55.1	11,468	28.0	93	0.10	2	18.4	84.7
19	73.8	2877	7.4	13	0.01	4	18.1	73.1	88	154.3	8046	25.6	35	0.05	4	18.8	30.4
20	187.5	1179	4.0	72	0.11	7	12.5	75	89	44.9	8822	20.9	33	0.03	5	18.9	87.1
21	167.8	860	2.8	193	0.28	1	15.7	64.6	90	138.6	15,988	49.0	9	0.01	2	15.9	87.7
22	114.7	5129	14.8	184	0.23	2	16.6	84.6	91	48	9785	23.4	508	0.53	2	19.6	78.2
23	79.9	4057	10.7	66	0.08	4	14.6	87.3	92	57.1	10,497	25.8	988	1.06	2	14.5	93.5
24	46.5	1522	3.6	69	0.07	1	14.8	94.2	93	58.9	13,868	34.3	68	0.07	2	20	55.2
25	227.4	3781	14.1	169	0.28	1	17.4	26.6	94	26.9	38,917	87.0	82	0.08	1	20.5	89.4
26	159.3	371	1.2	427	0.60	2	19.1	66.7	95	27	34,979	78.2	78	0.08	1	20.5	89.5
27	54	20,484	49.9	15	0.02	1	18.5	74.2	96	43.7	28,500	67.3	33	0.03	2	19.8	77.3
28	205.1	1047	3.7	71	0.11	4	14.9	62	97	36.2	34,898	80.4	58	0.06	2	20.3	77.1
29	106.4	1511	4.3	22	0.03	4	15.9	78.1	98	41.6	11,535	27.0	106	0.11	2	19.6	85.2
30	48.6	19,903	47.7	107	0.11	1	19.2	65.2	99	58.5	11,360	28.1	1285	1.39	2	17.6	87.8
31	137	1158	3.5	57	0.08	2	17.5	69.6	100	34.8	16,501	37.9	478	0.48	2	20.3	75.6
32	140.6	635	2.0	469	0.63	1	15.3	72	101	46.1	16,219	38.6	24	0.02	2	19.4	77.8
33	115.9	1471	4.3	656	0.83	1	18.2	53.6	102	46.7	1788	4.3	801	083	2	19.2	80.3
34	147.8	999	3.1	3	0.00	4	9.7	85.1	103	21.7	35,735	78.5	22	0.02	2	19.3	95.3
35	99.42	3438	9.5	92	0.11	2	19.2	70.2	104	35.9	11,906	27.4	17	0.02	4	18.7	89.4
36	61.9	20,816	52.0	84–18	0.09	1	20.1	75.6	105	80	6555	17.2	8	0.01	1	16.6	83.4

Table 4 (continued)

Sampled chimneys	Gas exit temp (°C)	CO mg/Nm^3	Emission rate (g/s)	SO$_2$ mg/Nm^3	Emission rate (g/s)	Degree of sooty	O$_2$ %	Combustion efficiency (%)	Sampled chimneys	Gas exit temp (°C)	CO\ mg/Nm^3	Emission rate (g/s)	SO$_2$ mg/Nm^3	Emission rate (g/s)	Degree of sooty	O$_2$ %	Combustion efficiency (%)
37	72.1	3417	8.8	1032	1.16	4	18.9	77	106	67.1	96	0.2	7	0.01	1	16	88.9
38	89.2	9896	26.7	8	0.01	1	16.2	75.8	107	34.3	16,536	37.9	56	0.06	1	20.3	76.3
39	159.5	1620	5.2	12	0.02	1	17.8	42.9	108	80.8	1549	4.1	21	0.02	1	19.2	58.8
40	39.7	25,289	58.9	46	0.05	1	20.2	47.7	109	58.4	32,099	79.3	19	0.02	1	19	77.5
41	57	5062	12.5	10	0.01	1	17.3	82.5	110	65	7180	18.1	23	0.03	1	19.4	67.1
42	112.2	892	2.6	3	0.00	2	8.2	90.1	111	36.3	18,039	41.6	37	0.04	1	20	81.7
43	181.4	771	2.6	67	0.10	4	16.5	55.8	112	33.4	27,226	62.2	74	0.07	1	20.5	68.7
44	112.4	5755	16.5	43	0.05	1	18.2	57.6	113	81.9	3630	9.6	26	0.03	1	19.5	47.3
45	77.2	4143	10.8	51	0.06	1	16.9	82.8	114	49.4	6838	16.4	20	0.02	2	19.1	82.2
46	118.6	6061	17.7	122	0.16	3	17.5	65.8	115	42.6	15,450	36.3	31	0.03	2	19.7	86.5
47	139.8	3	0.0	1253	1.69	9	15.9	72.3	116	46	12,777	30.4	36	0.04	3	19.9	82.8
48	93	2592	7.1	1686	2.01	6	12.6	90.4	117	84.3	15,406	41.0	12	0.01	2	17.9	80
49	48.4	1228	2.9	75	0.08	1	20.5	NA	118	53.9	12,802	31.2	21	0.02	2	19.2	86.1
50	58.6	3052	7.5	36	0.04	1	19.9	NA	119	82.6	15,184	40.2	12	0.01	4	17.9	80.6
51	47.5	9599	22.9	73	0.08	1	20.4	NA	120	48.6	14,130	33.9	16	0.02	1	18.2	88.6
52	81.3	27,064	71.5	46	0.05	2	20.1	NA	121	38.7	21,354	49.6	47	0.05	2	20.2	91.3
53	36.8	873	2.0	14	0.01	8	18.3	NA	122	47.4	16,732	40.0	12	0.01	2	17.9	94.3
54	50.9	7559	18.2	16	0.02	1	18.6	82.9	123	42.2	10,056	23.6	18	0.02	2	18.9	94.2
55	42.7	10,729	25.2	28	0.03	1	19.6	78.1	124	62.8	11,879	29.7	52	0.06	2	20.2	48.7
56	41.2	17,881	41.9	32	0.03	1	19.2	NA	125	44.1	19,628	46.4	24	0.02	2	19.4	88.6
57	54	11,639	28.4	15	0.02	1	18.4	83.5	126	75.3	9865	25.6	23	0.03	2	19.3	65.6
58	62.4	25,690	64.2	102	0.11	1	20.6	NA	127	53.3	14,225	34.6	42	0.04	1	20.1	67.5
59	43.7	25,699	60.7	91	0.09	2	20.5	NA	128	33.4	26,783	61.2	17	0.02	2	18.8	98.6
60	47.3	5732	13.7	15	0.02	2	18.5	NA	129	627	10,284	69.0	19	0.06	1	19	79.5
61	58.4	26,267	64.9	23	0.02	1	19.3	NA	130	46.2	12,879	30.6	82	0.09	1	20.5	56.5
62	34.4	7694	17.6	15	0.02	2	18.5	92.7	131	49.4	8948	21.5	41	0.04	3	20.1	77.4
63	85.8	10,070	26.9	14	0.02	2	18.3	70.2	132	83.8	13,764	36.6	18	0.02	1	18.9	66.4
64	47	16,627	39.7	16	0.02	5	18.7	87.1	133	39.7	17,286	40.3	34	0.03	2	19.9	86
65	40.8	7719	18.1	20	0.02	2	19.1	84.8	134	127.7	10,491	31.3	20	0.03	1	19.1	87.9
66	78.8	6323	16.6	14	0.02	3	18.2	72.4	135	42.3	9067	21.3	17	0.02	2	18.8	89.8
67	31.4	16,863	38.3	24	0.02	2	19.4	89.7	136	64	7884	19.8	73	0.08	2	18.8	76.7
68	47.7	13,562	32.4	16	0.02	1	18.7	84.8	137	41.4	23,267	54.5	52	0.05	2	20.2	67.4
69	230.8	1337	5.0	6	0.01	2	14.9	57.3	138	26.3	4999	11.2	2360	2.30	3	20.3	93.7

burning process of coal and also emitted gas concentrations at the stacks. The Spearman correlation analysis indicated that the statistical correlation values of combustion efficiency were 0.31 with CO and − 0.24 for SO_2 concentrations at the at the 95% significance level. Therefore, it might be concluded that there was low correlation between combustion process and the emitted gas concentraions at the stacks. The emission rate (Q) was also calculated on the basis of the actual measured stack gas concentrations, temperature at the stack gas exit, the molecular weight of each measured gas, and stack flow rates. Table 4 shows the overall results related to the chimney sampling procedure.

On the basis of the results from the sampled chimneys, the descriptive statistics (mean, CI at the 95% level, and the minimum and maximum values) of all the measurements, including CO and SO_2 concentrations, emission rates, gas exit temperatures, the degree of sooty, and O_2 levels, are presented in Table 5.

According to the statistics of the measured parameters, the mean of CO concentrations was approximately 11,000 mg/m^3 with the variation of the mean from approximately 9500 to 12,500 mg/m^3 at 95% CI among the measured CO releases. Similarly, the mean of SO_2 concentrations was nearly 173 mg/m^3 with ranging of the mean from 108 to 240 mg/m^3 at 95% CI among the measured SO_2 releases. In addition to the statistics of gas concentrations, the mean gas exit temperature was approximately 80 °C, the degree of sooty was 2 in the Bacharach scale, the mean of oxygen content was roughly 18%, and the overall mean of combustion efficiency was approximately 70% for the chimney samplings. On the basis of the measured gas concentrations, the released CO and SO_2 concentrations did not exceed the regulatory limits during the measurement periods.

AERMOD dispersion modeling outputs

According to the AERMOD analysis results, the dispersion maps of the estimated CO and SO_2 ground level concentrations were created for the maximum means of two different periods, namely, daily (24 h) for the CO and hourly for the SO_2 levels that were estimated from all specified receptors. The daily limits for ambient CO concentrations and the periodic limits of the heating season for ambient SO_2 concentrations have been officially described in the National Regulations (RAQAM 2008). Figure 7 illustrates the AERMOD analysis steps in three different layers.

Stage ***a*** refers to the first layer of the dispersion map that includes only elevation data of the study area, stage ***b*** illustrates all of the map points of the sampled chimneys that are subject to the sampling during the heating season, and stage ***c*** represents the dispersion map of the airborne pollutants released by those chimneys in the study area.

In this modeling study, the AERMOD also estimates the maximum daily modeled concentration by taking an average of 24 h (from 0 to 23 h) estimation for every assigned receptor and then reports the maximum daily concentration. The maximum monthly modeled concentration is calculated by taking an average of all daily modeled concentrations for each receptor for a specific month, and the highest monthly mean-modeled concentration among the assigned receptor is then used for the maximum monthly modeled concentration.

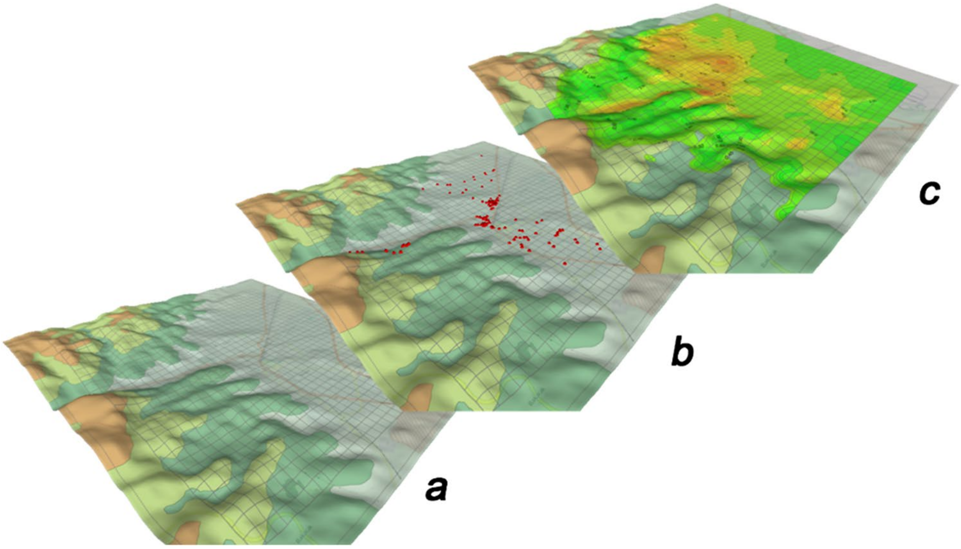
According to the AERMOD modeling result, the predicted highest daily mean CO concentration for the model was 41,533 µg/m^3 on February 29th at midnight for the downtown area and exceeded the official limits of 10,000 µg/m^3 specified in the national regulations (RAQAM 2008; NAQI 2016). Furthermore, a total of 20 apartment chimneys, ranging from 37,921 µg/m^3 to 10,058 µg/m^3, also exceeded the official limits in the heating season. The rest of the apartments had lower CO concentrations than the limits. The modeled CO dispersion showed that the higher CO concentrations were deposited mainly over the south-west of the downtown area, as presented in Fig. 8. This situation somehow made sense that CO emissions were released continually from the chimneys and did not spread out toward further locations by the downwind during the heating season and that the wind speed and direction had less effect on

Table 5 Descriptive statistics results of the chimneys

Measured parameters	Mean	CI[*]		Minimum	Maximum
		Lower Bound	Upper Bound		
CO, mg.m^{-3}	10,891.9	9358.6	12,425.3	3	38,917
CO emission rates, g/s	26.9	23.3	30.5	–	87
SO_2, mg.m^{-3}	172.9	107.8	238.1	3	2360
SO_2 emission rates, g/s	0.2	0.12	0.28	–	2.4
Gas exit temp., °C	79.8	68.6	90.9	20.2	627
Degree of sooty	2	2	3	1	9
O_2, %	18.3	17.8	18.7	8.2	20.6
Combustion efficiency,%	69.7	65.7	73.8	26.6	98.6

[*] Confidence Intervals (CI) at 95% significance level

Fig. 7 The AERMOD modeling process in different layers



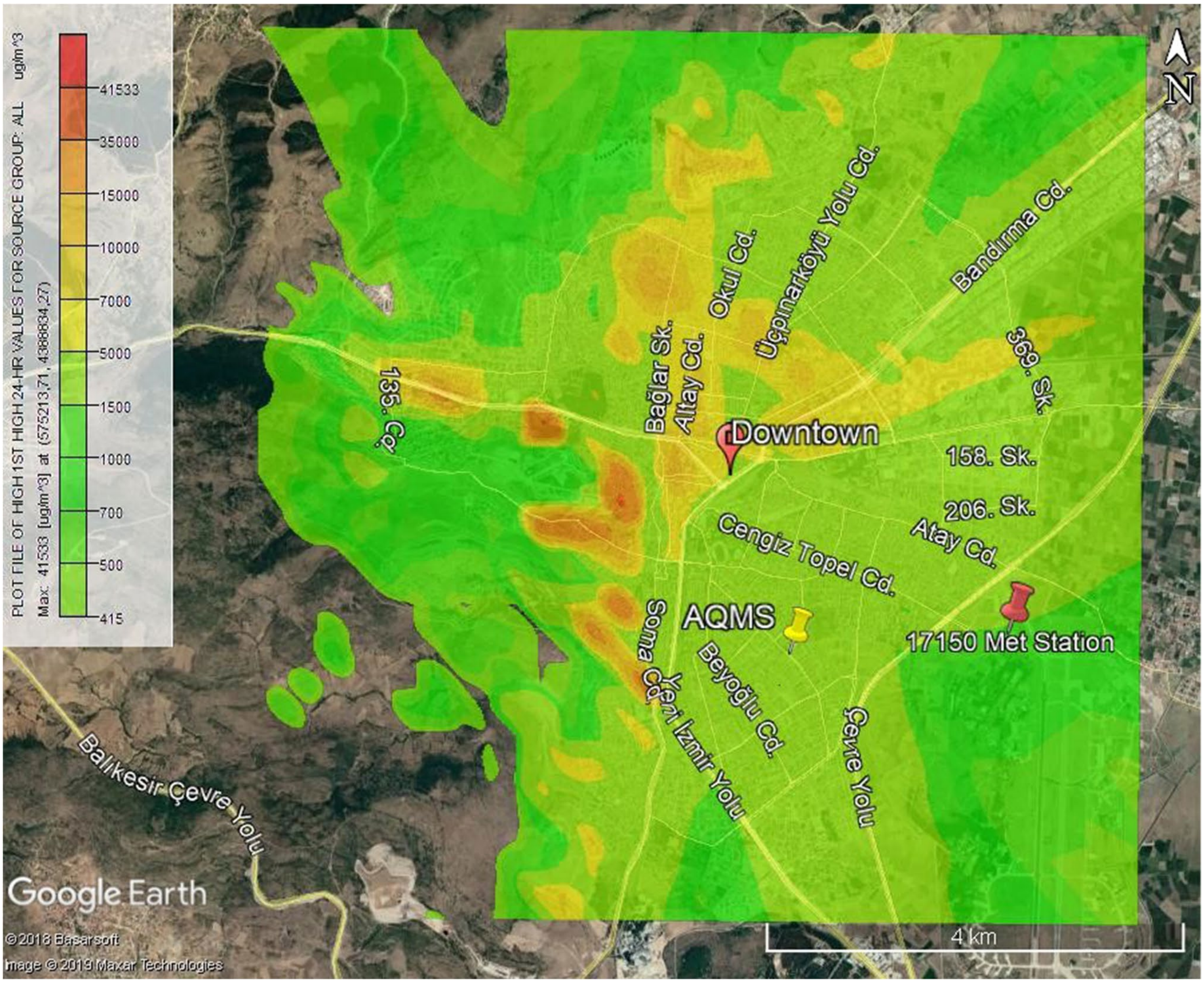


Fig. 8 The daily (24 h) maximum mean of CO dispersions over the downtown

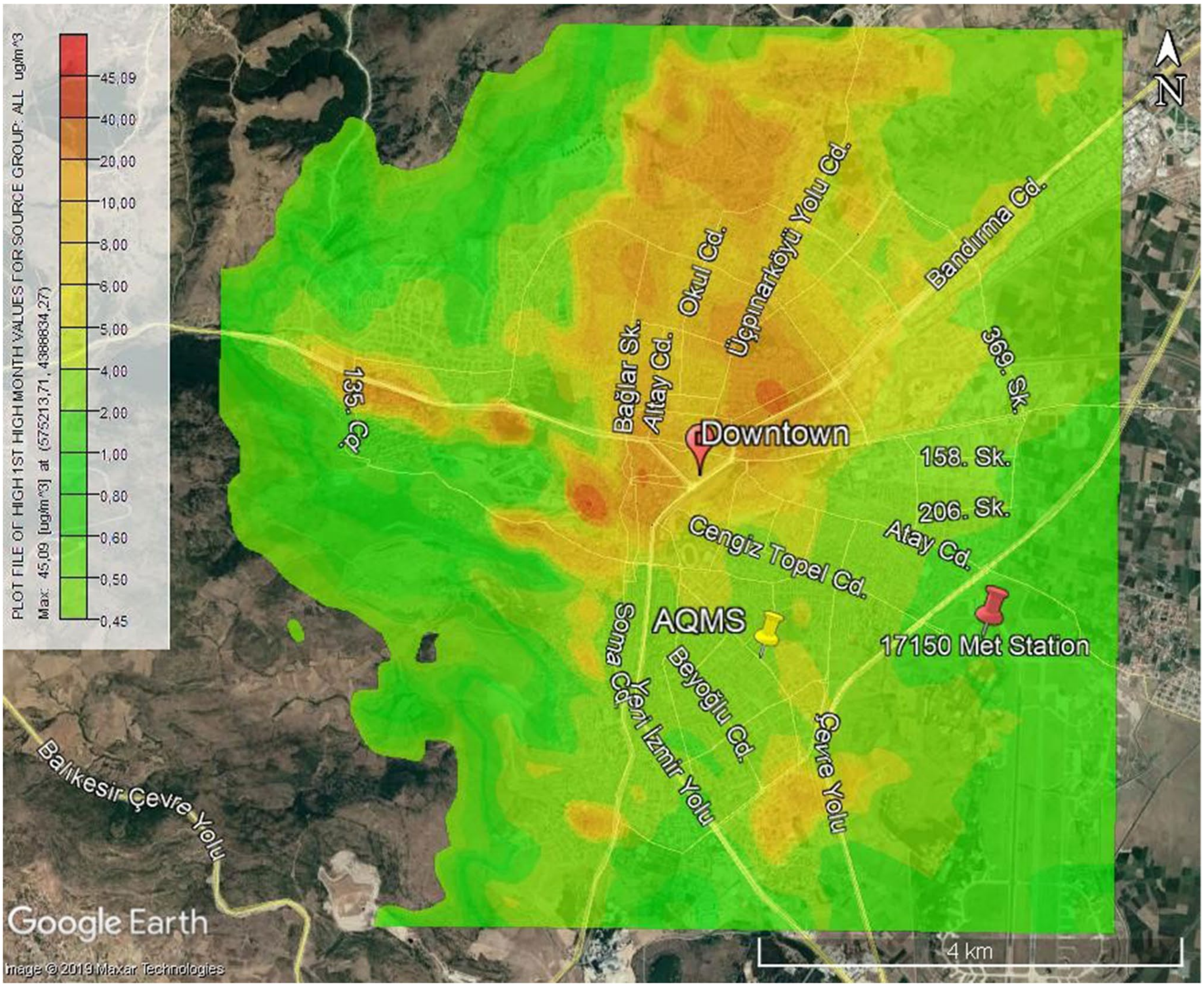


Fig. 9 The periodic-monthly SO_2 dispersions over the downtown

the CO dispersion because the topographical characteristic of downtown settlement is defined as semi-bowl-shaped in the north-western and south-western regions. It might be concluded that the ground-level CO concentrations were accumulated in the foothills of the south-western heights in the downtown.

According to AERMOD modeling outputs, the dispersion of SO_2 concentrations was also evaluated in this study. The regulation set the limit at 20 μg/m^3 for the maximum mean of monthly SO_2 concentrations in the heating season (RAQAM 2008; NAQI 2016). Three chimneys exceeded the ambient SO_2 limit concentrations from the model outputs. In detail, the highest predicted monthly SO_2 concentration was 45.1 μg/m^3 on February 29th at midnight in the heating season. This modeling prediction represents 2 months of periods that occurred during the heating seasons. Instead of estimating the ground-level SO_2 concentrations at the momentarily shorter period, for example, hourly or daily, the more meaningful scale might be the most extended time period that represents the heating season naturally. Therefore, the highest predicted periodic SO_2 concentration for the heating season was estimated in the range of 45.1 μg/m^3, 27.2 μg/m^3, and 23 μg/m^3, which indicated that only three chimneys exceeded the legal limits for the downtown area. Figure 9 depicts the AERMOD dispersion map for periodic SO_2 concentrations, including a few hotspots at the central part of the downtown.

As stated in the Sampling Site and Procedure section, according to the national regulation (RAQAM 2008), the regular heating season officially includes months from October through March for the study. Therefore, SO_2 emissions from the stacks have been modeled based on monthly averages including February and March, while CO emissions have been modeled in daily averages. In addition to the national regulation, long-time scale such as monthly or annual period have been previously used for ambient SO_2 modeling studies (Zou et al 2010; Sari and Bayram 2013; Tuygun et al 2017). Zou et al. (2010) and Gibson et al. (2013) also concluded that SO_2 modeling results based on long-time scales provide more reliable results than momentarily shorter time scales. For this reason, differences might be seen in the comparison of the points where the highest values are seen and also the areas where the predicted pollutant is effective in the dispersion maps of SO_2 and CO concentrations. Therefore, predicted SO_2 dispersions have been observed in a wider area and spread out more intensely in the northern part of the downtown. As illustrated in Fig. 9, the highest predicted SO_2 levels are seen in the both south-west and north-east of the central district of the downtown. Similar finding has been also reported by Sari and Bayram (2013).

The entire study area that represented all the chimneys was covered up by local SO_2 concentrations. The predicted SO_2 concentrations varied from less than 1 $\mu g/m^3$ to 18.8 $\mu g/m^3$ that remained within the allowed limits during the study period. Other studies have also reported the predicted SO_2 concentrations in previous studies. For instance, Economopoulos (1997) reported that the SO_2 level was 23.3 $\mu g/m^3$ in Athens, Greece, and Gibson et al. (2013) also confirmed that the predicted SO_2 concentrations were 4.9 $\mu g/m^3$ in Halifax, Canada and 8.7 $\mu g/m^3$ in Sidney, Australia.

Similarly, Meng et al. (2018) validated that the modeled SO_2 level was 10 $\mu g/m^3$ in Baoding, China. The modeled SO_2 levels reflected similar results with the SO_2 levels of other cities. Naturally, some cities had lesser SO_2 levels due to divergent source contributions and might be using a different type of fuel for heating purposes.

Consequently, when the AERMOD performance has been evaluated, air quality modeling results are satisfied for meteorological and air pollutant data. Other studies have reported that the AERMOD performs a good agreement between observed and modeled results (Perry et al. 2004; Dresser and Huizer 2011; Gibson et al. 2013; Abril et al. 2016). Therefore, the outputs of the AERMOD model used in the air quality modeling might have been valid for the study.

Conclusions

This study aimed to analyze the air quality levels that were caused by coals utilizing residential heating activities in a mid-size city of the south Marmara Region in Turkey. The city has been using coal for residential heating, while natural gas heating has continuously spread out throughout the city.

A total of 138 chimneys of heating boilers were sampled for CO and SO_2 pollutants, and the dispersion maps of those pollutants were prepared to visualize where the pollutants spatially spread out over the downtown area. The temporal variations of those pollutants occurred as expected that both pollutants had higher levels during the heating season or throughout the heating season due to space heating purposes. The similar results have been also reported for the same area by llten and Selici (2008), Tecer (2009), and Mutlu (2019) that air pollutants have shown increasing trend during the cold seasons. For the spatial analysis, the dispersion modeling asserted that the ambient CO and ambient SO_2 levels exceeded the regulation limits at the old settlements of the downtown where coals were still widely used during the heating season. Although, previous studies (Yun et al. 2020; Zhang et al. 2020) have recently suggested that PM pollutions due to coal-based residential heating had a significant contribution to the air quality and public health, also the mostly ambient CO levels due to its toxicity and the partly ambient SO_2 due to residential heating purposes should be still considered serious environmental issues for the cities.

Excessive ambient CO levels might be related to the combustion efficiency of older boiler systems in the study area. The mean value of the combustion efficiencies from the sampled systems was approximately 69%, with the lowest rate of almost 26% in the study area. Therefore, it is an essential point that the periodic maintenance or existing faults of the boilers must be followed throughout the heating season.

The ambient concentrations of SO_2 are directly related to the quality of used coal and its sulfur content; therefore, inspections of commercial coals must be regularly performed whether a fuel product meets the legal standards during a heating season. The transition to natural gas-operated heating systems requires more time and needs great infrastructure investment, and it cannot be set up immediately. Consequently, the use of coal for heating purposes remains a serious environmental concern during heating seasons when residential heating activities are compulsorily increased.

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Author's contributions The corresponding author conceived and designed the study. All data including air quality and meteorological have been analyzed in this study by the corresponding author and dispersion modeling steps have been carried out by the authors. All authors read and approved the final manuscript.

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Availability of data and materials The datasets analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors have declared no conflict of interest.

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