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Spatial-temporal variations in regional ambient sulfur dioxide concentration and source-contribution analysis: A dispersion modeling approach

Bin Zou [a,b,c], J. Gaines Wilson [d,*], F. Benjamin Zhan [b], Yongnian Zeng [a], Kongjiang Wu [a]

[a] School of Geoscience and Info-Physics, Central South University, Changsha, Hunan 410086, China
[b] Texas Center for Geographic Information Science, Department of Geography, Texas State University, San Marcos, TX 78666, USA
[c] School of Resource and Environmental Science, Wuhan University, Hubei 430079, China
[d] Department of Chemistry and Environmental Sciences, University of Texas at Brownsville, Brownsville, TX 78520, USA

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ABSTRACT

An understanding of the complexity of spatial-temporal variations in regional air quality and its respective source contributors is one of the priority research areas due to the adverse effects of air pollution on human health and the environment. In this paper, we integrate air dispersion modeling and Geographic Information System (GIS) based spatial analysis methods to characterize regional ambient air quality at a relatively fine geographical scale (1 km × 1 km) while ascertaining source contributors. The temporal variation analysis shows that sulfur dioxide (SO_2) pollution in Dallas County, Texas did not consistently increase or decrease from 1996 to 2002. The lowest and highest mean levels of annual SO_2 concentrations at all the receptors ($n = 2000$) were 0.39 $\mu g\,m^{-3}$ and 2.32 $\mu g\,m^{-3}$ in 2001 and 2002, respectively. Meanwhile, analysis results suggest that the annual SO_2 concentrations in a small part of Dallas County slightly declined with the highest value of −1.00 $\mu g\,m^{-3}$ over the 1996–2002 period, while most of the county experienced increased SO_2 concentration levels from 0.00 to 0.25 $\mu g\,m^{-3}$. In addition, the source apportionment analysis demonstrated that the variations in total annual SO_2 concentrations in Dallas County from 1996 to 2002 were significantly different from those by source classification. That is, compared to industrial emission sources, on-road vehicle emission sources caused variations in annual SO_2 concentrations with relatively larger extents (power of determinant = 0.42). However, extreme variations in concentrations were due to industrial emission sources (3.45% vs. 0.00%). Based on these observations, it can be concluded that the combination of air dispersion modeling and GIS-based spatial analysis shows promise to overcome the drawbacks of sparse intraurban air quality monitoring in characterizing the spatial-temporal micro-variations in regional ambient air quality and ascertaining roles of source contributors over long-term periods.

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1. Introduction

Ambient sulfur dioxide (SO_2) is regulated by the U.S. Environmental Protection Agency (U.S. EPA) as one of the six criteria air pollutants. Due to its potentially adverse effects on the human health (e.g. *low birth weight*) and the environment (e.g. *acid deposition*) (Zou et al., 2009a; U.S. EPA, 2008a), SO_2 has been given high priority in research activities in the fields of air pollution and environmental health research. Recent epidemiological findings have also confirmed the associations between low birth weight and maternal SO_2 exposure even at exceptionally low exposure levels (Bell et al., 2007). Therefore, it is critical to quantitatively evaluate regional ambient SO_2 exposure levels at fine scales within urban areas and ascertain source contributors in order to make relevant policy decisions for reducing health and environmental risks caused by deposition and exposure.

Current monitoring data from the U.S. EPA (2008b) have described the total amount of source-specific SO_2 emissions at different administrative unit levels (i.e. *national, state, county*). This data can help us understand the variation tendency of a specific contributor (e.g. *industrial emissions, on-road vehicle emissions*) over a certain geographic area over time. However, identifying the geographic distribution of SO_2 concentrations and their spatial-temporal dynamic variations based on statistic SO_2 emission data presents a considerable challenge. In addition, bias, even incorrect conclusions, may be introduced when evaluating air quality of

* Corresponding author. Tel.: +1 (956) 882 6646; fax: +1 (956) 882 6692.
E-mail address: jeff.wilson@utb.edu (J.G. Wilson).

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a location by only considering the emission data of a source around it, especially in an area without a sufficient number of air quality monitoring sites (Zou, 2010). For example, an area with high SO_2 concentrations might result from long-range transport although the total amount of emissions in that area may be low (Li, 2010).

Studies examining spatial patterns and or temporal tendencies of air pollution concentrations have been extensively reported in previous publications over the past decade. A key weakness of many of these studies is the fact that the spatial resolution of air quality monitoring sites in any particular study area was not sufficiently dense to characterize the true nature of small-area variability (Tayanc, 2000; Hung et al., 2005; Cyrys et al., 2008; Juneng et al., 2009; Pang et al., 2009). Further, it has been suggested that the coarse resolution at which these monitoring networks often operate may introduce additional error when evaluating health outcomes (Wilson et al., 2005).

Alternatively, air dispersion modeling methods for simulating air pollution concentrations by discretely located receptors (Holmes and Morawska, 2006) at different spatial resolutions have been rapidly developing over recent years. Orloff et al. (2006) successfully simulated the ambient hydrogen cyanide concentrations at five discrete locations near a gold heap leach field with the AMS/EPA Regulatory Model (AERMOD). Stein et al. (2007) simulated the spatial distribution of benzene concentrations with a grid size of 1 km × 1 km by using a hybrid modeling approach. In our earlier work (Zou et al., 2009b), we explored the spatial pattern of SO_2 concentrations in the Dallas area of Texas at a fixed time with AERMOD. These studies demonstrate the feasibility of using air dispersion models to disclose the spatial-temporal variations in air pollutant concentrations at a relatively higher resolution, addressing a critical problem in air quality monitoring and modeling.

In addition, recent studies have also shown that air dispersion modeling can provide a way to identify the relative contributions of different types of emission sources to the total ambient air pollution concentrations (Islam, 1999; Crabbe et al., 2000; Kuhlwein et al., 2002; Bullock and Breheme, 2002; Lin et al., 2004; Peace et al., 2006; Bell et al., 2007; Cheng et al., 2007). However, these studies either investigated the emission source contribution to urban air quality at a discrete receptor site (Lin et al., 2004) or had an issue with large-area estimated contributions of various emission sources to the total ambient air (Wang et al., 2005; Peace et al., 2006; Cheng et al., 2007). As a result, these spatial resolutions are too coarse for some applications. There exists little analysis of source-specific emission sources at higher resolution (e.g. 1 km × 1 km), let alone the examination of emission source contribution to air quality variations over longer-term periods of exposure.

In order to address the research gap described above, this study aims to develop a method to characterize ambient air pollution concentrations at a fine geographical scale and ascertain its source contributors. To achieve this goal, we first simulate ambient daily and annual SO_2 pollution concentrations across Dallas County from 1996 to 2002 corresponding to three source emissions (industrial emission sources only, on-road vehicle emission sources only, and industrial & on-road vehicle emission sources combined) using an air dispersion model. Next, we analyze the seasonal and yearly variations of simulated SO_2 concentrations and detect their spatial variation patterns over the entire study period at a resolution of 1 km × 1 km using GIS spatial analysis methods (i.e. Kriging, overlay). Finally, the contributions of different types of emission sources to the total SO_2 concentrations are determined with a series of statistical measures. The results visually demonstrate the dynamic variation patterns of ambient SO_2 pollution and further explain how these variation patterns would be influenced by emission sources over a longer time period. Fig. 1 illustrates the overall study design.

2. Study area

Dallas County is located in the Dallas-Fort Worth metropolitan area in Texas, United States (Fig. 2). The county has an area of 2352 km^2 with 1683 census block group polygons, and had a total population of approximately 2,218,899 in 2000 (U.S. Census Bureau, 2008). This area has attracted considerable attention due to concerns about poor air quality (McDade et al., 2000). The elevation of Dallas County is about 100–262 m above sea level (ASL) with a mean elevation of 155.7 m. The area is dominated by a humid subtropical climate with humid springs and hot summers. The annual mean precipitation is around 881 mm while most of the rainfalls occur in May with monthly average precipitation of 130 mm. The lowest and highest average monthly temperatures are 2 °C in January and 35.5 °C in August. The annual mean temperature is 18.7 °C. While S-SE is the predominant wind direction in Dallas County, the average annual wind speed in the area is 5.8 m s^{-1}.

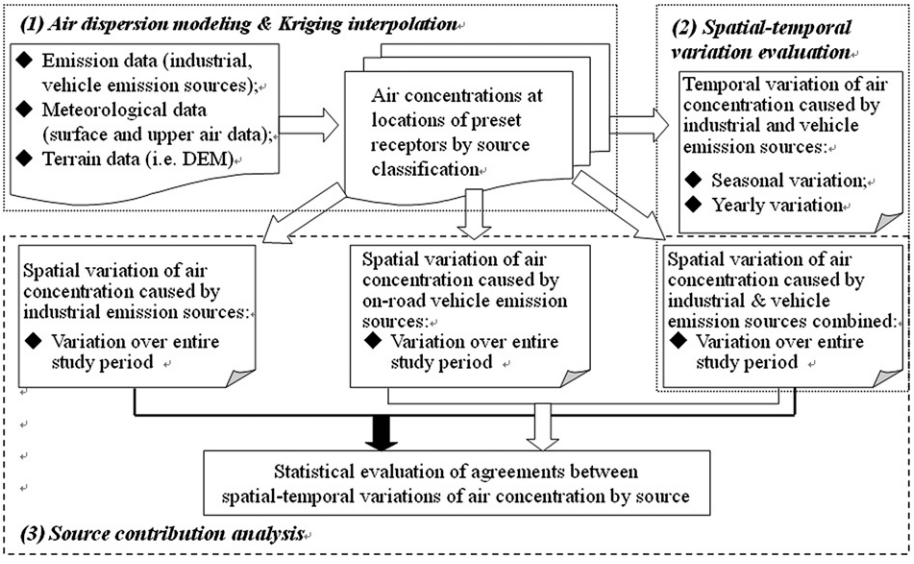

Fig. 1. An overview of the study design.

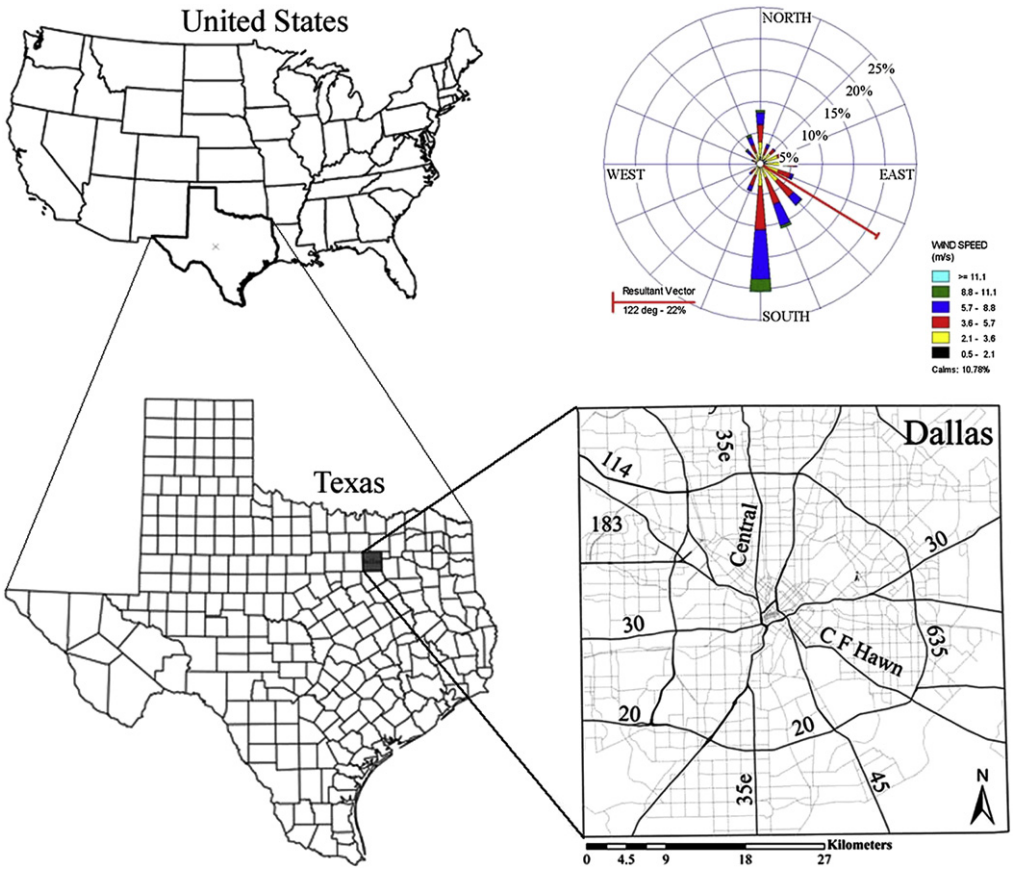

Fig. 2. The location of study area with wind rose diagram for 2002.

3. Materials and methods

3.1. Data

This study utilizes emission, meteorological, and terrain data. The SO_2 emission data in Dallas County for years 1996 and 2000 were directly extracted from the 1999 and 2002 National Emission Inventory (NEI) databases (U.S. EPA, 2008b). Meanwhile, the SO_2 emission data in Dallas County for other years (years 1997–1999, 2001–2002) were developed based on the 1999 and 2002 NEI and the yearly pollutant facilities summary reports provided by the U.S. EPA. The yearly SO_2 emissions were categorized as industrial emissions and on-road vehicle emissions (e.g. highways, major local streets). Meteorological data included the near-surface measurement and upper-air sounding data from 1996 to 2002, which were obtained from the National Climatic Data Center (NCDC, 2008) and the National Oceanic and Atmospheric Administration (NOAA, 2008), respectively. In addition, two 1° U.S. Geological Survey (USGS) digital elevation datasets at a scale of 1: 250,000 were used to characterize the topographic relief of the study area. For a detailed description of emission data, meteorological data and terrain data, please refer to our previous work (Zou et al., 2010).

3.2. Air dispersion modeling

Air dispersion modeling in this study was implemented by AERMOD, which has been recognized as a reliable model for estimating ambient concentrations of air pollutants (Venkatram et al., 2004; Perry et al., 2005). The settings and parameters for AERMOD were determined and processed with those (e.g. albedo, bowen ratio, and surface roughness) as described in our earlier work (Zou et al., 2010). After these preparations, the AERMOD model was run at daily and annual temporal scales for simulating the SO_2 concentrations at each receptor from different emission sources in Dallas County from 1996 to 2002. The validation process of AERMOD performance can also be found in our earlier work (Zou et al., 2010).

3.3. GIS-based spatial analysis

To understand the spatial pattern of ambient SO_2 concentrations in Dallas County and its yearly variations over the study period, spatial interpolation and overlay analysis were performed. For the spatial interpolation operation, ordinary kriging was used to produce the continuous annual SO_2 concentration maps from 1996 to 2002 with a resolution of 1 km × 1 km based on the SO_2 concentrations at the locations of preset receptors from air dispersion modeling. Ordinary kriging has been shown to be the best linear unbiased estimator (BLUE) (Jerrett et al., 2005). Next, overlay analysis, a raster calculation, was performed using functions in the ArcGIS spatial analysis module (version 9.3). As a result, we generated spatial variations of SO_2 concentrations at a scale of 1 km in Dallas County over the entire study period (1996–2002). The spatial interpolation and overlay analysis were separately implemented to ascertain the source contribution of different types of emission sources to the total SO_2 concentrations for each of the three emission scenarios.

3.4. Statistical evaluation

To detect how different emission sources would affect the annual SO_2 concentrations in Dallas County from 1996 to 2002, we

Table 1
Descriptive statistics of seasonal and annual SO_2 concentrations in Dallas County from 1996 to 2002.

Season	Statistical measure	1996	1997	1998	1999	2000	2001	2002
Spring	Min ($\mu g\,m^{-3}$)	0.06	0.10	0.10	0.10	0.02	0.03	0.08
	Max ($\mu g\,m^{-3}$)	2.80	4.51	3.73	260.35	117.88	123.23	95.86
	SD	0.33	0.59	0.58	11.85	3.76	8.57	10.33
Summer	Min ($\mu g\,m^{-3}$)	0.06	0.14	0.10	0.10	0.02	0.02	0.06
	Max ($\mu g\,m^{-3}$)	2.66	5.77	4.06	288.51	80.03	75.42	74.00
	SD	0.32	0.71	0.62	13.16	3.65	3.44	9.16
Autumn	Min ($\mu g\,m^{-3}$)	0.08	0.19	0.13	0.12	0.03	0.03	0.16
	Max ($\mu g\,m^{-3}$)	4.09	7.91	4.90	342.56	104.98	100.56	54.99
	SD	0.51	1.02	0.80	15.63	4.79	4.59	8.22
Winter	Min ($\mu g\,m^{-3}$)	0.09	0.19	0.15	0.09	0.04	0.03	0.17
	Max ($\mu g\,m^{-3}$)	3.77	6.37	4.87	310.39	105.74	99.70	74.14
	SD	0.47	0.86	0.80	14.16	4.82	4.55	9.53
	Annual ($\mu g\,m^{-3}$)	0.47	0.93	0.80	1.30	0.40	0.39	2.32

mapped the variation of source-specific annual SO_2 concentrations. Next, based on the classification standards for industrial & on-road vehicle emission sources combined-based variation map of annual SO_2 concentration (i.e. −1.00 to 0.00; 0.01–0.25; 0.26–1.00; 1.01–3.60; 3.61–8.50; 8.51–15.00; >15.00, unit: $\mu g\,m^{-3}$) utilizing 'natural break' points, all the spatial variation maps of annual SO_2 concentration by source classification were sorted into seven categories. The area proportion, mean, and SD of the annual SO_2 concentration in each source category were compared to initially detect the similarity of the variations of annual SO_2 concentration at different levels caused by different types of emission sources. As an additional quantitative measure of inter-source correlation, we employed a method developed by Wang et al. (2010) called *factor-detector*. This approach evaluates whether a geographical stratum (x_i) is responsible for an observed spatial pattern (y). The result of the *factor-detector* analysis is defined as PDx_iy. x_i is the full determinant of y if the value of PDx_iy is '1' while '$PDx_iy = 0$' means x_i has no impact on y. In this process, the source-specific variations in SO_2 concentrations were again reclassified into seven categories at 'natural break' points.

4. Results

4.1. Temporal variation of ambient SO_2 concentrations

Based on the simulated results from AERMOD, the mean, as well as the SD of seasonal (average) and annual SO_2 concentrations at all the locations of preset receptors were calculated to represent the levels of SO_2 pollution for the entire Dallas County in each year over the entire study period. The minimum and maximum values were also correspondingly extracted out among all these seasonal and annual SO_2 concentrations at all locations of preset receptors to indicate the variations between extreme upper and lower SO_2 concentrations in Dallas County from 1996 to 2002. The results are presented in Table 1 and Fig. 3.

As shown in Table 1, the mean annual SO_2 concentrations at all the locations of preset receptors from 1996 to 2002 fluctuated between 0.39 $\mu g\,m^{-3}$ and 2.32 $\mu g\,m^{-3}$ while the highest and lowest concentrations emerged in 2002 and 2001, respectively. This variation tendency was similar to those season-specific variations over the entire study period with the exception of the variation between spring in 2000 and 2001. Fig. 3 indicates that, among all the seasons in the study period, autumn and winter were the seasons with relatively higher mean annual SO_2 concentrations at all the locations of preset receptors in most of the study years while the relative lower concentrations mostly emerged in spring and summer. These results were only partly reversed in 1999 and 2001. In these two years, the mean of seasonal SO_2 concentrations at all the locations of preset receptors in the spring of 1999 was slightly higher than the mean in winter (i.e. 1.38 $\mu g\,m^{-3}$ vs. 1.26 $\mu g\,m^{-3}$). This seasonal phenomenon was again observed in 2002, with means of seasonal SO_2 concentrations at all the locations of preset receptors at 0.43 $\mu g\,m^{-3}$ and 0.41 $\mu g\,m^{-3}$ in the spring and autumn of 2001, respectively.

In addition, whereas the lowest annual SO_2 concentrations at all the locations of preset receptors in Dallas County did not show significant variations over the study period, the highest values in Table 1 demonstrate that there was substantial variability in maximum annual SO_2 concentrations around 1999. The highest levels of annual SO_2 concentrations in Dallas County after 1999 were elevated relative to years prior to 1999. The SD values in Table 1 also show that spatial variations between the seasonal SO_2 concentrations at the local locations of Dallas County gradually increased from 1996 to 1999 (e.g. with a change from 0.33 to 11.85 from the spring of 1996 to 1999), while these values appeared to randomly fluctuate from 1999 to 2002 (e.g. with 11.85, 3.76, 8.57, and 10.33 from the spring of 1999 to 2002).

4.2. Spatial variation of ambient SO_2 concentrations

Fig. 4 illustrates the spatial distributions of simulated annual SO_2 concentrations with a grid size of 1 km × 1 km in Dallas County and their variations from 1996 to 2002. As shown in Fig. 4(a), most areas of Dallas County in 1996 experienced relatively lower levels of SO_2 concentrations (e.g. ranging from 0.00 to 0.40 $\mu g\,m^{-3}$; 0.41 to 0.65 $\mu g\,m^{-3}$). After 1997, we see an increase in levels of SO_2 concentrations (e.g. ranging from 0.92 to 1.29 $\mu g\,m^{-3}$; 1.30 to 2.42 $\mu g\,m^{-3}$). The areas with extremely elevated annual SO_2

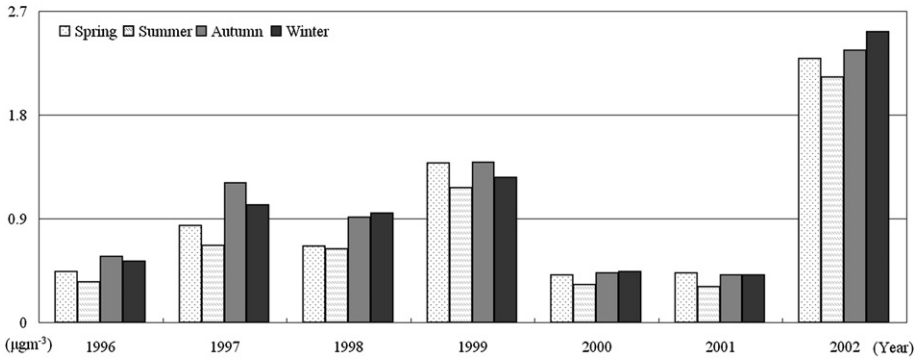

Fig. 3. Seasonal SO_2 concentrations in Dallas County from 1996 to 2002.

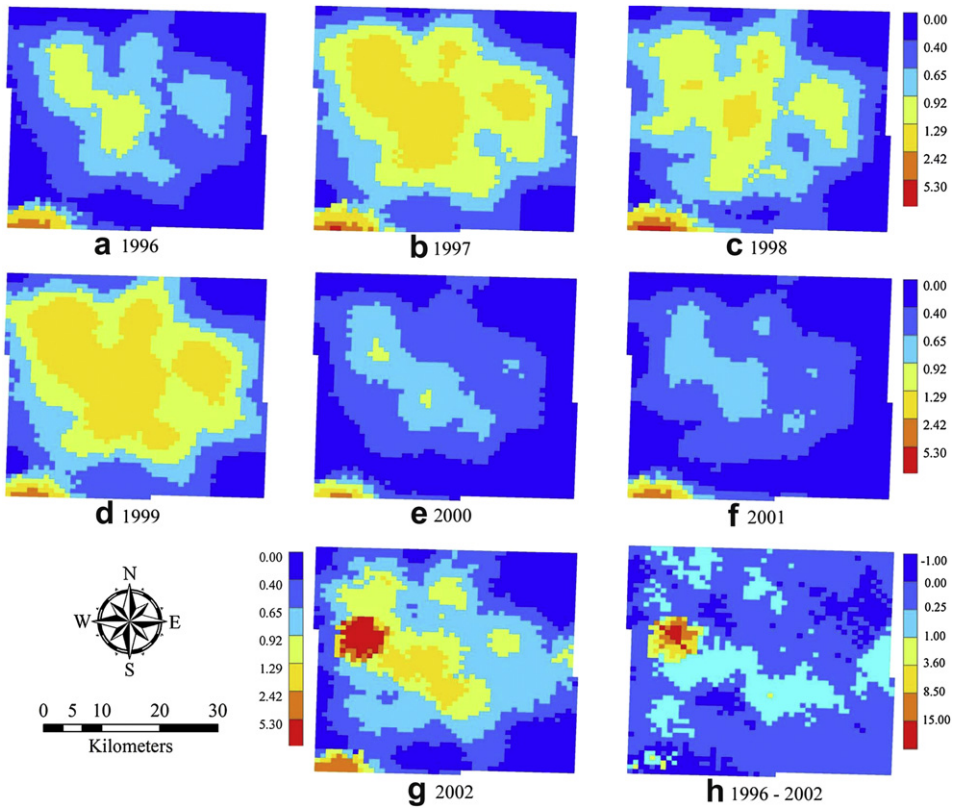

Fig. 4. Spatial distributions of simulated annual SO_2 concentrations in Dallas County and their variations, 1996–2002.

concentrations in Dallas County (i.e. greater than 5.30 $\mu g\,m^{-3}$) from 1996 to 1999 also appeared to be extending, especially in 1997 (Fig. 4(b)) and 1998 (Fig. 4(c)). However, the trend of increasing annual SO_2 concentrations in Dallas County stalled in 1999 and consistently fluctuated after 1999 (Fig. 4(d)). In 2000 (Fig. 4(e)) and 2001 (Fig. 4(f)), the original areas with relatively high levels of SO_2 concentrations (e.g. ranging from 0.92 to 1.29 $\mu g\,m^{-3}$; 1.30 to 2.42 $\mu g\,m^{-3}$) disappeared although the areas with SO_2 concentrations ranging from 2.42 to 5.30 $\mu g\,m^{-3}$ remained high. In contrast to 2000 and 2001, the geographic extent of the areas with higher SO_2 concentrations ranging from 2.42 to 5.30 $\mu g\,m^{-3}$ greatly increased in 2002, while a similar pattern was observed in other areas of lower concentration. Between 1996 and 2002, the areas with the highest concentration moved from the southwest corner of Dallas County to the northwest area of the County (Fig. 4).

In addition, although results in Fig. 4(h) indicate that the annual SO_2 concentrations in a small part of Dallas County declined slightly with an extreme value of $-1.00\,\mu g\,m^{-3}$ from 1996 to 2002, the annual SO_2 concentrations in most areas of Dallas County still increased. Among them, the areas with variation in increasing annual SO_2 concentrations ranging from 0.00 to 0.25 $\mu g\,m^{-3}$ ranked first, while the area of those districts ranging from 0.25 to 1.00 $\mu g\,m^{-3}$ and 1.00 to 15.00 $\mu g\,m^{-3}$ ranked second and third, respectively. The district with the greatest variation of annual SO_2 concentrations was found around the center of northwestern part of Dallas County, with values larger than 15.00 $\mu g\,m^{-3}$. The areas with relatively higher variations of annual SO_2 concentrations in Dallas County were largely along a northwest to southeast band, which is the predominant wind direction (i.e. 122 degree) as shown in Fig. 2.

4.3. Agreements on the spatial-temporal variations of ambient SO_2 concentrations by source classification

Fig. 5 illustrates the spatial distribution of source-specific SO_2 concentrations in Dallas County from 1996 to 2002 on a 1 km grid.

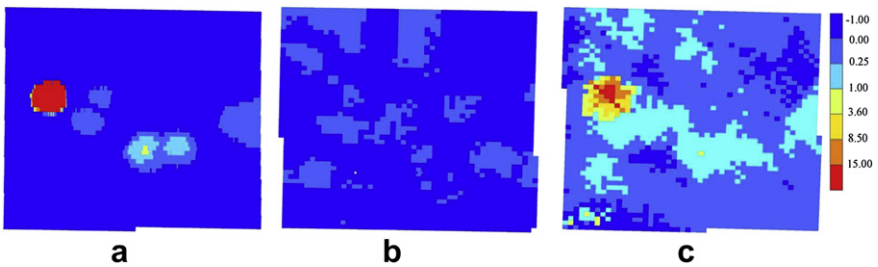

Fig. 5. Spatial-temporal variations in annual SO_2 concentrations in Dallas County from 1996 to 2002, including industrial emission sources (a), on-road vehicle emission sources (b), and industrial & on-road vehicle emission sources combined (c).

It is clear that the variations of annual SO_2 concentrations caused by industrial & on-road vehicle emission sources combined (Fig. 5(c)) are significantly different from those caused by industrial (Fig. 5(a)) or on-road vehicle emission sources alone (Fig. 5(b)). Fig. 5(a) shows that, while industrial emission sources caused the variations in high-concentration in areas, such as the northwestern part of Dallas County, they also resulted in reduced concentrations in other areas of Dallas County. The pattern in Fig. 5(b) shows that variations in concentrations from on-road vehicle emission sources increased over a large area of Dallas County from 1996 to 2002.

Table 2 further illustrates the results of variations in annual SO_2 concentration by emission source in Dallas County from 1996 to 2002. For low SO_2 concentration variability (i.e. levels 1–3), the total area exposed is most similar between on-road vehicle and industrial & on-road vehicle emission sources combined. However, for those areas with greater variability (levels 4–7) industrial – and industrial & on-road vehicle emission sources combined shared similar sizes of exposure area, while on-road vehicle emission sources were negligible. Standard deviations between the three groups confirm these observations. *PD* values from the *factor-detector* analysis again confirm these results, with a 0.38 between industrial & on-road vehicle combined and industrial emission sources, and a 0.42 between industrial & on-road vehicle combined and on-road vehicle emission sources.

5. Discussion

In contrast to studies using observed or simulated air concentrations at limited air quality monitoring or simulating sites to reflect the temporal variations of air quality in an region over a short time period (Gomiščck et al., 2004; Cheng et al., 2007; Cyrys et al., 2008), this study utilized simulated air concentrations at preset high-density receptors with to reveal the spatial-temporal variations of regional air quality over long-term periods. In this way, the drawbacks of sparsely observed or limited simulated data in disclosing variations of regional air quality may be overcome. That is, on one hand, the micro-variations in air quality for an area can be better understood with highly density preset receptors compared to sparse air quality monitoring or simulated sites. On the other hand, evaluation results of relative contribution of emission sources to regional air quality could be more theoretically significant using all the samples in the region at a fine scale but not with several sparse air quality monitoring or simulating sites on their own.

The temporal variation analysis of annual ambient SO_2 concentrations in Dallas County indicates that SO_2 pollution in this region did not consistently increase or decrease from 1996 to 2002. This result might be predominantly attributed to the unstable variations of the emission sources across this county, which fluctuated from 238 to 394 tons for industrial facilities, and from 2348 to 1587 tons for on-road vehicles. This also might be the reason in part for abrupt variations of maximum and SD of annual SO_2 concentrations in Dallas County before and after 1998. The analysis of the relationships between air quality and meteorological factors in this study indicates that high levels of air humidity, as well as low air temperature and wind speed might have resulted in the relatively higher seasonal SO_2 concentration in autumn and winter compared to spring and summer (see Fig. 6). This finding is similar to the results reported by Norisada et al. (1998) and Tayanc (2000), who found that meteorological factors can play an important role in regional air quality in the process of air dispersion.

Largely consistent with the results of temporal variation described above, we found that the spatial distribution of annual SO_2 concentrations at different levels in Dallas County also expanded (e.g. year 1997) or contracted (e.g. year 2001) with unstable variation from 1996 to 2002. These variations in spatial distribution of annual SO_2 concentrations could be partly attributed to the variations of the emission sources within the county. Furthermore, the decreased SO_2 concentrations in this study indicates that changes of the spatial pattern of emission sources (e.g. locations changes of industrial emission sources in 1996 and 2002 shown in Fig. 7) might be another factor causing the variations in spatial distribution of annual SO_2 concentrations at different levels, as well as the shift of the locations of maximum annual SO_2 concentrations in Dallas County over the study period. In addition, the reason for the areas with relatively higher annual SO_2 concentrations and variations in Dallas County occurred in a northwest to southeast direction, the predominant wind direction (122 degrees) shown in Fig. 2.

The source apportionment analysis suggests that the spatial variations in total annual SO_2 concentrations in Dallas County from 1996 to 2002 were not predominantly associated with any

Table 2
Summary statistical measures of the variations in annual SO_2 concentration at varying concentration levels from different emission sources in Dallas County from 1996 to 2002.

Levels[a]	Type	Area percentage (%)	Mean ($\mu g\ m^{-3}$)	SD
1	Industrial emission sources based	5.22	−0.0249	0.0121
	On-road vehicle emission sources based	17.29	−0.0937	0.0770
	Industrial & on-road vehicle emission sources combined based	11.89	−0.0993	0.1650
2	Industrial emission sources based	84.86	0.0470	0.0354
	On-road vehicle emission sources based	68.70	0.1032	0.0611
	Industrial & on-road vehicle emission sources combined based	64.43	0.1139	0.0705
3	Industrial emission sources based	6.47	0.6698	0.2417
	On-road vehicle emission sources based	14.01	0.4141	0.1516
	Industrial & on-road vehicle emission sources combined based	20.28	0.4029	0.1565
4	Industrial emission sources based	1.64	1.2654	0.2234
	On-road vehicle emission sources based	–	–	–
	Industrial & on-road vehicle emission sources combined based	1.21	1.7936	0.7411
5	Industrial emission sources based	0.30	5.8903	1.8940
	On-road vehicle emission sources based	–	–	–
	Industrial & on-road vehicle emission sources combined based	0.99	5.9687	1.2207
6	Industrial emission sources based	0.22	10.9195	1.9766
	On-road vehicle emission sources based	–	–	–
	Industrial & on-road vehicle emission sources combined based	0.78	11.2535	2.1034
7	Industrial emission sources based	1.29	58.7046	12.7057
	On-road vehicle emission sources based	–	–	–
	Industrial & on-road vehicle emission sources combined based	0.43	19.9989	3.4446

[a] The break points for different levels are same as shown in Fig. 5.

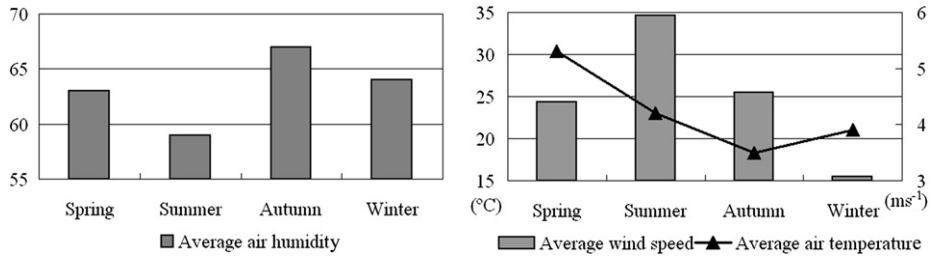

Fig. 6. Seasonal average air humidity, wind speed and air temperature in Dallas County from 1996 to 2002.

particular type of emission source. We have further shown that the contributions of industrial emission sources and/or on-road vehicle emission sources to the total annual SO_2 concentrations at relative lower levels (i.e. levels 1–3) in Dallas County were similar (Table 2). However, the industrial emission sources might significantly contribute to the total annual SO_2 concentrations at relatively higher levels (levels 4–7), especially for the annual SO_2 concentrations at the higher end. This finding can be attributed to the dense concentration of industrial sources in certain areas of the County.

In addition, although the simulated total annual SO_2 concentrations in Dallas County were a result of industrial and on-road vehicles emission sources only, the sum of the *PD* of industrial emission sources based on industrial & on-road vehicle emission sources combined-based annual SO_2 concentrations, as well as the one of on-road vehicle emission sources based on industrial & on-road vehicle emission sources combined-based annual SO_2 concentrations was still less than 1.00. This result suggests that the influences of industrial and on-road vehicle emission sources on total annual SO_2 concentrations in Dallas County are not independent. The combination of these sources amy be causing an additive effect. This may be the reason why the results shown in Fig. 5(a) cannot be generated by simply overlaying the results shown in Fig. 5(b) and (c).

In summary, our findings suggest that previous source-contribution studies (e.g. Cheng et al., 2007) did not sufficiently characterize the spatial and temporal complexity of the actual concentrations in a relatively large geographic area due to the fact that they utilized only a few points. Our findings further highlight that variability in air pollution is much more complex than we might have thought, and thus, more relevant and detailed studies are needed. However, we note that there are still several limitations that need to be addressed.

While a previous study has reported that the actual ambient SO_2 concentration could vary on a scale of tens of meters (Venkatram et al., 2004), we set the receptors at a resolution of 1 km × 1 km in Dallas County due to the limited computation resources. As a result, part of the extreme ambient SO_2 concentrations in several locations might be undetectable and the subsequent temporal variation analysis results based on the simulated annual SO_2 concentrations at locations of these receptors also might be unstable. However, it should be noted that the regional air quality is, in theory, more accurately assessed by the simulated ambient SO_2 concentrations at locations of these highly dense receptors compared to those in earlier studies only with sparse air quality monitoring or simulated sites (Tayanc, 2000; Cheng et al., 2007; etc.).

Recent work has shown that spatial analysis results can be influenced by the modifiable Areal Unit Problem (MAUP) which may introduce scale and aggregation issues (Ratcliffe and McCullagh, 1999; Tian et al., 2010). In the process of detecting the spatial-temporal variation of annual SO_2 concentration in Dallas County from 1996 to 2002 and examining its impact factors in terms of emission sources, we only produced the SO_2 concentration surface with a grid size of 1 km × 1 km. Although we believe such a fine grid should be enough to appreciate the intricacies of spatial-temporal variations of regional total SO_2 concentration and further ascertain source contributors, our results may be further improved by clearly ascertaining scaling effects (e.g. 1 km × 1 km, 2 km × 2 km, 3 km × 3 km) in future studies.

In addition, the ambient SO_2 concentration in Dallas County were not only caused by industrial and on-road vehicle emission sources,

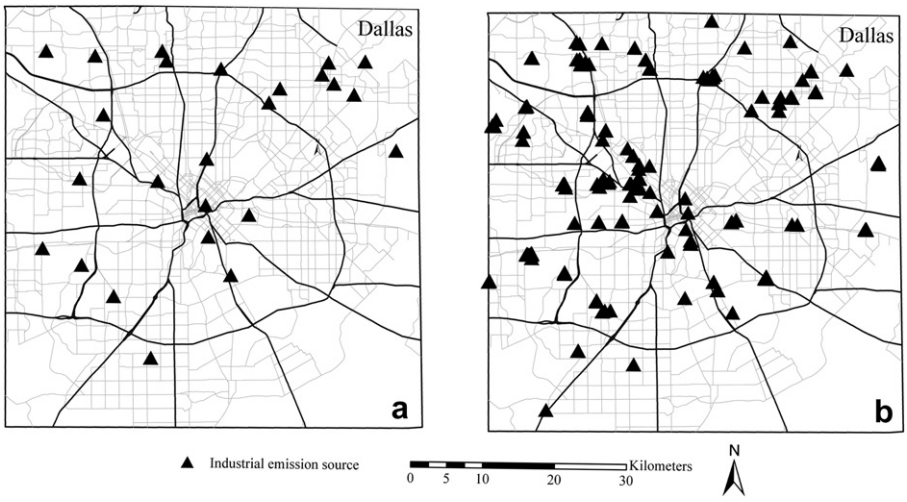

Fig. 7. Locations of industrial emission sources in Dallas County in years 1996 (a) and 2002 (b).

but also may be influenced by other area and biogenic emission sources, as well as from long-range transport and from adjacent areas. As a result, the ambient SO_2 concentration in this study might be underestimated and the other spatial-temporal variations of ambient SO_2 concentration resulting from these types of emission sources may remain un-differentiated. Meanwhile, we should note that the ambient SO_2 concentration in some study areas originate from both local emission sources and regional background SO_2 pollution, even though this is not the case for Dallas county from 1996 to 2002 due to the limited SO_2 emissions (Zou et al., 2010). In the source apportionment analysis, we only ascertained the contribution of the type of industrial and/or on-road vehicle emission sources to the regional total SO_2 concentration. Due to the complexity of air dispersion modeling, this operation makes it impossible to accurately determine the contribution of a certain industrial emission source (e.g. coal or gas power plant) or a certain portion of road emissions (e.g. diesel vs. petrol contribution, see Int Panis et al., 2002) to the total SO_2 concentration in the study area.

6. Conclusion

This study demonstrated that the analysis of spatial-temporal variations in regional air quality as well as the evaluation of different emission sources affecting air quality in an area through an air dispersion modeling approach can produce results that are more accurate than air quality data obtained from sparsely located monitoring sites. We also have shown the potential of utilizing air dispersion models and GIS-based spatial analysis to disclose micro-variations in air pollution concentrations at a fine geographic scale while parsing out source contributors. Our results show that most areas of Dallas County only experienced small variations of SO_2 concentration over the period from 1996 to 2002 and that annual SO_2 concentrations in some areas of the county greatly increased over this period. In addition, results from the source-contribution analysis indicated that the spatial-temporal variations in SO_2 concentration in Dallas County from 1996 to 2002 were not attributed to any particular type of emission source, but industrial emission sources could be the origin of extreme variations in SO_2 concentrations in some areas. The approach presented in this paper provides a general framework for effectively assessing air pollution exposure risk in a given urban area. This approach shows promise for use by policy makers, environmentalists, and epidemiologists in understanding the complex spatial-temporal variations of regional ambient air quality over long periods of time.

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