

# Characterizing air pollution patterns on multiple time scales in urban areas: a landscape ecological approach

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**Abstract** Quantifying the spatiotemporal patterns of air pollution in urban areas is essential for studying ecological processes, environmental quality, and human health in cities. To adequately characterize or monitor air pollution patterns, one important issue is scale because the concentrations of air pollutants are temporally dynamic and spatially heterogeneous. Our research addresses the scale issue in air quality monitoring and analysis by considering the following research questions: (1) How does the spatial pattern of ozone change with the temporal scale of analysis? (2) How does the spatial pattern of PM<sub>10</sub> change with the temporal scale of analysis? (3) What implications do these scale effects have for designing and evaluating air pollution monitoring networks? We systematically examined these questions based on data from official air pollution monitoring networks in the Phoenix metropolitan region, Arizona, USA. Our results showed that spatial patterns of both ozone and PM<sub>10</sub> may change substantially with the temporal scale of analysis. Ozone patterns at broader (but not finer) temporal scales were more consistent across years, and exhibited a more uniform, regionalized pattern. PM<sub>10</sub> patterns were less consistent across years than ozone, and exhibited a more localized effect. Spatial patterns of PM<sub>10</sub> also varied seasonally. Our study demonstrates that it is critically important to consider the temporal and spatial scales in designing or evaluating air monitoring networks in particular and in conducting air pollution research in general.

**Keywords** Air pollution · Spatial pattern of air pollutants · Monitoring network · Ozone · PM<sub>10</sub> · Scale effects · Phoenix metropolitan region

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## Introduction

Monitoring the spatial and temporal patterns of air pollutants in urban areas is necessary for protecting human health and ensuring environmental justice. To accurately assess air pollutants and identify populations at risk, a critically important first step is to determine how many air sampling stations are needed and where they should be placed. Although it would be desirable to have a dense network of air pollution monitors that covers the full spatial extent of an urban region, this is infeasible because of physical, fiscal, and technical constraints. Establishing an air monitoring site takes significant resources, and issues such as location, objective, power, and security all have to be considered (Maricopa County Air Quality Department 2011; Arizona Department of Environmental Quality 2011b). Thus, policy makers and resource managers need multiple sources of information in order to maximize their limited resources when designing or improving air monitoring networks. A fundamentally important but largely ignored issue in evaluating and designing air pollution monitoring networks is spatiotemporal scale. Scale is a central issue in ecological and geographic sciences and particularly in landscape ecology which studies the relationship between spatial pattern and ecological processes across a range of scales (Wu et al. 2000; Pickett and Cadenasso 1995; Turner 1989). Two key components of scale are grain size (corresponding to spatial or temporal resolutions) and extent (the spatial expanse or time duration of a study) (Wu et al. 2006). Spatial patterns, ecological processes, and their relationships are all scale-dependent, meaning that their characteristics and controls vary with the scale of observation or analysis (Levin 1992; Wu and Loucks 1995). Accurately assessing air pollution in an urban area requires the generation of time series of spatial patterns (maps) of air pollutants, and these patterns are most likely scale dependent as with ecological patterns. This scale dependence of air pollution patterns has important implications for the design of monitoring networks and the analysis of data obtained from them. Capturing spatial and temporally heterogeneous air pollution patterns can have important implications, including evaluating epidemiological effects or conducting social justice studies at different scales of exposure (Digar et al. 2011; Loo 2007). While the scale issues have been scrutinized extensively in ecology and geography, there is little landscape ecological work done on how scale matters in monitoring and analyzing the spatiotemporal patterns of air pollutants.

Thus, we attempted to address this research problem in the Phoenix metropolitan region, one of the fastest-growing urban areas in the United States and home to more than 4 million people (Luck and Wu 2002; Berling-Wolff and Wu 2004). With increasing anthropogenic activity, health standards for air pollution are frequently violated in this desert city (Bolin et al. 2000; Arizona Department of Environmental Quality 2011a). Ground-level ozone and particulate matter less than ten microns in size ( $PM_{10}$ ) are the two pollutants currently of most local concern, as the region is classified as being in non-attainment of standards for these pollutants (Arizona Department of Environmental Quality 2009; U.S. Environmental Protection Agency 2009a). Specifically, our study was designed to address the following research questions:

1. How does the spatial pattern of ozone change with the temporal scale of analysis (i.e. temporal extent)?
2. How does the spatial pattern of  $PM_{10}$  change with the temporal scale of analysis?
3. What implications do these scale effects have for designing and evaluating air pollution monitoring networks?

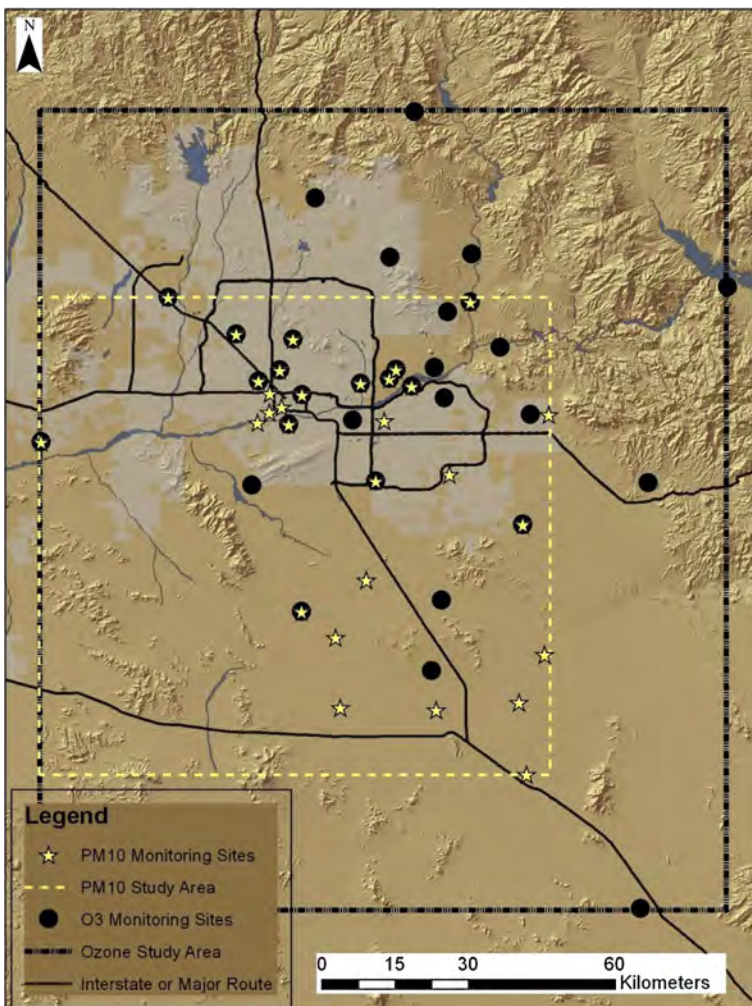
We hypothesized that, due to its chemical characteristics, ozone would be a regionally-scaled pollutant and its spatial pattern would be more uniform across space and more

consistent between sampling years. We also hypothesized that ozone would have a more apparent urban-to-rural gradient and be more stable outside of the urban area. By contrast, we hypothesized that the spatial pattern of  $PM_{10}$  would be more localized in relation to sources and less consistent across temporal scales.

## Methods

### Study area

The study is in the Phoenix metropolitan statistical area (MSA) in South-Central Arizona (Fig. 1). The MSA, within Maricopa and Pinal Counties, is a thriving area with more than 20



**Fig. 1** Topographical map of the Phoenix Metropolitan Statistical Area (*shaded*) and surrounding rural areas, depicting the ozone and  $PM_{10}$  study areas and monitoring sites. Note that some sites combined both ozone and  $PM_{10}$  monitors

self-governing municipalities. The rural areas of Maricopa and Pinal counties contain significant agriculture, including livestock and irrigated cropland. The region has experienced dramatic growth since the end of World War II, with population in the MSA expanding from 331,000 in 1950 to almost 4.2 million in 2010 (Wu et al. 2011). This growth has been exponential, with populations in Pinal and Maricopa Counties increasing by 99.9 % and 24.2 %, respectively, between the 2000 and 2010 census (U.S. Census Bureau 2011).

The Phoenix region is geographically situated in a river valley and is surrounded by mountainous topography. The region is located in arid, sub-tropical latitudes and has predominantly high atmospheric pressure, and thus light winds and weak atmospheric circulation. This prevailing lack of strong atmospheric circulation, in combination with the valley location, impedes the dispersion of pollutants out of the urban area (Ellis et al. 1999, 2000).

Industries and transportation in the Phoenix MSA region, such as agriculture, sand and gravel mining, construction, vehicle traffic, and unpaved roads in the urban periphery, in combination with the dry desert climate, create considerable sources for PM<sub>10</sub> pollutants (Maricopa County Air Quality Department 2009; Bolin et al. 2000). Ozone is a secondary pollutant and is not directly emitted; however, the abundant sources of ozone precursors, e.g. volatile organic compounds (VOCs), carbon monoxide (CO), and oxides of nitrogen (NO<sub>x</sub>), mixed with the commonly warm, sunny days, create an environment where active photochemical reactions produce significant amounts of ozone pollutants near the ground level (Ellis et al. 1999; Maricopa County Air Quality Department 2009).

For this study, the Phoenix MSA region was divided into ozone and PM<sub>10</sub> study areas (Fig. 1). We designed these study areas based on their geographic features and the location of existing pollution monitoring sites. We also explicitly chose these areas, i.e. the homogenous metropolitan area with a shallow buffer of nearby rural sites, for their assumed stationarity of data. Ozone is hypothesized to be a more regionally-scaled pollutant that is easily transported because of its chemical lifecycle, and as a secondary pollutant it can occur in broader areas not necessarily near its precursor sources. On a diurnal basis, precursors, e.g. CO, VOC, and NO<sub>x</sub>, react with sunlight to produce O<sub>3</sub> molecules. However, at night, O<sub>3</sub> in the nocturnal boundary layer will react with nitric oxide (NO) in a titration reaction that converts NO to NO<sub>2</sub> while ‘scavenging’, or destroying, O<sub>3</sub> molecules. Ozone pollution in the urban core, with ample NO sources, can often virtually disappear overnight, only to begin the cycle anew the next morning; while rural areas have more persistent ozone concentrations which can travel through the atmosphere (Gregg et al. 2003; Seinfeld and Pandis 2006; National Research Council 1991). Therefore, ozone concentrations are also hypothesized to be much more temporally variable within urban areas while more stable in rural areas. Thus, we designed the ozone study area to include rural areas further away from the urban center, increasing the number of sites for the statistical analyses, while still maintaining assumed stationarity. The ozone study area is approximately 2.3 million hectares in size and contains 32 pollution monitoring sites, including several in downwind uninhabited wilderness areas (Fig. 1).

PM<sub>10</sub>, in contrast, is hypothesized to be a more localized pollutant. The PM<sub>10</sub> study area is approximately 1 million hectares in size, and contains 30 pollution monitoring sites (Fig. 1). Because of the limitations of this assumed stationarity and the location of existing monitoring sites, the PM<sub>10</sub> study area is much smaller than the ozone study area. PM<sub>10</sub> is hypothesized to be a far more temporally variable and spatially localized pollutant than ozone, and the size of the study area was designed to be smaller to maintain a reasonable assumption of stationarity (Pohjola et al. 2002; Seinfeld and Pandis 2006).

## Data acquisition and processing

We obtained air pollution data for the study from the United States Environmental Protection Agency's (EPA) Air Quality System (AQS) database. These data were generated and submitted to AQS by local government air pollution agencies at the state, county, and tribal levels. This study utilizes data from 32 ozone and 30 PM<sub>10</sub> monitoring stations operated by these local agencies within the Phoenix MSA (Table 1). These air pollution monitors all complied with the EPA's Federal Reference Method or Federal Equivalency Method; thus the sampling equipment was approved for taking official air pollution measurements and rigorous maintenance and quality assurance plans for the equipment and data were required and verified (Code of Federal Regulations 2009; Maricopa County Air Quality Department 2011; Arizona Department of Environmental Quality 2011a).

We collected ozone data for the study in the time span of 2008 through 2010, with each of the three years being analyzed separately and compared with each other. The finest temporal resolution (or grain size) of these data is 1 h (i.e., raw data were one-hour averages). To examine the effects of different temporal scales on the air pollution pattern analysis, we focused on four temporal extents (i.e., time durations over which average values of measurements were derived): 1 h (at 15:00 on July 15), 8 h (15:00–22:00 on July 15), 1 month (July), and a season (April–October) (Table 2). The seasonal average was chosen instead of an annual average because many of the ozone monitoring sites only operate during this time period.

We also analyzed PM<sub>10</sub> data and compared them independently for the years 2008 through 2010. The temporal resolution for PM<sub>10</sub> was a 24-h average measured 1 day out of every six (1-in-6 day basis), as this is the operating schedule for some of the PM<sub>10</sub> monitors. Most PM<sub>10</sub> monitors operate on a finer time scale, collecting daily 24- or 1-h averages; however, all finer averages were rolled into a 24-h average and all data outside of the 1-in-6 day schedule were eliminated to create a consistent coarse resolution. These data were then analyzed at three different temporal extents: daily, monthly, and annual; daily and monthly extents included both winter and summer seasons (Table 2).

## Data analysis

It is desirable to use a number of methods when performing geostatistical or spatial analysis, such as variograms, covariances, or correlograms (Rossi et al. 1992). Comparing and contrasting the results from multiple methods and at multiple scales provide a more comprehensive understanding with more robust conclusions (Wu 2004; Jelinski and Wu 1996). In that spirit, this study uses several techniques to explore the data and address the research questions.

**Table 1** List of agencies operating monitoring stations within the study area. Agencies submit their data to the EPA's AQS database which was the source of data for this study

Agency	Type of agency	# O <sub>3</sub> stations	# PM <sub>10</sub> stations
Arizona Department of Environmental Quality	State	3	2
Fort McDowell Yavapai Nation	Tribal	1	1
Gila River Indian Community	Tribal	2	1
Maricopa County Air Quality Department	Local (County)	17	14
Pinal County Air Quality Control District	Local (County)	5	9
Salt River Pima-Maricopa Indian Community	Tribal	4	3

**Table 2** Multiple time scales used to analyze the spatiotemporal patterns of ozone and PM<sub>10</sub> in the Phoenix metropolitan region

Pollutant	Temporal resolution	Study years	Temporal extents				
			Seasonal (Apr–Oct)	Monthly (July)	8-h (July 15, 15:00–22:00)	1-h (July 15, 15:00)	
Ozone	1-h Averages, continuous sample grain	2008–2010	Seasonal (Apr–Oct)	Monthly (July)	8-h (July 15, 15:00–22:00)	1-h (July 15, 15:00)	
PM <sub>10</sub>	24-h Averages, 1-in-6 day sample grain	2008–2010	Annual	Monthly (Jan)	Monthly (Aug)	Daily (Jan 7 [2008, 2009], Jan 8 [2010])	Daily (Aug 22 [2008], 23 [2009], 24 [2010])

Note that for PM<sub>10</sub> the daily extent is applied to different days in the different sampling years based on the running time of the 1-in-6 day schedule

### Trend analysis

The first technique used was trend analysis, a useful method of exploring data when no a priori knowledge exists. For non-spatial data, a common procedure is to use regression to explore the relationship between independent and dependent variables. This procedure is also appropriate for spatial data, with the X-Y coordinates as the independent variable and the pollution concentrations, or Z-value, as the dependent variable (Fortin and Dale 2005).

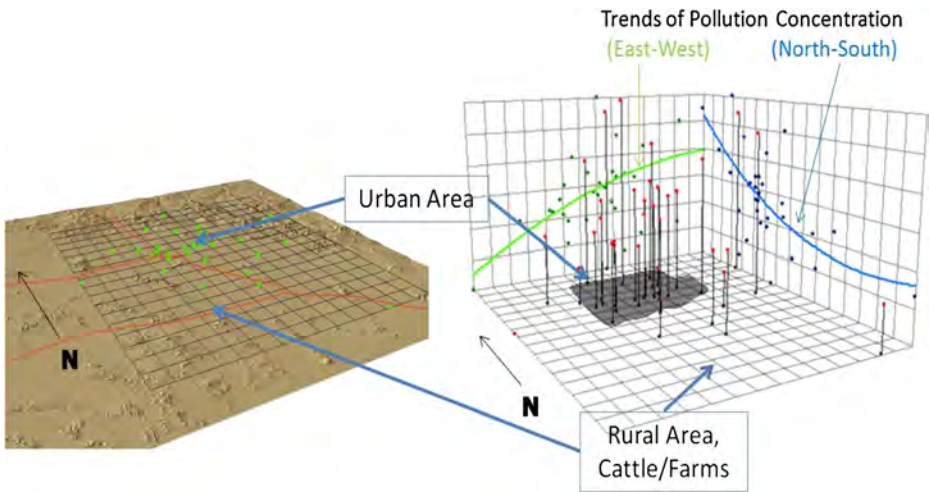
The data exploration was accomplished with the Trend Analysis tool in the ArcMap Geostatistical Analysis Extension, a Geographical Information System (GIS) application (ESRI 2010). Using the tool, the study area was overlaid with a grid within which monitoring sites were placed according to their X-Y coordinates. The measured pollution concentrations from each site were then displayed as vertical sticks in the Z axis (Fig. 2). The pollution concentrations were projected on the X-Z and Y-Z plane to give a graphical depiction in a spatially-explicit manner, i.e. north to south and east to west. A second-order polynomial (quadratic) multiple regression trend line was fitted to the two Z planes to show the spatial trend of the data (Fig. 2).

This analysis is a generalized ad hoc interpolation of the data with clear representation of the spatial trends. It is a global interpolation and not intended to model local spatial patterns of pollution. We compared multiple temporal extents and multiple years against each other to examine how those trends would change with scale.

### Correlation analysis

The second technique used was correlation analysis, similar to the method used by Ito et al (2001, 2004). We compared data from all 32 ozone and 30 PM<sub>10</sub> monitoring sites in a matrix format and calculated the coefficient of determination between each pair of sites. These correlations were cross-referenced with the distance between the sites and displayed in a correlogram. A trend line was also fitted to each correlogram.

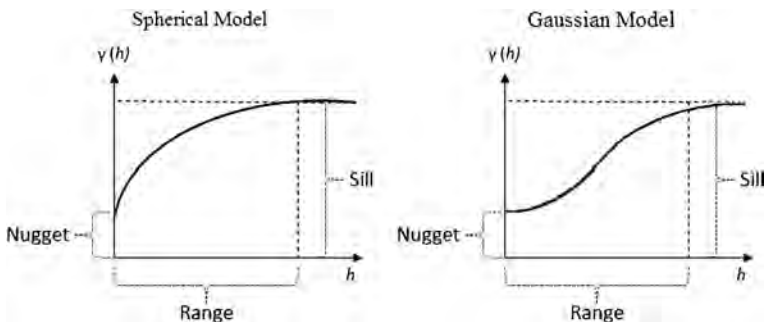
Correlograms provide a useful method of visualizing the spatial dependence between data points in relation to distance, although it is a general method that makes no determination of exogenous or endogenous processes effecting the pattern (Fortin and Dale 2005). The primary reason for using correlation analysis in this study was to explore how the spatial dependence of air pollution patterns would change with scale.



**Fig. 2** Illustration of the use of the Trend Analysis tool for depicting the spatial pattern of air pollutants in the Phoenix metropolitan region. The monitoring sites for ozone are marked in green on the gridded map on the left, and their concentrations are displayed in a 3-dimensional space on the right. The height of the Z-axis ‘stick’ is proportional to the pollution concentration over a given temporal extent. The urban areas of Phoenix are also depicted as a reference in relation to the surrounding rural areas

*Semivariance analysis*

The third technique used was semivariance analysis, which is useful for quantifying the structure of spatial autocorrelation, and necessary for determining the values of unmeasured locations using kriging (q.v. next section). Semivariance usually is plotted against the separation distances (or lag distance,  $h$ ) between two points in space to create a semivariogram (Fig. 3). The range in the semivariance plot indicates the distance within which spatial autocorrelation exists and beyond which statistical independence in the data begins (Griffith



**Fig. 3** Examples of generalized semivariograms—the spherical and Gaussian models. The key parameters of a semivariogram are: the nugget variance (variability due to local random effects and measurement errors), the range (distance up to which the spatial structure varies), the sill (plateau of semivariance values, or the end of spatial autocorrelation), and the structural variance, which is the difference between the sill and the nugget (variability due to spatial structure). The spatial lag, or distance between points, is  $h$ , and the semivariance value is  $\gamma(h)$ . This study found that ozone data fit the Gaussian model better, while  $PM_{10}$  data best fit the spherical model. Refer to Online Supplement 2 for examples of actual experimental semivariograms from this study

1992; Rossi et al. 1992; Fortin and Dale 2005). Semivariance between each pair of samples is computed based on the following equation:

$$\hat{\gamma}(h) = \frac{1}{2n(h)} \sum_{i=1}^n [z_i - z_{i+h}]^2 \quad (1)$$

where:  $\hat{\gamma}(h)$  is the semivariance for interval distance class  $h$ ,  $n(h)$  is the number of pairs of samples for the lag interval  $h$ ,  $z_i$  is the measured sample value at point  $i$ , and  $z_{i+h}$  is the measured sample value at point  $i + h$ .

The software, GS+: Geostatistics for the Environmental Sciences (Gamma Design Software 2006) was used for the semivariance analysis. Sample locations were formed into lag intervals with uniform distance. These lag intervals need to be small enough to capture the pattern, though if they are too small it will be unnecessarily patchy (Fortin and Dale 2005). Specifically, the maximum lag distance was set smaller than one half the spatial extent of the dataset (Meisel and Turner 1998). The shortest distance between sample points was used as the uniform distance with PM<sub>10</sub> data, though a slightly longer distance was used for ozone points to reduce excessive patchiness. Data were log or square-root transformed as appropriate to reduce skewness (Fortin and Dale 2005), and the  $h$ -lags were plotted in  $h$ -scattergrams to identify extreme outliers to be removed, as a necessary process described by Rossi, et al. (1992). The prepared data were modeled in isotropic semivariograms using the Gaussian model for ozone and the spherical model for PM<sub>10</sub>, as these models consistently produced the least error when paired with the respective parameter (Fig. 3, Online Resource 2). By definition of the GS + software, the sill never meets the asymptote in the Gaussian model; therefore range is estimated as the distance at which the sill is within 5 % of the asymptote (Gamma Design Software 2006). See Online resource 1 and 2 for further details on the parameters of the semivariance analysis.

### *Kriging*

Kriging is a geostatistical interpolation method to estimate values at unsampled locations based on the spatial autocorrelation structure quantified in the semivariance analysis (Cressie 1990; Fortin and Dale 2005). When additional sampling is too expensive or difficult to accomplish, as is often the case with air pollution monitoring, kriging provides an effective way of mapping out the spatial pattern of the pollutant over the large area. Our kriging of the maps of ozone and PM<sub>10</sub> concentrations over the study area was conducted using the Geostatistical Analysis Extension within ArcMap (ESRI 2010). All input settings were matched with those of the GS + software to maintain consistency with our semivariance analysis. Thematic maps were created at each temporal scale, for both ozone and PM<sub>10</sub>, so as to create a visual comparison of spatial patterns between scales.

## **Results**

### Spatiotemporal patterns of ozone

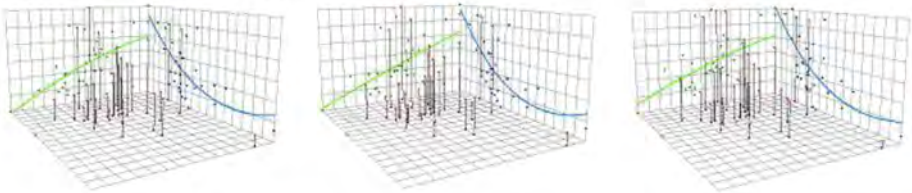
#### *Trend analysis of ozone*

Our analysis of data from the 32 ozone monitoring sites showed that the spatial trend of ozone concentration varied with different temporal extents in each of the three study years (2008–

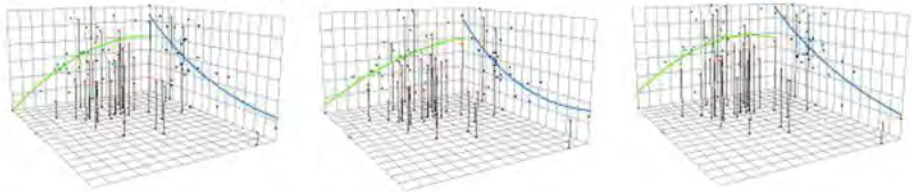


2010) (Fig. 4). The ozone pattern tended to be consistent across the 3 years on broader scales (i.e., the seasonal and monthly extents), but not on finer scales (i.e., the 8-h and hourly extents). On the seasonal and monthly scales, the highest ozone concentration consistently occurred in the northeastern section of the study area, but this was not the case on the finer scales (Fig. 4) Because the urban core is located toward the top middle of the study area and because the dominant wind direction is to the northeast (Pardyjak et al. 2009), the highest ozone concentration on the seasonal and monthly scales occurred in the rural mountainous areas downwind of the urban center. On shorter temporal scales (especially the hourly extent), the location of the highest concentration of ozone was much closer to the urban core, with the urban areas generally having higher ozone levels than the rural areas (Fig. 4).

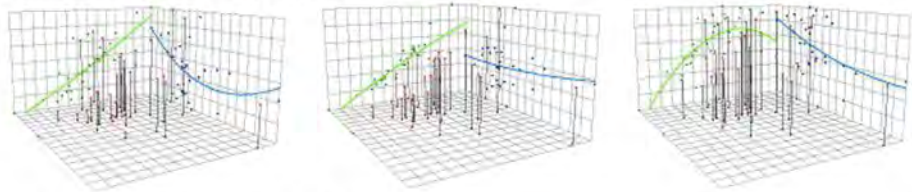
#### Seasonally Averaged Ozone



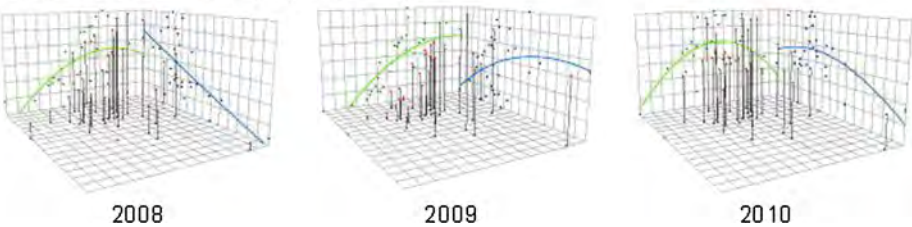
#### Monthly Averaged Ozone (July)



#### 8-Hour Averaged Ozone (July 15, 15:00-22:00)



#### Hourly Averaged Ozone (July 15, 15:00)



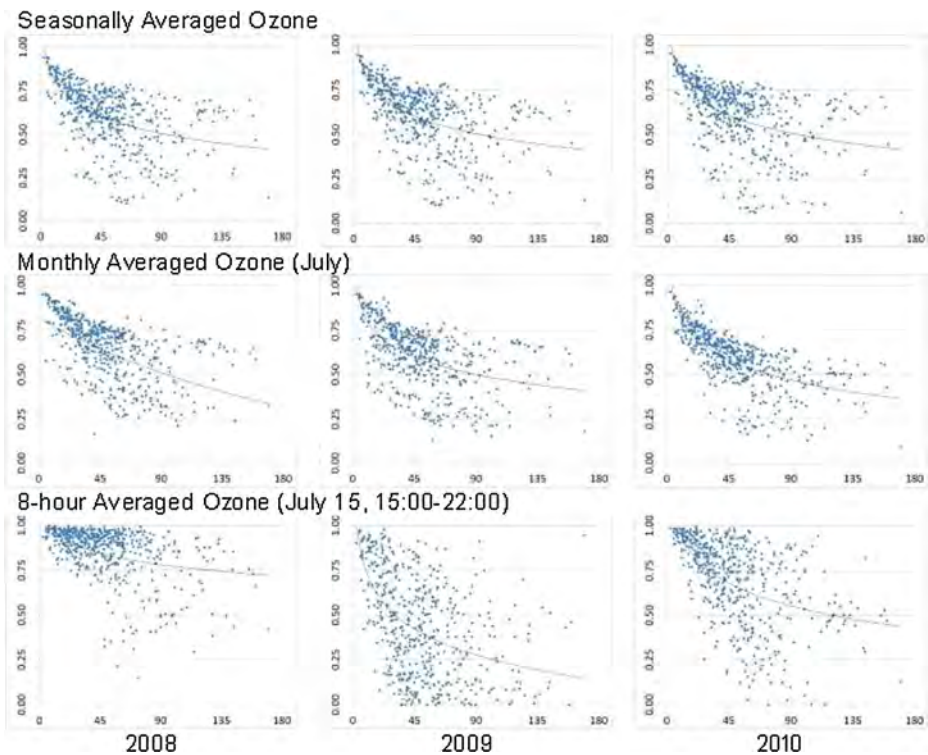
**Fig. 4** Trend analysis results showing spatial patterns of ozone concentration at different temporal extents and in different years in the Phoenix metropolitan region. The trend line on the 3D graphs depicts the concentration trend of pollutants across the study area and changes to the trend line between scales is the focus of the trend analysis method. Refer to Fig. 2 for details on the elements within each 3-D graph

### Correlation analysis of ozone

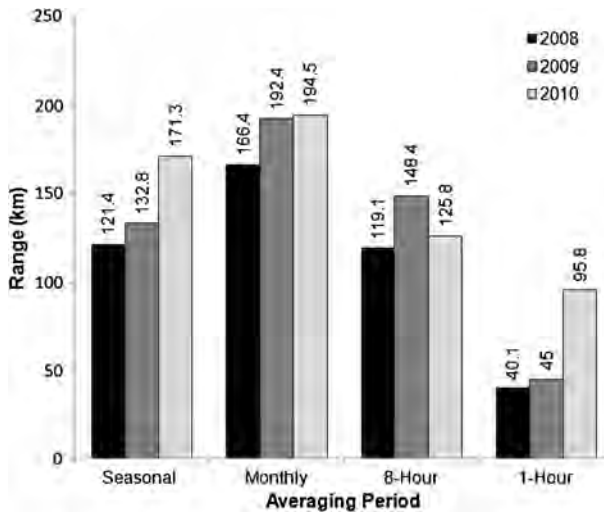
The degree of correlation in ozone concentration between monitoring sites generally decreased with increasing between-site distances, but the specific pattern differed between the long (seasonal and monthly) and short (8-h) scales (Fig. 5). The correlograms on the longer scales were also similar between years. However, the distance-based correlation pattern of ozone at the 8-h scale was different quantitatively from that on the longer scales, and highly variable between the three study years (Fig. 5). Except for the 8-h scale in 2009, the results of correlation analysis showed that the majority of ozone monitoring sites were highly correlated with each other (over 70 %) within a distance of 30 km.

### Semivariance analysis of ozone

The range—the distance over which ozone concentration was spatially autocorrelated—changed with temporal scales and between the three study years (Fig. 6; Online resource 1). In certain cases, the range for ozone was approaching 200 km, which is outside the spatial extent of the study area. The longest spatial autocorrelation range was found at the monthly scale, not at the longest temporal scale (seasonal). However, the range showed a consistent decreasing trend as the temporal scale became shorter than a month (Fig. 6).



**Fig. 5** Correlograms of ozone concentration on different temporal scales and in different years in the Phoenix metropolitan region. Each X axis represents distance from 0 to 180 km. Each Y axis represents the coefficient of determination ( $r^2$ ) from 0.00 to 1.00



**Fig. 6** Effects of temporal scale (extents) on autocorrelation ranges of ozone. Range is the distance (in km) over which spatial autocorrelation exists among the ozone monitoring sites, as determined in semivariogram analysis. Note that the 1-h scale has much shorter ranges than the other scales, and more variation between years, suggesting a major change in ozone patterns between the 1- and 8-h scales

### *Kriging interpolation of ozone*

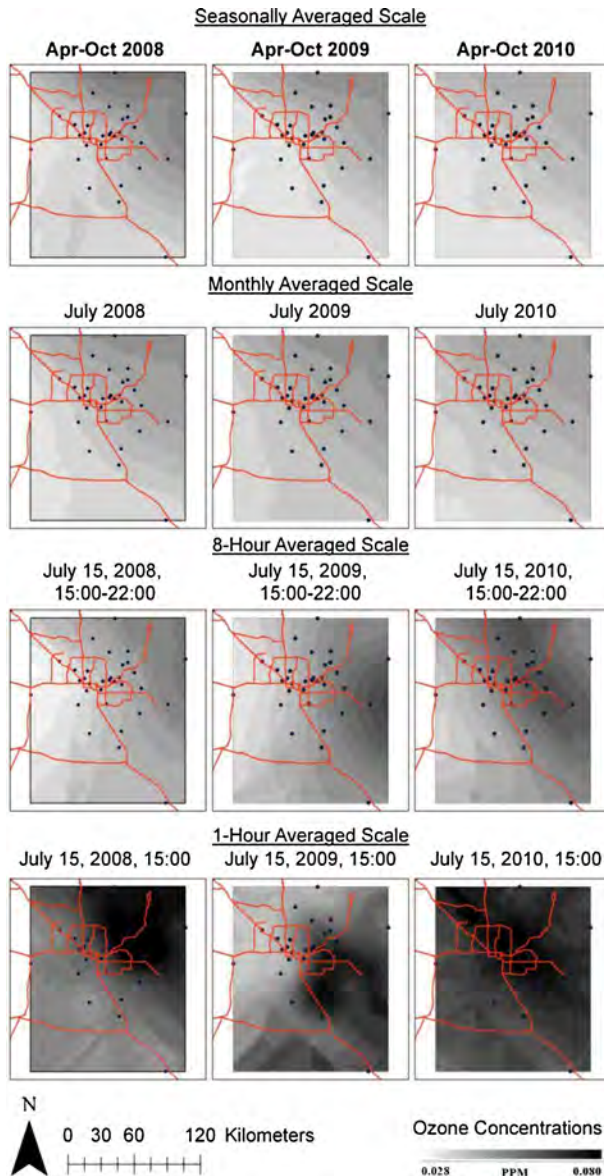
The krigged maps showed that ozone concentrations across the study area were relatively low at the seasonal scale, but increased appreciably with decreasing temporal scales (Fig. 7). The northeast mountainous region of the study area had higher ozone concentrations on the seasonal and monthly scales, with little variation in space and between years. This spatial pattern of ozone began to change at the 8-h scale as the areas of high ozone concentrations intensified with appreciable differences between years. At the 1-h scale, which is 3:00–4:00 P.M. in a summer afternoon, ozone levels were almost at their highest for the entire region (Khoder 2009). At this fine scale, the spatial pattern of higher ozone concentrations occurred in both the urban and rural areas, and also varied considerably between years (Fig. 7).

### Spatiotemporal patterns of $PM_{10}$

#### *Trend analysis of $PM_{10}$*

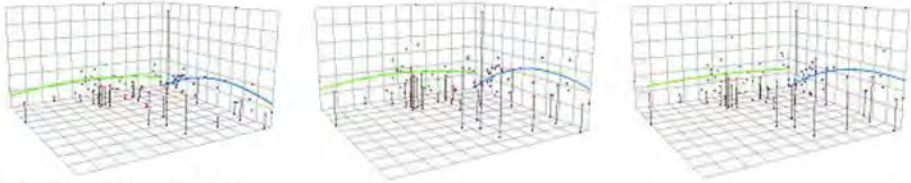
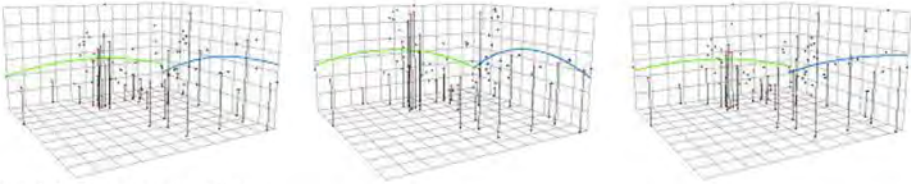
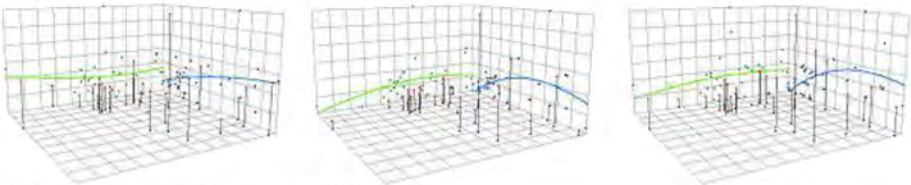
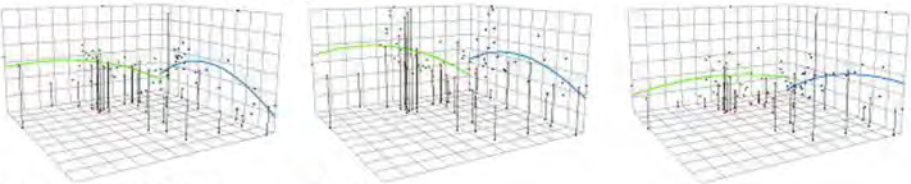
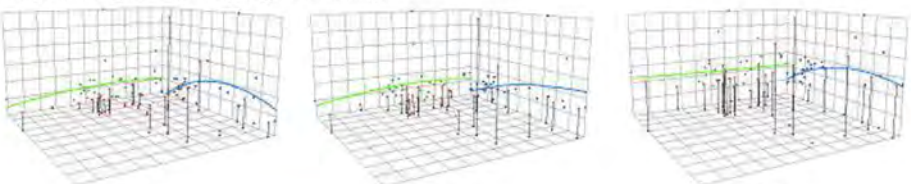
Data for  $PM_{10}$  were analyzed for summer and winter seasons on three different temporal scales: annual, monthly, and daily. The spatial pattern of  $PM_{10}$  was more variable between years than that of ozone, but the general trend shown in spatial pattern was similar (Fig. 8). The spatial trend of  $PM_{10}$  did not change appreciably between the temporal scales, but differences were noticeable between summer and winter. In particular,  $PM_{10}$  levels tended to be higher in the urban areas in winter, but in the rural areas in summer (Fig. 8). The spatial pattern at the annual scale closely resembled that of August, implying that the summer pattern was predominant most of the year.

One site, located in rural Pinal County south of the Phoenix metropolitan area, had higher  $PM_{10}$  concentrations than all other sites, regardless of scale or season. This site, known as the Cowtown monitor, was surrounded by agriculture operations (including cattle feedlots) and



**Fig. 7** Krigged maps of ozone concentrations, each of which is bordered by the ozone study area shown in Fig. 1. *Black dots* represent the ozone monitoring sites; highways are represented as lines. Ozone concentrations range from 0.028 to 0.080 PPM

not far from housing developments (Arizona Department of Environmental Quality 2010). The Cowtown monitor was sited as a hotspot monitor of local agglomerated sources, and as such had particulate concentrations that were much higher than other monitors in the region (Arizona Department of Environmental Quality 2010; U.S. Environmental Protection Agency 2009b). The Cowtown monitor was included in the trend analysis, but excluded as an outlier in the semivariogram analysis.

Annually Averaged PM<sub>10</sub>Monthly Averaged PM<sub>10</sub>  
(January)Monthly Averaged PM<sub>10</sub>  
(August)Daily Averaged PM<sub>10</sub> (January 7-8)Daily Averaged PM<sub>10</sub> (August 22-24)

2008

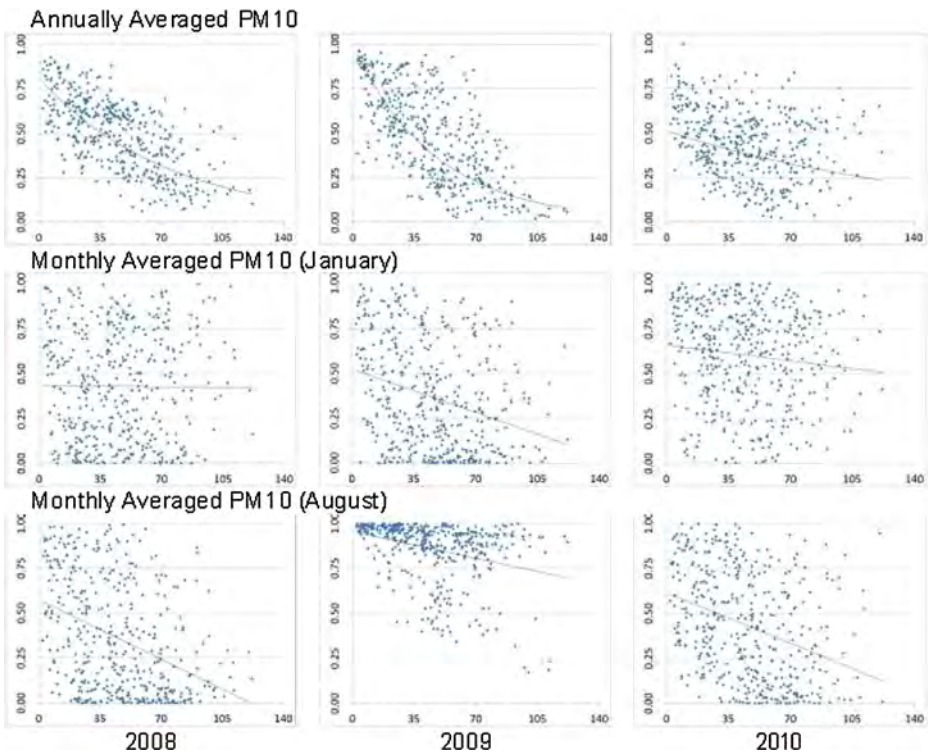
2009

2010

**Fig. 8** Spatial patterns of PM<sub>10</sub> concentration at different temporal extents and in different years. Refer to Fig. 2 for details on the elements within each 3-D graph

### Correlation analysis of PM<sub>10</sub>

The PM<sub>10</sub> correlation analysis was only conducted at the annual and monthly (winter and summer) scales because there was only a single value at the daily scale. The distance-based correlation patterns of PM<sub>10</sub> were more variable between scales and between years than those of ozone (Fig. 9). For the annual-scale pattern, high-levels of correlation (>70 %) appeared

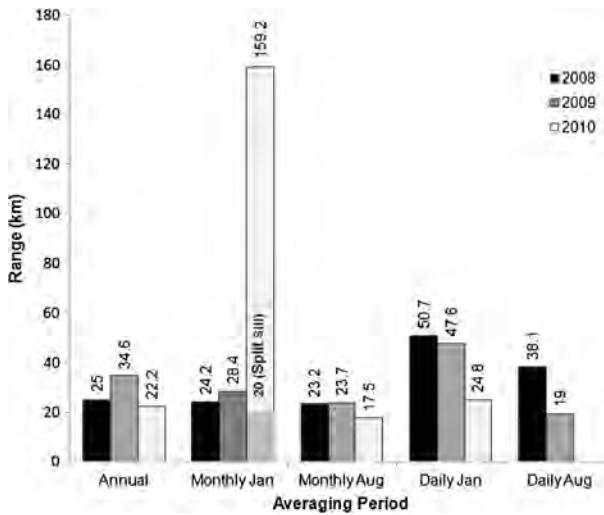


**Fig. 9** Correlograms of  $PM_{10}$  concentration on different temporal scales and in different years in the Phoenix metropolitan region. Each X axis represents distance from 0 to 140 km. Each Y axis represents the coefficient of determination ( $r^2$ ) from 0.00 to 1.00

within 10 to 20 km. At the monthly scale, however, the correlation disappeared. August 2009 is an extreme exception, however, with most of the correlations being above the 80 % level, even so far as 120 km. August 2009 had hotter and drier weather than August 2008 or 2010.

#### *Semivariance analysis of $PM_{10}$*

The ranges of  $PM_{10}$  were, in general, less than 50 km in distance for different temporal scales and study years (Fig. 10)—much shorter than those of ozone. Unlike ozone, the ranges of  $PM_{10}$  tended to get longer with shorter temporal scales, with ranges longer in winter than summer. Major differences in the  $PM_{10}$  patterns occurred between the daily and monthly scales. January 2010, an exceptionally rainy month, exhibited a multi-scale nested semivariogram (Robertson and Gross 1994). The first sill evident in the semivariogram was reached at 20 km, similar to the other sample years. The second sill was estimated by the GS + software to be reached at 159 km, which is outside of the study area. The semivariogram on August 24, 2010, was also different from the other sample years with an apparent linear pattern with no sill. The study area was experiencing a weather event on that day with windy conditions out of the north, which is unusual.



**Fig. 10** Effects of temporal scale (extents) on spatial autocorrelation ranges of  $PM_{10}$ , as determined in semivariogram analysis. The semivariogram from Jan 2010 exhibited two nested sills giving a multi-scalar range. The Aug 2010 daily semivariogram did not display a sill, as the data appeared to be linear in nature

### *Kriging interpolation of $PM_{10}$*

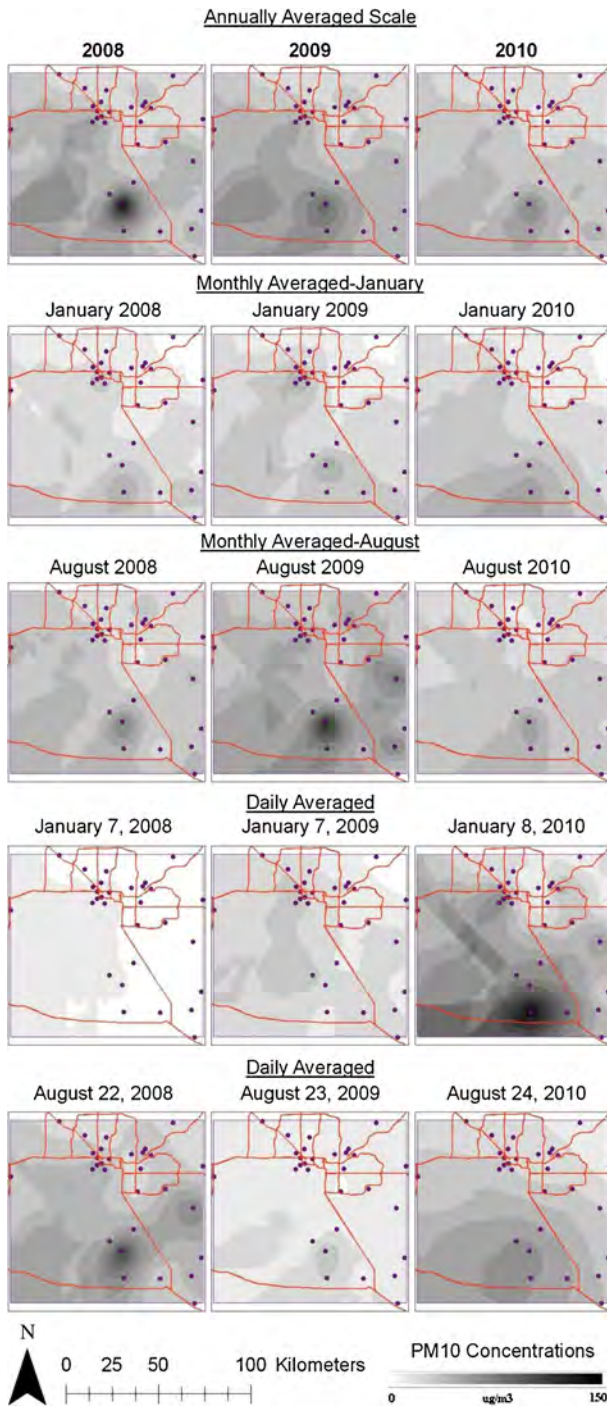
The Krigged maps of  $PM_{10}$  showed that concentrations tended to be higher in the southern agricultural portion of the study area, while the urban areas in the northern portion had the lowest concentrations, especially at higher elevations (Fig. 11). The overall spatial pattern at the annual scale was fairly consistent between the three study years, all showing a  $PM_{10}$  ‘hotspot’ in the south-central portion of the study area (the Cowtown monitor as mentioned previously).

At the monthly temporal scale, the  $PM_{10}$  pattern varied between winter and summer, with the summer pattern more closely resembling the annual pattern and having a distinct urban/rural gradient.  $PM_{10}$  winter concentrations in the southern agricultural areas were lower and more comparable with the northern urban areas. In January 2009, the urban area had the highest  $PM_{10}$  concentrations in the study area. The pattern between study years at the monthly scale was also similar to each other, although there appeared to be more variation between the summer months. At the daily scale, the spatial pattern of  $PM_{10}$  showed the greatest variability between scales and between years (Fig. 11).

## **Discussion**

### Changing ozone patterns on different temporal scales

Our study has shown that spatial patterns of ozone in the Phoenix metropolitan region may change substantially with the temporal scale of analysis. For example, the results of trend analysis and Kriging indicated consistently higher concentrations in the northeast portions of the study area on the longer time scales (i.e. seasonal and monthly), but this pattern dissipated on shorter time scales (i.e., the 8-h and 1-h scales). Likewise, the correlograms showed high degrees of correlation with strong trends at the seasonal and monthly scales, but not at the finer



**Fig. 11** Kriged maps of  $\text{PM}_{10}$  concentrations, each of which is bordered by the  $\text{PM}_{10}$  study area shown in Fig. 1. *Black dots* represent the  $\text{PM}_{10}$  monitoring sites, and highways are represented as *lines*. The  $\text{PM}_{10}$  Concentration color scale ranges from 0 to  $150 \mu\text{g}/\text{m}^3$



scales. Our results from semivariance analysis further indicated that the spatial autocorrelation ranges for ozone were quite sensitive to the temporal scale of analysis.

These patterns were not unexpected, given the meteorological conditions in this region of Arizona and the chemical lifecycle of ozone and its relation to other urban pollutants, such as  $\text{NO}_x$ . Ozone requires the mix of VOCs,  $\text{NO}_x$  or CO, and sunshine to be created, but excessive  $\text{NO}_x$  also scavenges  $\text{O}_3$  molecules at night when the ozone reaction stops. Thus urban areas with high  $\text{NO}_x$  sources often have a reduction in ozone concentrations overnight. In contrast, rural areas often maintain steady concentrations of ozone after dark and over time, as there is not a sufficient amount of  $\text{NO}_x$  to scavenge it and the other sinks of  $\text{O}_3$ , such as dry deposition, occur much slower (Gregg et al. 2003). These dynamics likely account for the spatial patterns of higher ozone concentrations in the downwind rural areas of the Phoenix metropolitan region. Also, the average wind direction in the region is from west to east, and there are also daytime anabatic winds which push ozone out of the urban areas and up against the mountains east of the urban valley (Ellis et al. 1999). A nighttime katabatic wind will drain some of the pollution back into the urban area, but the long-term effect is to have higher ozone concentrations in the eastern mountains.

These results confirm our hypothesis that ozone is a regional-scaled pollutant with long-distance ranges for spatial autocorrelation (i.e. more uniform across space), at least over the longer seasonal and monthly temporal scales. At the shorter 8-h and, especially, 1-h scales, however, this hypothesis is no longer valid as ozone exhibits short-ranged patterns more strongly influenced by local factors. A key message here is that the spatial patterns of ozone do change with temporal scales.

#### Changing $\text{PM}_{10}$ patterns on different temporal scales

The spatial patterns of  $\text{PM}_{10}$  also varied with temporal scales and between study years. In particular, major scale effects occurred between summer and winter months, with summer showing a much higher rural-to-urban pollution gradient than winter. The correlation analysis showed that  $\text{PM}_{10}$  concentrations had little correlation over long distances at the monthly scale, and this result was corroborated by the generally much shorter ranges from semivariance analysis. Once again, meteorological factors and source locations were likely the dominating determinants for the patterns of  $\text{PM}_{10}$ .

$\text{PM}_{10}$  is not as easily transported as finer particles because it is heavier and tends to settle out of the atmosphere sooner (Chung et al. 2012). Nevertheless, some meteorological conditions such as wind speed and relative humidity have a strong effect on  $\text{PM}_{10}$  concentration levels, as well as the strong influence that nearby sources in the Phoenix valley have on  $\text{PM}_{10}$  concentrations (Wise and Comrie 2005). However in the wintertime, the southwestern deserts are often subjected to atmospheric stagnation events. The atmosphere over this desert region is typically dry and cool during the winter, and as sunset approaches, the ground surface begins to cool faster than the atmosphere above it. The rapid cooling of the ground and boundary layer atmosphere, resulting in temperature inversion, can create stable atmospheric conditions at low altitudes (Pardyjak et al. 2009). This nighttime temperature inversion also creates stagnant atmospheric conditions that contribute to trapping particulate pollution close to its sources (Pardyjak et al. 2009). As the Phoenix metropolitan area is geographically located in a valley, this effect is compounded and likely accounts for the smaller urban-to-rural gradient observed in the winter. According to Wise and Comrie (2005), with the typically dry atmospheric conditions in the region (summer and elsewhere), the observed patterns at the annual scale are likely due to the effect of local sources of  $\text{PM}_{10}$ .

In general, the spatial patterns of  $PM_{10}$  showed more consistency between years than we originally anticipated, but the considerable effects of temporal scale confirmed our hypothesis. Also, the results seem to support our hypothesis that  $PM_{10}$  is a local pollutant influenced mainly by nearby sources, though we found that seasonal meteorology is as important to  $PM_{10}$  patterns. In addition, the winter to summer pattern dynamics were as informative as the spatiotemporal dynamics between different temporal scales.

### Sample size and kriging

The use of Kriging techniques when interpolating data from an air monitoring network with a small number of sampling sites has inherent risk involved. Kriging has reduced accuracy with small sample sizes and different alternatives to this method have been suggested (Diem 2003). For example, the study by Diem and Comrie (2002) specifically addressed the problem of a sparse sample size by using a linear regression model to improve the accuracy of the interpolation. However, linear regression models have their own disadvantages, such as the necessity of significant resources and high-quality data (Diem and Comrie 2002). Although we recognize the problems with Kriging to create accurate high-resolution pollution surfaces with a small sample size, this study has focus primarily on the landscape-level pattern and its changes between temporal scales. As such, we believe that our results are adequately robust for this purpose.

### Implications of scale in air pollution analyses

The findings of this study have important implications for the design and evaluation of air pollution monitoring networks in large urban regions. In general, the temporal scale of observation and analysis may substantially affect what air pollution patterns will be revealed. These scale effects, if not adequately understood, may influence people's perception and misguide governmental policy decisions. To overcome this problem, researchers and decision makers need to better understand the multi-scale patterns of air pollution in time and space, and this scale multiplicity must be considered explicitly in designing or evaluating air monitoring networks.

More specifically, air pollution monitoring networks should be designed so that both grain size (the spatial and temporal resolutions of the monitoring network) and extent (the time duration and spatial expanse of the network) are appropriate. For example, in the US, much emphasis is often placed upon a community or region to comply with Federal air pollution health standards, with each standard having differing averaging intervals such as annual, 24 h, or 8 h. If the region's government focuses on only a few single sites or local areas that are exceeding specific standards, the density of monitors may be much higher than the rest of the region (Nejadkoorki et al. 2011). This may lead to a deficient monitoring network that is unable to capture the spatiotemporally heterogeneous patterns of air pollution over the entire region. With these implications in mind and building upon the results from this study, we are now conducting a comprehensive evaluation of the air pollution monitoring network in the Phoenix metropolitan region, which will identify its deficiencies and redundancies based on integrated data on environmental settings, demographics, and air quality measurements.

Scale multiplicity of air pollution patterns may also affect environmental justice research. The studies of environmental justice, or equity, seek to identify unique socioeconomic population groups exposed to disproportionate amounts of pollution risk. As shown in this study, pollution patterns may change when the temporal scale of analysis is changed. For example, if an environmental justice study only utilizes peak 1-h values to find populations

affected by acute pollution exposure, it risks missing those population groups affected by chronic exposure to monthly or annual pollution patterns. To cope with this problem, a multi-scale approach is needed (Wu 2004, 2007). Part of our ongoing research is to take such an approach, and as such, we are using the multi-temporal scale kriging results from this study to explore a number of environmental equity-related research questions in the Phoenix metropolitan region. For example, do certain population groups experience disproportionately higher pollution risks? How would the detection of such potential environmental injustices change with the scale of analysis?

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