Spatial-Temporal Distribution of Tropospheric Ozone in the Carolina Piedmont Megapolitan Area

Bradley J.F. Bereitschaft, University of North Carolina Greensboro

Tropospheric ozone, a key component of photochemical smog, is a significant hazard to biological systems. Ozone is of particular concern in highly populated urban regions where large numbers of people may be exposed to unsafe concentrations, resulting in impaired respiratory function and an increased risk of heart disease. This article explores the spatial-temporal distribution of tropospheric ozone within the Carolina Piedmont megapolitan area over the ten-year period 1998 -2007. Analyzing both the spatial distribution and temporal trends of tropospheric ozone levels at this scale is particularly useful for understanding how large urban agglomerations influence regional air quality through the "sharing" of air pollution. The influence of various localized variables known to affect the surface concentration and distribution of ozone, including climatic conditions and the emission of point and non-point precursor chemicals, also investigated. were

Introduction

Tropospheric ozone, a key component of photochemical smog, is a significant hazard to biological systems (Burnett et al., 1994; Krupa et al., 1995). Known to impair respiratory function and increase certain risks associated with heart disease, ozone is of particular concern in highly populated urban regions (Bell et al., 2004; Schlink et al., 2006). In an effort to mitigate this hazard, considerable research has been conducted to understand the factors that contribute to the formation, movement, and distribution of ground-level ozone.

Since the dawn of the industrial age, anthropogenic production and release of ozone precursor emissions (primarily nitrogen oxides (NOx) and volatile organic compounds (VOC)) from point (e.g., industrial operations, power plants) and non-point sources (e.g., vehicular exhaust, building emissions) has contributed significantly to rising tropospheric ozone levels (Berntsen et al., 1997; Syri et al., 1999). Tropospheric ozone is readily produced in the atmosphere when nitrogen oxides and VOCs undergo a series of chemical reactions in the presence of sunlight (Atkinson, 2000). The

photochemical production and spatial distribution of tropospheric ozone, however, is not only influenced by abundance of precursor emissions, but also site-specific topography, wind speed and direction, intensity of ultraviolet (UV) radiation, temperature and other local climatic variables (Fuglestvedt and Jonson, Guicherit and 1995: Roemer. 2000: Stathopoulou et al., 2008).

The distribution of ozone has been studied using a variety of techniques over a number of spatial scales. Caballero et al. (2007) estimated the spatial variability of ozone over a 5872 km² area along the southeast coast of Spain using multiple linear regression. Values of ozone concentration were estimated using regressed relationship between altitude, distance to precursor sources, and ozone. Liu and Rossini (1996) used the spatial interpolation technique of kriging to predict mean outdoor ozone concentrations at particular home sites within the Toronto metropolitan area. They found predicted ozone values obtained using kriging were more accurate than using a simple nearest-neighbor (proximity polygon) approach. Similarly, Matejicek, Engst, and Janour

(2006) used inverse distance weighting (IDW) and ordinary kriging to conduct a spatial analysis of ozone and NO₂ levels over Prague. In a review of ozone-mapping studies, Diem (2003) found that 19 of 50 studies used some form of kriging to produce ozone surfaces, making it the most popular modeling technique.

This study uses ordinary kriging to model the distribution of surface ozone at a newly conceptualized megapolitan region, a scale of urban agglomeration proposed by Lang and Knox (2008). Megapolitan regions include multiple metropolitan statistical areas and their surrounding countryside. My analysis focused on the Carolina Piedmont megapolitan region. one of twenty such regions defined by Lang and Knox (2008). The main objectives were to characterize the spatial and temporal trends in the distribution of tropospheric ozone within and around the Carolina Piedmont megapolitan area during the ten year period 1998 – 2007, and in doing so to better understand to what extent ozone levels in adjacent metropolitan areas may influence one another and the surrounding countryside.

Study Area and Methodology

The Carolina Piedmont megapolitan region spans 50 counties in North Carolina and South Carolina (Figure 1). The area includes census-defined urbanized micropolitan statistical areas, 10 metropolitan statistical areas (MSAs), and four adjacent combined statistical areas (CSAs) (Raleigh-Durham-Carv. NC. Greensboro-Winston-Salem-High Point, NC, Charlotte-Gastonia-Salisbury, NC, and Greenville-Spartanburg-Anderson, SC). At the heart of this region of near contiguous urbanization is the Interstate-85 corridor that facilitates intra-urban travel between major urban nodes within the region and inter-urban travel between the Carolina Piedmont and its neighboring megapolitan regions: Atlanta, GA to the south, and Washington-Baltimore, VA/MA to the north. The area is generally confined to the piedmont region of North and South characterized topographically by gently rolling hills, and located between the Appalachian Mountain range to the west and the Carolina

coastal plain to the east. The climate of the region is subtropical throughout with warm, humid summers, mild winters, and moderate annual precipitation.

As of 2007, the projected population of the Carolina Piedmont region was 7,353,520, an increase of approximately 14 percent from 2000 (derived from U.S. Census data). This growth is twice the rate of the national average of 7 percent over the same period. Not all areas of region. however, have experienced population growth. Seven counties have had negative growth over the last eight years. These counties line the periphery of the study area and are predominately rural. Counties with the greatest gains in population are in close proximity to the I-85 corridor or are located within the region's four CSAs. These four CSAs serve as the major population nodes within the Carolina Piedmont. Thus, while the region is growing as whole, there also appears to be a significant rural to urban shift in population.

2.2 Data Acquisition

Ozone data was obtained from the Environmental Protection Agency's (EPA) AirData online database. AirData provides a user-friendly interface to query summaries of air pollution measurements throughout the United States. The database contains both emission data (amount of pollutants released annually from point and nonpoint sources) and ambient pollutant concentration data gathered at over 4000 certified monitoring stations. Annual countylevel ozone precursor emission data was obtained for 1990 and 1996 - 2002, point-source precursor emission data for 1990, 1996, 1999, and 2002, and complete monitor-specific ozone concentration data from 1998 - 2007. These years included the full range of emission data available through AirData.

A total of 71 ozone monitoring stations were used, including 43 within, and 28 surrounding, the Carolina Piedmont study area (Figure 1). The additional 28 monitors were used to eliminate edge effects within the study area and to understand the distribution of ozone immediately beyond the borders of the Carolina

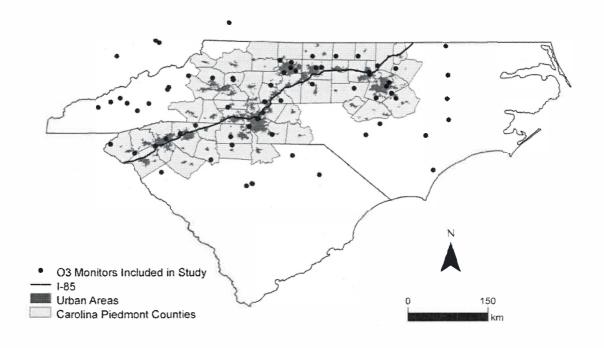


Figure 1. Carolina piedmont counties and urban areas included in the megapolitan regional study area

Piedmont. Data regarding annual 4th maximum 8-hour ozone concentrations and number of annual 8-hour ozone exceedances for the years 1998 through 2007 were obtained for each monitoring station for the period in which they operated (not all stations were in operation for the full 10 years). Annual ozone exceedances refer to the number of 4th highest 8-hour ozone concentrations that were greater than the EPA standard threshold of 0.075 ppm.

Additional climatic data regarding regional maximum temperature, average temperature, and precipitation was obtained by special request through the Southeast Regional Climate Center (SERCC). SERCC provided annual climate averages for North and South Carolina for the years 1998 through 2007. Using the Geostatistical Analyst extension in ArcGIS,

ordinary kriging was used to produce ozone surface layers indicating the spatial distribution of 4th maximum 8-hour ozone concentrations across the Carolina Piedmont megapolitan area for every other odd year from 1999 through 2007 (Figure 2). The density of ozone monitors within the Carolina Piedmont provided low prediction standard errors relative to the area outside. Where the distribution of ozone exhibited anisotrophy or noticeable spatial spatial model was trends, the adjusted accordingly. The semivariance of ozone concentrations in 1999, for example, varied considerably between the northwest-southeast direction and the northeast-southwest direction, indicating anisotrophy. The distribution of ozone also exhibited a secondary trend with ozone concentrations greatest near

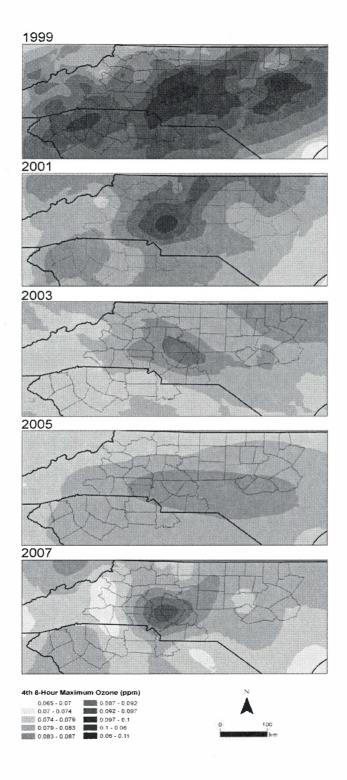


Figure 2. Annual 4th maximum 8-hour ozone concentrations every odd year 1999-2007.

the center of the study area and diminishing toward the edges. This calibration process, carried out within Geostatistical Analyst for every surface created, was used to minimize prediction errors when possible.

Ordinary kriging was also used to produce a surface of average number of annual ozone exceedances during the period 1998-2007. Ozone exceedance indicates what areas are not in compliance with EPA ozone

standards. According to the EPA's 2008 standards, an exceedance occurs when the 4th highest ozone concentration over an 8-hour period is above 0.075 ppm. (Stone, 2008) The location and magnitude of VOC and NOx precursor emissions from known point sources (e.g. factories, power plants) are shown in Figures 3 and 4 respectively.

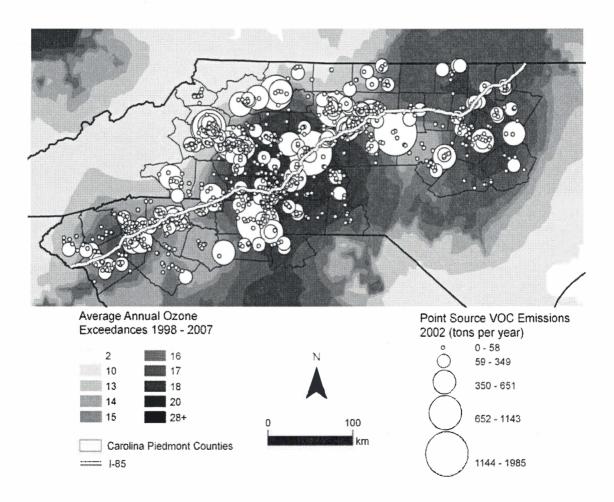


Figure 3. Average annual ozone exceedances 1998-2007 overlain with the location and magnitude of point-source emissions of volatile organic compounds (VOCs) in 2002.

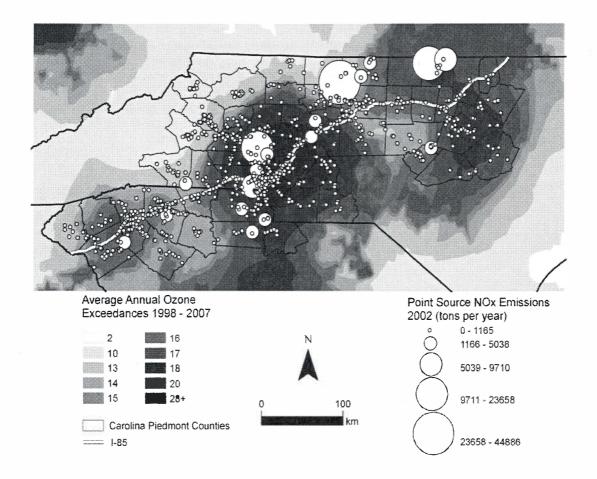


Figure 4. Average annual ozone exceedances 1998-2007 overlain with the location and magnitude of point-source emissions of nitrogen oxide (NOx) in 2002.

The emissions data was obtained for 2002, the last year such data was available. The point sources were mapped on top of a background layer of average annual ozone exceedances to allow visualization of the potential spatial relationships between precursor emissions and ozone levels. These two figures however, do not include non-point source emissions (e.g. vehicle tailpipe emissions), which are far more spatially diffuse. Figure 5 indicates total precursor emissions (point and non-point source) by county in 2002, with an overlay layer of average ozone concentrations.

In order to identify highly populated areas exposed to numerous ozone exceedances over the period 1998-2007, it was first necessary to produce a fine-grained map detailing the distribution of population throughout the Carolina Piedmont region. An intermediate map was produced using census-block level population data (2000) for the entire bi-state region. Highly populated areas, consisting of census blocks with greater than 386 people per square kilometer (the census definition of urban) were used to produce an urban map layer. A map of ozone exceedances for the

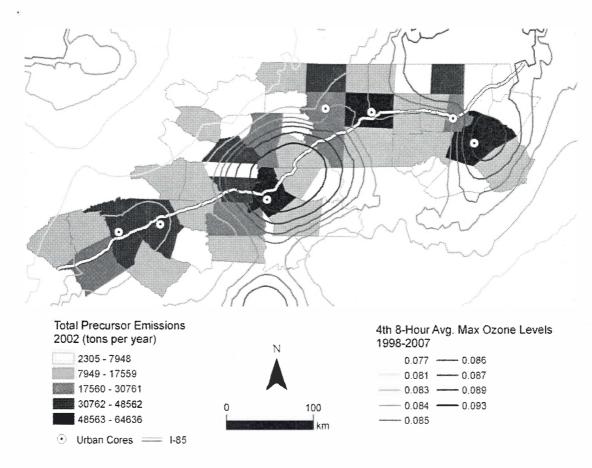


Figure 5. Total precursor emissions by county with an overlay map of average ozone concentrations 1998-2007.

period 1998-2007 was then used as an overlay to identify urban exposure to ozone exceedences.

Discussion

From 1998 to 2007, the average 4th maximum 8-hour ozone concentration recorded throughout the Carolina Piedmont decreased from 0.0965 ppm to 0.0821 ppm, a 17.5 percent reduction (Figure 6). By comparison, the national average 4th maximum 8-hour ozone concentration also decreased markedly from 0.0857 ppm to 0.0763 ppm, a 12.2 percent reduction. Similarly, the number of 8-hour ozone exceedances decreased from 1998 to

census blocks exposed to ozone exceedances.

2007 in the Carolina Piedmont (36.6 to 13.7) and throughout the U.S. (17.3 to 7.5). In the Carolina Piedmont region, the average annual ozone concentration ranged from a high of 0.0963 ppm in 2002 to 0.0745 ppm only two years later in 2004. Average annual ozone concentrations in the Carolina Piedmont were consistently higher than the national average.

Figure 2 provides a visualization of the changes in ozone concentrations within the Carolina Piedmont region from 1999 to 2007. As illustrated in Figure 6, ozone concentrations over this time period exhibit a general decline,

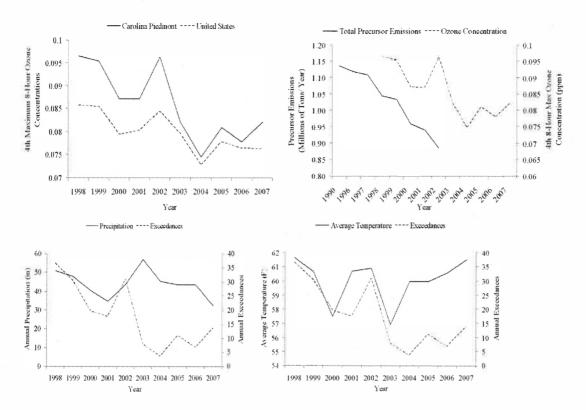


Figure 6. Summary of piedmont ozone concentrations vs. U.S., precursor emissions vs. ozone concentration, and precipitation and temperature in comparison to annual ozone exceedences.

though they spiked briefly in 2002. It is clear that there existed three "hotpspots" of relatively high ozone concentrations in 1999. These ozone hotspots were generally located over the Greenville-Spartanburg, Charlotte, and Raleigh-Durham metropolitan areas. By 2001, overall ozone concentrations had decreased, with one well defined node of high ozone concentration over the Charlotte area. This area of high ozone extends northward into Greensboro-Winston-Salem-High Point, likely affecting the area's annual ozone concentrations and the number of ozone exceedances. Charlotte remains the main source node of ozone concentrations in 2003, 2005 and 2007, though concentrations are noticeably reduced in 2003 and

2005. It is possible to better identify and gauge the impact of these ozone source nodes by observing ozone levels averaged over the entire 1998-2007 period. Figures 3 and 4 indicate that the highest levels of ozone exist over the Charlotte metropolitan area, with secondary nodes over Raleigh-Durham (the "Triangle") and Greenville-Spartanburg. The effective ozone airshed of Charlotte extends well beyond the borders of its municipal or metropolitan boundaries. Both point-source and non-point source emissions of ozone precursors (NOx and VOCs) decreased from 1990 to 2002. During this period, point-source emissions decreased by 17 percent, while non-point source emissions decreased by 35 percent.

In addition, 45 of 50 counties within the Carolina Piedmont experienced a decrease in precursor emissions. Although available data overlaps only over a five year period, both total precursor emissions and average ozone concentrations exhibit an overall decline (Figure 6). The location of point-sources of VOCs and NOx in 2002 can be observed in Figures 3 and 4 respectively. The largest number of point sources and greatest emissions appear to be clustered around the I-85 corridor, where industrial activity is most concentrated. Counties with the highest overall emissions are generally located around urban core areas where both elevated levels of industrial and vehicular activity take place. Counties with the highest non-point precursor emissions are all highly urbanized and located within the region's four largest metropolitan areas. In most counties, a significant majority (>70 percent) of ozone precursor emissions are from non-point sources. making their contribution to local ozone concentrations of particular importance (unpublished data, EPA, 2002). The spatial relationships between total precursor emissions by county and average ozone concentrations (1998-2007) are illustrated in Figure 5.

The urban areas exposed to the greatest number of average annual ozone exceedances period over the 10 year study are generally located within the northeast quadrant of the Charlotte metropolitan Northeast area. Charlotte. Kannapolis, and portions Huntersville, experienced an average 25 or more ozone exceedances per year. This area represents approximately 6 percent of the Carolina Piedmont's population. Outside the Charlotte area, most urbanized clusters within the Carolina Piedmont (representing approximately 75 percent of the population) have experienced between an average of 10 and 20 ozone exceedances per year. Less than one percent of the population, located at the far-west edge of the study area, experienced less than ten 8-hour exceedances per year.

The link between climatic fluctuations and annual ozone levels were also explored. Figure 6 indicates fluctuations in average annual

precipitation within the Carolina Piedmont megapolitan area. When compared with the changes number of ozone in exceedances, it appears that the two variables have a negative relationship (i.e., as annual precipitation increases, ozone exceedances decrease). As annual temperature increases. number of ozone exceedances also increase 6). The relationship temperature and ozone exceedances begins to widen after 2003, possibly as a result of the decrease in precursor emissions. Nevertheless. positive association between annual temperature and ozone exceedances is largely maintained throughout the 10 year period.

Conclusions

The spatial distribution of ozone throughout the Carolina Piedmont area is clearly influenced by the concentration of human settlement and activity. This is expected given that the majority of ozone precursor emissions are from nonpoint sources, which are most abundant in highly-populated urban areas. Produced in lower quantities, the impact of point-source emissions, such as those from factories, power plants, and other industrial processes, are likely have less impact on local concentrations.

The use of kriging to estimate annual ozone concentrations and exceedances has provided an effective means of visualizing the distribution of ozone within and between the metropolitan areas of the Carolina Piedmont. The spatial distribution of ozone exceedances indicates that the Charlotte area experienced the highest levels of tropospheric ozone in the bistate region. Ozone produced over Charlotte, however, was not confined to the area's urban core, but rather extended well into neighboring urban areas as well as the surrounding countryside. The "Triad," a combined statistical area of 1.5 million approximately 100 km northeast of Charlotte, is close enough to receive considerable amounts of transported ozone and ozone precursors from Charlotte. Though the interaction of airsheds is highly variable on an hourly to annual basis, and

dependent on prevailing wind directions and weather patterns, these data suggest that considerable ozone "sharing" occurs between individual metropolitan areas within the Carolina Piedmont. The ozone models created in this study, however, are only snapshots of annual variations and cannot account for the full range of inter-urban ozone transportation that might occur under specific daily, monthly, or seasonal conditions.

The reduction in ozone concentrations and number of ozone exceedances within the Carolina Piedmont region between 1998 and 2007 are likely related to both a general decrease in precursor emissions as well as fluctuations in climatic conditions, including annual temperature and precipitation. While the overall trend is towards a reduction in ambient ozone levels at the surface, which is expected given the reduction in precursor emissions, it is possible to see that prevailing weather conditions can upset this trend on an annual basis. For example. in 2002 ozone concentrations and ozone exceedances spike to their highest level during the 10-year period, despite a decrease in ozone levels over the previous four years. Precursor emissions, however, continue to decrease in 2002 as they had for the previous seven years. The elevated ozone levels experienced in 2002 may have resulted from the brief spike in temperature and lower precipitation totals over the previous one to two years.

It is interesting to note that while the population in the Carolina Piedmont rose by 14 percent from 1998 to 2007, non-point source ozone precursors (produced primarily from mobile sources) actually declined by 17 percent (at least during the five-year overlap period 1998-2002; see Figure 6). Given that one would expect emissions to increase with a rise in population and associated vehicular traffic, the decline in non-point source emissions suggests that more stringent vehicle emissions standards applied by the EPA in 1994, and again in 2004, are having a beneficial effect. The current 6-EPA-reported point-source year lag in emissions, however, precludes comparison with the most recent ozone concentration and exceedances data, which are updated monthly.

One of the benefits of kriging is the ability to obtain prediction errors at every location within the predicted surface. The prediction standard error for the average 4th maximum 8hour ozone concentration (1998-2007) ranged from 0.00435 ppm to 0.00656 ppm. The most reliable predictions are primarily within the bounds of the Carolina Piedmont megapolitan area, where the majority of ozone monitors are located. Given the size of the study area, it would have been ideal to have more than 71 data points, though unfortunately no additional sources of ozone data are known. This suggests the difficulty of accurately producing ozone surface layers at smaller scales, such as the city or metropolitan-level, with the available data.

This case study demonstrates that the megapolitan region is a useful scale at which to study the distribution of tropospheric ozone. Though tropospheric ozone and other air pollutants can disperse great distances, there is clearly a zone of influence within which a particular urban center can significantly impact the air quality of the surrounding region. Urban areas that produce very little ozone can still experience numerous ozone exceedances and high ozone concentrations due to the influence of neighboring communities. This is of particular relevance in megapolitan regions where inter-urban dispersion of ozone and the "sharing" of air pollution is common. From an air quality perspective, it is therefore reasonable to investigate these large urban agglomerations as a single functional unit, in order to better understand the movement and distribution of pollutants as well as the underlying factors that influence their production.

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