



## A comparison of trajectory and air mass approaches to examine ozone variability

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### ABSTRACT

Back trajectory analysis is a commonly-used tool for understanding how short-term variability in surface ozone depends on transport into a given location. Lesser-used but equally effective methods are air-mass based approaches that are primarily driven by changes in temperature and humidity conditions. We compare and combine these two fundamentally different approaches by evaluating daily near-surface afternoon warm-season ozone concentrations from 2001 to 2006 in and around the Shenandoah Valley of Virginia. Analysis of variance is used to compare summer afternoon ozone levels between air masses as identified by the Spatial Synoptic Classification to clusters of 72-h back trajectories estimated by the HYSPLIT model.

Ozone concentrations vary significantly across both air masses and trajectory clusters at all ozone monitors. Concentrations are highest for air masses characterized by dry, warm conditions and for air originating from the north and west of the study area or circulating over the mid-Atlantic region. In many cases, the interaction between synoptic types and back trajectory clusters produce results not evident from the examination of simple trajectories or air masses alone. For example, ozone concentrations on Moist Moderate days are 30 ppb higher when air parcels travel moderate distances into the Shenandoah Valley from the west than when they travel longer distances from the north or northeast. Combining air mass and trajectory approaches provides a more useful characterization of air quality conditions than either method alone.

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### 1. Introduction

Day-to-day surface ozone variations at a given location are influenced by a variety of meteorological factors, including advection of ozone from upstream sources and *in situ* production. Ozone transport is often examined by tracing the trajectory of a hypothetical air parcel into the location of interest (e.g., Jiang et al., 2003; Taubman et al., 2006; Suthawaree et al., 2007; Delcloo and De Backer, 2008). These back trajectories are primarily calculated from the observed wind/pressure field and often have been used as the basis for ozone climatologies (Moody and Galloway, 1988; Dorling et al., 1992a,b; Moy et al., 1994; Dorling and Davies, 1995; Moody et al., 1995, 1998; Harris and Oltmans, 1997; Brankov et al., 1998; Eneroth et al., 2003).

Although air flow impacts ozone levels, ozone production depends on a variety of other meteorological factors, such as actinic radiation levels, temperature, and humidity (e.g., Comrie, 1990; Comrie and Yarnal, 1992; Liu et al., 1994; Poissant et al., 1996;

Xu et al., 1997; Seinfeld and Pandis, 1998), factors that are not explicitly considered with back trajectory approaches. Air mass classifications use a suite of meteorological variables as inputs, including both wind and thermal-moisture variables. Air mass-based approaches have been successfully applied to examine ozone variability in the United States (Comrie and Yarnal, 1992; Greene et al., 1999; Lennartson and Schwartz, 1999; Rohli et al., 2004), Canada (Heidorn and Yap, 1986; McKendry, 1994; Cheng et al., 2007), the United Kingdom (McGregor and Bamzels, 1995), New Zealand (Khan et al., 2007), Hong Kong (Tanner and Law, 2002), and Taiwan (Cheng et al., 2001). However, air mass approaches are not used as often as trajectory-based methods in air quality research. Despite the geographic diversity of these studies, some climatological consistency is evident. High ozone events tend to be associated with the approach of a slow-moving anticyclone from the west (Comrie, 1990; Cheng et al., 2007) and linked to high temperature, low humidity conditions and clear skies (Greene et al., 1999; Ellis et al., 2000). Some cases even report locally high ozone linked to specific orographic effects that create a subsidence inversion (Cheng et al., 2001; Tanner and Law, 2002). The impact of wind direction tends to vary by location because of the presence of high ozone regions nearby, advection of air masses associated with

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high ozone levels, and unique local topographic factors (e.g., Lenartson and Schwartz, 1999; Greene et al., 1999; Ellis et al., 2000; Rohli et al., 2004).

Our goal is to examine, both separately and in combination, the efficacy of air mass and back trajectory approaches in accounting for afternoon near-surface ozone concentrations in Virginia. Our study is focused on the Shenandoah Valley of Virginia and surrounding areas as a component of the larger Shenair Initiative, a multi-institutional effort to explore air quality variability in the region. Using data from ozone monitors in the Shenandoah Valley and from stations throughout Virginia, we examine the variability of ozone based upon the resident air mass, the general characteristics of the 72-h back trajectory terminating on that day, and the combined effects of both air masses and trajectories.

## 2. Data

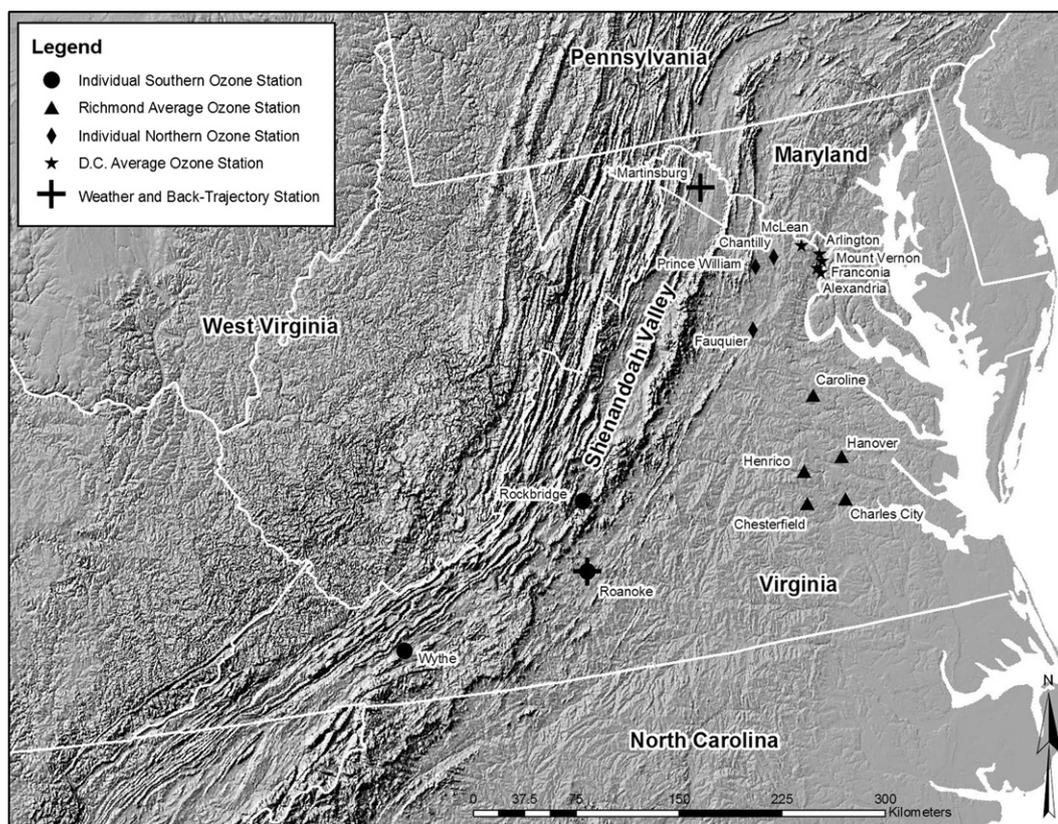
The Shenandoah Valley of Virginia is a 320-km long northeast-to-southwest oriented valley extending from the eastern West Virginia panhandle southwestward to north of Roanoke, Virginia (Fig. 1). The Valley is predominantly rural with an overall population of approximately one million. The hourly ozone data used in this study were obtained from the Environmental Protection Agency's Air Quality System database, an on-line archive of ambient air quality monitor observations (<http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm>). Because ozone production depends in part on photochemical reactions, ozone has strong diurnal and seasonal components. In an effort to limit these influences, we sampled ozone at 1800 UTC (1400 local time) from April 1 through October 31, 2001–2006. This period of record was selected for consistency with a different component of the larger

Shenair project that involves relating air quality to respiratory health in the region.

Ozone data were available from 16 monitoring stations in and around the Shenandoah Valley (Fig. 1). One topic of interest to the Shenair Initiative is the extent of regional ozone transport into the Valley as compared to local ozone production. Two potential external sources for transport include the Washington, D.C. and Richmond metropolitan areas. Although both cities are east of the Valley and prevailing winds are westerly, easterly transport will occur given the appropriate synoptic conditions. To simplify portions of our analysis, we averaged the ozone readings from Alexandria, Arlington, Franconia, McLean, and Mount Vernon into the “Washington” group and the observations from Caroline, Charles City, Chesterfield, Hanover, and Henrico into the “Richmond” group. These stations were grouped because of their proximity and statistically significant inter-station Pearson correlations (greater than 0.89 for the Washington group and from 0.77 to 0.89 for Richmond). The six remaining monitors were analyzed individually with Chantilly, Fauquier, and Prince William linked to a first-order weather station in the northern Shenandoah Valley and Roanoke, Rockbridge, and Wythe associated with a southern valley station.

On a station-by-station basis, average 2 p.m. warm-season ozone concentrations vary from about 47 to 54 ppb (Table 1). Over the period of record, all of the stations recorded values over 100 ppb except for the two rural, southern monitors. We selected monitoring sites based upon data completeness (all stations were at least 95% complete). Station-days with missing observations were excluded from the analysis.

Average monthly ozone levels are comparable from April through August, with a slight increase in mid-summer (Fig. 2a).



**Fig. 1.** Map of meteorological stations and ozone monitors used in this study. The ozone monitors indicated by stars are combined into an average value for Washington, D.C. and the monitors indicated by triangles are averaged for Richmond, Virginia. The remaining ozone monitors are examined individually.

**Table 1**

Summary statistics for 2 p.m. ozone concentrations (ppb) for each monitor. The five locations under Washington, D.C. are averaged together into a single value for Washington and the five stations under Richmond, VA are similarly averaged.

Station	Number	Percent missing	Minimum	Maximum	Mean	Standard deviation
Washington, D.C.	1284	0.00	0.80	130.00	50.65	21.42
Alexandria	1239	3.50	0.00	138.00	47.49	21.76
Arlington	1226	4.50	0.00	149.00	51.62	22.99
Franconia	1188	7.50	0.00	137.00	52.94	22.21
McLean	1257	2.10	1.00	131.00	50.00	21.16
Mount Vernon	1270	1.10	1.00	145.00	51.61	22.23
Chantilly	1260	1.90	1.00	117.00	50.37	19.10
Prince William	1237	3.70	1.00	113.00	50.16	18.39
Fauquier	1251	2.60	0.00	105.00	46.86	16.58
Richmond, VA	1280	0.30	5.00	113.80	52.01	17.05
Caroline	1232	4.00	2.00	115.00	51.10	17.53
Charles City	1238	3.60	4.00	134.00	51.87	18.52
Chesterfield	1252	2.50	2.00	123.00	51.03	17.71
Hanover	1267	1.30	5.00	122.00	53.71	18.62
Henrico	1250	2.60	2.00	137.00	52.29	19.22
Rockbridge	1260	1.90	2.00	91.00	47.21	14.88
Roanoke	1273	0.90	3.00	110.00	50.79	16.51
Wythe	1263	1.60	1.00	99.00	50.23	14.13

There is a precipitous decline in ozone in September and October, however. The lack of symmetry about the summer incoming radiation or temperature maxima suggests that additional physical–chemical processes are responsible. During summer, the highest average ozone values are observed at the northernmost monitors in Chantilly and Prince William. However, ozone values are typically lowest at these stations in spring and autumn. Ozone levels are higher in the more urbanized north from June–August, but the southern area has higher ozone levels than the north in April, May, and October (Fig. 2b). This same general pattern is reflected in the data for Washington and Richmond, in which Washington is similar to the overall northern valley pattern while Richmond is comparable to the southern stations.

Significant interannual variability is evident, with mean ozone concentrations about 10 ppb higher in 2001 and 2002 than the subsequent four years (Fig. 3a). In this case, there is no difference between northern and southern valley stations during the high ozone years (Fig. 3b). Climatologically, Virginia suffered through a prolonged and spatially extensive drought from 2001 through early 2003. The high solar radiation levels coupled with high temperatures typical of summertime droughts in this region produced an environment conducive to ozone production. A period of heavy rainfall began in spring, 2003, which, coupled with rainfall from several tropical systems in late summer and autumn, resulted in 2003 being the wettest year on record throughout much of Virginia, effectively ending the drought rather quickly. Thus, 2003–2006 exhibited much lower mean ozone levels than the drought years.

The daily Spatial Synoptic Classification (SSC) was developed from four-times per day surface weather observations at Martinsburg, West Virginia and Roanoke, Virginia, representing the north and south valley, respectively. Data used in the model include air temperature, dew point temperature, sea-level pressure, the south–north and west–east components of the wind vector, and cloud cover. Although the SSC is developed for the entire year, we only use April–October data in this research because of our emphasis on ozone, which is generally much lower in the cool season. These data have very few missing observations (less than 3%)—days with incomplete data are not included in this analysis.

Air parcel back trajectories were calculated for Martinsburg and Roanoke from wind/pressure initialization fields of the Eta Data Assimilation System (EDAS). Ideally, we would use a consistent data

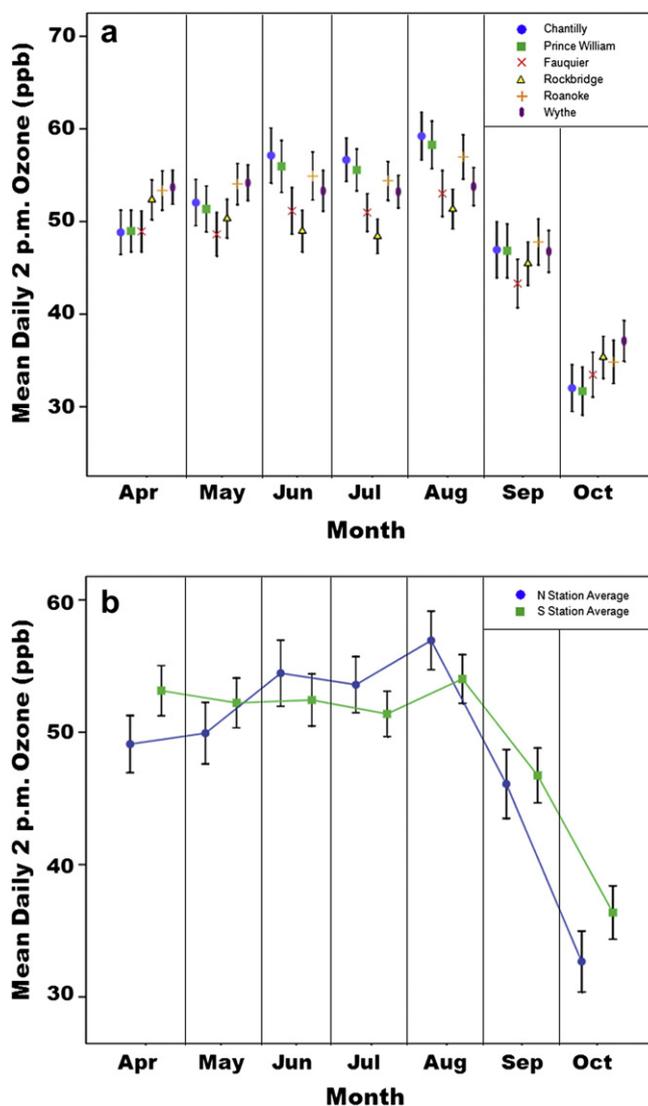


Fig. 2. Monthly variability of 2 p.m. daily ozone concentrations (ppb) for individual stations (a), the average of northern and southern stations (b).

set over our entire period of record, but in 2004 the spatial resolution improved from 80 km to 40 km. Accordingly, we compared the back trajectories at both resolutions during an overlap period from January 1–April 30, 2004. We found no statistically significant differences between the resulting trajectories. Therefore, we used the spatially finer data set to calculate all trajectories starting in 2004. Trajectories were calculated from 1997 to 2006 to provide a large sample size for subsequent analysis, but trajectories are only linked to ozone observations from 2001 to 2006. Subsequent analysis (see Section 3.2) required complete 72-h trajectories. From 1997 to 2006, complete trajectories were available 69% of days at Martinsburg and 73% at Roanoke, but for the 2001–2006 period, the trajectory data set was 87% and 91% complete, respectively.

### 3. Methods

#### 3.1. Spatial Synoptic Classification (SSC)

The SSC is a daily classification of weather types derived from surface weather observations (Sheridan, 2002, 2003). It is a hybrid classification by which each day in the record is classified based on

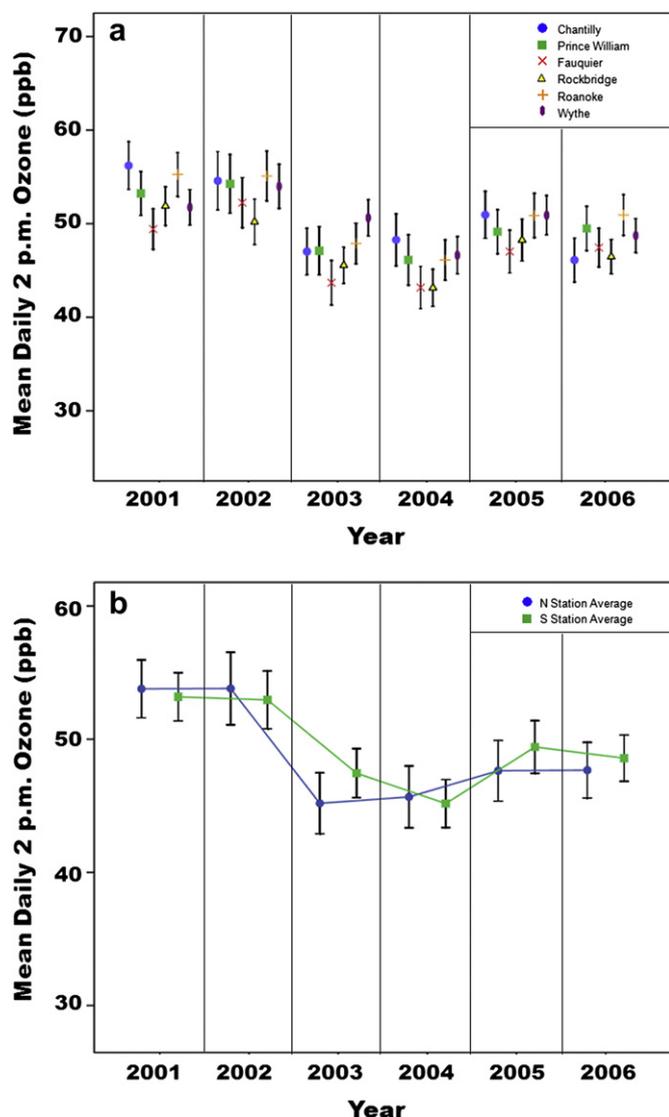


Fig. 3. Interannual variability, 2001–2006, of 2 p.m. daily ozone concentrations (ppb) for individual stations (a) and the average of northern and southern stations (b).

the proximity of the observations to a paradigm air mass type for that station and time of year. Similar to the classic system developed by Bergeron (1930), the SSC identifies six mutually distinct air mass types:

**Dry Polar (DP)**—a cold, dry air mass typically associated with a polar anticyclone advected into the region from the north. DP has seasonably cool and sunny conditions in the Shenandoah Valley in summer.

**Dry Moderate (DM)**—air associated with near-normal temperatures that typically arrives in the region from the west. In some cases, DM represents Pacific air that has dried and warmed adiabatically upon descent over the eastern Rockies.

**Dry Tropical (DT)**—although DT is traditionally linked to air that originates in a desert source region, the Shenandoah Valley rarely receives pure DT air from the Sonoran/Chihuahuan Deserts of southwestern North America. In Virginia, DT is more commonly associated with air from the west or south that has undergone significant anticyclonic subsidence and has developed the hot and dry characteristics typical of desert air masses.

**Moist Polar (MP)**—a cool, humid air mass commonly associated with overcast skies and (frequently) precipitation. In the Shenandoah Valley, MP often occurs in conjunction with frontal overrunning and the advection of Atlantic air from the northeast or east to the north of the frontal boundary.

**Moist Moderate (MM)**—humid but warmer than MP air. In an idealized sense, MM air masses are found south of MP air but closer to the surface front. In reality, MM includes a variety of synoptic patterns that lead to moderate temperatures and elevated humidity.

**Moist Tropical (MT)**—warm, humid air that typically arrives from source regions in the Gulf of Mexico or the Atlantic Ocean. During summer, as warm, humid conditions prevail over much of the southeastern United States and frontal passages are limited, MT air becomes the predominant weather type in this region.

**Transition (TR)**—a day in which a frontal passage occurred and thus cannot be classified into a single air mass type. Transitions are identified based upon the change in dew point temperature, wind direction, and pressure over the course of a day, regardless of the direction of these changes.

Because the paradigm air mass conditions vary over the course of the year, the SSC allows for the identification of, for example, MT air in winter and DP air in summer. As such, it is a *relative* air mass classification that identifies synoptic types based upon the expected distribution at that time of year. Nonetheless, MT is still more common in Virginia in the summer than DP. For more details on the SSC methods and typical characteristics of each air mass, please see Sheridan (2002, 2003).

The SSC was calculated from archived weather observations taken at the first-order weather stations in Martinsburg and Roanoke from 1948 to present. We downloaded the daily SSC calendar for April–October, 2001–2006 from the SSC web site at <http://sheridan.geog.kent.edu/ssc.html>.

### 3.2. Back trajectories

Three-dimensional back trajectories were calculated using version 4.8 of the Air Resources Laboratory's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (Draxler and Hess, 2004). Wind initialization fields from the Eta Data Assimilation System were used as model input (Draxler and Hess, 2004; Draxler and Rolph, 2006). Trajectories were computed hourly for 72-h periods such that the trajectory terminated at Martinsburg or Roanoke at 1800 GMT (commensurate with the ozone observations) at an elevation 500 m above ground level. Only complete 72-h trajectories were included. HYSPLIT back trajectories provide a three-dimensional air parcel path into the receptor site (in our case, Martinsburg or Roanoke) as derived from the three-dimensional gridded wind/pressure field.

To facilitate comparison between the SSC classification and HYSPLIT trajectories, we used a two-stage cluster analysis technique to categorize the back trajectories into distinct groups, or clusters. Cluster analysis has been commonly used to classify natural trajectory groups (e.g., Moody and Galloway, 1988; Dorling et al., 1992a,b; Lee et al., 1994; Dorling and Davies, 1995; Cape et al., 2000; Berto et al., 2004; Jorba et al., 2004; Russell et al., 2004). The two-stage hierarchical/non-hierarchical approach was first employed in climatological analysis by Davis and Kalkstein (1990) in an effort to address some of the reproducibility and bias issues associated with simple hierarchical cluster analysis methods. In this research, each hourly trajectory endpoint consists of a latitude, longitude, and elevation. The horizontal components were converted to distances from each station, and the vertical component

was adjusted relative to the 500 m terminal elevation. Thus, each trajectory is characterized by 3 dimensions  $\times$  72 h or 216 independent variables. To insure that the variables were equally weighted in the subsequent classification, each variable was converted into z-score units by subtracting the mean from each observation and dividing by the standard deviation. Next, we employed complete linkage cluster analysis to 1) identify natural groups within the data set and 2) calculate seeds for the subsequent analysis. The number of clusters was determined using a variety of measures, including the Pseudo- $F$ , Pseudo- $T^2$ , and Cubic Clustering Criterion statistics. Finally, the means of each cluster (for each variable) were used as starting seed values for the subsequent convergent  $k$ -means cluster analysis. This non-hierarchical approach is an optimization strategy that allows for unrestricted reshuffling of observations between clusters until a convergence criterion is reached (in this case, no further reassignment of trajectories). Complete-linkage clusters that occurred less than 3% of the time were not used as seeds of  $k$ -means clusters, so our resulting groupings represented clusters of commonly-observed trajectories. For both cities, a 10-cluster solution was chosen for the final classification.

Clusters were computed using 72-h back trajectories for the entire year (rather than just the ozone season) because of the need to apply these results to other aspects of the larger Shenair Initiative. We nevertheless found that all 10 trajectory clusters occurred at each station between April and October.

To determine if the resulting trajectory groups differed from each other, we used Hotelling's  $T^2$  statistic on the two horizontal components and one vertical component of each trajectory every 6 h. This statistic compares the distances between trajectory groups at each 6-h endpoint (i.e., 6 h before termination, 12 h before termination, etc.). This method was applied so that all three spatial components were weighted equally. A Type I error rate of 0.05 was assumed for statistical significance.

One of the primary goals of this research is to examine linkages between trajectories and air masses with respect to ozone levels. These comparisons are made by linking each trajectory cluster to the air mass observed on the day the trajectory terminated. Thus, in the first 2 days of the 72-h trajectory, when in many cases the trajectory was not near the Shenandoah Valley, the air mass type was not considered. Although we would expect a different group of trajectories if a three-day cluster sequence at Martinsburg was DP–DP–DP as compared to MT–TR–DP, by examining every possible three-day combination we would have very small samples and little statistical power. We acknowledge that air mass and trajectory sequencing is an important issue that merits future study.

### 3.3. Ozone comparisons

As our dependent variable is continuous and our independent variables are categorical, we used a one-way analysis of variance (ANOVA) to assess ozone differences. ANOVA was used to test for: 1) ozone differences between SSC air masses, 2) ozone differences between trajectory clusters, and 3) ozone differences between air mass/trajectory group combinations. In the latter case, the combination of air mass and trajectory was incorporated via an interaction term. First, we ran Levene's test for homogeneity of variance across groups—if variances were different, then the sum of squares was adjusted according to the Welch and Brown–Forsythe tests. For cases when significant differences between groups were detected (i.e., the null hypothesis of equal means was rejected), we ran both the Bonferroni and Games–Howell *post-hoc* tests to identify which groups differed from which other groups (Field, 2005).

Separate analyses were run for each of the six individual ozone monitors in and around the Shenandoah Valley as well as for the

Washington and Richmond groups. The three northern monitors and Washington were linked to the SSC and trajectories at Martinsburg, and the three southern stations and Richmond were associated with Roanoke.

## 4. Results and discussion

### 4.1. Ozone and the SSC

Afternoon ozone concentrations differ significantly between SSC air mass types in the northern Shenandoah Valley (Fig. 4 and Table 2). Mean ozone levels approach 70 ppb under DT air masses but are less than 25 ppb for MP conditions. ANOVA *post-hoc* tests (as summarized in the last two columns of Table 2) show that DT (and sometimes MT) air masses have the highest ozone concentrations at each station, whereas MP is consistently linked to the lowest ozone levels. Furthermore, DM air typically has higher ozone levels, whereas MM air is associated with lower ozone levels. These results highlight the importance of both solar radiation and temperature in ozone production. Dry air masses are typically associated with less cloud cover, and tropical air masses are characterized by the highest temperatures (Hondula et al., 2009). Thus, DT air provides optimal conditions for high ozone levels—hot, dry air with high pressure resulting in clear skies and the potential formation of subsidence inversions that trap pollutants within the planetary boundary layer. Cool, moist, overcast air masses are the least favorable conditions for ozone production.

In general, ozone levels are quite consistent between stations within a given air mass. This reflects the regional consistency of ozone concentrations over space as well as the spatial generalizability of the SSC. The lone exception in the northern valley is for MT air in which ozone concentrations between Washington and Fauquier differ by 15 ppb. One possible explanation is the enhancement of ozone production in the presence of sea salts (advected in MT air) over Washington's urbanized environment. Chemical interactions involving high nitrous oxide levels near urban areas have been shown to increase the formation of ozone and its precursors on warm and humid days with some afternoon sunshine (Knipping and Dabdub, 2003; Tanaka et al., 2003). While the same process is also occurring in more rural areas, the lower

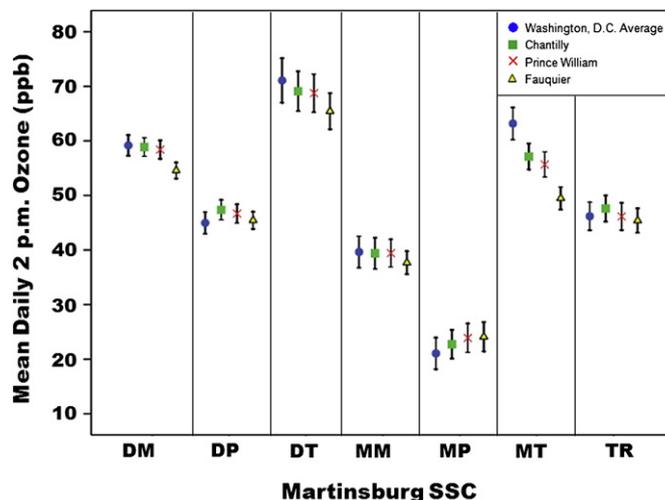


Fig. 4. Variability of ozone by SSC air mass at Martinsburg for the northern Shenandoah Valley. Error bars are 95% confidence intervals. Air mass abbreviations are as follows: DM = dry moderate, DP = dry polar, DT = dry tropical, MM = moist moderate, MP = moist polar, MT = moist tropical, TR = transition. Overall mean ozone levels are significantly different across air masses (Table 2).

**Table 2**

Analysis of variance results for ozone variability by SSC air mass. MSE is the mean square error. All of the results are statistically significant at the 0.05 level. Air masses are listed with respect to the descending value of ozone in column 5 with distinct groups identified by parentheses (e.g., for Washington, ozone is significantly higher for DT than for both MT and DM, which are not significantly different from each other). The number of significantly different subsets (SS) is listed in the last column.

Station	Number	MSE between	MSE within	Groupings by decreasing ozone values	SS
<i>Analysis of variance results for northern stations</i>					
Washington Average	1251	33 065.095	299.228	(DT), (MT,DM), (TR,DP), (MM), (MP)	5
Chantilly	1227	25 388.035	239.176	(DT), (MT,DM), OR,DP), (MM), (MP)	5
Prince William	1206	22 545.036	225.374	(DT), (MT,DM), (TR,DP), (MM), (MP)	5
Fauquier	1219	18 036.309	183.968	(DT), (DM,MT,DP), (TR,MM), (MP)	5
<i>Analysis of variance results for southern stations</i>					
Richmond Average	1275	18 515.09	203.221	(DT), (DM,MT), (DP,TR), (MM), (MP)	5
Rockbridge	1256	18 184.742	132.509	(DT), (DM), (MT,DP,TR), (MM), (MP)	6
Roanoke	1268	20 295.97	176.177	(DT), (DM), (MT), (DP,TR,MM), (MP)	5
Wythe	1258	12 013.855	142.529	(DT), (DM), (MT), (DP,TR,MM), (MP)	6

background concentrations of nitrous oxides do not result in markedly higher ozone production.

Results for the southern Shenandoah Valley are quite similar (Fig. 5 and Table 2). For all ozone monitors, DT has the highest concentrations and MP the lowest. Furthermore, the ordering of *post-hoc* air mass groups is quite similar between stations and in comparison to the northern valley results. In the south, the ozone differences between DT and MP are smaller, primarily because MP levels are up to 10 ppb higher (DT levels are comparable). As with Washington in the north, we also found a similar elevation of ozone concentrations in urbanized Richmond during MT events.

#### 4.2. Ozone and back trajectories

Two-stage cluster analysis identified 10 trajectory cluster groups for Martinsburg (Fig. 6). Hotelling's  $T^2$  test determined that these cluster distributions are significantly different from each other (Table 3). Clusters are primarily distinguished by the initial source region, trajectory length, direction of approach into the station (clockwise, counterclockwise, or neutral; ascending or descending), and the extent of recirculation over the region. To aid in our discussion and presentation, we used these attributes to assign a working name to each cluster. Furthermore, we ordered the clusters beginning with southwesterly flow into the station and sequentially rotated subsequent clusters clockwise (i.e., the first cluster is southwest flow, the next is west, etc.). The last three

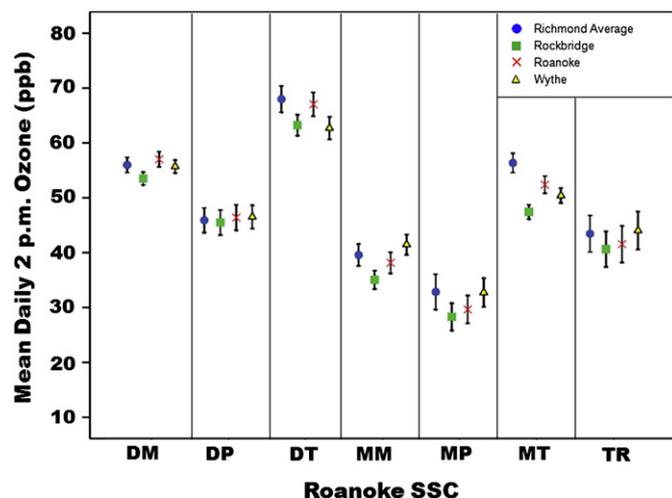


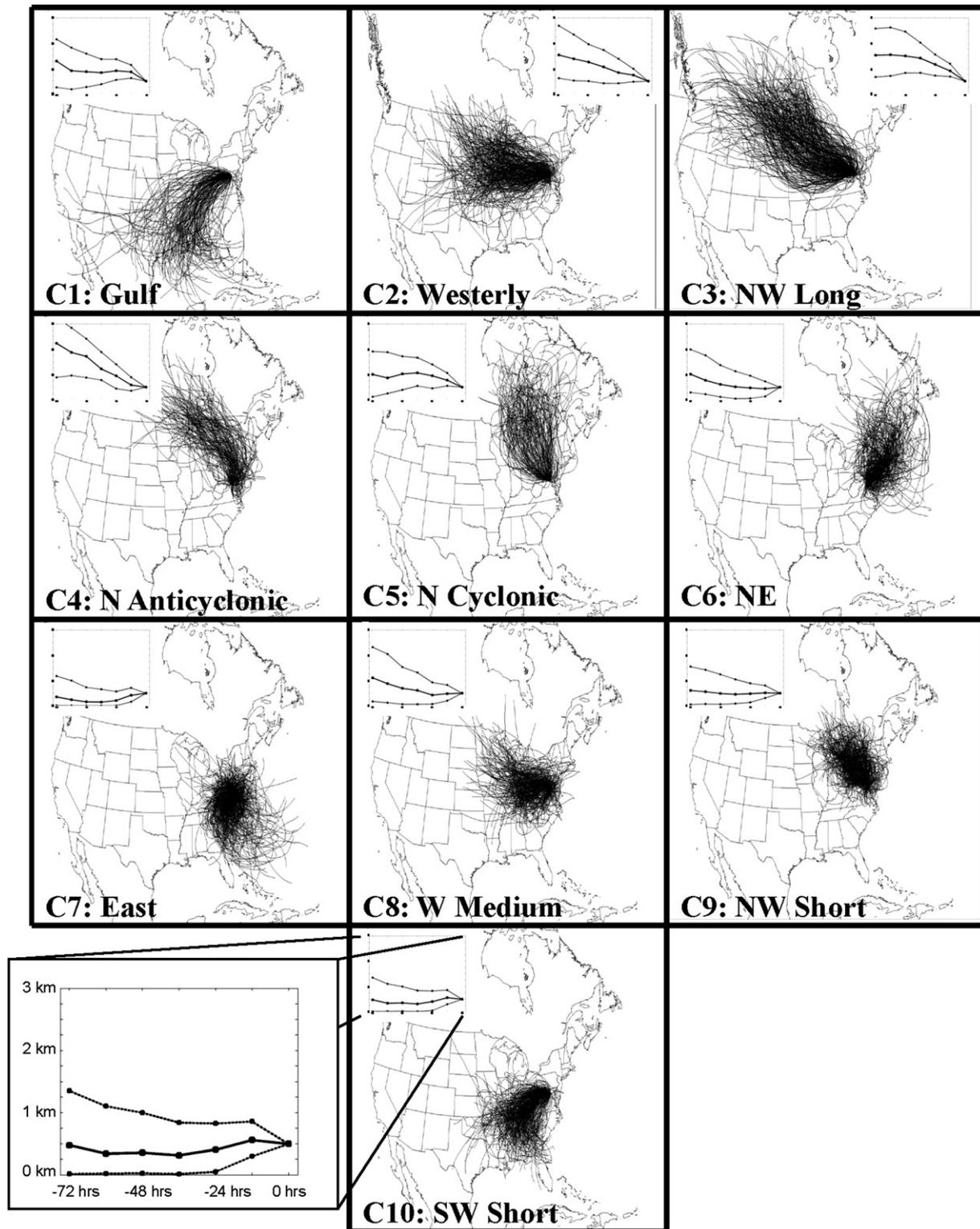
Fig. 5. Variability of ozone by SSC air mass at Roanoke for the southern Shenandoah Valley (as in Fig. 4). Overall mean ozone levels are significantly different across air masses (Table 2).

clusters (West Medium, Short Northwest and Short Southwest) are associated with more localized flow and recirculation of air with weak vertical motion over the study region—a synoptic situation commonly linked to a nearby, slow-moving or stationary anticyclone.

Visual examination of the Martinsburg trajectory groups indicates that cluster analysis successfully distinguishes between different three-dimensional flow regimes (Fig. 6). There is some spatial overlap between clusters, but the median flow paths are fairly distinct in three dimensions. For example, clusters 4 and 5 (North Anticyclonic and North Cyclonic) both have trajectories approaching Martinsburg from the north over the previous 24–36 h. However, the general curvature of the trajectories is different, as are the vertical profiles, with North Anticyclonic showing strong subsidence compared to the North Cyclonic cluster. Recall that with our method, vertical motion is weighted equally to each horizontal component in the cluster analysis. The 10 Roanoke clusters are similar to those for Martinsburg and thus are not duplicated here.

As with the SSC, there are statistically significant overall differences in 2 p.m. ozone concentrations between trajectory clusters in the northern valley (Fig. 7 and Table 4). Ozone levels are highest for clusters with generally shorter trajectories (clusters 8–10: West Medium, Short Northwest, and Short Southwest). Ozone is high because of both transport from urban/industrial regions in the Midwest combined with *in situ* photochemistry. Those clusters with longer trajectories tend to have mean ozone levels below 55 ppb. Northerly trajectories have lower concentrations, possibly because of lower temperatures and less transport from regions with high ozone levels. Northeast trajectories (cluster 6) exhibit the lowest ozone levels despite flow paths over highly industrialized areas of the northeastern United States. As with the SSC results, the between-cluster ozone variability is greater than the inter-monitor variability, but the differences are smaller. For most trajectory groups, ozone is highest over Washington and declines southward toward Fauquier, an inter-station trend that is particularly strong for westerly trajectories.

Ozone results for the southern valley are comparable to the north but are more poorly differentiated despite their statistical significance (Fig. 8 and Table 4). Again, clusters with shorter trajectories (clusters 8–10: West Medium, Short Northwest, and Short Southwest) have the highest ozone levels, but in this case the ozone values are more comparable to clusters with longer trajectories (clusters 1–5), regardless of direction. Most of the between-cluster distinction arises because clusters 6 and 7 (Northeast and East) have mean ozone levels well below 50 ppb. With respect to inter-station variability, urbanized Richmond has the highest ozone levels and Rockbridge has the lowest. It is interesting that the monitor at Wythe, our most isolated site in southwestern Virginia, has higher ozone levels than the other locations when flow is from an easterly direction (clusters 6 and 7). This result is probably related to the



**Fig. 6.** Clusters of 72-h back trajectories for Martinsburg, West Virginia (northern Shenandoah Valley). The inset on each map shows the median vertical component of the trajectory (in km above ground level) and the 25th and 75th percentiles. Clusters are approximately ordered so that the trajectories shift clockwise from cluster 1. Shortest trajectories are associated with the last three clusters (clusters 8–10).

accumulation of ozone (via production and advection) of Atlantic air parcels as they pass over urban areas and heavily forested regions of central Virginia.

In general, SSC appears to be a better overall ozone discriminator than trajectory cluster (compare Tables 2 and 4), though both

methods exhibit statistically significant differences in ozone levels. This result is further verified through a direct comparison of the mean square errors for the subset of days at each station for which both SSC and cluster data are available (ANOVA not shown). This difference between the SSC and trajectories probably accounts for

**Table 3**

Results of Hotelling  $T^2$  test on the overall differences between trajectory clusters, for Martinsburg (MRB) and Roanoke (ROA). Both analyses are statistically significant at the 0.05 level; df = degrees of freedom.

Station	Value	F	Hypothesis df	Error df
MRB	9.545	72.151	324.000	22 043.000
ROA	8.268	65.998	324.000	23 276.000

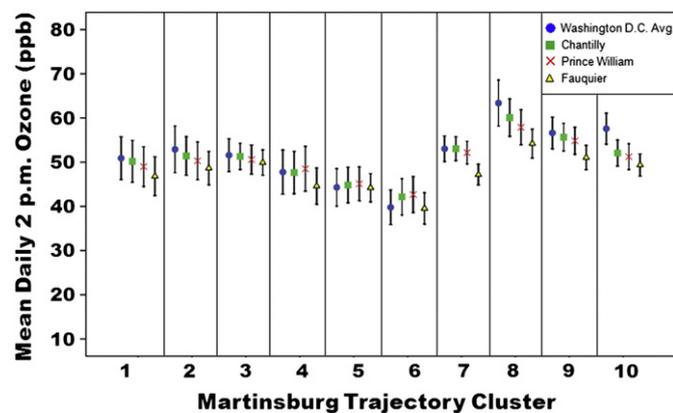
the higher number of subsets identified by the SSC approach (last column of Tables 2 and 4).

**4.3. Ozone and air mass/trajectory interactions**

As an example of interactions between SSC and HYSPLIT back trajectories, we present the results of the one-way ANOVA with interactions for Chantilly, an ozone monitoring station in northern Virginia relatively close to the trajectory terminus at Martinsburg. Results show that SSC, back trajectory, and their interaction all have significantly different mean ozone levels (Table 5). The crossing of lines on a plot of the marginal means suggests an important and possibly significant interaction effect (Fig. 9).

For some air masses, ozone levels are fairly independent of trajectory. Overall ozone concentrations are lowest for MP and highest for DT air regardless of the trajectory. However, for other air masses, knowledge of SSC type or trajectory alone does not provide enough information to fully understand ozone potential.

For example, MM, associated with relatively cool and generally overcast conditions, is not typically a high ozone air mass. This is especially true when the MM air arrives into Martinsburg from the north or northeast (clusters 5 and 6). Mean ozone values for MM air vary from about 20 ppb for North Cyclonic and Northeast trajectory clusters (clusters 5 and 6) to more than 50 ppb for West Medium (cluster 8). With cluster 5 (North Cyclonic), the general cyclonic curvature of the trajectory suggests the presence of positive vorticity and rising motion that would enhance cloud cover in the arriving air mass. For cluster 6 (Northeast), the short path-length of the trajectory from nearby Atlantic moisture sources would likewise mitigate against ozone production and transport. MM air arriving from the north and east has very low ozone concentrations despite trajectories passing over urban areas. MM air is generally not conducive to high ozone production, so in this case, the impact of moisture-laden air trumps that of ozone advection. North Cyclonic and Northeast trajectories have fairly low elevations over the 72 h resulting in high humidity considering the source regions.



**Fig. 7.** Variability of ozone by trajectory cluster for the northern Shenandoah Valley. Error bars are 95% confidence intervals. Overall mean ozone levels are significantly different across trajectory groups (Table 4). Please refer to Fig. 6 for the trajectory group associated with each cluster number.

**Table 4**

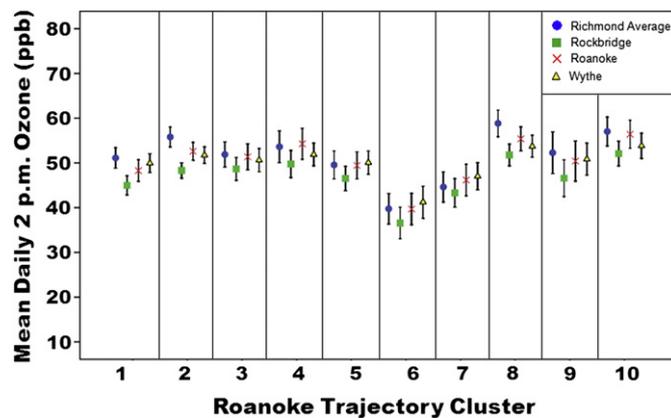
Analysis of variance results for ozone variability by trajectory cluster for the northern and southern Shenandoah Valley (as in Table 2).

Station	Number	MSE between	MSE within	Groupings by decreasing ozone values	SS
<i>Analysis of variance results for northern stations</i>					
Washington Average	1116	5632.848	442.518	(8, 10, 9), 2, 7, 3, 1, (4, 5, 6)	4
Chantilly	1097	3253.153	361.711	(8, 9, 7, 10, 2, 3), 1, (4, 5, 6)	4
Prince William	1077	2132.986	342.731	(8, 9, 7, 10, 2, 3, 1, 4, 5, 6)	4
Fauquier	1091	2249.766	267.776	(8, 9, 3, 10, 2, 7, 1), (4, 5, 6)	3
<i>Analysis of variance results for southern stations</i>					
Richmond Average	1159	3544.954	268.198	(8, 10, 2, 4, 9, 3), 1, 5, (7, 6)	5
Rockbridge	1140	2064.401	206.077	(10, 8, 4, 3, 2, 9, 5), 1, (7, 6)	3
Roanoke	1154	2593.796	253.428	(10, 8, 4, 2, 3, 9), 5, 1, (7, 6)	4
Wythe	1146	1305.936	192.681	(10, 8, 4, 2, 9, 3, 1, 5), (7, 6)	3

By comparison, cluster 4 (North Anticyclonic) is associated with strong subsidence and has ozone readings on MM days twice those of cluster 5 (North Cyclonic) despite very similar trajectory patterns. In contrast, ozone levels exceed 50 ppb for the West Medium cluster (8). These trajectories are generally traveling at a low speed over the Ohio Valley and Midwestern source regions where ozone can be advected and enhanced via *in situ* production in drier air. Despite fairly low elevations, these trajectories are comparatively short, and there is evidence of significant recirculation over the mid-Atlantic, which combines ozone advection with some *in situ* production. Thus, while knowledge that an MM air mass occurred at Chantilly on a given day suggests the presence of below normal ozone levels, if the air arrived via a slow-moving air mass from the west, ozone levels would be much higher than expected. The results shown for Chantilly are representative of those found for other monitors in the northern part of our study region.

As another example, MT air is typically associated with high ozone levels, particularly under westerly or northwesterly trajectories. The combination of air mass conditions conducive to ozone formation (high temperatures and incoming solar radiation) and advection from ozone-rich regions results in high ozone at Chantilly. However, MT air associated with easterly or southerly flow (i.e., ozone-poor source regions) results in low ozone levels despite the warm conditions that would usually favor ozone formation.

For the southern Shenandoah Valley, SSC remains the most important ozone discriminator (Fig. 10 and Table 5). DT air has the



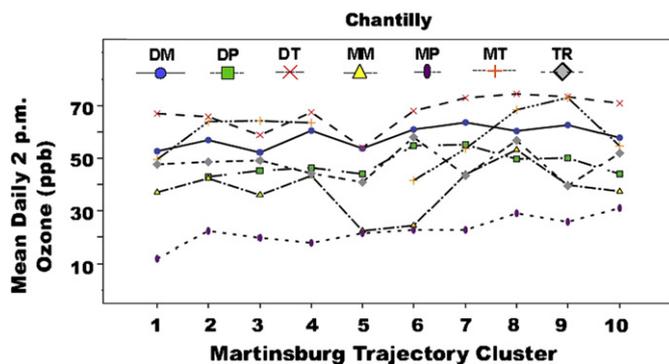
**Fig. 8.** Variability of ozone by trajectory cluster for the southern Shenandoah Valley. Error bars are 95% confidence intervals. Overall mean ozone levels are significantly different across trajectory groups (Table 4). Please refer to Fig. 6 for the trajectory group associated with each cluster number.

**Table 5**

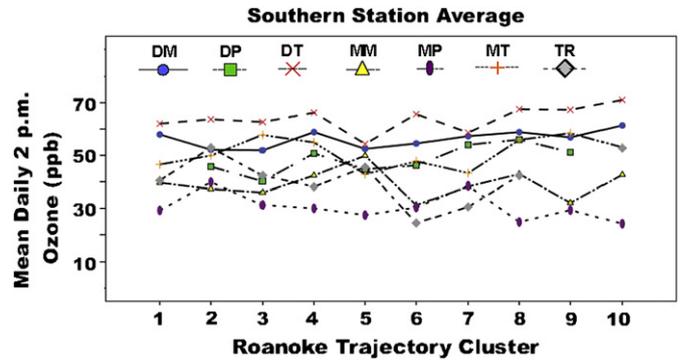
Analysis of variance results for the interaction between SSC air mass and trajectory cluster (as in Table 2). Statistically significant differences at the 0.05 level are evident for every case except the one indicated by an asterisk (\*).

Station	Dependent	MSE	F-statistic
Washington Average	SSC	15 013.50	53.26
	Cluster	971.98	3.45
	SSC × Cluster	494.48	1.75
Chantilly	SSC	11 110.32	48.91
	Cluster	711.37	3.13
	SSC × Cluster	481.28	2.12
Prince William	SSC	10 852.39	49.98
	Cluster	609.73	2.81
	SSC × Cluster	414.58	1.91
Fauquier	SSC	7889.32	44.75
	Cluster	375.84	2.13
	SSC × Cluster	292.83	1.66
Richmond Average	SSC	8234.27	44.77
	Cluster	390.44	2.12
	SSC × Cluster	301.68	1.64
Rockbridge	SSC	8900.30	77.01
	Cluster	234.62	2.03
	SSC × Cluster	269.24	2.33
Roanoke	SSC	9998.46	64.44
	Cluster	341.24	2.20
	SSC × Cluster	329.90	2.13
Wythe	SSC	6274.23	47.30
	Cluster*	188.20	1.42
	SSC × Cluster	253.08	1.91

highest ozone levels regardless of trajectory cluster, with mean concentrations typically between 60 and 70 ppb. Ozone levels are lowest on MP days in 7 of the 10 clusters. Most of the interesting interactions occur on days with moderate ozone levels. For example, mean ozone concentrations vary markedly on days classified as Transitions (which are typically frontal passages). When Transitions occur on days with Westerly trajectories (cluster 2), ozone exceeds 50 ppb, but on Transition days with Northeast (cluster 6) or East (cluster 7) trajectories, ozone is at or below 30 ppb (lower than any other SSC group). Therefore, knowledge that a Transition occurred, while useful, does not in and of itself provide sufficient information for a good estimate of ozone conditions. Frontal passages may be associated with cleansing of the resident air mass and improving air quality. This is the case when the transition occurs on a day with east or northeast



**Fig. 9.** Mean ozone as a function of both SSC air mass and trajectory cluster for the northern Shenandoah Valley. There are statistically significant differences by air mass, trajectory, and (in many cases) their combination (Table 5). Note that all possible combinations do not always occur (e.g., Moist Tropical air mass and North Cyclonic trajectory cluster). Please refer to Fig. 4 and the text for the air mass abbreviations and to Fig. 6 for the cluster numbers.



**Fig. 10.** Mean ozone as a function of both SSC air mass and trajectory cluster for the southern Shenandoah Valley. There are statistically significant differences by air mass, trajectory, and (in many cases) their combination (Table 5). Note that all possible combinations do not always occur (e.g., Dry Polar air mass and Gulf trajectory cluster). Please refer to Fig. 4 and the text for the air mass abbreviations and to Fig. 6 for the cluster numbers.

trajectories observed at 1800 GMT, conditions most typical of frontal overrunning in this region. However, transition to north-west flow results in poor ozone air quality, most likely a by-product of the advection of ozone-rich air from upwind urbanized Mid-western sources.

Another example of a significant interaction is the case of MT air. Mean MT ozone levels vary from about 40 to 60 ppb, depending on the trajectory cluster. MT ozone concentrations are highest for the shorter trajectory clusters (8–10). These represent the classic summertime high ozone scenario in this region, in which air flow is weak under the dominance of a slow-moving subtropical anticyclone. *In situ* ozone production is large given the high temperatures and relatively abundant sunlight, and ozone levels tend to increase from day-to-day. However, MT air can also occur with northerly or easterly trajectories (clusters 5–7), and in these cases, ozone levels are markedly lower. Air flow is stronger than under clusters 8–10, so here the presence of warm, humid conditions alone is insufficient to generate high ozone levels.

## 5. Conclusions

In Virginia's Shenandoah Valley, weather changes play a critical role in the daily variations of 2 p.m. warm-season ozone levels. Air mass conditions (here described using the SSC) provide a very useful discriminator of ozone concentrations because they implicitly account for many of the factors related to ozone formation (e.g., temperature, humidity, sunlight). We have likewise demonstrated that there is a strong relationship between ozone and 72-h back trajectories. In this case, short trajectories with recirculation over the study region are linked to high ozone levels, whereas trajectories originating from northern and eastern source regions contain lower ozone levels. The differences in these trajectory groups indicate how a wide variety of circumstances can influence ozone levels. Transport from ozone source regions can advect ozone into the region, but ozone production will be enhanced under clear skies and high temperature conditions often associated with DT air masses. Furthermore, anticyclonic flow enhances the opportunity for ozone enhancement via subsident drying and warming coupled with the recirculation of high ozone air over the study region.

Most importantly, we find that the interactions between SSC and trajectory clusters provide more information than could be obtained from either method alone. In many cases, for a given air mass on the trajectory's terminal day, air parcels arriving into the region along different trajectories have significantly different ozone

signatures. Our results indicate that, when characterizing the ozone climatology of a station or region, there is sufficient “value added” in considering both the thermal/moisture/sky cover conditions and the path of the air parcels into the station.

One reason that both methods are important is that the groupings are only partially redundant. For example, most MT air masses are associated with southerly trajectories that tap moisture sources in the Gulf of Mexico or Atlantic Ocean. But during summer, MT air can occasionally arrive from the north (given that high dew point temperatures can extend into southern Canada on some days). It is these atypical MT trajectories that are associated with unusually high ozone levels (e.g., Figs. 9 and 10). In related research, Hondula et al. (2009) found that half of the trajectories linked to a given SSC air mass are atypical of the air mass paradigm (e.g., DT air arriving from the north or east). *In toto*, these results indicate that one cannot assume, given a proper air mass classification of a given day, that the air likely arrived into the station along a specific trajectory. Although this assumption would produce some inherent error in a climatological analysis, those errors would be enhanced in an air quality application (in this case, for ozone) because pollutant levels are heavily influenced by both advection and *in situ* air mass production. Our findings here that both trajectory and air mass are acting in tandem to influence ozone levels are currently being extended to other pollutant species (PM, SO<sub>2</sub>, and CO) over the same region. We strongly encourage researchers to investigate the joint relationship between air mass type and back trajectories in other regions.

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