Can Meteorologically Adjusted Ozone Trends Estimate the Impact of the NOx SIP Call?

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Abstract

Breathing in ground-level or tropospheric ozone can trigger a variety of health problems including chest pain, coughing, throat irritation, and congestion. It increases problems with bronchitis, emphysema, and asthma. The ability to determine the impact of ozone precursor emission controls on ground-level ozone trends is complicated by the impact of meteorology, which can be either conducive to ozone formation or not. How do you know if emission controls are really working? The major precursors to ground-level ozone formation are volatile organic compounds (VOCs) and nitrous-oxides (NOx). The EPA Nitrogen Oxides State Implementation Plan Call (NOx SIP Call) began in 2001 in an effort to mitigate the formation of ground-level ozone. Since ozone is strongly affected by the influence of meteorological variables, many different approaches have been taken to determine the trend in ozone by removing the effects of varying meteorology. The purpose of this project was to build a time series model that removes the effects of meteorology, autocorrelation, and seasonal trends based on ozone and meteorological data from the Maryland Department of the Environment and the New Jersey Department of Environmental Protection. This data spans April through October of 1997-2006 for Maryland and Washington, DC and 1997-2005 for New Jersey. As the result of our analysis, a series of models were combined with a filtered time series model and back trajectory modeling to estimate the reduction in ground-level ozone over this ten-year period. These results suggest an improving trend in ozone concentrations over this time period in New Jersey and Maryland.

1. Introduction

Tropospheric ozone is one of the most important pollutants in today's world. The primary factors influencing ground-level ozone formation are solar radiation, nitrogen oxides, volatile organic compounds, light wind and high temperature (NRC, 1991, Chapter 4). One of the important chemical reactions driving ozone formation is the decomposition of nitrogen dioxide (NO_2) by ultraviolet radiation (UV) into nitrogen oxide (NO) and monatomic oxygen (O), which then combines with diatomic oxygen (O_2) to form ozone (O_3):

$$NO_2 + UV \rightarrow NO + O$$
$$O + O_2 \rightarrow O_3$$

Because of its effects on human health and agriculture, government officials have sought to control ozone by setting emissions standards on its precursors, nitrogen oxides (NOx) and volatile organic compounds (VOCs). The Nitrogen Oxides State Implementation Plan Call (NOx SIP Call) was implemented in 2001 and requires the reduction of NOx emissions at electric utilities. As shown in Figure 1, 61% of the supplementary control systems implemented by the NOx SIP Call were put into place in 2003 and 2004. While many of these plans have had some effect on the trend of ozone over time, it is a challenge to interpret the success of these plans because of the strong effect of varying meteorological conditions on ozone concentration. Therefore, it is necessary to determine an ozone trend adjusted for varying meteorology in order to determine how effective emission controls, such as the NOx SIP Call, have been. In this project, we have designed a model that accounts for varying meteorology from ten years of data

provided by the Maryland Department of the Environment and nine years of data provided by the New Jersey Department of Environmental Protection, to determine the impact of the NOx SIP Call in the state of Maryland and New Jersey.

A multitude of statistical techniques exist to account for meteorological variation (Thompson et Al., 2000), notably time series filtering (Rao & Zurbenko, 1994), semi-parametric modeling (Milanchus et al., 1998), regression tree analysis (Huang & Smith, 1999), dynamic linear modeling and general additive modeling (Zheng et. al, 2006), among many others (more listed in References). However, we chose time series linear regression due to its simplicity and straightforwardness of interpretation. Through careful selection of explanatory variables, we were able to construct a model that explained approximately seventy percent of the variance in eight-hour ozone concentrations. This resulted in a statistically significant estimate of the residual trend in ozone concentration over the ten-year period of study.

2. Methods

a. Quality Control and Data Conditioning

The data from both the Maryland Department of the Environment and the New Jersey Department of Environmental Protection included both one-hour observations and the forward rolling eight-hour averages. Each year contained observations beginning on April 1st and ending on October 31st for 1997 to 2006 for Maryland and 1997 to 2005 for New Jersey. We began by reading all of the ozone data into statistical analysis software (SAS) and then sorting both statistics by site. The daily maximum for both one-hour and eight-hour observations were then extracted and matched with daily meteorological data from airports and the Clean Air Status and Trends Network sites. In Maryland, Baltimore Washington Thurgood-Marshall International Airport (BWI) and the EPA Clean Air Status and Trends Network (CASTNET) site from

Beltsville, Maryland (BEL116) were used (Fig. 2a). In New Jersey, data was taken from Philadelphia International Airport (PHL), LaGuardia Airport (LGA), and from the CASTNET site at Washington's Crossing (WSP144) (Fig. 2b). High temperature, resultant wind speed and resultant wind direction were taken from the airports. High temperature was converted to degrees Kelvin and the wind speed and wind direction were converted into cardinal wind components (North, South, East, and West). Taken from CASTNET sites were the maximum solar radiation and the average relative humidity, among other variables from both sites. In using the weather data from these non-collocated sites we assumed the variation in those parameters across the region would be negligible.

The ozone data was also checked for data completeness; only sites having ninety percent or better data completeness in each of the ten years were considered for the analysis. There were fourteen sites in Maryland and eleven sites in New Jersey that had ninety percent or greater data completeness. Most of the sites with insufficient data completeness began recording data in the middle of their respective periods between 1997 and 2006 or 1997 and 2005.

b. Correlation of Meteorological Variables

Both the one-hour and eight-hour concentrations were log-transformed to increase correlation with the meteorological variables. Using SAS we built the correlation matrix (Table 1), which showed us that solar radiation, high temperature and average relative humidity were the most useful predictors in our model. Since relative humidity is a measure of the amount of water vapor in the atmosphere, we understand that if more moisture is present in the atmosphere then there would be increased condensation. It may be possible that water vapor condensing on NOx and VOC's limits the amount ozone that can form. Before building the general linear model we examined the relationship between the ozone concentration and several meteorological

variables. From this list of significantly correlated meteorological variables—including previous day's and current day's wind vectors, interaction terms, and second and third order terms—we used SAS to perform stepwise selection at significance .01 to determine which variables would be appropriate for the model.

c. Seasonal Trend

To be certain that we removed all the possible variability resulting from meteorological variables we decided to examine the model residuals by month. The result yielded by this examination was unexpected and caused the addition of a term representing the day of the year in the model. This variable is represented as such: D=1 on April 1st, D=31 on May 1st and so on. This variable was highly significant when adopted into the model, the full results of which will be discussed later on.

d. Estimation of Overall Trend

To estimate the overall trend, we added a 'year' term, *Y*. This allowed us to calculate the change in background concentration over time without further adjustment for seasonal variation.

We proceeded to adjust for autocorrelation present in the general linear model. To account for weekly fluctuations in ozone trends we used an autoregressive error model with a seven day lag using the method of maximum likelihood; lagged terms that were not significant were removed using the Yule-Walker method. The explanatory variables adopted into this model are those that were chosen via stepwise selection in the general linear model.

e. Filtered Time Series Regression

Because the New Jersey ozone data was taken year round, we decided to also perform a filtered time series regression using a Kolgomorov-Zurbenko (KZ) filter on this data. The

regression was performed on lagged meteorological variables versus the long term ozone trend which was part of the results from the filter.

f. Back Trajectory Modeling

We also performed several back trajectories using NOAA's Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT). We used this model to determine where particles of air would be coming from forty-eight hours before the ozone observation occurred. This was done for good and very unhealthy days, before and after the application of the NOx SIP Call. While we took a simple approach to the Maryland back trajectories, we decided to use the HYSPLIT model differently on the New Jersey data. The possibility exists that by modeling based only whether the air quality was good or very unhealthy days that the results would be biased to particular weather patterns. We selected days used in New Jersey HYSPLIT modeling on the following criteria: PHL High Temperature greater than 90°F, PHL Maximum Solar Radiation greater than 850 Wm^{-2}, and LGA Relative Humidity less than 50%.

3. Results

a. General Linear Model

The correlation matrix (Table 1) allowed us to place all the variables in the stepwise selection, and the correlations of different variables indicated which would be most strongly represented in the model. As we expected, the temperature and the average relative humidity showed strong correlation to the ozone data in the plot as well as mild curvature, which explains the significance of their respective second-order terms. The stepwise selection found that the high temperature, relative humidity and maximum solar radiation were significant and offered the most explanatory power. Though the resultant wind speed and calculated wind vectors were often significant, they provided little explanatory power (partial R-square of less than one

percent) and were subsequently dropped from the model. Therefore, we used the remaining variables to build a general linear model:

$$\log(O_3) = \beta_0 + \beta_1 T + \beta_2 T^2 + \beta_3 H + \beta_4 H^2 + \beta_5 S$$

Where *T* is daily high temperature, *H* is average relative humidity, and *S* is maximum solar radiation.

b. Seasonal Trend

An unexpected finding occurred in examining model residuals by month (Fig 3ab); this plot showed that early in the ozone season the model under-predicts the ozone in the spring and over-predicts the ozone in the fall. All of the sites displayed a similar seasonal trend. In order to neutralize this seasonal trend, we added a day of the year term into the model (Fig 4ab). Not only was this variable highly significant when adopted into the model, it explained approximately ten percent of the variability in the residual eight-hour ozone concentration. We surmise that the seasonality of the ozone trend is related to the affect of biogenic VOC's released by plants in the spring and fall, when most plants are blooming and dying respectively.

c. Overall Trend

The full model, now including the day of the year term (to account for seasonal affects) and the year term is as follows:

$$\log(Max8hrO_3) = \beta_0 + \beta_1 T + \beta_2 T^2 + \beta_3 H + \beta_4 H^2 + \beta_5 S + \beta_6 D + \beta_7 Y$$

Therefore, if the year slope is negative, the concentration of ozone is decreasing over time. Also, because the concentration is expressed on a logarithmic scale, the value for β_7 approximates the average change in ozone concentration per year (a value of β_7 = -.01 indicates a decrease of approximately one percent per year). This model was tested for autocorrelation using the Durbin-Watson statistic and multicollinearity using the variance inflation factor. Though

multicollinearity was not a significant problem, the presence of autocorrelation was significant enough to justify use of an autoregressive error model. After incorporating the linear model into the autoregressive error model the explanatory power of the models increased dramatically. Of the sites analyzed in Maryland and Washington D.C., nine showed statistically significant decrease in ozone trends between 1997 and 2006 (Table 2). The three sites in Washington, D.C., in addition to the two remaining sites in Maryland, did not show a significant change. No site indicated an increase in ground-level ozone concentration. The largest decrease occurred at the Fair Hill site with an estimated reduction of fifty-two percent. Of the sites analyzed in New Jersey, nine also showed a significant decrease in ozone trends between 1997 and 2005 (Table 3). The explanatory power of the models ranged from sixty-seven to seventy-four percent (adjusted R-Square). Therefore, we conclude that the NOx SIP Call has decreased ozone concentrations in Maryland and New Jersey.

d. Filtered Time Series Regression

The filtered time series regression broke the ozone data into three different trend lines. The raw data was reduced to a short term trend line (background noise), seasonal trend (biogenic affect), and a long term ozone trend (Fig. 5a-d for example). The interesting finding in the long term trend is the somewhat sinusoidal pattern in the trend. In all the sites, there is a 2.5 to 3 year cycle in the ozone concentration (Fig. 5d for example). Another interesting finding from this method is that the maximum correlation in the regression occurs when the high temperature is lagged by 138 days (Fig. 6 for example). Multiple reasons have been posed for this pattern in the long term trend including the possible impact of the El Nino Southern Oscillation and the sunspot cycle, but there is not enough support here for either of these reasons. While the results

from this analysis are intriguing, this filtering technique should be performed on a longer period of data before attempting to conclude on a reason for the occurrence of this trend.

e. HYSPLIT modeling

1) MARYLAND BACK TRAJECTORY MODELING

The HYSPLIT model also returned several trajectories for good and bad days, before and after the NOx SIP Call, which point to decreased ozone concentrations for the state of Maryland. Two particular examples will be examined here. On July 14, 1997, the highest recorded ozone observation at the Edgewood site was 136 ppb. These 'very unhealthy' levels of ozone were traced back to the Midwest and the Ohio River Valley (Fig 7). Similarly, on June 26, 2003, the highest recorded observation was 129 ppb; and again HYSPLIT found the air to be coming from the Ohio River Valley (Fig 8). It was a similar result for many of the bad days, and those unhealthy ozone days that were dissimilar resulted from stagnant air preventing at the surface preventing ventilation that would have dissipated the already present ozone and its precursors from previous days. These results from the HYSPLIT indicate that the reductions from the application of the NOx SIP Call have decreased ozone concentrations in Maryland, and many of the days with terrible air quality have come from the Midwest and Ohio River Valley between 24 and 48 hours beforehand. It should be noted that this conclusion is anecdotal, as the only discriminating variable in selecting days for the model is the highest ozone concentration on a specific day, without regards to similar weather patterns between good and bad days.

2) NEW JERSEY BACK TRAJECTORY MODELING

In order to remove a potential bias to a particular weather pattern, we selected days for the back trajectory based upon particular criteria given earlier. The result allowed us to see a difference between surface transport of ozone (or its precursors) and long range transport over

the mountains. As an example we took the back trajectories from July 2nd through July 4th, 2002 (Fig. 8 a-c). Each of these days passed the selection process by the criteria mentioned earlier, but it was only July 2nd that was an unhealthy ozone day. It is interesting to note first that this unhealthy day had the lowest high temperature and maximum solar radiation of the three days and also the highest relative humidity (Table 4). We also notice a significant difference in the trajectories themselves. Each of the red trajectories represents a trajectory ending at 10 meters off the ground at the ending location (marked by the star). The ending location is the Jackson site, the design value site for the state. The blue trajectory and the green trajectory represent trajectories ending at 100 meters and 500 meters above the ending location respectively. The one common trait between these three days is that the higher altitude trajectories all pass over the mountains into the Ohio River Valley. However, on July 2nd, the surface trajectory remains on the lee side of the mountains. More specifically, the first 12 hours back place the trajectory over Interstate Route 95, and afterwards over the Chesapeake Bay. From this observation we can conclude that when the surface winds remain trapped on the lee side of the mountains the ozone concentration at the Jackson site may be increased dramatically. However, this is only one example, and while many other days showed similar conclusions, there should be an ensemble of days (selected by the same criteria) modeled using HYSPLIT to back these observations.

4. Conclusion

a. Overall Trend

It can be concluded that ozone has decreased in concentration in Maryland and New Jersey since the implementation of the NOx SIP Call in 2001. However, while the concentration of ozone over time has decreased, we note that the models built in this study do not take into account the emissions of NOx or VOCs, and therefore do not specifically indicate the effect of the NOx SIP

Call. This is evident because the adjusted ozone trend drops off dramatically after the implementation of the supplementary control systems beginning in 2001. Yet, because it does take into account the varying effects of meteorology, the autoregressive error model strongly suggests that the emission reductions resulting from the NOx SIP Call in the Midwest and Eastern part of the U.S have resulted in decreased tropospheric ozone concentrations in the States of Maryland and New Jersey. As we continue to work on the models, we intend to add terms representing NOx emissions and VOC emissions (both biogenic and anthropogenic). Also, as we continue to work with the HYSPLIT model we hope to account for variation not explained by the autoregressive error model, and in particular during periods of highest ozone concentration by comparing days with similar weather patterns. We also intend to explore the NOx trend itself in comparison to the ozone trend over time, which we intend to publish in a future paper.

b. Seasonal Trend

We present two possible explanations for ozone seasonality: The first is the possible influence of stratospheric ozone that mixes in as the inversion layer breaks down in the morning, with the mixing being strongest in April and weakest at the end of October. Second is the biogenic VOC emissions from plants are strongest in spring, and weakest in fall. We hope to incorporate the Biogenic Emission Inventory System (BEIS) model to further account for seasonal fluctuations in VOCs that may contribute to seasonal ozone trends.

5. Acknowledgements

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7. **Figures and Tables**

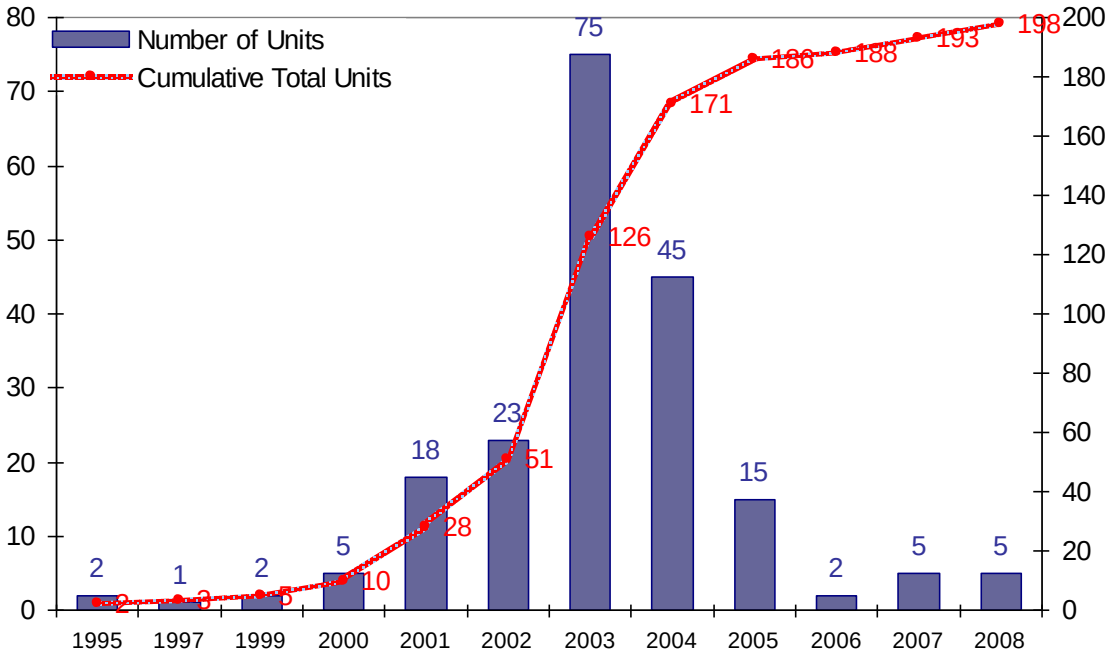


Figure 1) Selective Catalytic Reduction Units installed from 1995 to 2008- 61% of the units were installed in 2003 and 2004. Graph from the Institute of Clean Air Companies

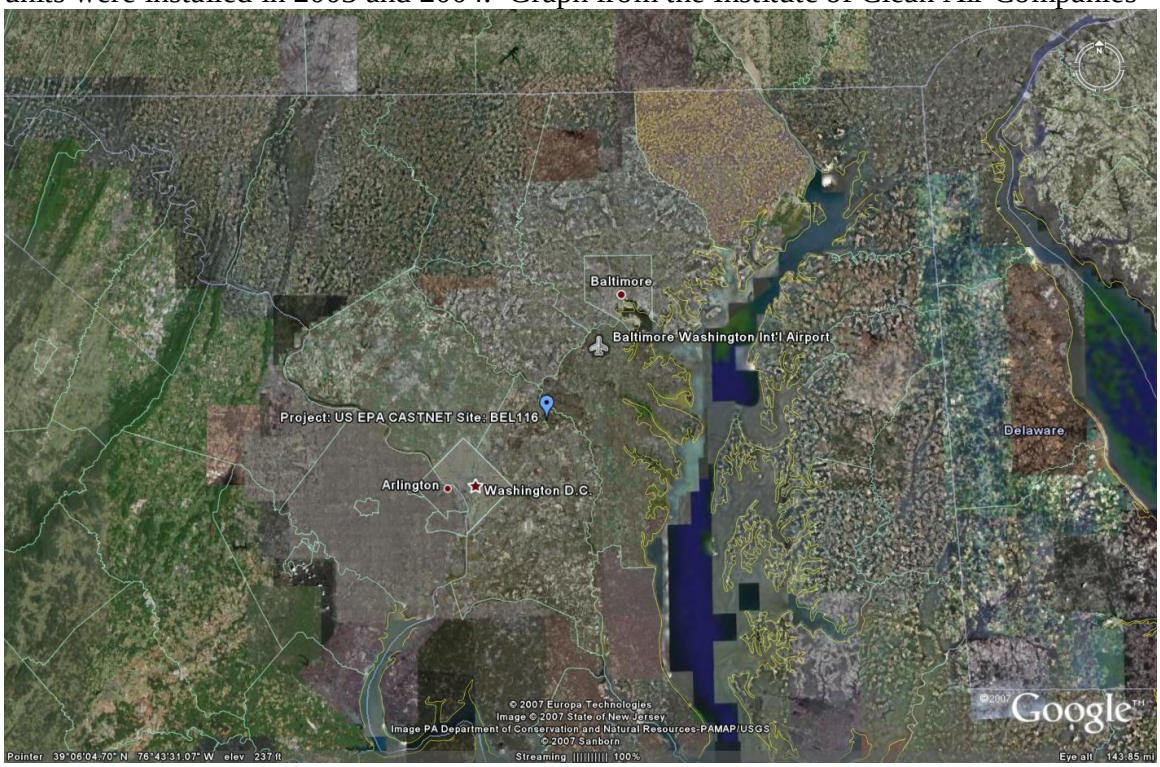


Figure 2a) Map of Maryland with BWI and BEL 115 highlighted.

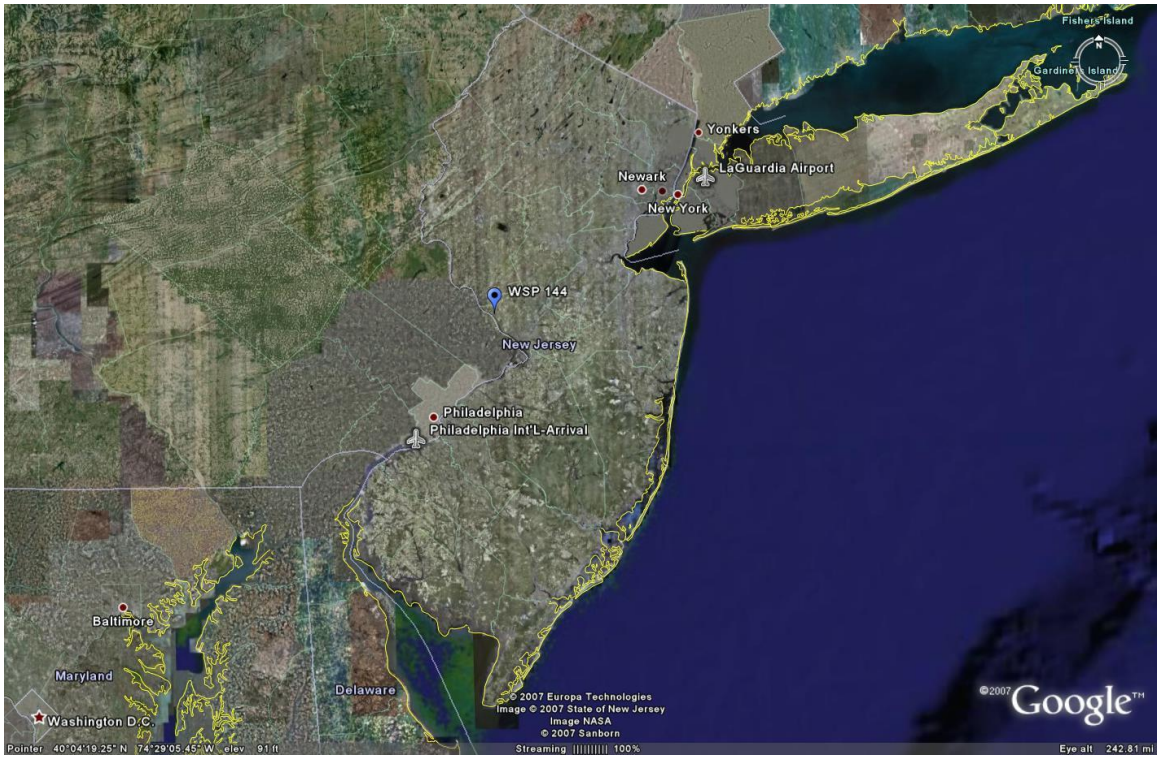


Figure 2b) Map of New Jersey with LGA, PHL and WSP 144 highlighted.

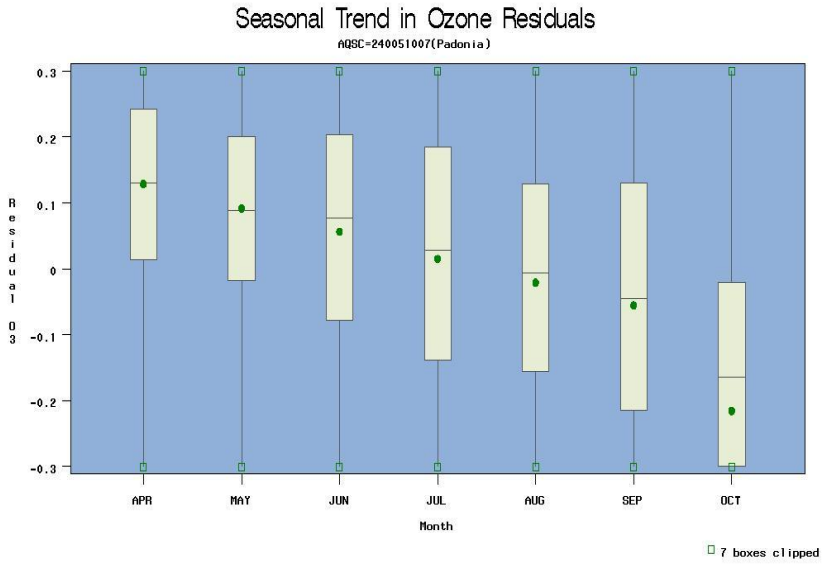


Figure 3a) Seasonal ozone trend for Padonia, Maryland. Similar trends resulted at the other sites in Maryland

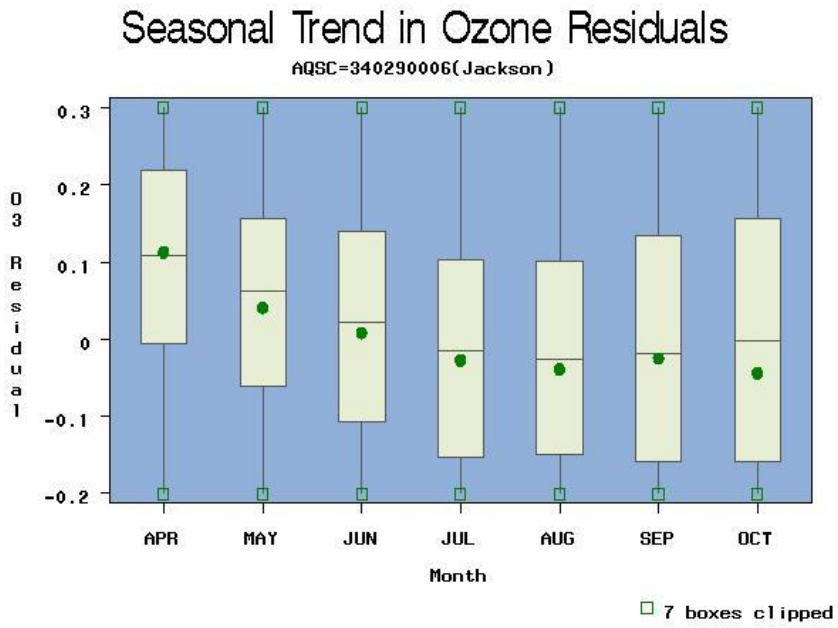


Figure 3b) Seasonal ozone trend for Jackson, New Jersey. Similar trends resulted at the other sites in New Jersey

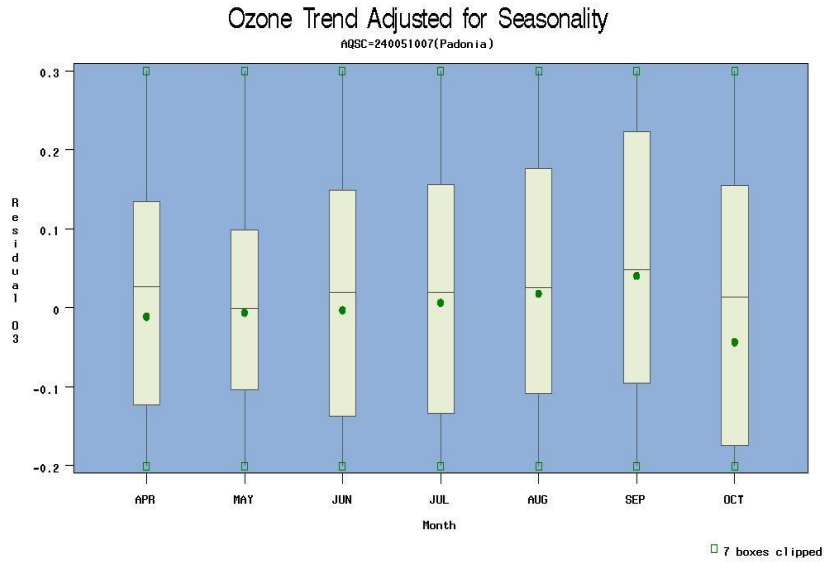


Figure 4a) Ozone trend with seasonality neutralized for Padonia, Maryland. Similar trends resulted at the other sites in Maryland

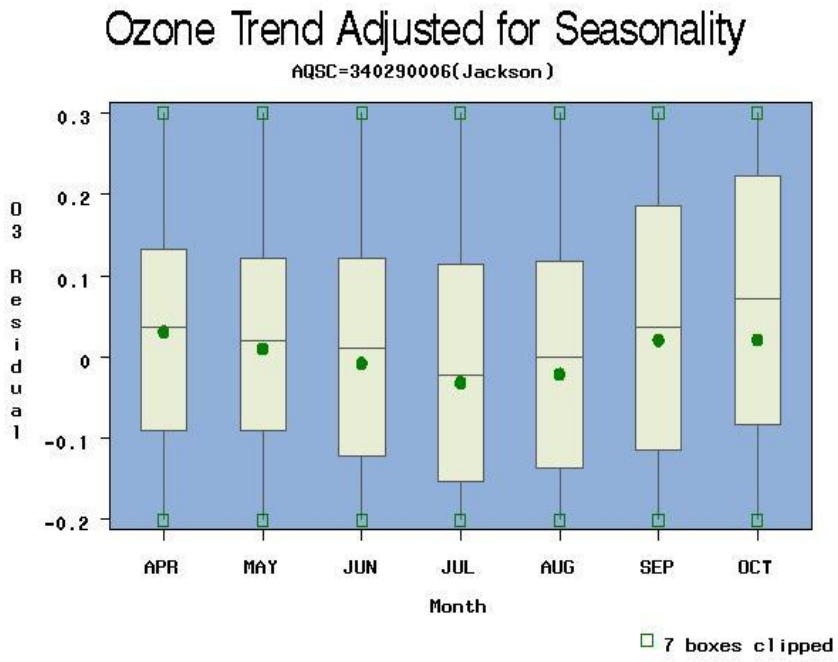
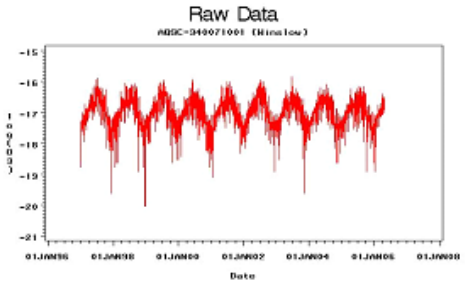
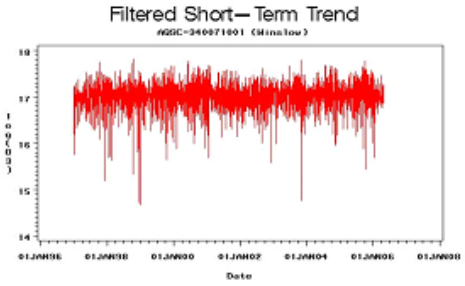


Figure 4b) Ozone trend with seasonality neutralized for Jackson, New Jersey. Similar trends resulted at the other sites in New Jersey

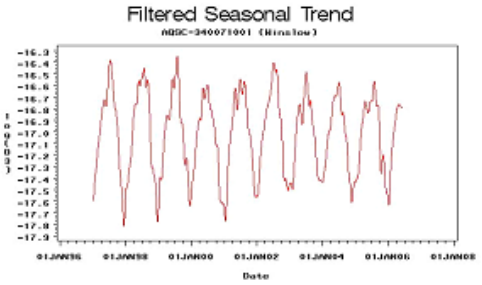
5a

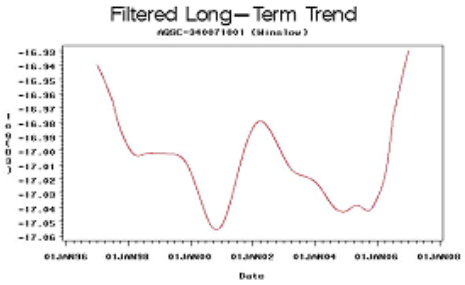




5b

5c





5d

Figure 5a-d) KZ Filter application on ozone data from Winslow, New Jersey. The upper left is the raw data plot, the upper right is the short term trend (background noise), the lower left is the seasonal trend, and lower right is the long term ozone trend.

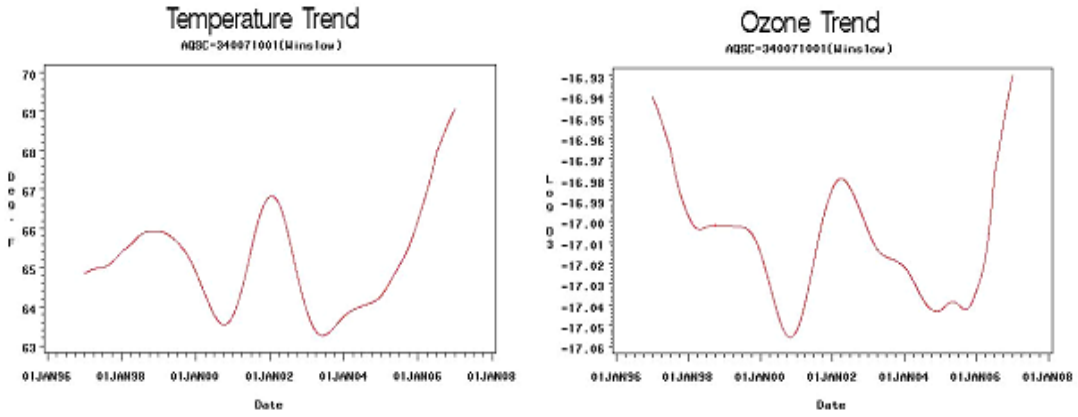


Figure 6) Plots of lagged temperature (lagged by 138 days) and the long term ozone trend at Winslow, New Jersey. There were slightly different lags in temperature that allowed for the maximum correlation.

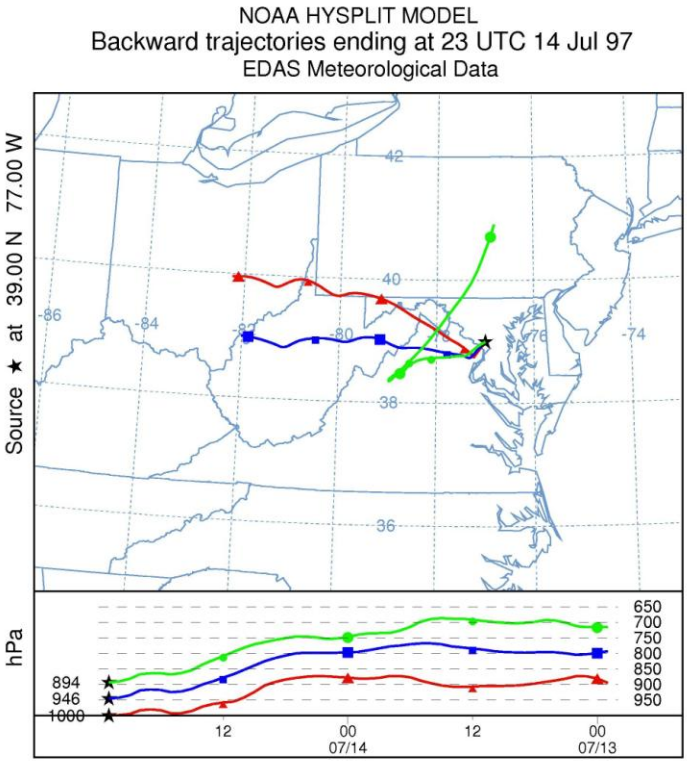


Figure 7) HYSPLIT model results for July 14, 1997, ending at Baltimore, Maryland

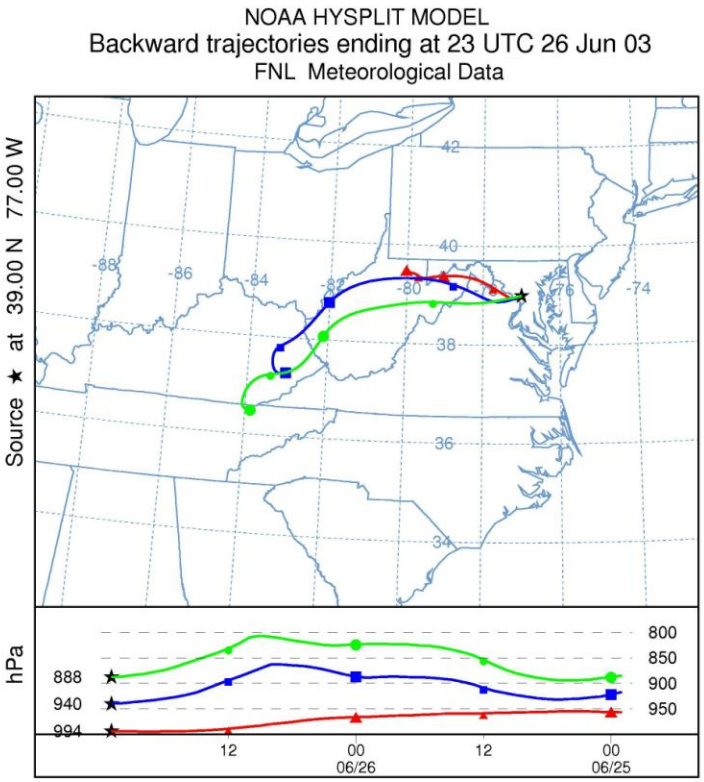


Figure 8) HYSPLIT model results for June 26, 2003, ending at Baltimore, Maryland.

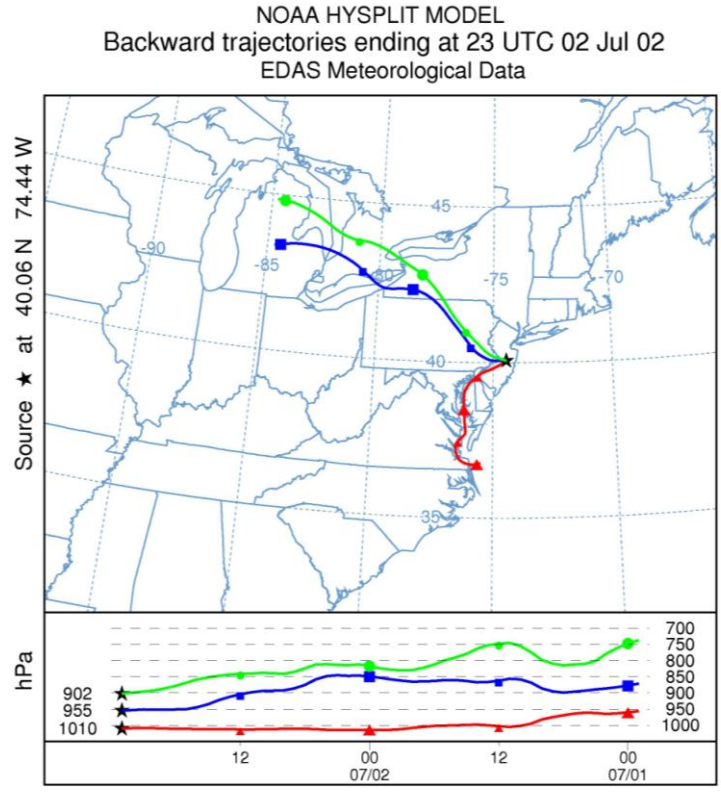


Figure 9a) HYSPLIT model results for July 2nd, 2002 ending at Jackson, NJ.

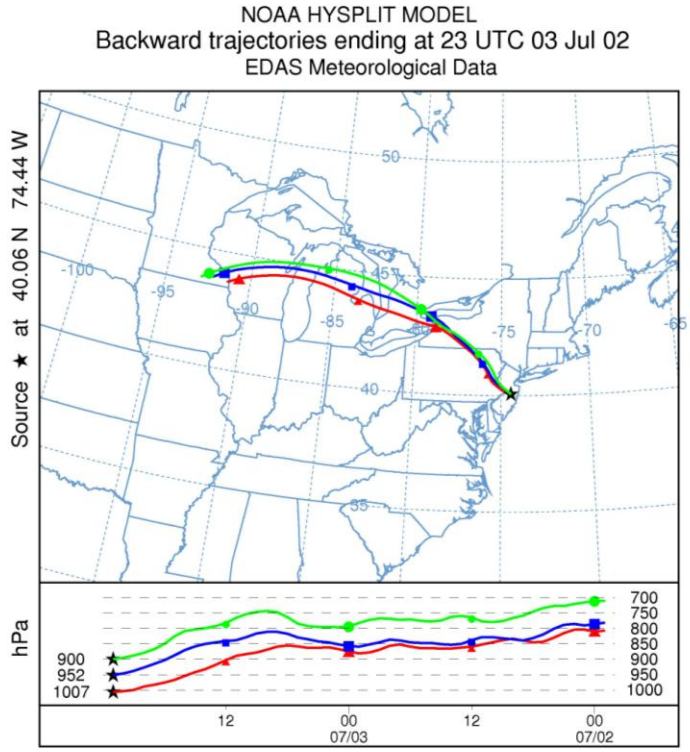


Figure 9b) HYSPLIT model results for July 3rd, 2002 ending at Jackson, NJ.

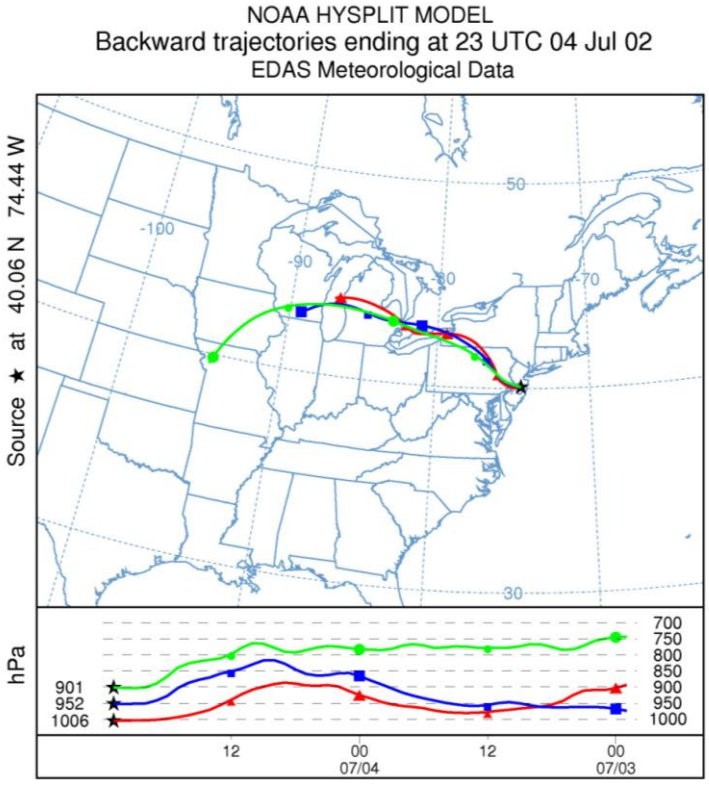


Figure 9c) HYSPLIT model results for July 4th, 2002 ending at Jackson, NJ.

Table 1) Correlation Between Meteorological Parameters and Ozone Concentration, Padonia, Maryland (Pearson Coefficient of Correlation, R)

	Ozone	**T**	**S**	**H**	**W_{North}**	**W_{South}**	**W_{East}**	**W_{West}**
Ozone	1	.685	.601	-.317	-.278	.17	-.176	.022
T		1	.477	-.0966	-.379	.248	-.284	-.055
S			1	-.542	-.181	.068	-.363	.207
H				1	-.013	.010	.271	-.351

T = Maximum Temperature, S = Maximum Solar Radiation, H = Average Relative Humidity
W = Directional Component of Resultant Wind Vector

Table 2) Maryland model results for each ozone station. *- significant change at 0.01 level

Site AQSC	**Site Name**	**Annual Trend**	**Estimated Total Change**	**Net Change (Lower 99% C.I.)**	**Net Change (Upper 99% C.I.)**
110010025	Takoma Park	-.68%	-6.60%	-27.53%	+14.34%
110010041	McMillan Reserve	+.14%	+1.36%	-11.72%	+14.44%
110010043	River Terrace	-.98%	-9.41%	-20.19%	+1.36%
240030014	Davidsonville	-2.40%	-21.57%*	-28.84%	-14.31%
240051007	Padonia	-1.16%	-11.04%*	-17.59%	-4.49%
240053001	E. Maryland	+.64%	+6.57%	-2.98%	+16.12%
240130001	South Carroll	-1.09%	-10.42%*	-18.50%	-2.33%
240150003	Fair Hill	-6.97%	-51.47%*	-61.86%	-41.08%
240170010	So. Maryland	-1.73%	-16.05%*	-25.00%	-7.11%
240251001	Edgewood	-.51%	-4.96%	-12.15%	+2.23%
240251001	Aldino	-1.01%	-9.70%*	-15.84%	-3.56%
240290002	Millington	-1.41%	-13.24%*	-20.27%	-6.21%
240313001	Rockville	-2.61%	-23.20%*	-36.23%	-10.18%
240330002	Beltsville	-2.33%	-21.02%	-32.02%	-10.03%

Table 3) New Jersey model results for each ozone station. *- significant change at 0.01 level

Site AQSC	Site Name	Annual Trend	Estimated Total Change	Net Change (Lower 99% C.I.)	Net Change (Upper 99% C.I.)
340170006	Bayonne	-2.44%	-19.72%*	-27.94%	-11.49%
340070003	Camden	-0.61%	-5.33%	-15.36%	+4.70%
340273001	Chester	-1.90%	-15.71%*	-23.24%	-8.19%
340230001	E. Brunswick	-1.19%	-10.16%*	-18.66%	-1.71%
340010005	Galloway	-3.13%	-24.55%*	-31.98%	-17.12%
340150002	Gloucester	-1.66%	-13.88%*	-22.34%	-5.41%
340290006	Jackson	-1.42%	-12.00%*	-21.93%	-2.06%

340210005	Lawrence	-1.67%	-13.96%*	-15.60%	-6.67%
340190001	Raritan	-0.87%	-7.52%	-15.60%	+.58%
340110007	Vineland	-1.66%	-13.88%*	-23.16%	-4.59%
340071001	Winslow	-1.72%	-14.34%*	-21.52%	-7.16%

Table 4) New Jersey Data for 7/2-7/4/2002

Day	**7/2/2002**	**7/3/2002**	**7/4/2002**
Concentration	0.125	0.094	0.082
High Temperature	95F	97F	99F
Maximum Solar Radiation	869.7 Wm^{-2}	906.7 Wm^{-2}	889.2 Wm^{-2}
Average Relative Humidity	47%	43%	44%