



Observation-Based Assessment of the Impact of Nitrogen Oxides Emissions Reductions on Ozone Air Quality over the Eastern United States

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ABSTRACT

Ozone is produced by chemical interactions involving nitrogen oxides (NO_x) and volatile organic compounds in the presence of sunlight. At high concentrations, ground-level ozone has been shown to be harmful to human health and to the environment. It has been recognized that ozone is a regional-scale problem and that regionwide control strategies would be needed to improve ozone air quality in the eastern United States. To mitigate interstate transport of ozone and its precursors, the U.S. Environmental Protection Agency issued a regional rule in 1998 known as the “ NO_x State Implementation Plan (SIP) Call,” requiring 21 states in the eastern United States to reduce their summertime NO_x emissions by 30 May 2004. In this paper, the effectiveness of the new emission control measures mandated by the NO_x SIP Call is assessed by quantifying the changes that occurred in the daily maximum 8-h ozone concentrations measured at nearly 50 locations, most of which are rural (33 sites of the Clean Air Status and Trend Network and 16 sites of the Air Quality System), over the eastern United States. Given the strong dependence of ozone formation and accumulation on meteorological conditions, the incidence of the latter is first mitigated, and meteorologically adjusted ozone concentrations are extracted using a multiple regression technique. By examining the differences between the cumulative distribution functions of the meteorologically adjusted ozone concentrations, it is shown that ozone concentrations in the eastern United States are now on average 13% less than those prior to the NO_x SIP Call. Using back-trajectory analyses, it is also shown that emission controls on the electricity-generating units located in the Ohio River Valley have contributed toward the improvement of ozone air quality in downwind regions, especially east and northeast of the Ohio River Valley.

1. Introduction

Ozone is a major component of the smog that forms during the summer over the eastern United States. At high concentrations, ozone has been shown to be harmful to human health and vegetation. Ozone is not directly emitted into the atmosphere, but instead is pho-

tochemically produced by the interaction of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight. Because of well-known adverse effects on human health, the U.S. Environmental Protection Agency (EPA) has promulgated a National Ambient Air Quality Standard (NAAQS) for ozone.

In the mid-1990s, most northeastern states struggled with the attainment of the ozone NAAQS. At that time, computer modeling and analyses conducted by Ozone Transport Assessment Group (OTAG)—a partnership among the EPA, states, and industry—revealed the importance of interstate transport of ozone and its

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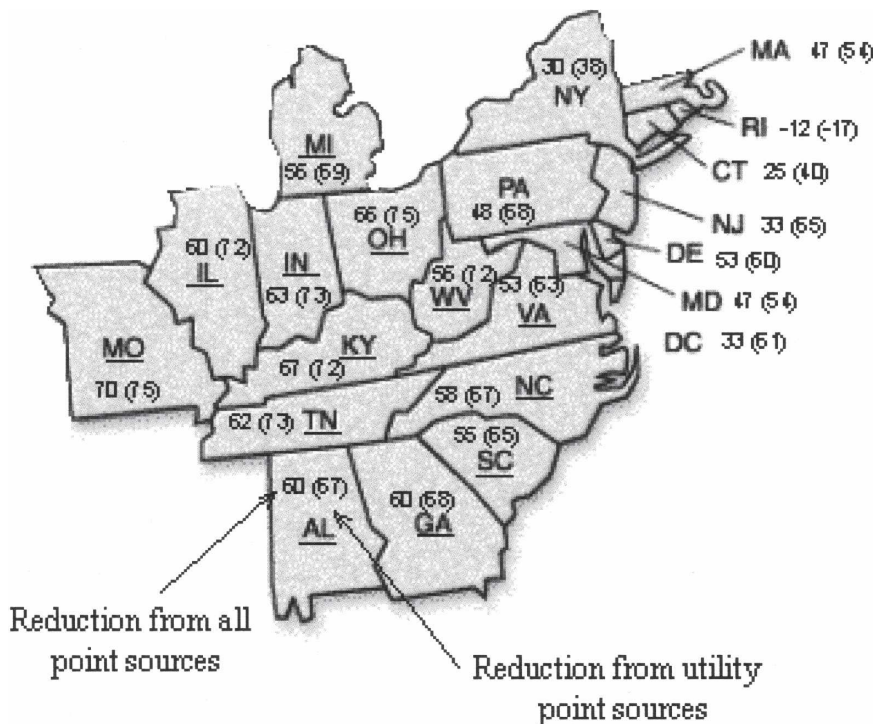


FIG. 1. Map of states affected by the SIP Call plan, the mandated summertime NO_x reductions (% of the 2007 base case), and their compliance deadline (states whose names are underlined had their deadline delayed past May 2003).

precursors in addressing the ozone problem over the eastern United States. Corroborating the findings of Sillman et al. (1990), the OTAG final report (Ozone Transport Assessment Group 1997) stated that emissions along the Ohio River Valley (ORV) in the central part of the OTAG domain appeared to be associated with many regional-scale ozone episodes and that emission reduction from that region would benefit many downwind areas. Hence, building on the OTAG results regarding ozone transport, EPA issued a new regulation in September 1998 requiring 22 states in the eastern United States and Washington, D.C., to reduce their summertime NO_x emissions. To be more specific, the new rule, known as the NO_x State Implementation Plan (SIP) Call and hereinafter simply referred to as the SIP Call, required the state air pollution control agencies to identify in their SIP the emission control measures they would implement to meet the new state-wide NO_x budgets. The EPA, using the so-called 2007 baseline emissions (i.e., the projected emissions for 2007 after the implementation of all-existing laws and regulations) as a reference, derived these new budgets. Although states had to comply with the mandated budgets, they were given the flexibility to develop their own control strategies. Reducing NO_x emissions was

thought to be a better option than further restricting VOCs (the other ozone precursor) to address regional-scale ozone problems, because the latter were shown to have little effect on rural ozone (Sillman et al. 1990). Also, modeling studies (Rao 1993; Rao et al. 1996; Ozone Transport Assessment Group 1997) indicated that most of the eastern United States is NO_x limited. The main targets of the SIP Call are the electric utility point sources and other industrial nonutility sources (Streets et al. 2001). Initially, 1 May 2003 was set as the deadline for the submission of the SIP Call to the EPA. Litigation delayed the initial deadline until 31 May 2004 and reduced the number of states involved to 21. Figure 1 depicts the 21 states subject to the NO_x SIP Call and specifies the amount of reduction each of them had to achieve. Further detailing these changes, Fig. 2 displays the gridded (1° latitude × 1° longitude) NO_x emissions from electrical generation units on 1 July 1997 and the percent reduction in the NO_x emissions between 1 July 1997 and 1 July 2004. Each map was derived from the information contained in the Continuous Emission Monitoring System database (which was available online at <http://cfpub.epa.gov/gdm>). It is evident that the Ohio River Valley and the western Kentucky–Tennessee areas were “hot spots” in terms of NO_x

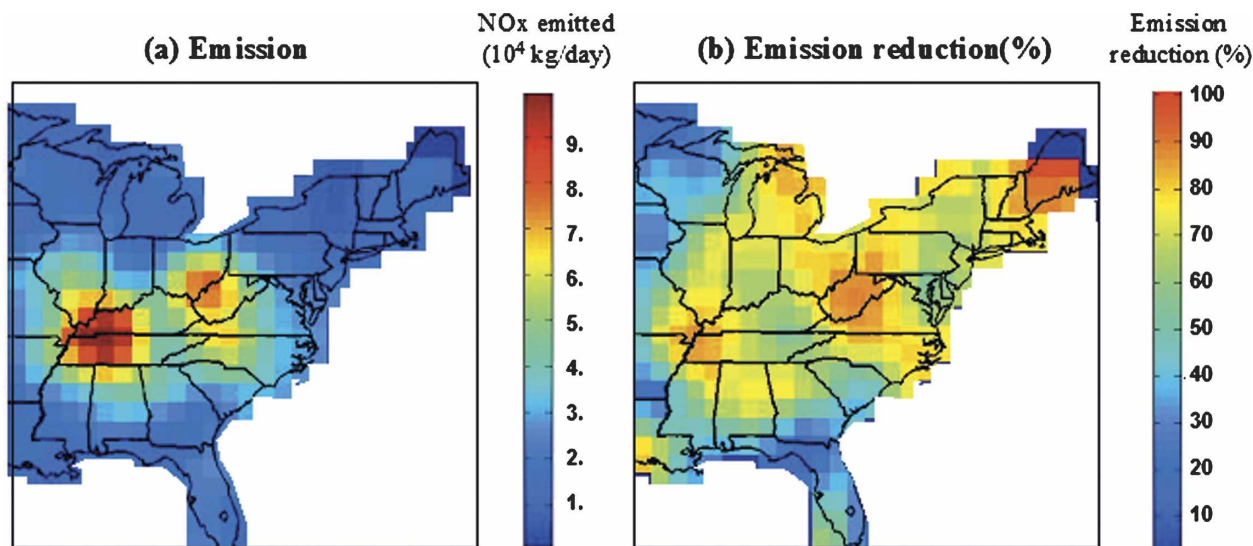


FIG. 2. (a) Map of power industry NO_x emissions (kg day⁻¹) in the eastern United States on 1 Jul 1997, and (b) emission reduction (%) from 1 Jul 1997 to 1 Jul 2004 (i.e., since implementation of NO_x emissions reduction regulations).

emissions in 1997. Yet, these are the areas that experienced the largest change after the implementation of the SIP Call, surpassed only by the New England area (Maine), where NO_x emission reductions occurred not as a direct result of the application of emission control technology, but from a shift from high-NO_x to low-NO_x fuels (i.e., a switch from oil to natural gas).

According to Environmental Protection Agency (2005), compliance with the NO_x emission reductions mandated by the SIP Call was nearly 100% in 2004. The object of this paper is to examine the effectiveness of these NO_x reductions in improving ozone air quality in the eastern United States. To this end, we quantify the changes between the daily maximum 8-h ozone concentrations measured during the ozone seasons of 1997 and 1998 and during the 2003 and 2004 seasons. The 1997 and 1998 seasons are used to depict the situation before the SIP Call was issued. Data collected in 2003 and 2004 are used to capture the results of the SIP Call implementation. However, because the implementation deadline was delayed and some states were still making the transition to compliance in 2003, the changes we identify may not be whole. Note that other NO_x emission control programs, such as the Acid Rain Program, have also been in place since the late 1990s prior to the SIP Call. Therefore, even if, as stated in Environmental Protection Agency (2005), the effects of the Acid Rain Program on ozone are considerably less than those of the SIP Call (4% reduction resulting from the Acid Rain Program vs 10% resulting from the SIP Call), our results represent the effect of all emission changes that

have occurred from 1997 to 2004, not those solely due to the implementation of the SIP Call.

Because meteorological conditions greatly affect ozone formation and accumulation, it is important to moderate their influence on ozone concentrations prior to assessing the impact of any regulatory program (Cox and Chu 1993; Flaum et al. 1996; Kuebler et al. 2001). Accordingly, the concentration changes we report here were obtained after moderating the influence of meteorological conditions on ambient ozone concentrations. Following Brankov et al. (1998), we use back-trajectory analysis in an attempt to specifically assess the changes that occurred throughout the eastern United States following the reduction in emissions from sources in the ORV, the source region identified as the origin of regional ozone episodes impacting the northeastern United States. Because of the uncertainty associated with individual trajectories, we consider an ensemble of trajectories in this study in examining the potential source–receptor relationships. In addition, we compute changes in the standard deviation of the synoptic-scale variations (i.e., strength of the weather-induced variations imbedded in the ozone time series, which affects regional-scale pollutant transport) in the meteorologically adjusted ozone concentrations that occurred between 1997 and 2004.

2. Data

The daily maximum 8-h ozone concentrations measured at the Clean Air Status and Trend Network

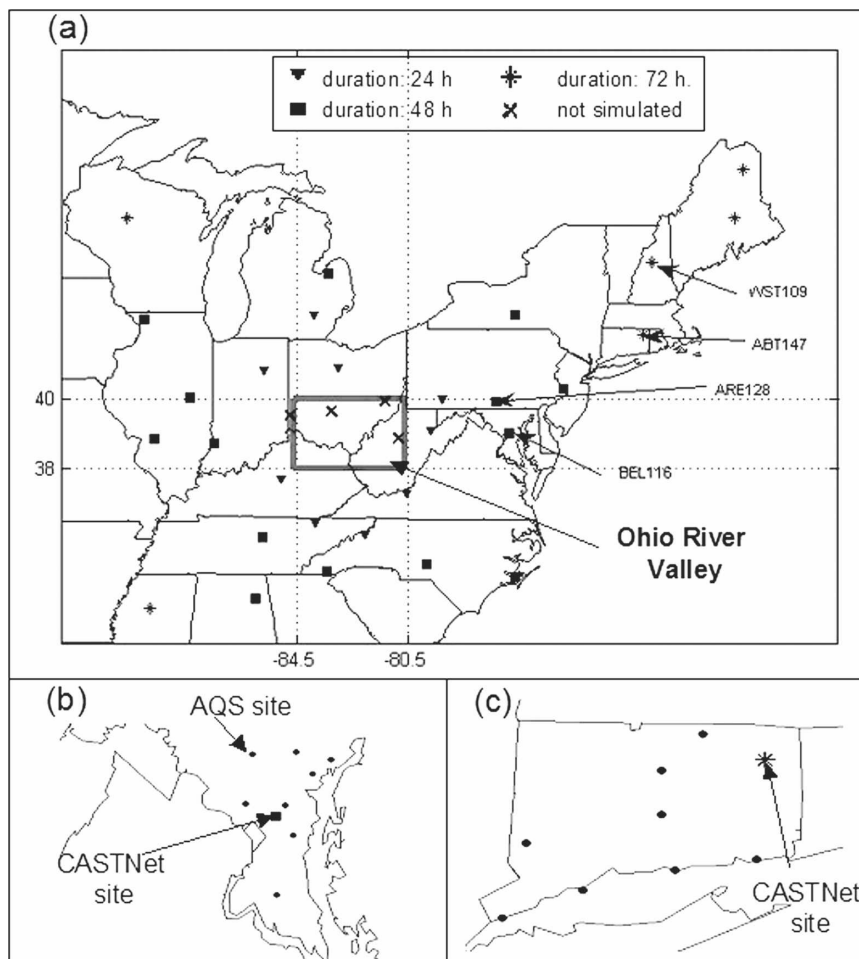


FIG. 3. (a) Location of the CASTNet observation sites included in the study, delineation of the Ohio River Valley, and the duration of transport modeled. Location of AQS sites plus corresponding CASTNet sites in (b) Maryland and (c) Connecticut.

(CASTNet) sites are analyzed in this study. These data were available on the EPA Web site (online at <http://www.epa.gov/castnet/>). CASTNet data are appropriate for the assessment of the NO_x SIP Call because the observation sites are mostly in rural areas where, as shown by Sillman et al. (1990), ozone production depends strongly on NO_x concentrations and is nearly independent of VOCs. Measurements collected at sites east of -92° (eastern United States) with at least 80% completeness or better for the two ozone seasons of 1997 and 1998 and for the seasons of 2003 and 2004 were selected. An ozone season encompasses the 5-month period from 1 May to 30 September. Data completeness is evaluated on the two 5-month periods as a whole, not month by month. Site selection was also determined on the availability of the meteorological variables (solar radiation, temperature, and relative humidity) used to derive meteorologically adjusted ozone

data. Thirty-three CASTNet sites met the completeness requirement. In addition, to compare the results characterizing CASTNet sites with those obtained with EPA Air Quality System (AQS; see information online at <http://www.epa.gov/ttn/airs/airsaqs/detaildata/>), the latter of which are used to determine compliance with the ozone NAAQS, we also examined ozone concentrations at eight AQS sites in Connecticut and eight sites in Maryland. Each AQS site in this study also met the completeness requirement of 80% or better imposed on the CASTNet data and was matched with the closest and most meteorologically representative National Weather System weather station to derive meteorologically adjusted concentrations. Figure 3 identifies location of the 33 CASTNet sites included in this study (Fig. 3a), as well as that of the AQS sites in Connecticut (Fig. 3b) and Maryland (Fig. 3c). The full identification code is provided for the four CASTNet sites

whose results are individually displayed later in this paper.

3. Methods of analysis

a. Accounting for the influence of meteorological conditions on ozone concentrations

Ambient ozone concentrations depend on the concentrations of ozone precursors (NO_x and VOCs) and the prevailing meteorological conditions. For identical precursor concentrations, dry and sunny conditions will lead to more ozone formation than rainy and overcast conditions. Hence, the influence of different meteorological conditions needs to be mitigated for properly when assessing the impact of the SIP Call. The method utilized to accomplish this task entails the decomposition of time series of ozone concentrations and relevant meteorological variables into pairs of time series reflecting fluctuations occurring at two distinct temporal scales [seasonal and longer-term fluctuations (baseline) and weather-induced or synoptic-scale fluctuations] and the use of a multiple regression framework to quantify the incidence of governing meteorological conditions on each temporal scale (Milanchus et al. 1998; Rao and Zurbenko 1994; Kuebler et al. 2001; Ibarra-Berastegi et al. 2001; Wise and Comrie 2005).

More specifically, the iterative moving-average filter described in Zurbenko (1986) and Rao and Zurbenko (1994) used with a half-window width of 15 days and five iterations splits a time series into two, with one having variation at frequencies less than 0.4 month^{-1} (i.e., the synoptic-scale or weather-induced variations) and the other having variations characterized by frequencies greater than 0.4 month^{-1} (i.e., the baseline or seasonal plus longer-term variations). As recommended by Rao et al. (1997), we work with log-transformed ozone data to account for the multiplicative effects of weather on the ozone baseline.

1) IDENTIFICATION OF METEOROLOGICAL VARIABLES INDUCING OZONE BASELINE FLUCTUATIONS

By systematically calculating the correlation between ozone baseline values from 1984 to 1995 and a variety of meteorological variables at six sites in the United States (four of which are in the SIP Call region), Milanchus et al. (1998) showed that ozone baseline fluctuations are better correlated with total daily solar radiation than with temperature. Applying a phase shift (time lag) to the baseline temperature time series (of up to 23 days at more northern locations) could increase the correlation with the ozone baseline, but not quite to the levels reached with solar radiation. Milanchus et al.

(1998) also showed that including a second meteorological covariate linked to humidity (i.e., ozone removal) increases the correlation between ozone concentrations and meteorological conditions. They found that the combination of solar radiation and specific humidity was better than any other combination of two variables in explaining the influence of meteorological conditions on ozone baseline fluctuations. The observations at the CASTNet sites included in this study corroborate the findings of Milanchus et al. (1998) and Chan et al. (2000). Ozone daily baseline fluctuations are best reproduced at all 33 CASTNet sites by solar radiation and specific humidity. The median percent of variance explained by the meteorological adjustment methodology is 62% with this combination, versus 55% when temperature and specific humidity are used. Because solar radiation is not commonly measured at the AQS sites, daily maximum temperature is used as a surrogate, as in Zalewsky (1995) and Flaum et al. (1996).

2) IDENTIFICATION OF METEOROLOGICAL VARIABLES LINKED TO OZONE SYNOPTIC-SCALE FLUCTUATIONS

Following a systematic multiregression approach and considering various pairs of explanatory variables, Milanchus et al. (1998) found that synoptic-scale fluctuations are best explained by solar radiation and dewpoint depression. Of interest is that ozone baseline and synoptic-scale fluctuations are explained by different humidity covariates; ozone baseline is most correlated with specific humidity, whereas the ozone synoptic signature is better reproduced by dewpoint depression. One explanation for this difference may be that specific humidity exhibits a strong seasonal (baseline) signature with only modest short-term (synoptic scale) fluctuations, while dewpoint depression, on the other end, mostly exhibits short-term fluctuations with very limited seasonality. In other relevant studies, Eder et al. (1993), Vukovich (1995, 1997), and Rao et al. (2003) attribute synoptic-scale variability in ozone concentrations in the eastern United States to fluctuations between stagnation events versus ventilating frontal systems. Tying the results of Zalewsky (1995) and Milanchus et al. (1998) to Eder et al. (1993) and Vukovich (1995, 1997) may provide a deeper understanding of the synoptic-scale fluctuations in ozone.

3) MODERATING THE INFLUENCE OF METEOROLOGICAL CONDITIONS ON OZONE CONCENTRATIONS

A multiple linear regression between the ozone baseline component and its two covariates (total daily solar

radiation for CASTNet sites and maximum temperature for AQS sites in conjunction with specific humidity) allows determination of the part of ozone baseline fluctuations explained by meteorological conditions, because the influence of other meteorological variables not included in the regression is considered to be negligible. Conversely, the residuals of the baseline regression represent the portion of ozone baseline fluctuations that cannot be explained by meteorological conditions. In the same way, the residuals of the multiple regression between the ozone synoptic-scale component with total solar radiation and dewpoint depression (maximum temperature and dewpoint depression for AQS sites) allows determination of the part of ozone synoptic-scale features that appear nearly independent of the meteorological variables considered here. The sum for each day of the baseline and synoptic-scale residuals represents (in the logarithmic scale) the portion of ozone fluctuations not explained by meteorological conditions but reflects the changes in ozone we assume to be attributable to emission changes mandated by the SIP Call. Last, daily meteorologically adjusted (met-adj.) values are calculated as the exponential of the daily residuals to which is added the overall mean of the log-ozone data at each site. The met-adj. values, indicative of the scale of the original data and bearing the same units, are used to assess the effectiveness of the SIP Call. Year-round data from 1988 to 2004 were used site by site to derive the met-adj. ozone concentrations.

b. Synoptic-scale variation in meteorologically adjusted ozone

In a modeling effort aimed at identifying the effects of different types of NO_x emission controls on different temporal scales of variation discernable in ozone time series, Hogrefe et al. (2000) showed that a uniform “across-the-board” reduction of anthropogenic emissions would affect the intraday and diurnal variations of ozone, pointing to a high degree of local control. More important to us, they showed that reducing emissions from large elevated point sources has a limited effect on short-term time scales but has substantial effects at longer time scales (synoptic and longer), pointing to a regional control of ozone variations (i.e., pollution transport-based control). Following the results of Hogrefe et al. (2000), we calculated the standard deviation of the synoptic-scale variation in met-adj. ozone concentrations of the “pre-” and the “post-” SIP Call ozone seasons, as well as the difference between the two estimates.

c. Assessment of changes in the meteorologically adjusted daily maximum 8-h ozone concentrations resulting from implementation of the SIP Call

The method chosen to extract the changes in ambient ozone concentrations resulting from the SIP Call first requires calculation of two cumulative distribution functions (CDFs) characterizing the met-adj. 8-h daily maximum ozone observations for the pre- and the post-SIP Call periods, respectively. More specifically, observations collected in the ozone seasons of 1997 and 1998 are used to characterize conditions prevailing in the pre-SIP Call period because the SIP Call was issued in September 1998. Observations collected in the ozone seasons of 2003 and 2004 are used to characterize the post-SIP Call period. Note that all new pollution control measures mandated by the SIP Call were not fully implemented before 31 May 2004 (for further details, see Fig. 6 in Environmental Protection Agency 2005), so the phrase post-SIP Call is not totally accurate.

Once these two CDFs are established, a global estimate of the change is calculated as the mean of the differences between corresponding percentiles reported on the pre- and the post-SIP Call CDFs, divided by the average concentration (in the appropriate percentile interval) of the pre-SIP Call period. Only the 5th to the 95th percentiles were examined, with the CDFs' tails excluded to free us from the effects of extreme values in the data. Working with relative differences (%) simplifies the comparison of sites with different ozone burdens. Calculating differences between all individual percentiles allow us to closely reproduce the shape of each CDF, and greatly facilitates comparison of CDFs formed from different sample sizes.

d. Assessing the impact of emission reductions in the Ohio River Valley on ozone air quality over the eastern United States

The OTAG final report (Ozone Transport Assessment Group 1997) indicated that electrical generation units and other point sources of NO_x in the ORV region were significant contributors to the ozone pollution problem in the northeastern states and the SIP Call was intended to reduce transport of ozone and its precursors. Examination of the continuous emission monitoring data reveals that the ORV region has experienced large NO_x emission reductions following the implementation of the SIP Call. In an effort to link emission changes in the ORV to ozone air quality in downwind areas, calculation of the difference between the pre- and post-SIP Call situations was repeated for those days when a given monitoring site was thought to

be downwind of the ORV, and again for days when transport to the site was from other directions. The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT; release 2005) model was used for this purpose (detailed information on HYSPLIT was available online at <http://www.arl.noaa/ready/hysplit4.html>). The HYSPLIT model can simulate the processes of transport, dispersion, and deposition of pollutants through the atmosphere. For this study, only the air parcel trajectories routines were utilized; dispersion and deposition of pollutant material were not considered. HYSPLIT was set to calculate back trajectories initiated at 1900 UTC each day of the four ozone seasons considered and for each CASTNet site shown in Fig. 3. In the back-trajectory setting, meteorological data are processed in reverse-time order (i.e., the model proceeds upwind), permitting identification of the days when the considered observation site was downwind of the ORV. The latitude and longitude of each monitoring site were successively used as starting locations in HYSPLIT model, with the starting altitude set at 250 m above the ground level for all sites. The duration of each simulation was limited to 24 and 48 h for sites less than 400 or 800 km from the ORV centroid, respectively. Simulation time was extended to 72 h for the sites farther than 800 km from the ORV. Figure 3 recapitulates the simulation time utilized for each site and identifies the ORV centroid.

The HYSPLIT model was applied with Eta Model Data Assimilation System (EDAS) archived files or the National Centers for Environmental Prediction–National Center for Atmospheric Research (NCEP–NCAR) global reanalysis fields (both were available online at <ftp://gus.arlhq.noaa.gov/pub/archives/>). EDAS fields were used primarily because their spatial and temporal resolution is better than that of the NCEP–NCAR reanalysis fields (80 km and 3 h vs 2.5° latitude \times 2.5° longitude and 6 h). NCEP–NCAR reanalysis fields were used to simulate 2004 because EDAS fields were not yet available through the web for 2004. They were also used for about 5 days of the ozone season of 1997 and 60 days of the 1998 season for which EDAS fields contained missing data that led to premature terminations of the trajectories.

Uncertainty in the trajectories was taken into account by running the ensemble form of HYSPLIT in which multiple trajectories automatically depart from the selected starting location by offsetting the meteorological data by a fixed grid factor. In this study, the offset grid factor was set equal to one-half (0.5) of a meteorological grid point in the horizontal direction, which corresponds to a shift of about 150 km in both the x and y directions with the NCEP–NCAR reanaly-

sis fields. For this study, no offset was imposed in the vertical direction (default value = 1) because we focus only on the plane position (longitude, latitude) of the trajectories and not their heights. In our setting of the ensemble form of HYSPLIT, nine offset trajectories were calculated for each initial time, that is, for each day simulated. A site was considered downwind of the ORV on a given day if at least one of the nine trajectories initiated led back to the ORV area and intercepted the longitude–latitude rectangle marking its location (see Fig. 3), regardless of vertical coordinate (height). The ORV location (rectangle) includes six of the largest electric generation units in the valley. Another possibility for including trajectory uncertainty in our analysis would be to initiate trajectories at different hours within a given day. In this context, HYSPLIT was set to calculate four trajectories within a window of 3 h for each day simulated. This latter method resulted in little difference in the back-trajectory termination locations.

4. Results

a. Importance of adjusting the ozone levels for meteorological variability

To illustrate the differences between the results one may infer from the raw data versus the met-adj. 8-h ozone observations, Fig. 4 displays the CDFs characterizing the pre- and post-SIP Call periods at site ABT147 (in Connecticut). Figure 4a displays the raw data, that is, ozone not adjusted for meteorological conditions while Fig. 4b depicts the met-adj. information. At this site, it appears that meteorological adjustment has reduced the spread of the CDF. In addition, it seems that meteorological conditions at the ABT147 site were favorable to ozone formation in 1997–98 but were not as conducive to ozone formation during 2003–04, as one can infer from the fact that the distance between the pre- and the post-SIP Call CDFs established from the raw data (see Fig. 4a) is larger than that derived from the met-adj. data (see Fig. 4b). Using the methodology described above to calculate the changes in the daily maximum 8-h ozone, it appears that the ozone improvement between the pre and post-SIP Call situations would be estimated at 17% if deduced from the raw data versus only 12% if deduced from the met-adj. data. Examination of the differences between changes estimated from the raw or met-adj. ozone data reveals that the met-adj. change is smaller than that assessed from raw data along most of the Atlantic Ocean coast (i.e., from North Carolina to Connecticut). On the other hand, the changes in met-adj. values are larger than those assessed from the raw data in the Midwest (Wisconsin and Illinois) and Tennessee. Elsewhere, the

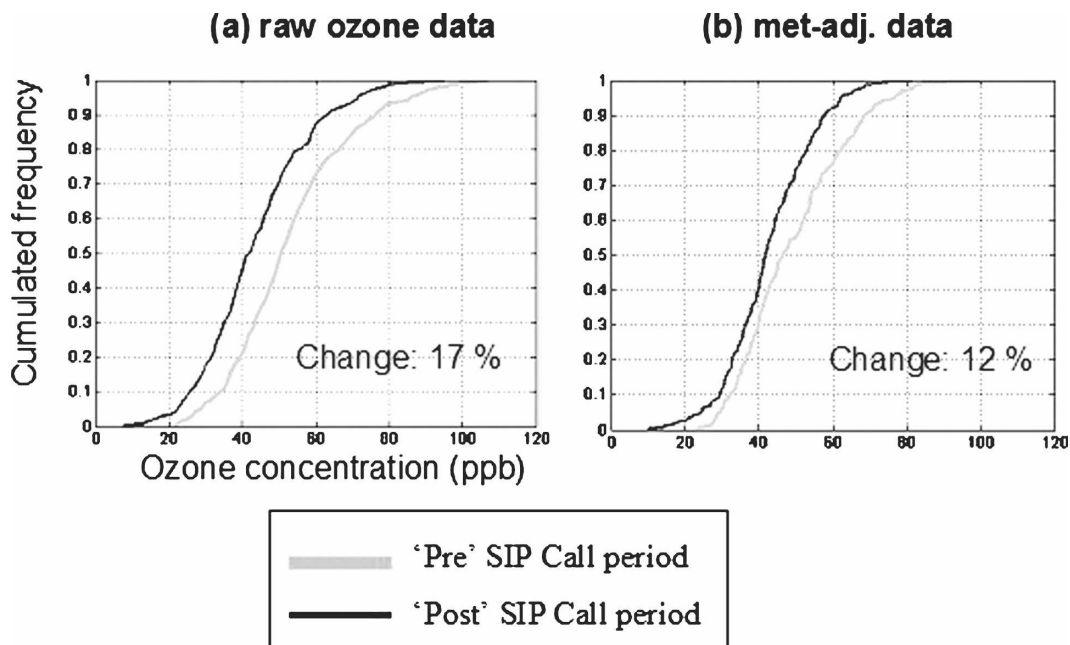


FIG. 4. Cumulative distribution functions of the daily maximum 8-h ozone concentrations at ABT147 site during the pre- and post-SIP Call periods, based on (a) raw and (b) met-adj. data.

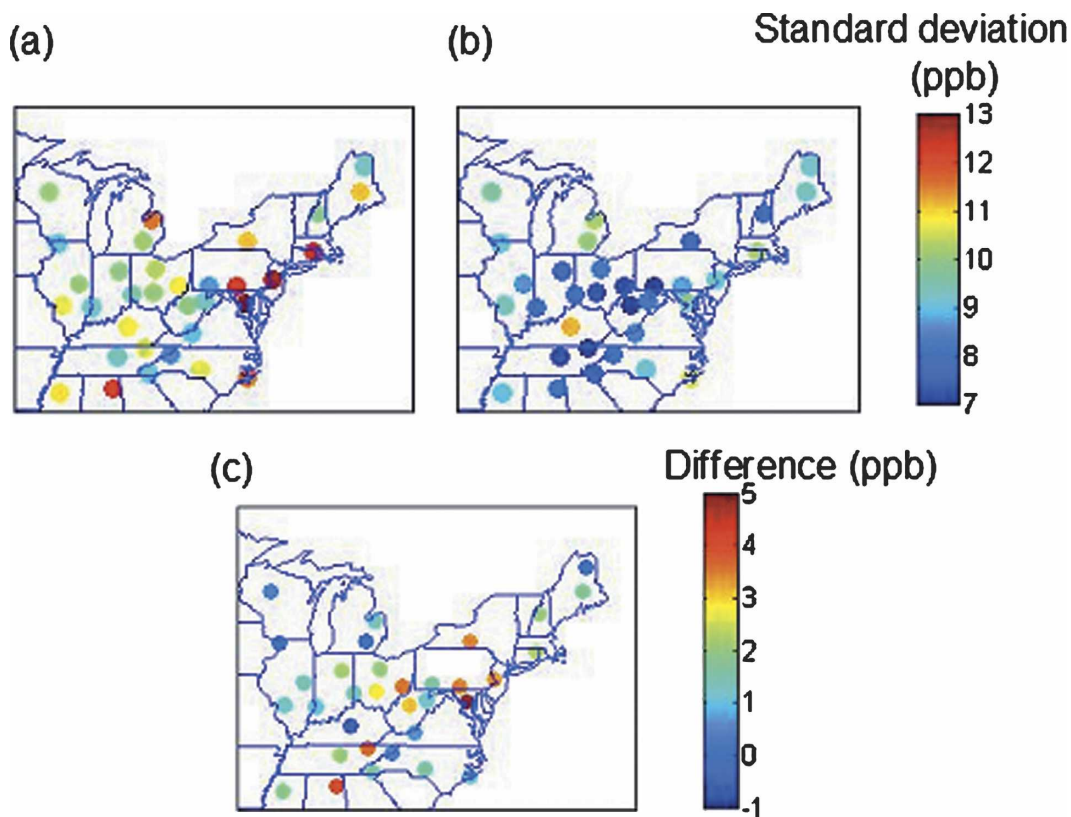


FIG. 5. Synoptic-scale variation (std dev) in meteorologically adjusted ozone at study sites: (a) pre-SIP Call conditions, (b) post-SIP Call conditions, (c) difference between (a) and (b).

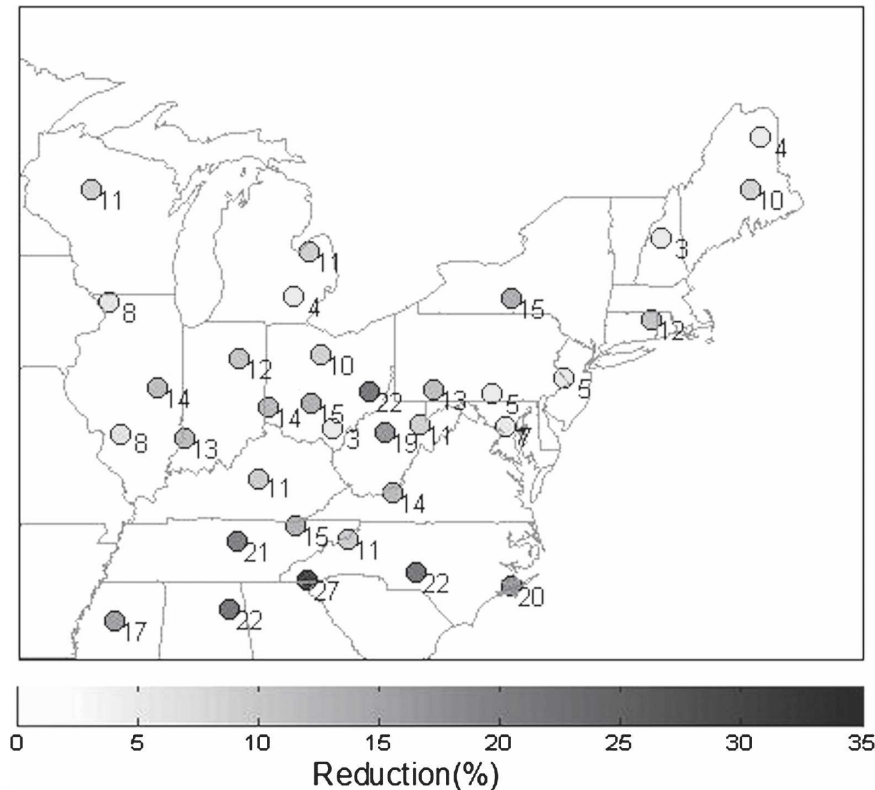


FIG. 6. Reduction (%) of the met-adj. daily maximum 8-h ozone concentrations resulting from implementation of the SIP Call – CASTNet sites.

changes estimated from met-adj. and raw data are very similar.

b. Changes in the strength of the synoptic-scale signal in meteorologically adjusted ozone

As indicated before, the primary objective of the NO_x SIP Call is to reduce the regional-scale transport of ozone and its precursors by reducing NO_x emissions from point sources. Synoptic-scale variation in met-adj. ozone (characterized by the standard deviation) decreased at all but 2 of the 33 monitoring sites in the study region between the pre- and post-SIP Call ozone seasons (Fig. 5). These observation-based results corroborate the model-based results of Hogrefe et al. (2000) that the SIP Call has indeed affected the energy content of the synoptic-scale forcing in ozone, reflecting the influence of the long-range transport of ozone pollution.

c. Changes in the meteorologically adjusted daily maximum 8-h ozone concentrations after the implementation of the SIP Call

Figure 6 displays the change in the daily maximum 8-h ozone concentrations at all 33 CASTNet sites in-

cluded in this study. All sites experienced a reduction in ozone concentrations after the implementation of the SIP Call ranging from 4% to 27%, (on average 13%) of the ozone concentrations observed prior to the SIP Call. Our estimate of the ozone improvement from the pre- to post-SIP Call period is consistent with the 14% improvement reported in Environmental Protection Agency (2005) for the same period using a different meteorological adjustment technique. The largest improvement is evident over the southern part of the domain (northern Mississippi, northern Alabama, Tennessee, and North Carolina), as well as at sites in the vicinity of the ORV. Ozone reductions also occurred for New England and the urban corridor in the Northeast (eastern Pennsylvania, Maryland, and New Jersey), but to a lesser magnitude.

Improvement in ozone air quality is evident at all AQS sites in Connecticut and Maryland, as depicted in Fig. 7, which is interesting because the AQS sites involved in this study are situated in rural locations as well as residential, suburban, and commercial environments. Thus, the benefits of the SIP Call seem to extend beyond the countryside. Also interesting is the large variability of ozone improvements evaluated at nearby

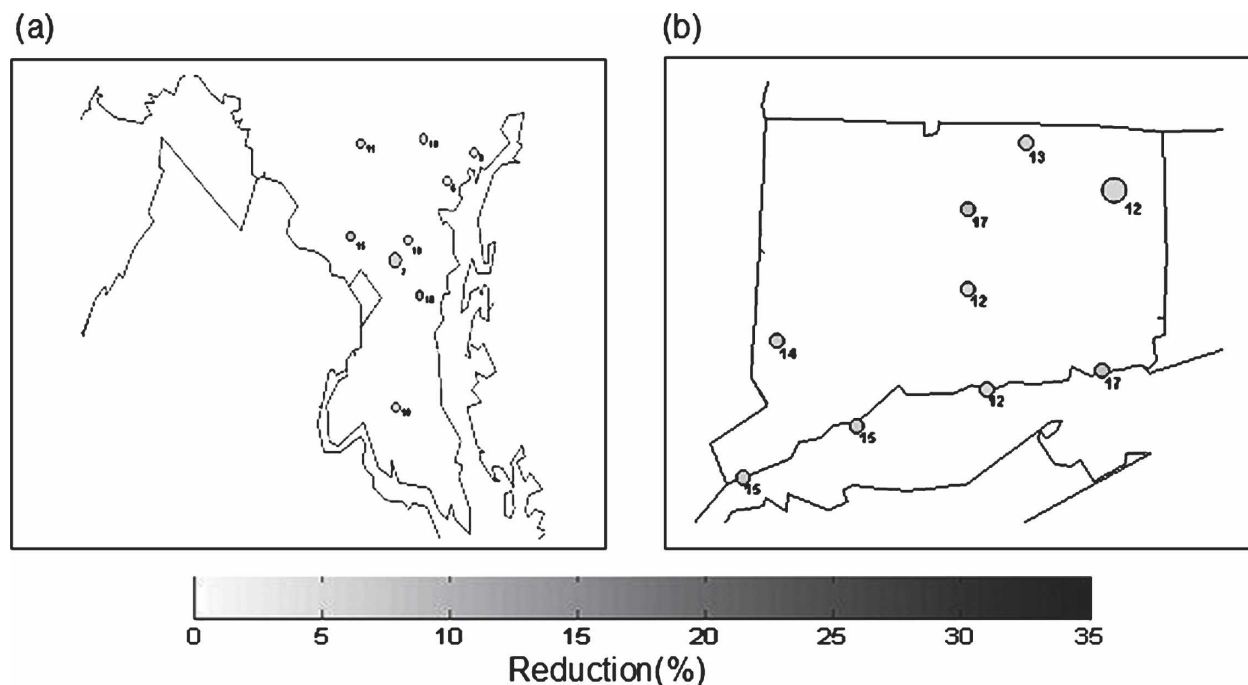


FIG. 7. Reduction (%) of the met-adj. daily maximum 8-h ozone concentrations resulting from the implementation of the SIP Call-AQS sites in (a) Maryland and (b) Connecticut.

sites. The range of ozone changes in the state of Maryland (from 6% to 18%), for instance, is the same as that found at widely separated CASTNet sites (from 3% in New Hampshire to 15% in New York) throughout the northeastern United States. Note that CASTNet estimates of change in both Maryland and Connecticut are between the minimum and maximum changes determined at the AQS sites in the corresponding states.

d. Assessing pollution transport

HYSPLIT was used to calculate back trajectories from each CASTNet site, except for the four sites located in the ORV. Our objective is to use the results of this back-trajectory simulation to separate the days when the trajectories led back to the ORV from the days when they did not. First, Fig. 8 depicts the overall variability of transport direction from/to site ABT147 during the ozone season of 1998. All trajectories (one trajectory per day) that led back to the ORV appear in red, while trajectories toward other directions are displayed in a different color. Obviously, the 50 trajectories drawn extended in all directions, and show that the site is successively under the influence of various transport regimes during the ozone season. Illustrating our handling of the spatial uncertainty in HYSPLIT, Fig. 9 displays the nine trajectories calculated for 21 May and 28 July 2003. Each of these trajectories describes the

transport direction after offsetting the meteorological fields (EDAS fields in this case) half a grid point in the *x* and *y* directions, that is, about 40 km. Although the size of the offset was similar, impacts on transport are very different. While the offset did not affect the trajectories of 28 July very much, it greatly affected the

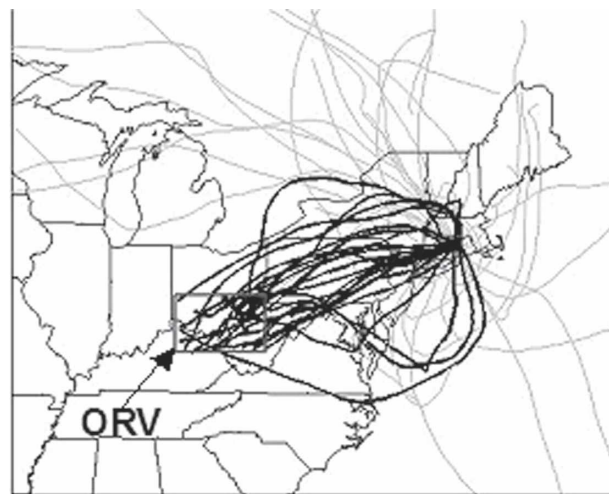


FIG. 8. Examples of back trajectories modeled from site ABT147 during the ozone season of 1998. Thick black lines indicate trajectories that led back to the Ohio River Valley, gray lines indicate trajectories that did not lead back to the Ohio River Valley.

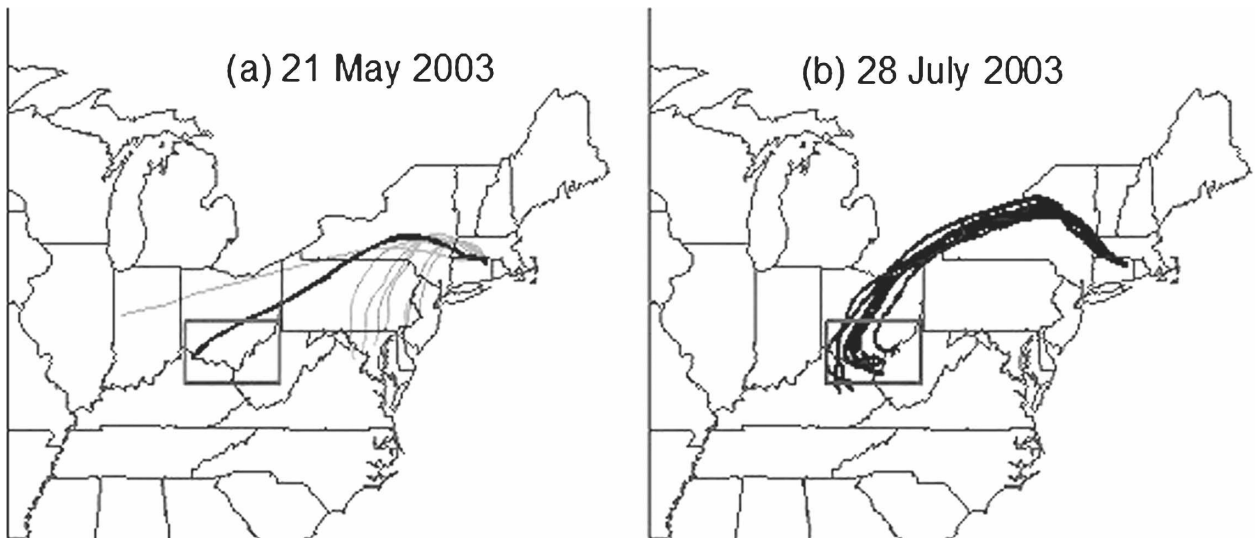


FIG. 9. Modeling spatial uncertainty with HYSPLIT: trajectories calculated for (a) 21 May and (b) 28 Jul 2003 1998. Thick black lines indicate trajectories that led back to the Ohio River Valley; gray lines indicate trajectories that did not lead back to the Ohio River Valley.

results on 21 May, because only one trajectory intercepted the ORV. Varying levels of stability in wind speed and direction explain the latter results. The spreading observed for the 21 May trajectories and the spatial steadiness of 28 July results suggest the possibility of assigning different certainty levels to each day

simulated, as far as determining the influence of emissions from the ORV is concerned. Because of the very limited number of days leading back to the ORV, especially for some sites, we did not develop this possibility. A site was said to be downwind of the ORV on a given day even if one trajectory led back to this area.

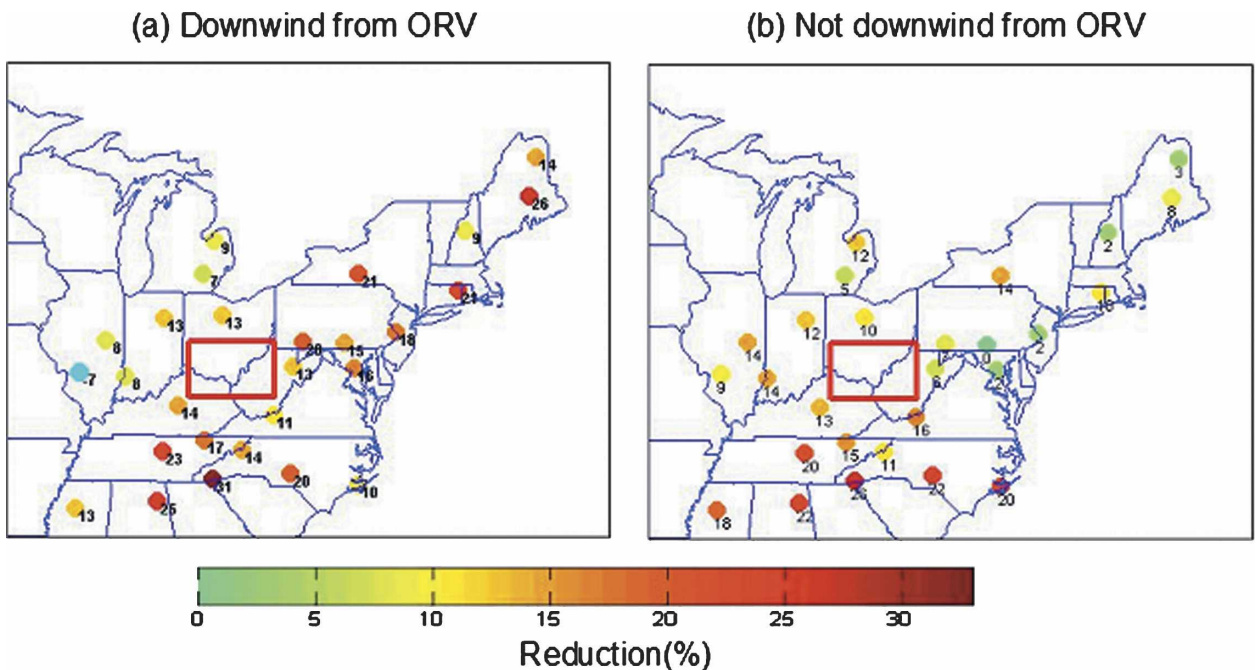


FIG. 10. Comparison of the reduction (%) of the met-adj. daily maximum 8-h ozone concentrations when (a) downwind vs (b) not downwind from the Ohio River Valley.

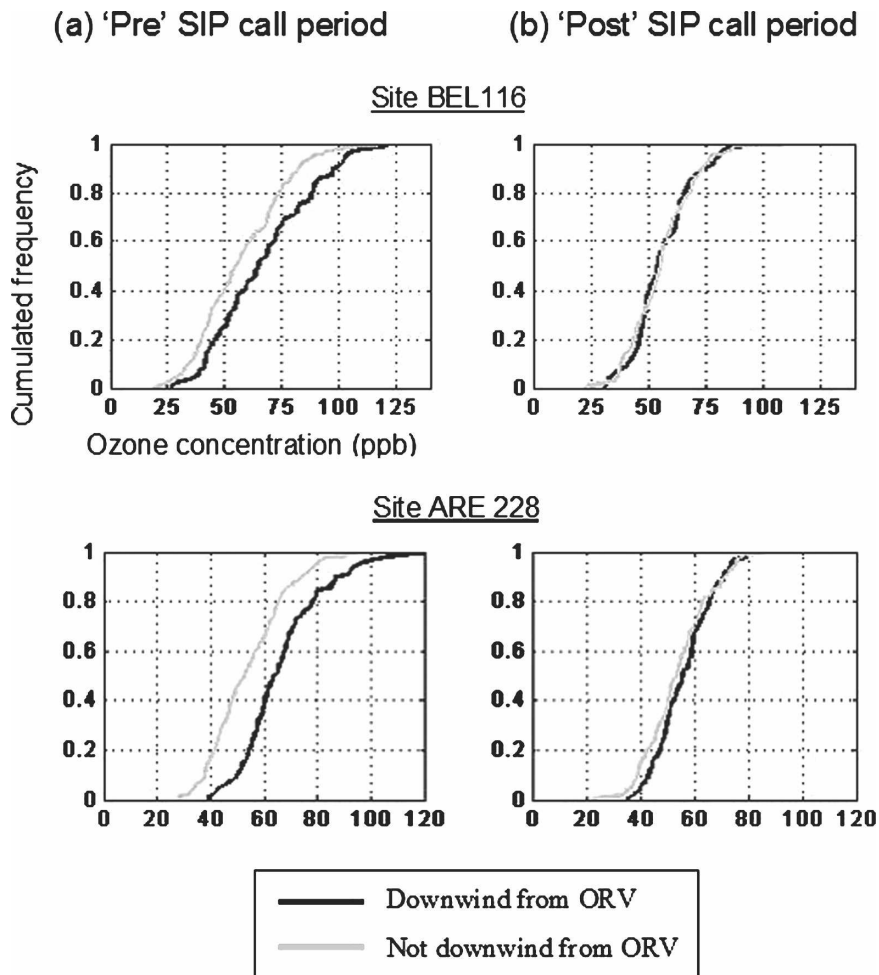


FIG. 11. Cumulative distribution functions of the met-adj. daily maximum 8-h ozone concentrations at sites BEL116 and ARE228 for (a) pre-SIP Call and (b) post-SIP Call conditions.

e. Changes in the meteorologically adjusted daily maximum 8-h ozone concentrations resulting from controls on point sources in the ORV

The results from HYSPLIT allowed identification of days when each ozone measurement site was downwind of the ORV. The segregation of all days simulated into a “downwind” or “not downwind” category allows us to assess the impact of reduced emissions in the ORV on ozone air quality at different locations in the eastern United States. Note that widespread NO_x emission reductions were applied throughout the whole SIP Call region, not just in the ORV where they were, nonetheless, remarkably intense. Therefore, days labeled as “downwind from the ORV” should, strictly speaking, be referred to as downwind from the ORV and other major NO_x sources (i.e., urban areas between the ORV and an observation site). Similarly, the changes in

ozone concentrations that we attribute to emission reduction in the ORV should be understood as also imputable to changes that occurred elsewhere along the path of the trajectories leading back to ORV, as well as to local changes. When identifying an overall reduction in met-adj. ozone concentrations at any given location, we do not distinguish the impact of local emission reductions from that of interstate transport.

Two sites in the northwestern corner of the domain (Wisconsin and northern Illinois) had to be discarded from our analysis because the number of days identified as being downwind was insufficient to provide meaningful results (less than 10 downwind days for each sites during the pre-SIP Call period). Figure 10 displays, side by side, the changes calculated for each site in the downwind (Fig. 10a) and not downwind (Fig. 10b) situations. This separation of the transport directions does not seem to have much impact in the region south of

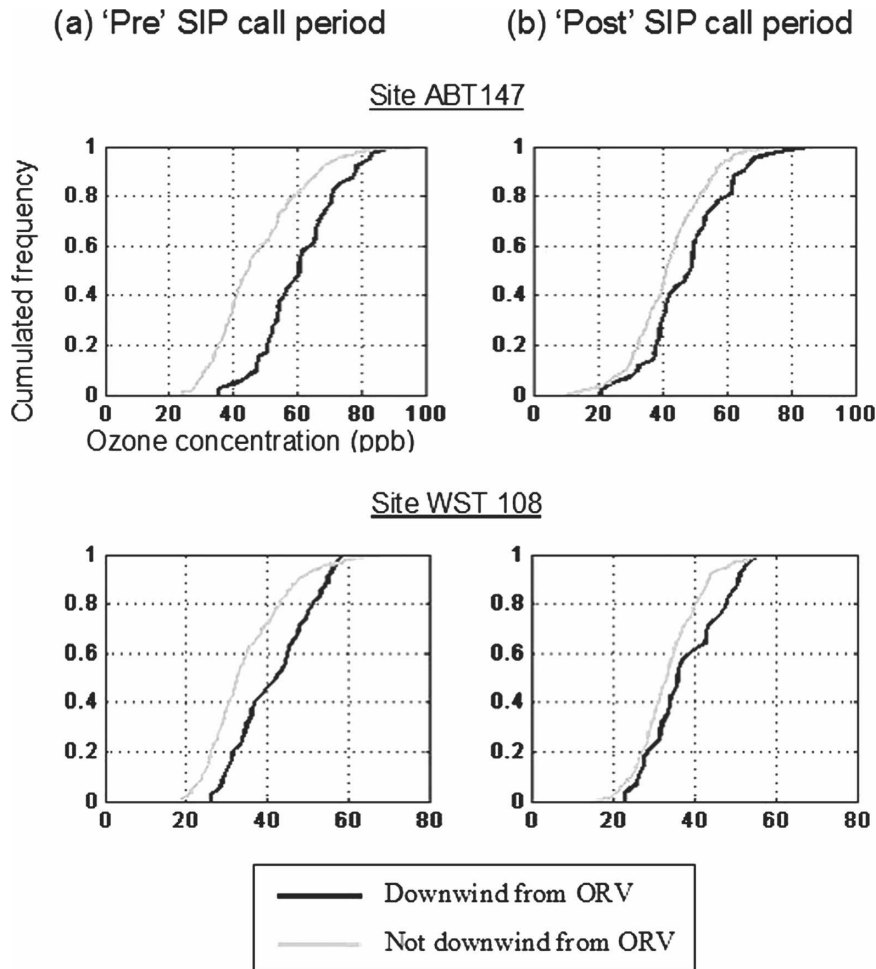


FIG. 12. Cumulative distribution functions of the met-adj. daily maximum 8-h ozone concentrations at sites ABT147 and WST 108 for (a) pre-SIP Call conditions and (b) post-SIP Call conditions.

the ORV. However, calculated reductions in ozone concentrations in the east and northeast of the ORV are larger when the sites are downwind than when the transport is from other directions. Following the SIP Call, high-performance emission control devices are now utilized in all states affected by the new regulation, including the ORV. Therefore, a likely explanation for the latter result is that, prior to the SIP Call, transport from the ORV was causing higher-than-normal ozone concentrations downwind, consistent with the findings of the Ozone Transport Assessment Group (1997) and Sillman et al. (1990). As illustrated in Fig. 11, while ozone concentrations at BEL116 (Maryland) and ARE228 (eastern Pennsylvania) used to be higher than average when that site was downwind of the ORV, one can no longer differentiate between the downwind versus the not downwind situation because of the implementation of the SIP Call. In other words, pollution

transport from the ORV now resembles transport from all other regions. Reinforcing the same finding but to a lesser measure, Fig. 12 displays the CDFs of ozone at sites ABT146 (Connecticut) and WST209 (New Hampshire), both of which are in the northeast. While ozone concentrations are still higher than average when these sites are downwind of the ORV, the difference between the downwind and not downwind scenarios has been greatly reduced since the implementation of the SIP Call.

5. Summary

Recognizing the OTAG's work on long-range transport of ozone and its precursors, the EPA issued the NO_x SIP Call requiring 21 states in the eastern United States and Washington, D.C., to reduce their summertime NO_x emissions to help to alleviate the ozone trans-

port affecting the states in the eastern and northeastern United States. In this paper, we examined the effectiveness of the emission control measures from the NO_x SIP Call by quantifying changes that occurred in the daily maximum 8-h ozone concentrations measured at 33 CASTNet sites and 16 AQS sites in the eastern United States. The method used to calculate changes relies on the establishment of two cumulative distribution functions (CDFs) characterizing the ozone observations in the pre- and post-SIP Call periods and evaluation of the mean differences between the 5th to the 95th percentiles reported on each CDF. The ozone seasons of 1997–98 were used to define the pre-SIP Call conditions, while the seasons of 2003–04 define the post-SIP Call conditions. Because other NO_x emission control programs, such as the Acid Rain Program, were in place after 1997 but prior to the SIP Call program, our results characterize the global effect of all NO_x controls implemented during the 1997–2004 period and not just from SIP Call. However, the changes in ozone air quality stemming from the NO_x SIP Call are considerably more pronounced than those resulting from the other NO_x emission reduction plans (Environmental Protection Agency 2005).

Because ozone formation and buildup depend not only on the concentrations of precursors (NO_x and VOCs) but also on the prevailing meteorological conditions, the effects of meteorological conditions on ozone must be moderated to properly assess the efficacy of emission controls. In this paper, a multiple regression method was used to accomplish this task. The results reveal a strong decline in the daily maximum ozone 8-h concentrations at all sites studied in the post-SIP Call period, with the largest decreases (around 25%) in northern Mississippi, northern Alabama, Tennessee, and North Carolina. Prior to the SIP Call, large NO_x emissions from the ORV were thought to be a major contributor to poor ozone air quality in the eastern and northeastern United States. The results of trajectory analyses permitted corroboration of the latter statement, because they showed that emission reductions in the ORV resulted in a substantial improvement in ozone air quality east and northeast of the ORV. Ozone concentrations at the eastern monitoring sites are no longer higher when these sites are downwind of the ORV than when they are affected by transport from other directions. In the northeast, transport from the ORV still seems to lead to higher-than-average ozone concentrations, but to a considerably lesser extent than before the SIP Call. In other words, emission controls implemented under the NO_x SIP Call have been effective in meeting the objectives of reducing interstate

ozone transport and helping to improve ozone air quality throughout the eastern United States.

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