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ESTIMATING THE AREA OF INFLUENCE OF OZONE PRODUCED BY LOCAL PRECURSOR EMISSIONS FOR A SUMMER PERIOD WITH A RANGE OF PHOTOCHEMICAL ACTIVITY

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1. INTRODUCTION

Regional transport of ozone (O3) and its precursors is suspected to significantly affect O3 control strategies and the effectiveness of local controls throughout much of the eastern U.S. A Federal Advisory Committee Act (FACA) work group is studying the identification of Areas of Influence (AOI's), essentially O3 airsheds, around which to design controls for subregions of the eastern U.S. It is difficult, however, to quantify the production, life-time and effect of transport on regional/urban O3 concentrations ([O3]). It is also difficult to quantify the sensitivity of the transported O3 to hydrocarbon (VOC) or nitrogen oxide (NOx) emissions or to emissions reductions in any given region or urban area. At least two types of methodologies have been proposed to evaluate AOIs. They include sensitivity studies to evaluate the change in [O3] in the AOI when emissions in the source region are changed (Yang et al, 1997); and Process and Mass tracking studies to evaluate the actual transport of mass and the production of O3 on route from the source region to the AOI (Lo and Jeffries, 1996; Yarwood et al., 1997). Each of these approaches has limitations: sensitivity studies have predictive value but they lack explanatory value; ie, they do not provide adequate insights into the processes and feedbacks that cause the change in [O3] in the AOI. They may ignore feedbacks in the system which effectively buffer [O3], thereby underestimating the size of the AOI. Further, computed sensitivities may vary considerably with the base case scenario conditions, so it is unclear how sensitivities should be calculated or applied in light of changes in future scenario conditions. Process and Mass tracking studies do have considerable explanatory value; they can explain in great detail how precursor and O3 transport determine [O3] in the AOI for a particular scenario. They do not, however, have predictive power; ie, they do not predict exactly how emissions reductions will affect [O3] in the AOI. We propose that these methods have complementary strengths and must be used together both to define the AOI and to understand the impact of emissions reductions in the AOI. In this study, we use sensitivity simulations to evaluate the AOI of precursor emissions in selected source regions, and we use a process analysis to explain the results of those sensitivity simulations.

2. MODEL AND SCENARIO CONDITIONS.1995

We used a version of the Regional Acid Deposition Model (RADM) (Chang et al., 1990), a complex, three-dimensional grid model, that we modified to calculate and output integrated reaction and process rates (Tonnesen and Dennis, 1997). We used 21 vertical layers and a 20 km grid resolution in a domain extending from Kentucky to Maine, one-way nested in an 80 km grid that extended from southern Texas to New Brunswick, Canada. Meteorological inputs are described in Li et al. (in this volume). We conducted a series of emissions source modulation experiments using a 25-day base case scenario during July and August, 1988, that includes two frontal passages which clean out the system, and two periods of stagnant conditions in which peak model [O3] exceeds 200 ppb. For the source modulations we reduced either NOx emissions, VOC emissions, or both NOx and VOC emissions by 50% in selected model grid cells. We then evaluated the area of influence of these source cells by calculating changes in [O3] and its precursor concentrations, and the change in the sensitivity of [O3] to precursors in areas downwind of the source cell. For example, we calculated the change in hourly [O3] using: Delta = Modulation-Base Case, so negative deltas represent decreases relative to the base case. We conducted individual source modulations in the

Figure 1. Model domain and areas of source modulation.

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four urban areas shown in Figure 1: a 40 by 120 km area around Pittsburgh, a 60 by 80 km area near Baltimore., a 40 by 60 km area near Philadelphia, and a 60 by 80 km area around New York City; and a combined modulation using all four areas.

3. RESULTS.

We evaluated the response of [O3] and odd oxygen production, P(Ox), by examining the hourly perturbation in the plumes moving downwind from the urban areas. We discuss briefly here the results for the combined 50% VOC and NOx reduction. The size of the AOI will be determined in part by the magnitude of perturbation defined to be significant. Considerable study is still required to determine what constitutes a significant response in relation to control strategy development, but for this exploratory study we used a change in hourly [O3] of more than 2 ppb as a significant response, and then observed the [O3] response plume for the 25 day simulation period. In all model simulations with NOx emissions reductions, nighttime and morning [O3] increased near urban centers and other large NOx sources because of decreased titration of O3 by the reduced NO emissions. As a result, early morning [O3] typically increased by several ppb from levels below 20 ppb in the base case. Then, as the day progressed, the increases in urban [O3] gradually became smaller, and then switched to [O3] decreases by late morning to mid afternoon. (The only exception to this was several cells in the center of NY City for which [O3] increased at all hours.) Daytime [O3] levels always decreased at greater distances downwind of the source modulation areas; maximum reductions occurred late in the afternoon at the time of peak [O3]. The [O3] reductions did not persist through the night-time in the surface layer because of the effects of titration, deposition, and chemical reactions of [O3], but the [O3] reductions did persist through the nighttime above the surface layer. As the nighttime inversion broke up in the mid-morning, the [O3] reduction became evident again in the surface layer.

We describe here the results of the combined VOC and NOx reduction for a simulation from July 28 to August 1, 1988. This was one of the stagnant, high [O3] episodes with light south-westerly to north-westerly winds. During this period, the Pittsburgh AOI (defined by a plume of 2 ppb reduction in hourly [O3]) first extended south-eastward through most of Maryland on July 28 and 29; then north-eastward to about 200 km north of NY City on July 30 and 31; and finally south-eastward to the North Carolina border on August 1. The plumes of 2 ppb [O3] reduction from Baltimore, Philadelphia and NY City showed similar spatial extent and temporal variability. The plume for the combined 4 area source modulation extended from NC, across the eastern boundary over the Atlantic Ocean, and north into Maine. It is convenient to show the AOI over a period of several days by integrating the [O3] response plume over time to show cumulative response. Figure 2a shows the integrated [O3] response as ppb-hours for the four urban areas combined. The dark shades over the urban areas show net increases in [O3]. The maximum [O3] increase was 1840 ppb-hours over southern Long Island; this represents an average hourly [O3] increase of 18 ppb. It is useful to determine if this increase is primarily due to low [O3] levels increasing to values still below the [O3] standard, or more problematic increases in high [O3], so we also calculated the response using cumulative hours of [O3] above 80 ppb (not shown here); in this case there was no increase in [O3] for Pittsburgh, Philadelphia, or Baltimore. The increased [O3] was limited to a smaller area around New York City, and was again largest in the south Long Island cell, with an average hourly [O3] increase of 9 ppb. This shows that most of the increases in [O3] occurred during early morning hours and was caused by reduced titration of O3 resulting from the NO emissions reductions, and was primarily an increase from near zero to less than 80 ppb.

The white areas in Figure 2a show areas with integrated [O3] reductions greater than 100 ppb-hours. We note that there may be considerable temporal variation in the hourly [O3] that is not evident in the integrated 108 hours response. The largest integrated [O3] reductions, greater than 500 ppb, occurred over Rhode Island. The cumulative [O3] plume extended from northern Virginia to central Maine, and beyond the eastern boundary over the Atlantic Ocean. High [O3] levels result from local P(Ox) and from transport of O3 produced in upwind cells. An evaluation of the P(Ox) response plume, or the P(Ox) AOI, is useful in explaining the [O3] AOI. Figure 2b shows the cumulative P(Ox) response for the same time period as Figure 2a. The change in [O3] shown in Figure 2a results directly from the change in P(Ox) in Figure 2b. Production rates are integrated over five days, so a -60 ppb response indicates an average reduction in P(Ox) of 12 ppb/day. For each of the four urban areas, the P(Ox) response plume extended approximately 200 km from the source modulation region. For the most part, the plumes did not overlap, so there was little interaction between P(Ox) plumes. If the plumes did overlap, we would expect non-linear interactions in P(Ox) for the combined modulation. The precursor response plumes are not shown, but they extended over approximately the same area as did P(Ox). Figure 2b shows that the AOI of [O3] is significantly enlarged because P(Ox) itself has an AOI that extends about 200 km downwind from the source modulations. As a result, the [O3] AOI would be some 200 km larger than that of a reacting tracer that decays with the same loss rate as [O3].

Figure 3 shows a 36 hour time-series for a 400 by 400 km area around the source modulation areas. Figure 3a shows average reductions in [O3] of 2 to 3 ppb at 6 PM, and reductions in P(Ox) of 0.8 ppb/hr at noon. In general, reductions in P(Ox) are caused by reductions in

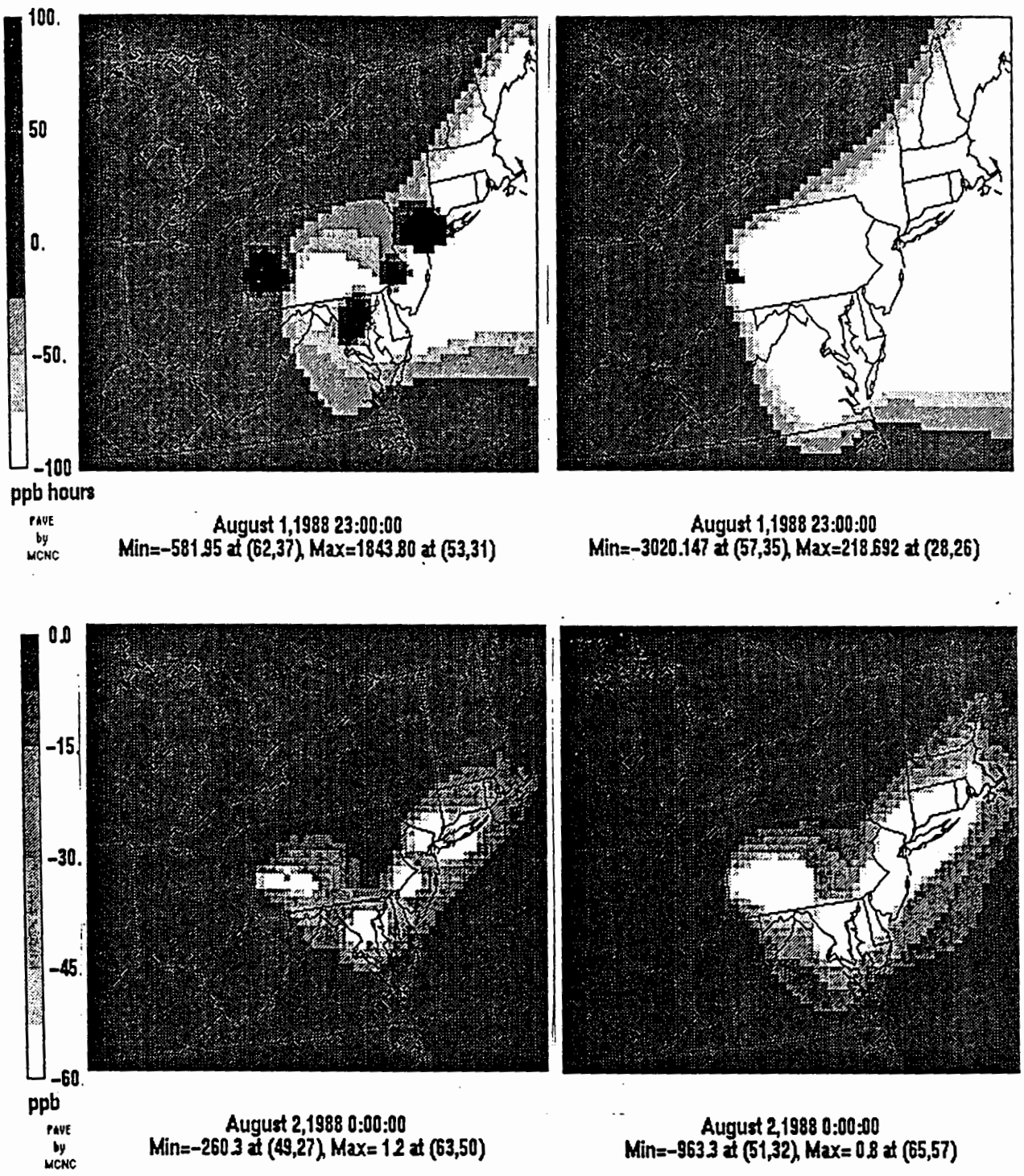

Figure 2. Cumulative response to the combined VOC and NOx source modulation integrated over 108 hours for two different base case scenarios: on the left, (a) and (b) show the cumulative [O3] and P(Ox) response for the high O3 scenario; on the right, (c) and (d) show the cumulative [O3] and P(Ox) response for the low O3 base case scenario.

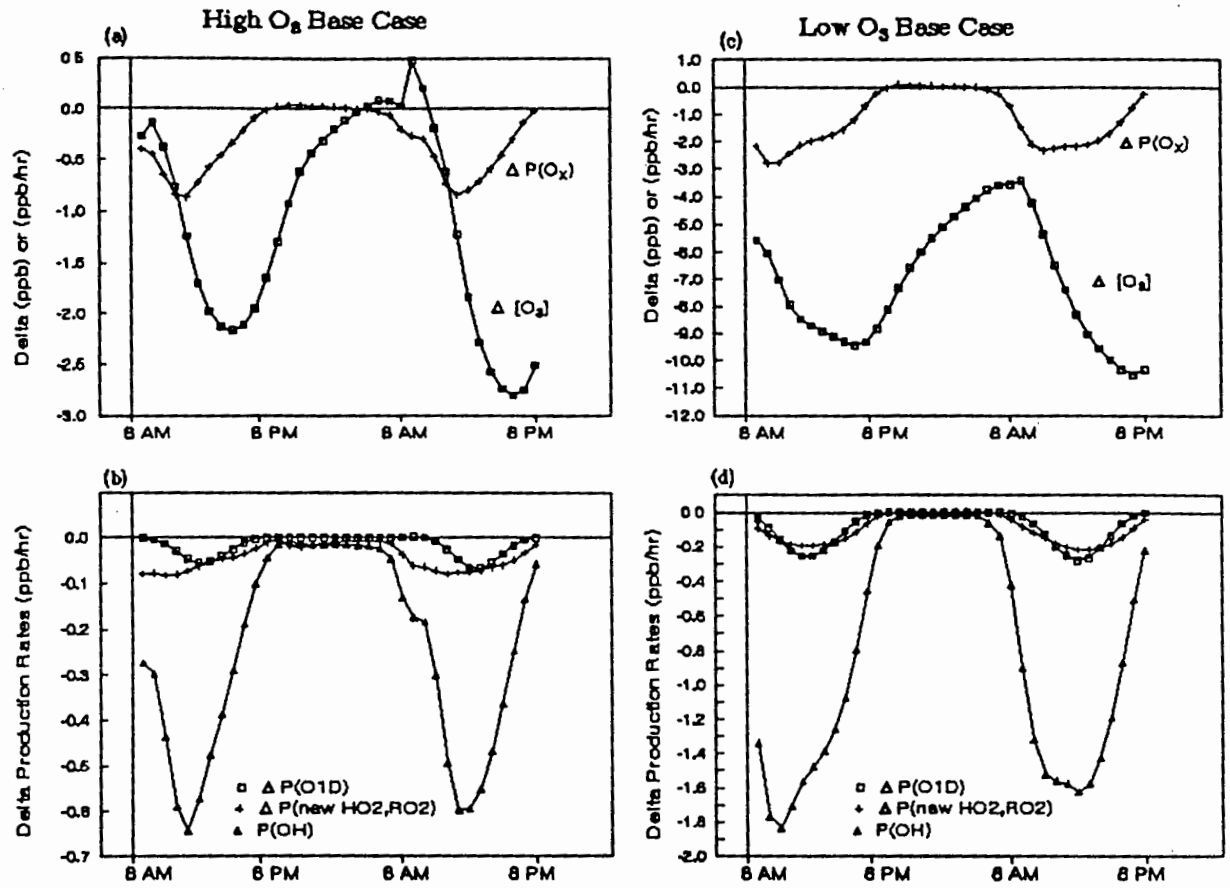

Figure 3. Time-series showing the photochemical response to the source modulation for a 36 hour period, averaged over a 400 by 400 km sub-domain: (A) change in P(Ox) and [O3] for the emissions modulation of the high O3 scenario and (B) the low O3 scenario; (C) effects on radical initiation and total OH production for the high O3 scenario and (D) for the low O3 scenario.

NOx and VOC precursor levels, but the P(Ox) reduction can be amplified by feedback effects due to reduced radical initiation via O3 photolysis as a result of the [O3] reduction. In this case, the P(Ox) response plume was determined primarily by the precursor response plumes with a lesser contribution from the reduced O3 radical initiation. This is illustrated in Figure 3b which shows that the change in radical initiation by photolysis of aldehydes, P(new HO2,RO2), is larger than the change in radical initiation by photolysis of O3, P(O1D).Total OH production, P(OH), is determined by the product of radical initiation and OH chain length, so the approximately 0.1 ppb/hr reductions in radical initiation result in reductions in P(OH) of up to 0.6 ppb/hr. Changes in P(Ox) in Figure 3a result from and are proportional to the changes in P(OH) in Figure 3b.

Figure 4 shows the O3 production efficiency time series. In the high O3 base case, P(Ox)/P(HNO3) averaged 9 ppb/ppb. In the high O3 source modulation, total P(Ox) decreased, but P(Ox)/P(HNO3) increased to 11 ppb/ppb. Thus, the decrease in [O3] and P(Ox) was smaller than we might have expected because the O3 production efficiency increased with the NOx emissions reduction. The ratio of P(Ox)/P(HNO3) is also correlated with P(Ox) and [O3] sensitivity to NOx and VOC, (Tonnesen and Dennis, 1997). A ratio of 9 ppb/ppb indicates conditions of equal sensitivity to VOC and NOx. The ratio averaged over this sub-domain increased from 9 to 11 ppb/ppb with the combined VOC and NOx reduction indicating that the source modulation case has become relatively more sensitive to VOC reductions.

We expect that the characteristics of the base case scenario will affect [O3] sensitivity in several ways: the production efficiency of O3 per unit of precursor emissions and the sensitivity of [O3] to VOC and NOx vary considerably with the scenario conditions; O3 loss rates and chemical lifetime varies with [O3] and other species concentrations and with deposition rates. These and other photochemical feedback processes tend to buffer the [O3] level. For example, as we reduce [O3] and precursor levels, loss rates of O3 decrease and O3 production efficiency increases making [O3] relatively insensitive to changes in precursor emissions. Reductions in transport of [O3] and precursors are

accompanied by compensating increases in O3 production efficiency downwind, so we expect [O3] sensitivity studies will underestimate the [O3] AOI. We tested this hypothesis by evaluating the AOI using a low [O3] base case scenario with full biogenic emissions but zero anthropogenic emissions. This base case had high O3 production efficiency, low O3 loss rates, [O3] levels ranging from 30 to 80 ppb. We then performed a source modulation for the same source areas and adding anthropogenic emissions of the same magnitude as for the reduction described in Section 3.1 above. Figure 2c shows the cumulative [O3] response calculated using: Delta = Low O3 Base Case - Modulation. Comparing Figure 2c to 2a, we note first that the AOI is considerable larger, extending from North Carolina throughout Maine. The maximum [O3] reduction is also larger, 3020 ppb-hours in the low O3 Base Case compared to 582 ppb-hours in standard Base Case. Finally, with the exception of one Pittsburgh cell, the large [O3] increases due to early morning titration effects were compensated by larger decreases in P(Ox) and [O3] later in the day.

Figure 2d shows the cumulative P(Ox) response for the low O3 Scenario. Comparing Figure 2d to Figure 2b, we see that the same emissions modulation produced very different AOIs for P(Ox). The largest reduction in cumulative P(Ox) in the base scenario was 260 ppb compared to 963 ppb in the low O3 scenario. The P(Ox) response plumes also overlap for Philadelphia, Baltimore, and New York City; this suggests that there will be non-linear interactions in the P(Ox) response in the plume. Figure 3c shows the [O3] and P(Ox) time-series for the 400 by 400 km sub-domain. Reductions are about a factor of 4 larger than in the high O3 base case. Figure 3d shows a large reduction in radical initiation due to lower O3 photolysis which amplified the [O3] reduction caused by precursor reductions. Finally, Figure 4 shows that the low O3 source modulation had a much higher O3 production efficiency, 28 ppb/ppb, compared to 9 ppb/ppb in the high O3 base case.

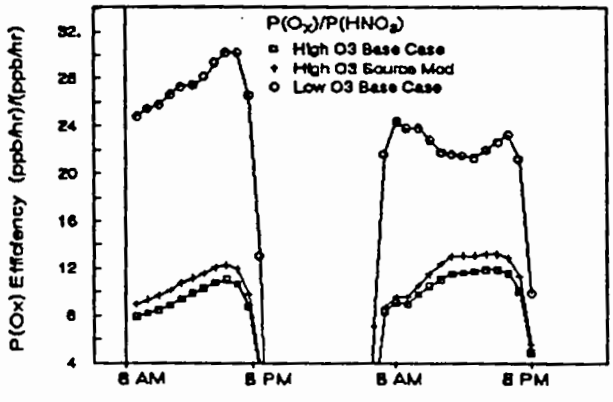

Figure 4. O3 production efficiency for a 36 hour time-series, averaged over a 400 by 400 km area.

4. CONCLUSIONS.

The AOI of precursor emissions is determined by a combination of P(Ox) and transport of [O3]. Transport of precursor emissions creates an P(Ox) AOI that can extend on the order of hundreds of km downwind from the source area even under conditions of stagnant meteorology and light winds. The P(Ox) AOI is also affected by feedbacks from the [O3] response that affect radical initiation rates. The [O3] AOI extends on the order of hundreds of km beyond the P(Ox) AOI. The area and size of the AOI depends on wind speed and direction, but also varies depending on the methodology and level of precursor emissions used. A combination of methodologies and a range of scenarios should be used to evaluate AOIs for the full range of conditions for which changes in air quality are likely to occur.

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16. ABSTRACT

Regional transport of ozone (O3) and its precursors is suspected to significantly affect O3 control strategies and the effectiveness of local controls throughout much of the eastern U.S. A Federal Advisory Committee Act (FACA) work group is studying the identification of Areas of Influence (AOI's), essentially O3 airsheds, around which to design controls for subregions of the eastern U.S. It is difficult, however, to quantify the production, life-time and effect of transport on regional/urban O3 concentrations ([O3]). It is also difficult to quantify the sensitivity of the transported O3 to hydrocarbon (VOC) or nitrogen oxide (NOx) emissions or to emissions reductions in any given region or urban area. At least two types of methodologies have been proposed to evaluate AOIs. They include sensitivitiy studies to evaluate the change in [O3] in the AOI when emissions in the source region are changed (Yang et al., 1997); and Process and Mass tracking studies to evaluate the actual transport of mass and the production of O3 on route from the source region to the AOI (Lo and Jeffries, 1996; Yarwood et al., 1997). Each of these approaches has limitations: senesitivity studies have predictive value but they lack explanatory value; ie, they do not provide adequate insights into the processes and feedbacks that cause the change in [O3] in the AOI. They may ignore feedbacks in the system which effectively buffer [O3], thereby underestimating the size of the AOI. Further, computed sensitivities may vary considerably with the base case scenario conditions, so it is unclear how sensitivities should be calculated or applied in light of changes in future scenario conditions. Process and Mass tracking studies do have considerable explanatory value; they can explain in great detail how precursor and O3 transport determine [O3] in the AOI for a particular scenario. They do not, however, have predictive power, ie, they do not predict exactly how emissions reductions will affect [O3] in the AOI. We propose that these methods have complementary strengths and must be used together both to define the AOI and to understand the impact of emissions reductions in the AOI. In this study, we use sensitivitiy simulations to evaluate the AOI of precursor emissions in selected source regions, and we use a process analysis to explain the results of those sensitivity simulations.

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