Ozone Source Apportionment Results for Receptors in Non-Attainment Counties in the Great Lakes Region

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ABSTRACT

Photochemical model source apportionment probing tools are valuable from a scientific and regulatory perspective for tracking source contribution to ozone. The use of source apportionment is more desirable than the “zero-out” approach to determine geographic and emissions sector culpability. Zeroing out emissions for large regions such as entire States fundamentally changes the atmospheric chemistry and makes interpretation of the results difficult. Source apportionment of high ozone concentrations at non-attainment monitor locations is useful for regulators in developing effective emissions control scenarios.

The Comprehensive Air-Quality Model with extensions is a state of the science photochemical model containing multiple ozone source apportionment tools. Ozone contributions are tracked using the anthropogenic pre-cursor culpability assessment (APCA) source apportionment tool for 18 geographic regions and 6 emissions sector categories: electrical generating point sources, non-electrical generating point sources, on-road, off-road, area, and biogenics. A future year emissions scenario for 2012 is used for this application to present a better understanding of what is contributing to high ozone concentrations in the future when certain emissions controls and sector growth is taken into consideration. Results are given for receptors in counties near the Great Lakes that may not meet the 8-hr ozone NAAQS by 2012.

The highest contributors to high modeled ozone concentrations include on-road, off-road, and point source emissions of nitrogen oxides. The Chicago and Detroit receptors are dominated by local emissions. The Cleveland receptor has a large contribution from emissions in down-State Ohio in addition to large local contribution. Chicago emissions also dominate contributions to high ozone at receptors to the north along the western and eastern shores of Lake Michigan. Source apportionment results for the finer compared to coarser grid resolution show a higher percentage of local contribution to ozone at the finer grid resolution.

INTRODUCTION

Probing tools are valuable from a scientific and regulatory perspective for one-atmosphere modeling. Use of source apportionment is more desirable for regulatory applications than the use of the "zero-out" approach to determine geographic and emissions sector culpability for long-term modeling simulations. Zeroing out emissions for large regions such as entire States fundamentally changes the atmospheric chemistry and makes interpretation of the results difficult.

CAMx is a state of the science photochemical model that contains a variety of ozone source apportionment tools, including the original ozone source apportionment tool (OSAT) and the anthropogenic pre-cursor culpability assessment (APCA) tool. The APCA tool assesses regional and emission sector contribution to ozone formation and provides information that is most policy relevant. When ozone is formed under VOC limited conditions due to biogenic VOC + anthropogenic NOx then OSAT attributes it to the biogenic VOC sources. When ozone is formed under NOx-limited conditions due to biogenic VOC + anthropogenic NOx then OSAT attributes it to the anthropogenic NOx sources. APCA is designed to provide more control strategy relevant information and recognizes that there are source categories such as biogenics that can not be controlled so the model only attributes ozone to biogenics when it is due to the interaction of biogenic VOC + biogenic NOx. In the case where ozone formed to biogenic VOC + anthropogenic NOx under VOC-limited conditions, OSAT attributes it to biogenic VOC, but APCA redirects the attribution to anthropogenic NOx. In NOx-limited conditions both OSAT and APCA attribute the ozone to anthropogenic NOx.[1] The APCA tool is chosen to track ozone contribution for this modeling study.

Contribution to high ozone is tracked using the APCA source apportionment tool for 18 geographic regions and 6 emissions sector categories: electrical generating units (EGUs), non-electrical generating units (non-EGUs), on-road mobile, off-road mobile, area, and biogenic. A scenario representing emissions in 2012 is used for the source apportionment application to present a better understanding of contributions to ozone when certain emission controls and sector growth is taken into consideration. Contribution is shown for monitor locations in the Great Lakes region that may not show attainment of the 8-hour ozone standard in 2012: Chicago, Cleveland, and Detroit. Additional information is given for other receptors along the shore of Lake Michigan that are impacted by emissions from Chicago.

METHODS

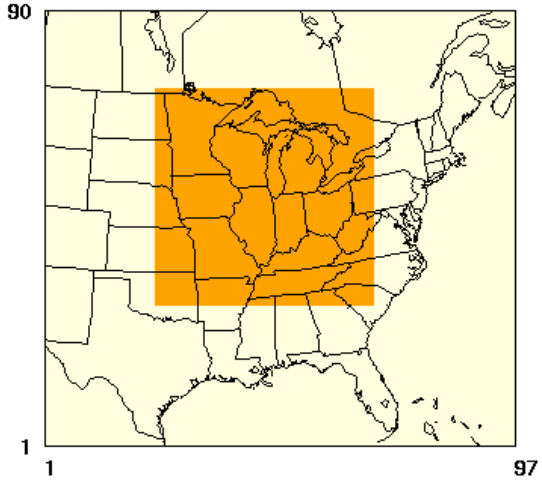
The Comprehensive Air Quality Model with Extensions (CAMx) version 4.30 uses state of the science routines to model ozone formation and removal processes over regional and urban scales.[2,3,4,5] The model is applied with an updated carbon-bond IV (CB4) gas phase chemistry module.[1,6] CAMx is applied using the PPM horizontal transport scheme and an implicit vertical transport scheme with the fast CMC chemistry solver.[1] The photochemical model is initiated at midnight Eastern Standard Time and run for 24 hours for each episode day. The summer 2002 simulation is initiated on June 2 and run through

August 31. The first 11 days of the simulation are not used in any analysis to minimize the influence of initial concentrations.

Boundary conditions represent pollution inflow into the model from the lateral edges of the grid and initial conditions provide an estimation of pollution that already exists. The initial and boundary conditions are based on monthly averaged species output from an annual (calendar year 2002) application of the GEOS-CHEM global chemical transport model.[7,8] Boundary conditions vary by month and in the horizontal and vertical direction. Where an initial or boundary concentration is not specified for a pollutant the model will default to a near-zero concentration.

The meteorological, emissions, and photochemical models are applied with a Lambert projection centered at (-97, 40) and true latitudes at 33 and 45. The 36 km photochemical modeling domain consists of 97 cells in the X direction and 90 cells in the Y direction covering the central and eastern United States (Figure 1). The 2-way nested 12 km domain covers most of the upper Midwest region with 131 cells in the X and Y directions. CAMx is applied with the vertical atmosphere resolved with 16 layers up to approximately 15 kilometers above ground level.

Figure 1. 36 km (large box) and 12 km (small dark box) modeling domain



Meteorological input data for the photochemical modeling runs are processed using the National Center for Atmospheric Research (NCAR) 5th generation Mesoscale Model (MM5) version 3.6.1.[9,10] Important MM5 parameterizations and physics options include mixed phase (Reisner 1) microphysics, Kain-Fritsch 2 cumulus scheme, Rapid Radiative Transfer Model, Pleim-Chang planetary boundary layer (PBL), and the Pleim-Xiu land surface module. Analysis nudging for temperature and moisture is only applied above the boundary layer. Analysis nudging of the wind field is applied above and below the boundary layer. These parameters and options are selected as an optimal configuration for the central United States based on multiple MM5 simulations using a variety of physics and configuration options.[11,12]

Emissions data is processed using EMS-2003. The EMS-2003 model is selected for its ability to efficiently process the large requirements of regional and daily emissions processing. In addition to extensive quality assurance and control capabilities, EMS-2003 also performs basic emissions processes such as chemical speciation, spatial allocation, temporal allocation, and growth and control of area, point, and mobile source emissions.[13,14] Anthropogenic emission estimates are made for a weekday, Saturday, and Sunday for each month. The biogenic emissions are day-specific. Volatile organic compounds are speciated to the Carbon Bond IV (CB4) chemical speciation profile.[15] Anthropogenic emissions are based on 2002 estimates and projected to 2012 based on growth factors and known control programs implemented between 2002 and 2012. Biogenic emissions are estimated with EMS-2003 using the BEIS3 model.[16] The BELD3 land use dataset is input to the biogenic model for fractional land-use and vegetative speciation information.[17,18] Other inputs to the biogenic emissions model include hourly satellite photosynthetically activated radiation (PAR) and 15 m (above ground level) temperature data output from MM5.[19]

The ozone monitors used in the source apportionment analysis are shown in Table 1. The source apportionment data is the average contribution over all modeled hours where predicted ozone at the monitor is greater than a threshold concentration value. Two different thresholds are used to examine different distributions of high modeled 8-hour ozone: 75 and 85 ppb.

Table 1. Ozone monitors included in source apportionment analysis

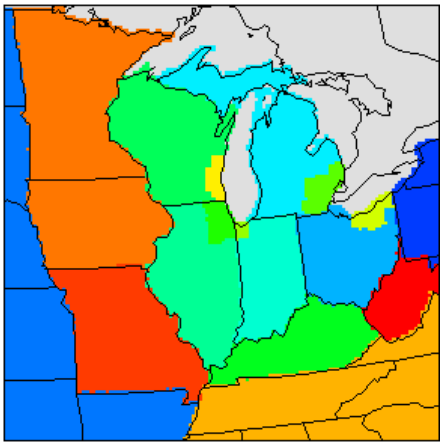
Metropolitan Area	County	State	AIRS ID
Chicago	Cook	IL	1703100321
Detroit	Macomb	MI	2609900091
Cleveland	Geauga	OH	3905500041
Milwaukee	Milwaukee	WI	5507900851
Holland	Allegan	MI	2600500031
Kenosha	Kenosha	WI	5505900021
Green Bay	Door	WI	5502900041

The geographic regions tracked for ozone contribution are listed in Table 2 and shown graphically in Figure 2 over the 12 km modeling domain. The contribution from the lateral and top boundaries of the model is also tracked for each receptor location.

Table 2. Complete list of source regions tracked for ozone contribution

Canada	Illinois Chicago non-attainment (NA) Counties
Northeast States (MANE-VU)	Detroit NA Counties
Central/Western States (CENRAP+ WRAP)	Indiana Chicago NA Counties
Ohio	Cleveland NA Counties
Michigan	Milwaukee NA Counties
Indiana	Southeast States (VISTAS)
Illinois	Minnesota+Iowa
Wisconsin	Missouri
Kentucky	West Virginia

Figure 2. Source regions tracked in the 12 km grid domain



Six emissions source sectors are tracked for contribution to ozone: onroad mobile, offroad mobile, area, electrical generating units, non-electrical generating units, and biogenics. Offroad mobile emissions include sources such as construction equipment, locomotives, commercial marine vessels, and airports. Two distinct groups of stationary point sources are tracked for contribution to ozone: electrical generating units and non-electrical generating units.

An option in CAMx is employed to force elevated point sources into particular regions rather than placement based on coordinates and the 12 km geographic region map. This ensures that elevated emissions are placed in the appropriate geographic region and not incorrectly grouped with another region when a grid cell contains the boundary for more than one region. A good example of this is the Ohio River Valley where many large stationary point sources exist along State boundaries and could be grouped into the wrong region based on the 12 km grid cell source region map. This option improves the confidence in the source apportionment results for stationary point sources.

RESULTS AND DISCUSSION

The percent contribution of high ozone (greater than 85 ppb) from NOX, VOC, and boundaries are shown in Table 3. An interesting feature of NOX and VOC contribution is

that VOC contribution is highest at the Chicago monitor and decreases at monitors further north along Lake Michigan. The lowest VOC contributions are seen at the rural sites downwind from Chicago along Lake Michigan. The Detroit monitor is similar to Chicago in terms of NOX and VOC contribution. The Cleveland monitor has a much smaller contribution from VOC to high ozone than Detroit, Chicago, and even Milwaukee. Kenosha is located between Chicago and Milwaukee and has NOX and VOC contribution to ozone that is between the values for Chicago and Milwaukee.

The boundary contribution represents ozone entering the model through the lateral and top boundaries, which may represent a hemispheric or global background contribution. The emphasis of the modeling exercise is the estimation of potentially controllable pollution in the central and eastern United States, meaning minimal effort was made to determine the validity of ozone concentrations entering the model from boundaries.

Table 3. Percent contributions from NOX, VOC, and boundaries

City	State	NOX	VOC	boundary
Chicago	IL	63.9	17.0	19.0
Holland	MI	70.0	6.7	23.3
Detroit	MI	57.6	15.9	26.5
Cleveland	OH	68.1	5.5	26.3
Green Bay	WI	79.5	6.1	14.5
Kenosha	WI	67.3	14.8	17.9
Milwaukee	WI	72.7	11.4	15.9

The percent contribution to high ozone (> 85 ppb) from emission source sectors is shown in Table 4. The source sector contribution is shown for onroad mobile, offroad mobile, area, stationary point, and biogenic sources. The emissions sector contributing the highest amount of ozone varies from site to site, but the biggest controllable contributors are mobile and stationary point sources. The contributions from offroad mobile sources are comparable to onroad mobile and stationary point source emissions at most of the monitor locations presented here.

Table 4. Percent contribution from emissions source sectors and boundary

City	State	biogenic	onroad	offroad	area	point	boundary
Chicago	IL	6.0	17.7	20.8	9.3	27.3	19.0
Holland	MI	6.8	18.2	18.7	6.7	26.3	23.3
Detroit	MI	5.7	25.6	17.5	9.3	15.5	26.5
Cleveland	OH	4.9	24.2	20.4	6.5	17.7	26.3
Green Bay	WI	8.2	17.8	24.4	7.7	27.5	14.5
Kenosha	WI	6.0	19.8	25.5	10.1	20.6	17.9
Milwaukee	WI	6.4	18.5	22.6	9.8	26.9	15.9

The contribution from electrical generating units and non-electrical generating units to the total stationary point source category is shown in Table 5. The contribution from EGUs is highest at the Cleveland monitor and lowest at Chicago, Milwaukee, and Green Bay. The Detroit monitor has a fairly even contribution from EGUs and non-EGUs to the stationary point source category.

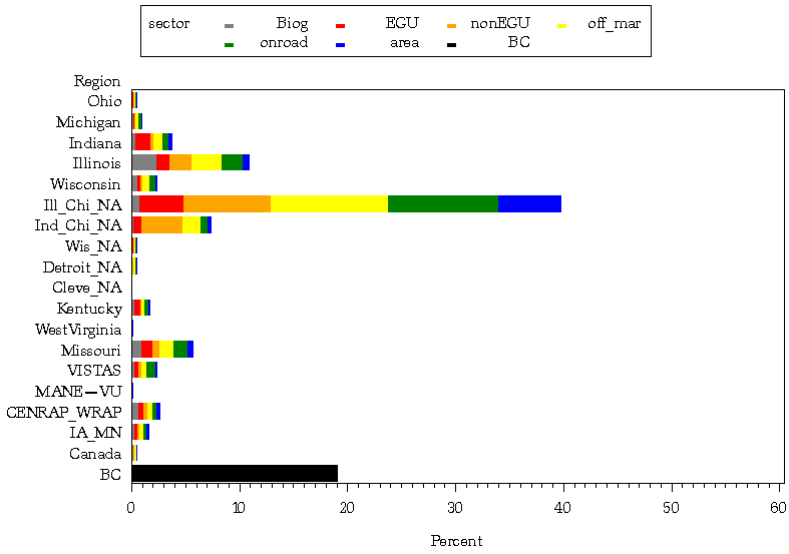
Table 5. Percent contribution from stationary point Source EGUs and non-EGUs

City	State	EGU	nonEGU
Chicago	IL	39.7	60.3
Holland	MI	44.0	56.0
Detroit	MI	46.8	53.2
Cleveland	OH	63.0	37.0
Green Bay	WI	38.3	61.7
Kenosha	WI	43.9	56.1
Milwaukee	WI	39.0	61.0

The contributions to high ozone (>85 ppb) from regions and sectors tracked with ozone source apportionment are shown for the Chicago monitor in Figure 3.

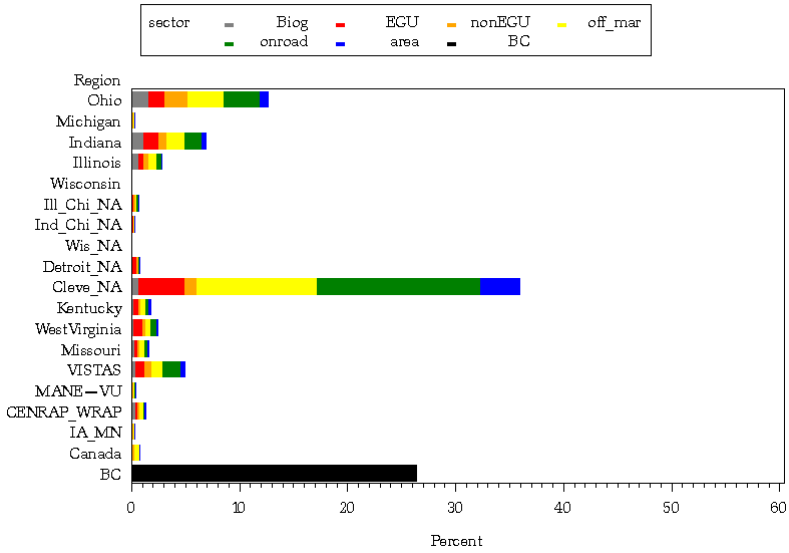
The contribution is clearly highest at the Chicago monitor from sources in the Illinois Chicago non-attainment counties. The next largest geographic contributors include the rest of the State of Illinois (which does not include the Chicago non-attainment counties), the Chicago non-attainment counties in Indiana, down-State Indiana, and Missouri. There are clearly high contributions from local sources and from transport out of the south and southwest. The relative mixes of sources that contribute to the Chicago monitor are fairly consistent from each of the source regions.

Figure 3. Contribution (%) to high ozone at the Chicago monitor



The percent contribution to high ozone (> 85 ppb) at the Cleveland location is shown in Figure 4. The counties that make up the Cleveland non-attainment area make up the largest controllable emissions contribution. The rest of the State of Ohio is the next largest contributor, followed by other States in the Ohio River Valley region. The onroad and offroad mobile source sectors dominate the local contribution from the Cleveland non-attainment counties. Transport from other regions tends to be split between all source groups tracked in the model.

Figure 4. Contribution (%) to high ozone at the Cleveland monitor



The contributions from VOC and boundaries in Table 6 suggest the use of a higher threshold precludes the inclusion of more regional ozone into the assessment. VOC contributions are higher and boundary contributions are lower using 85 ppb cutoff, suggesting an increased local characteristic to the hours included in the analysis. The contributions from NOX to high ozone remain the same for both 75 and 85 ppb thresholds.

Table 6. Percent contribution from all sources using two different thresholds

Category	75 cutoff	85 cutoff
NOX	65.3	65.6
VOC	10.3	12.5
boundary	24.4	21.9

Table 7 shows the contribution to high ozone using the 75 and 85 ppb thresholds from local sources. It also shows the local source contribution at 12 and 36 km grid resolution at the 85 ppb threshold. Table 7 also shows higher local contributions to high ozone when using a higher threshold. This is likely due to more regional ozone formation being included in the average contribution estimate when using a lower threshold since it will include more days in the assessment.

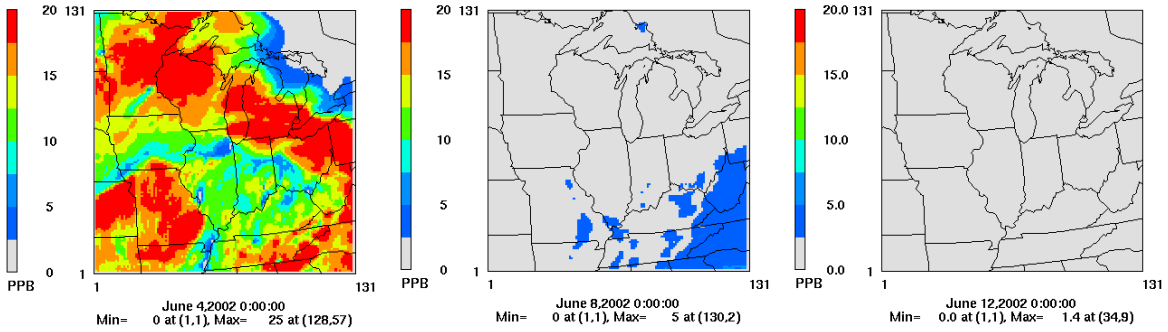
Table 7. Percent contribution from local sources using two different thresholds and grid resolutions

	12 km		36 km
City	75 cutoff	85 cutoff	85 cutoff
Chicago	33.6	39.7	32.2
Detroit	32.1	39.3	33.2
Cleveland	30.2	35.9	26.6

The contribution from local sources increases at the Chicago, Detroit, and Cleveland monitor locations going from 36 km to 12 km grid resolution. This effect from grid resolution is likely due to less dilution of large NOX emissions with the 12 km grid cells leading to higher NOX and ozone concentrations in these urban areas.

Since the initial conditions are tracked as part of the APCA source apportionment tool, the impact from start-up conditions may be assessed quantitatively to help determine how many days are necessary to run the model before initial condition impact on ozone formation is most minimal. In the past a spin-up period of two to three days was used to eliminate initial condition effects for ozone modeling. CAMx source apportionment runs show 1-hour ozone attributed to initial concentrations does not exceed 6 ppb anywhere in the domain by the 7th day of the episode or 1.5 ppb by the 11th day of the episode.

Figure 5. Contribution to ozone from initial conditions: day 3 (left), 7 (middle), and 11 (right)



CONCLUSIONS

The Chicago, Detroit, and Cleveland monitor locations have high ozone contributions from local mobile and stationary point source nitrogen oxides. Chicago area emissions have a large contribution to monitors further north along the shore of Lake Michigan. The offroad mobile source sector contribution is comparable to the contribution from onroad mobile and stationary point sources at the Chicago, Detroit, and Cleveland monitors used for this study.

The use of fine grid resolution tends to increase the relative impact on ozone from local emissions sources. The higher ozone concentration threshold of 85 ppb also tends to increase the local source impact. The recommended ozone attainment test approach recommends starting with a cutoff threshold of 85 ppb for peak ozone values included in the attainment analysis, so the 85 ppb threshold may be more relevant to regulatory applications.[20] The benefit from regional emission reductions may not be fully realized in the attainment test since days with large regional contributions are de-emphasized based on the use of an 85 ppb minimum threshold.

A limitation to this analysis is that all ozone formation regimes may not be captured at each monitor by modeling the summer of 2002. Multiple high ozone episodes were observed at each monitor in this analysis during the summer of 2002 so these results do represent high ozone events in the Great Lakes region.

This type of source apportionment analysis highlights the similarity and differences in contribution to high ozone from geographic and source sector groups at several metropolitan areas in the Great Lakes region. The contribution information is useful for the development of efficient control programs and improves the understanding of local and regional impacts on high ozone formation.

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KEY WORDS

Ozone, source apportionment, CAMx, photochemical model, OSAT, APCA