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Apportionment of Ozone above the Background Concentration to Emission Sources

Executive Summary

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COORDINATING RESEARCH COUNCIL, INC.

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EXECUTIVE SUMMARY

Air quality models are used to design emission control strategies to meet regulatory standards. Source apportionment involves assigning fractions of a pollutant concentration to the sources, chemical species, and/or geographic areas that contribute to the concentration. If this can be done accurately, then controls can be focused on the most important contributors to violations of the standards.

A new source apportionment method has been developed, termed the Path-Integral Method (PIM). This method focuses on the anthropogenic increment of a pollutant concentration. The anthropogenic increment is the difference in concentration between a base-case simulation with all emissions included and a background simulation without the anthropogenic emissions. The anthropogenic increment is important for regulatory purposes because this is the portion of a pollutant concentration that can be reduced by emission controls within the modeling domain. The PIM can allocate the anthropogenic increment to sources whereas other methods cannot.

The objectives of this project were (1) to apply the PIM to an existing modeling data set and (2) to determine the contributions of light-duty vehicles (LDVs) and other major sources to the anthropogenic ozone (O_3) increment. A secondary objective was to evaluate and quantify the uncontrollable (non-anthropogenic) O_3 background concentration.

For the base case, we used a scenario developed previously in CRC Project A-76-3 for 2030 to represent low emission vehicle (LEV) III controls and gasoline with 10 ppm sulfur. Simulations were conducted for July with the Comprehensive Air Quality Model with Extensions (CAMx) using the Carbon Bond 2005 (CB05) chemical mechanism. The full 36-km modeling domain included the continental U.S., and we analyzed results for a 12-km sub-domain covering the eastern U.S. Within this sub-domain, we focused on a rural site at Great Smoky Mountains National Park (GRSM) and four urban sites at Atlanta, GA, Washington, DC, Cincinnati, OH, and New York, NY. The background case contained no U.S. anthropogenic emissions but did contain anthropogenic emissions in Canada and Mexico. We apportioned the anthropogenic increments of O_3, formaldehyde (FORM), nitrogen dioxide (NO_2), acetaldehyde (ALD2), and propionaldehyde plus higher molecular-weight aldehydes (ALDX) to five source categories: on-road LDVs, other on-road vehicles (non-LDVs), off-road vehicles, area sources, point sources.

For our simulation of year 2030, the monthly maximum of the maximum daily average 8-h (MDA8) O_3 is ~40 - 118 ppb and ~20 - 50 ppb in the base and background cases, respectively. The concentration range in the background case is in good agreement with other recent modeling studies. The anthropogenic MDA8 O_3 increment is ~ 5 – 91 ppb. The monthly average FORM is 1.5 – 8.3 ppb and 1.0 – 6.3 ppb in the base and background cases, respectively, with an anthropogenic increment of 0 – 4.3 ppb. The monthly maximum hourly NO_2 concentration is 0 – 81 ppb, 0 – 30 ppb, and -2 – 81 ppb for the base case, background case and anthropogenic increment, respectively.

The PIM determines the source contributions by integrating first-order sensitivity coefficients over a range of emissions, a path, from the background case to the base case. Viewed from the starting to the ending point of the integration, the path represents the growth of anthropogenic emissions into the background. Alternatively, viewed in the reverse direction from the ending to the starting point, the path represents a specific emission-control strategy leading to zero anthropogenic emissions. The sensitivity coefficients were calculated by the decoupled direct method, which has previously been implemented in CAMx. For the numerical integration we used a Gauss-Legendre formula with 3 integration points. Comparison of the sum of the source contributions with the anthropogenic increment showed that this formula gives accurate results.

We obtained source contributions for 3 emission-control paths, termed the diagonal, point-sources-first (PtF), and point-sources-last (PtL) paths. Along the diagonal path, emissions from all anthropogenic sources are reduced together by the same factor. On the first leg of the PtF path, the point-source emissions are reduced to zero while other emissions are held constant at their levels in the base case. On the second leg, the other anthropogenic emissions are reduced to zero along a diagonal path. The PtL path is the same as the PtF path except that the two legs are reversed.

Figure ES-1 gives the source contributions at the GRSM and Washington sites obtained with the diagonal path. Results are shown for the monthly maximum of the MDA8 O_3, the monthly average FORM, and the monthly maximum hourly NO_2 concentrations. These different metrics were chosen because they are relevant to regulations. Considering the results in Figure ES-1 and results for the other 3 sites, point sources have the largest contribution to the anthropogenic O_3 increment (ΔO_3) at all sites and for all paths. After point sources, either off-road or non-LDV sources contribute the most to ΔO_3, and either area sources or LDVs contribute the least, depending on the site. The ranking of the sources by their contribution to ΔO_3 is similar to the ranking by their NO_x emissions in the 12-km domain, indicating that primarily the NO_x emissions from the sources control their contributions to ΔO_3.

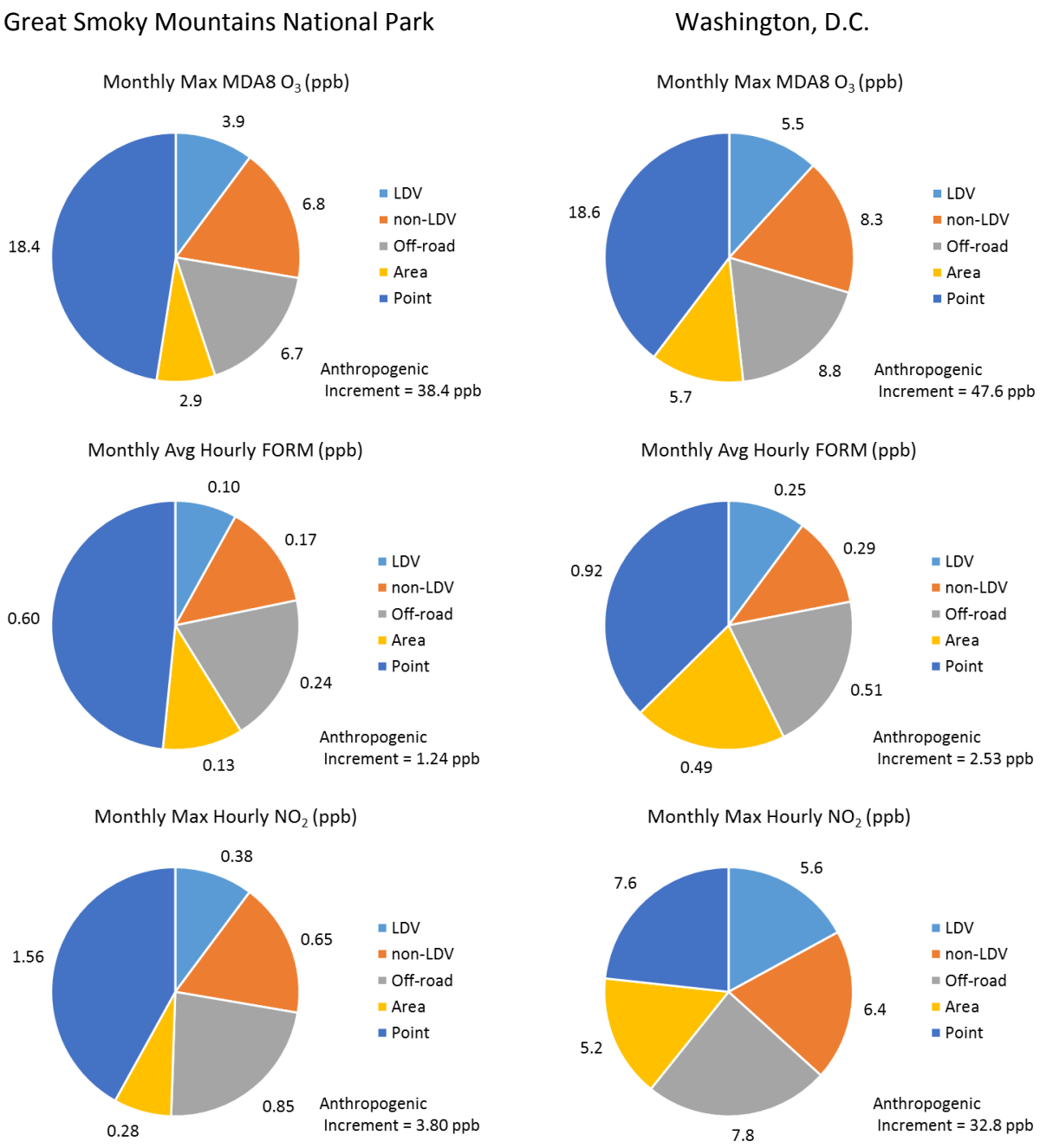

Figure ES-1. Source contributions to O_3, formaldehyde (FORM), and NO_2 at a rural and urban site, as obtained from the PIM using the diagonal path. The contributions are an apportionment of the anthropogenic increment to the pollutant (base case minus background).

At the five sites, point sources are generally the largest contributor to ΔFORM for the diagonal and PtL paths but not always for the PtF path. Point sources are not major emitters of FORM or other VOC's. The contribution of point sources to the FORM concentration is due to the indirect effect of the NO_x emissions in enhancing oxidation of VOC's from other anthropogenic sources and biogenic VOC's. LDVs are the smallest or second smallest contributor to ΔFORM for all sites and paths.

Point sources are the largest contributor to ΔNO_2 at 4 of the sites, regardless of path, and the largest or second-largest contributor at Washington, DC. Either area sources or LDVs are the smallest contributor at each site. As for ΔO_3, the ranking of the sources by their contribution to ΔNO_2 tends to follow the ranking by their NO_x emissions, in this case because the emitted NO is the direct precursor to NO_2.

We also apportioned the anthropogenic increments ΔALD2 and ΔALDX to sources with the diagonal path. Using the monthly average concentration as the metric, point and area sources are generally the largest contributors to ΔALD2 and ΔALDX at the five sites, and LDVs are usually the smallest contributor. Off-road and non-LDV sources have intermediate contributions, with the former always contributing more than the latter.

For comparison to the PIM results, we calculated source contributions using the Ozone Source Apportionment Technology (OSAT). OSAT employs tracers in CAMx to track the fate of the VOC and NO_x introduced into the modeling domain and the O_3 formed from the VOC and NO_x. Both OSAT and PIM identify point sources as the largest contributor to monthly maximum and average MDA8 O_3. The OSAT source contributions are generally larger than the PIM contributions for the LDV, non-LDV, off-road, and area sources but smaller than the PIM contributions for point sources. For OSAT, the sum of the anthropogenic source contributions generally over-estimates ΔO_3. In OSAT, there is no requirement that these two quantities must be equal, whereas in PIM there is.

A major advantage of the PIM is that it allocates the anthropogenic increment rather than the total concentration, which ensures that the increment is neither over- nor under-allocated to the anthropogenic sources. The other major advantage is that it is applicable to any species simulated by the model. Another advantage is that there is no need for auxiliary assumptions related to the chemistry, e.g. a criterion for assigning O_3 formation to NO_x or VOC emissions. Further, the PIM allows both positive and negative source contributions. Lastly, once calculation of sensitivities is implemented in the model, further modifications are not necessary, regardless of changes to the chemical mechanism or whether source contributions are needed for a different pollutant (e.g., FORM instead of O_3). The major disadvantage of the PIM is the computational effort, which is significantly greater than tracer methods such as OSAT.