Accounting for Transport of International Emissions in CMAQ Modeling of the VISTAS Region

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Anthropogenic and natural aerosols from emissions in Mexico, Canada, Central America, Asia, and Africa have been shown to affect air quality and regional haze in the continental United States. States may consider the fraction of the ambient aerosol that is beyond the regulatory control of the U.S as they evaluate emission control strategies to achieve reasonable progress in reducing regional haze in Class I areas. To support the VISTAS states in evaluating the contribution from international emissions, the VISTAS RPO carried out analyses to estimate how much of the haze at Class I areas in the VISTAS region in 2002 and 2018 (as projected by the CMAQ model) results from international emissions.

CMAQ modeling of regional haze in the VISTAS region for 2002 and 2018 was carried out on a grid of 12x12-km cells that covers the VISTAS states (AL, GA, FL, MS, SC, NC, TN, KY, VA, WV) and states adjacent to them. This grid is nested within a larger national CMAQ modeling grid of 36x36-km grid cells that covers the continental United States, portions of Canada and Mexico, and portions of the Atlantic and Pacific Oceans along the east and west coasts. Transport of pollution from the national domain into the VISTAS modeling domain is reflected in hourly-varying boundary conditions around the VISTAS domain, which are derived from the CMAQ simulations in the 36-km national domain. The national domain incorporates emissions from northern Mexico and southern Canada, and therefore explicitly calculates the effects of these international emissions on concentrations at the boundaries of the VISTAS modeling domain.

Effects of emissions from more distant sources that are outside of the 36-km national domain were estimated using outputs from a global air pollution model. The Big Bend Regional Aerosol and Visibility Observational (BRAVO) study had found that the performance of national-scale modeling was enhanced by using monthly averages of outputs from a global air pollution model, GOCART, to generate SO$_2$ and sulfate boundary conditions for its 36-km national modeling grid (Schichtel, et al., 2005), instead of using the EPA default concentrations that are normally used. VISTAS elected to use a similar approach, using the outputs of the GEOS-Chem global air pollution model, which also contains a detailed representation of ozone-NO$_x$-VOC-PM chemistry.

Under EPRI sponsorship, Harvard University had already used the GEOS-Chem model, with 2°x2.5° horizontal resolution, to estimate natural conditions and transboundary influences on elemental and organic carbon aerosols in the United States in 1998 (Park et al., 2003). After some emission inventory and model improvements, similar information was developed for sulfate and nitrate aerosols in 2001 (Park et al., 2004).
Under contract to VISTAS (which was acting on behalf of all the RPOs), Harvard then simulated global air pollution for the year 2002 (the VISTAS modeling year) with \(4^\circ \times 5^\circ\) horizontal resolution and 3-hour temporal resolution (Jacob, 2005). The coarser grid scale was considered sufficient to provide boundary conditions outside of North America, largely over the oceans. Although the VISTAS simulation was generally similar to the 2001 EPRI simulation mentioned above, the VISTAS modeling incorporated the following substantive changes in addition to the change in grid scale: (1) use of U.S. anthropogenic emissions from the EPA NEI 1999 inventory, except for ammonia; (2) use of forest fire information specific to 2002; (3) inclusion of the secondary organic aerosol (SOA) formation mechanism from Chung and Seinfeld (2002); and (4) inclusion of prototype soil dust and sea salt simulations. An additional model improvement, which has since been implemented in the standard version of GEOS-Chem, was the application of surface emissions and dry deposition to the entire mixed layer column diagnosed by GEOS (the Global Earth Observation System of NASA) rather than to just the surface layer of the model. Details of the emissions, meteorology, model configuration, and model performance evaluation for the VISTAS simulations are described in the report to VISTAS (Jacob, 2005).

For North America, the VISTAS GEOS-Chem modeling used monthly-average emissions, so the model results do not reflect diurnal or weekday-weekend variability in emissions, and also diminish the impacts of episodic emissions, such as the July 2002 wildfire in Ontario, Canada. The Mexican emissions were from the inventory of the BRAVO Study, while Canadian emissions for the year 2000 were used. Emissions from Caribbean islands were not included, which could impact model estimates at the Everglades.

The surface meteorological data inputs to GEOS-Chem had 3-hr temporal resolution and the upper air meteorological inputs had 6-hr resolution, with a horizontal resolution of \(1^\circ \times 1^\circ\) or better and 48 layers vertically.

The output of the GEOS-Chem modeling, as delivered to VISTAS, contained concentrations of all visibility-related components of particulate matter with 3-hr temporal resolution and \(4^\circ \times 5^\circ\) horizontal resolution over multiple layers of the atmosphere.

Even though the coarse spatial resolution degraded the performance of the model over North America, relative to results from prior modeling for other years using finer grids, the concentrations simulated by GEOS-Chem maintained the synoptic-scale structure and did not have a continental-scale bias. At a finer scale, the concentrations simulated by GEOS-Chem for the surface layer in 2002 were generally within a factor of two of regional-averages (\(~500\) km scale) of measurements of the various chemical components of particulate matter in the United States and, except for organic carbon, satisfactorily simulated the seasonal variability in the observations.

In the meantime, Harvard University had also carried out new finer-grid simulations for 2001 for EPRI, using a \(1^\circ \times 1^\circ\) grid over North America, nested in a coarser grid that
covered the globe. The final version of these simulations included modifications to the modeling grid to improve delineation between domestic US emissions and those of foreign sources at the national boundaries, and revision (reduction) of the Canadian and Mexican sulfur emissions. These GEOS-Chem 1°x1° grid scale simulations for 2001 and their results are described in the paper by Park et al. (2006). Over appropriate geographic and temporal scales, the results of these simulations for 2001 were similar to those of the 2002 VISTAS simulations, which gave confidence in the VISTAS GEOS-Chem results.

The concentration fields that resulted from the GEOS-Chem simulations were not directly usable as boundary conditions for the 36-km grid scale CMAQ simulations. GEOS-Chem and CMAQ use different horizontal and vertical grid scales and different map projections, and have different labels for chemical constituents. The GEOS-Chem outputs were processed by Daewon Byun at the University of Houston to adjust for these discrepancies and to develop boundary conditions for the 36-km CMAQ modeling. Since the CMAQ modeling has temporal resolution of one hour, the boundary conditions were given for every hour. However, since the GEOS-Chem outputs had 3-hr resolution, the hourly boundary conditions were the same for three consecutive hours. An example of the resulting boundary conditions for sulfate at the surface is shown in Figure 1.

![Average Sulfate Boundary Conditions](image)

**Figure 1.** GEOS-CHEM projections of sulfate boundary conditions for the national CMAQ modeling grid on July 1, 2001, as indicated by the colors along the boundary of the modeling domain.
Using the boundary conditions provided by the global simulations, VISTAS calculated the impacts of international anthropogenic emissions on haze at US Class I areas in two different ways, which are summarized in Table 1:

- First, as the difference between the results of two GEOS-Chem simulations: (1) with 2002 international emissions (natural and anthropogenic) but only natural US emissions, and (2) with only natural emissions worldwide. Such differences could be calculated with the 4°x5° grid of the VISTAS simulations or the 1°x1° grid of the EPRI simulations. Since the chemical reactions in these GEOS-Chem simulations do not reflect the contribution to the chemistry from anthropogenic emissions in the US, the result is an estimate of what the international contribution would be if the US emissions were only from natural sources (the 2064 goal) while the emissions of the rest of the world remained at 2002 levels. (See the upper half of Table 1 for portrayal of this calculation.)

- Second, as the difference between the results of two CMAQ 36-km grid simulations of the US, both using 2018 OTW Base F emissions, with boundary conditions based on GEOS-Chem simulations: (1) with all global sources active, and (2) with U.S. anthropogenic emissions and all global natural emissions (Mexican and Canadian anthropogenic emissions within the 36-km CMAQ domain were also removed). Since the GEOS-Chem simulations are based on 2002 emissions, the result of this calculation is an estimate of what the international contribution would be in 2018 if international emissions remain at 2002 levels. (See the lower half of Table 1 for portrayal of this calculation.)

Both cases underestimate the total international contribution because the GEOS-Chem simulations assumed that all fires outside of the US were natural, i.e., that there are no international fire emissions due to anthropogenic burning.

Table 1. Approaches for estimating international contributions using GEOS-Chem results.

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<th>Case</th>
<th>Natural Sources</th>
<th>Anthropogenic Sources</th>
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<td>GEOS-Chem</td>
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<td>CMAQ with GEOS-Chem boundary conditions</td>
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<td>(1)</td>
<td>International and US</td>
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The CENRAP RPO applied a third method to estimate the impacts of international emissions. They used the PSAT (PM Source Apportionment Technology) module of the CAMx regional air quality model to track the 2002 contributions of three non-domestic source categories (Canadian emissions in the CAMx modeling domain, Mexican emissions in the CAMx modeling domain, and boundary conditions) at all Class I areas. The boundary conditions applied at the boundaries of the CAMx 36-km scale grid were based on the outputs of the GEOS-Chem modeling described above. The international impacts in 2002 were then calculated as the sums of the PSAT-determined contributions of these three source categories. One major difference between this approach and the VISTAS approaches is that the PSAT international impacts included the contributions from fire emissions. This will result in larger OC and EC international impacts compared to the VISTAS approach.

To illustrate the magnitudes of the international contributions to two of the principal components of the visibility-impairing aerosol, Figures 2 and 3 show the annual averages of estimates of international contributions to ammonium sulfate and organic carbonaceous mass (OCM) at Class I areas in the VISTAS region and in adjacent states. Results are presented for all three of the methods described above. Two sets of GEOS-Chem results are shown – for 2002 with the VISTAS 4°x5° grid and for 2001 with the EPRI 1°x1° grid.

![Figure 2. Estimates by four methods of impacts of international emissions on annual average ammonium sulfate concentrations at Class I areas.](image-url)
Figure 3. Estimates by four methods of impacts of international emissions on annual average organic carbonaceous mass concentrations at Class I areas.

For ammonium sulfate there is reasonable agreement (better than a factor of two or so everywhere except at Everglades) between all pairs of methods, while the results are more variable for OCM. The impacts estimated by the VISTAS 4°x5° GEOS-Chem simulations tend to be smaller than those of the other methods. Not surprisingly, the biggest impacts from international emissions tend to occur at Class I areas that are near the Canadian and Mexican borders.

The projected contributions from international emissions on the 20% worst visibility days in 2002 are compared to annual average contributions in Figures 4 and 5. Results obtained using both the PSAT and CMAQ models are shown. At the VISTAS sites, CMAQ projected slightly higher international contributions to sulfate on the 20% worst visibility days than the annual average contributions. Compared to the corresponding annual average international contribution, PSAT generally showed lower contribution to sulfate on the 20% worst days at the mountain sites and equal or slightly higher contributions at the coastal sites. For organic carbonaceous mass, there were no clear trends between 20% worst and annual average days in either the PSAT or CMAQ results.
Figure 4. Estimates by the PSAT and CMAQ models of the impacts of international emissions on ammonium sulfate mass concentrations at Class I areas on the 20% worst visibility days and on the annual average in 2002.

Figure 5. Estimates by the PSAT and CMAQ models of the impacts of international emissions on organic carbonaceous mass concentrations at Class I areas on the 20% worst visibility days and on the annual average in 2002.

Figures 6 and 7 put the international impacts into the context of current measured concentrations. They compare the international impact estimates with current monitored concentrations on the 20% worst visibility days at two VISTAS Class I areas chosen as
examples, Shenandoah (SHEN) and St. Marks (SAMA). At both locations, the international impacts on concentrations on such days are small, typically less than 5% of the measured concentrations.

At the same two Class I areas, the magnitudes of international contributions relative to the uniform rate of progress (URP) goal for 2018 are shown in Figures 8 and 9, where all the haze index values were calculated using the new IMPROVE algorithm. The numerical value given at 2004 (e.g., 29.31 dv in Figure 8) is the baseline period haze index value (the average over the five years from 2000 to 2004). Because the CMAQ results are scaled with Relative Response Factors (RRFs), the initial CMAQ haze index matches the measured baseline value. In each plot, the open triangle at 2018 indicates the estimated haze index, in deciviews, as predicted by the CMAQ model using the Base G1 emission inventory. The X indicates the haze index in 2018 if international emissions are removed from the 2018 CMAQ simulation, so the difference between the open triangle and the X represents the international contribution.

![Figure 6](image_url)

**Figure 6.** For the 20% worst visibility days in 2002 at Shenandoah, CMAQ-simulated international contributions (right bars) and domestic contribution to measured PM concentrations (measured component minus international contribution) (left bars).
Figure 7. For the 20% worst visibility days in 2002 at Saint Marks, CMAQ-simulated international contributions (right bars) and domestic contribution to measured PM concentrations (measured component minus international contribution) (left bars).

Figure 8. Illustration of the effect of removing the international emissions contribution to the CMAQ-modeled haze index on 20% worst days at Shenandoah in 2018. Open triangle = estimate with all emissions; X = estimate with international emissions removed.
At Shenandoah, Figure 8 shows that the modeled 2018 haze index is already below the Uniform Rate of Progress line. Removing the international contribution would lower the 2018 haze index by another 2.26 dv. Although the international contribution is small today, it becomes increasingly important as US emissions are reduced. (For these analyses, the international emissions are assumed to remain constant from the baseline period to 2018).

At Saint Marks, Figure 9 shows that the modeled 2018 haze index is above the Uniform Rate of Progress line. Removing the international contribution would reduce the 2018 haze index by 1.32 dv, to a value that lies just below (indicating greater reduction than) the Uniform Rate of Progress line.

Information of the type portrayed for 2018 in Figures 8 and 9 is summarized in Figure 10 for all VISTAS Class I areas and Class I areas in adjacent states. The quantity represented is the ratio (in percent) of two metrics of improvement in the haze index from the baseline period to 2018: the CMAQ-modeled improvement in haze index divided by the improvement reflected by the Uniform Rate of Progress line (the magenta line in Figures 8 and 9). For example, in Figure 9 the ratio at St Marks with all emissions considered is $(26.31 - 23.38)/(26.31 - 22.88) = 0.85 = 85\%$. A ratio of less than $100\%$ means that the model-predicted improvement is not sufficient to meet the goal of the Uniform Rate of Progress; i.e., the predicted haze index in 2018 is above the Uniform Rate of Progress line.
Figure 10. Percent of Uniform Rate of Progress haze index reduction achieved in 2018 according to the CMAQ modeling with the Base G1 inventory. Results are given for improvements calculated using the new IMPROVE algorithm (squares) and the old IMPROVE algorithm (diamonds). Solid symbols indicate results when all emissions are considered; open symbols indicate results when effects of international emissions are removed.

Figure 10 shows that removing the international contribution is sufficient to raise the reductions ratios at all Class I areas in the figure above 100%. This means that when the international contributions are considered, all Class I areas exceed the Uniform Rate of Progress in 2018. This conclusion holds whether the old or new IMPROVE algorithm is used calculate the haze index.

Figures 11 through 14 display the Uniform Rate of Progress for the four Class I areas in North Carolina and show the modeled predictions for 2018 with all emissions considered and without the contribution from international emissions. These figures have the same format as Figures 8 and 9.
Figure 11. Illustration of the effect of removing the international emissions contribution to the CMAQ-modeled haze index on 20% worst days at Great Smoky Mountains in 2018. Open triangle = estimate with all emissions; X = estimate with international emissions removed in 2018. Great Smoky Mtns. also represents Joyce Kilmer-Slickrock.

Figure 12. Illustration of the effect of removing the international emissions contribution to the CMAQ-modeled haze index on 20% worst days at Linville Gorge. Open triangle = estimate with all emissions; X = estimate with international emissions removed in 2018.
**Figure 13.** Illustration of the effect of removing the international emissions contribution to the CMAQ-modeled haze index on 20% worst days at Shining Rock. Open triangle = estimate with all emissions; X = estimate with international emissions removed in 2018.

**Figure 14.** Illustration of the effect of removing the international emissions contribution to the CMAQ-modeled haze index on 20% worst days at Swanquarter. Open triangle = estimate with all emissions; X = estimate with international emissions removed in 2018.
References


