# 9) Reconciliation of Source Apportionment Results and Conclusions

The primary goal of BRAVO was to determine the contribution of major source regions in the United States and Mexico to Big Bend's haze. In support of this goal, the NPS/CIRA group conducted a number of sulfate source apportionment analyses. The analyses focused on sulfates because they were the largest contributor to haze, accounting for 55% of the haze on average. In addition, most sulfates are from anthropogenic industrial sources and have well-understood atmospheric processes that can be simulated in available air quality models.

The source apportionment techniques included source-oriented modeling using the Regional Modeling System for Aerosols and Deposition (REMSAD) air quality model, receptor-oriented modeling merging airmass transport and receptor data, and a hybrid technique which merged source attribution results from the source-oriented models with the receptor data. *EPRI* [2004] also used the source-oriented Community Multiscale Air Quality (CMAQ) model to estimate source attribution to Big Bend's sulfate. The receptor techniques used several wind fields and airmass transport models. All receptor techniques were validated by testing them against the BRAVO perfluorocarbon tracer data and synthetic (modeled) Big Bend sulfate concentrations and source attributions. The source attribution techniques that passed the various validation tests are summarized in Table 9-1.

This section examines and reconciles the differences between the source attribution approaches and derives a reconciled set of results. These results are then used to provide the best estimate for the sulfate source apportionment during the BRAVO period, the Big Bend high sulfate episodes, and for the days with the lowest sulfate concentrations. In addition, the contribution of the sulfur source regions to Big Bend's haze is estimated.

## 9.1 Average Source Attribution Results

The average source attribution for the source- and receptor-oriented techniques to Big Bend's sulfate during the BRAVO period are presented in Table 9-2. The source areas include Mexico, Texas, the eastern and western U.S., and the Carbón facilities in Mexico. This table also provides the average, range, and standard deviation across all techniques for each source region. Overall, the different techniques for Mexico, Carbón, and the western U.S. are in good agreement with a standard deviation of 5%. The eastern U.S. and Mexico standard deviations are larger at 11 and 12%, respectively. The REMSAD source attributions differed the most compared to the other techniques. The REMSAD Carbón and Mexico contributions were significantly lower, while the contribution from the eastern U.S. was significantly larger than with the other techniques. Significant is defined as more than one standard deviation from the mean. The TrMB-EDAS method also had significant differences with a larger Texas contribution and smaller eastern U.S. contribution than the mean. However, these two source regions also had large uncertainties and the confidence intervals encompass the mean estimates.

Differences are also evident between the source and receptor methods. The source-oriented models had smaller Mexican and Texas contributions and larger eastern U.S. contributions than the receptor techniques. For example, the source-oriented models attributed ~28% of Big Bend's sulfate to Mexican sources while the receptor models attributed ~50%. For the eastern U.S., the source-oriented models estimated their contribution at ~40% compared to ~20% for the receptor models.

**Table 9-1. Summary of the source attribution techniques used in the BRAVO study. Only the receptor techniques that passed all validation studies are shown.**

| | Model | Model Orientation | Transport Model | Wind Field(s) | Airmass Max. Age | Domain | Boundary Conditions | Reference |
|---|---|---|---|---|---|---|---|---|
| 1 | [1]REMSAD | Source | REMSAD | MM5 | Indefinite | U.S. and Mexico | GOCART Global Model | section 6 |
| 2 | [2]CMAQ | Source | CMAQ | MM5 | Indefinite | Texas and surrounding States and N. Mexico | REMSAD sulfate modified by IMPROVE and CASTNet measured $SO_4$ & $SO_2$ | *EPRI*, 2004 |
| 3 | [3]FMBR–MM5 | Receptor | Monte Carlo Model | MM5 | 10 days | Most of U.S. and Mexico | None | section 8.2.4 |
| 4 | [3]FMBR–EDAS | Receptor | Monte Carlo Model | EDAS/FNL | 10 days | Most of U.S. and Mexico | None | section 8.2.4 |
| 5 | [4]TrMB–MM5 | Receptor | Monte Carlo Model | MM5 | 5 days | North America | None | Section 8.2.3 |
| 6 | [4]TrMB-EDAS | Receptor | HYSPLIT | EDAS/FNL | 5 days | North America | None | Section 8.2.3 |
| 7 | Synthesized REMSAD | Hybrid | REMSAD | MM5 | Indefinite | U.S. and Mexico | GOCART Global Model | Section 8.2.2 |
| 8 | Synthesized CMAQ | Hybrid | CMAQ | MM5 | Indefinite | Texas and surrounding states and N. Mexico | REMSAD sulfate modified by IMPROVE and CASTNet measured $SO_4$ & $SO_2$ | Section 8.2.2 |

1. REMSAD - Regional Modeling System for Aerosols and Deposition model
2. CMAQ - Community Multiscale Air Quality model
3. FMBR - Forward Mass Balance Regression (FMBR)
4. TrMB - Trajectory Mass Balance

Model Orientation:

Source – Eulerian grid model that simulates source emissions
Receptor – technique that used receptor data with no information on the emission rates
Hybrid – technique that used both receptor data and source emission rates.

**Table 9-2. Big Bend's relative sulfate source attribution results by the various air quality and receptor modeling techniques. The relative contributions are the ratio of the average source attributions to the average predicted concentration at Big Bend during the entire BRAVO period. The bolded values are all within one standard deviation of the mean attribution across all techniques.**

| **Source Region** | **CMAQ[1] (%)** | **REMSAD (%)** | **FMBR - MM5 (%)** | **FMBR - EDAS/FNL (%)** | **TrMB - MM5 (%)** | **TrMB - EDAS(%)** | | **Range (%)** | **Average (%)** | **Std Dev** |
|---|---|---|---|---|---|---|---|---|---|---|
| Carbón | - | 14 | **26** ± 6 | **20** ± 7 | **23** ± 12 | **22** ± 12 | | 14–26 | 21 | 5 |
| Mexico | **32** | 23 | **55** ± 14 | **52** ± 14 | **45** ± 20 | **48** ± 20 | | 23–55 | 43 | 12 |
| Texas | **19** | **16** | **24** ± 8 | **24** ± 12 | **19** ± 13 | 30 ± 20 | | 16–30 | 22 | 5 |
| Eastern U.S. | **39** | 42 | **20** ± 10 | **24** ± 10 | **23** ± 9 | 16 ± 14 | | 16–43 | 28 | 11 |
| Western U.S. | **6** | **9** | **0** ± 5 | **0.** ± 9 | 14 ± 15 | **6** ± 17 | | 0–14 | 6 | 5 |
| Outside of domain | **5** | **7** | | | | | | 5–7 | 6 | 1 |

1. The CMAQ model run to attribute sulfate to Carbón was not conducted due to time constraints .

Detailed analysis of the source and receptor modeling techniques identified potential systematic biases in both types of techniques. It was found that the REMSAD model likely overestimated the eastern U.S. contributions and underestimated the Mexican contributions (see chapter 6). The CMAQ modeling attempted to correct for these biases by adjusting the REMSAD predicted sulfate concentrations used at its boundaries with observed sulfate and sulfur dioxide concentrations. This generally lowered the concentration on the eastern boundary of the CMAQ modeling domain. In Mexico, CMAQ used an upper $SO_2$ emission rate from Carbón I & II which was about 160% larger than that used by REMSAD. Also, the other Mexican $SO_2$ emissions in the CMAQ domain were doubled to compensate for the apparent underestimation of Mexico's contribution to sulfate throughout Texas as estimated by REMSAD. The resulting CMAQ source attributions were larger from Mexico and smaller from the eastern U.S. compared to REMSAD. However, comparisons of the predicted to measured sulfate indicated that the eastern U.S. contributions were still overestimated and Mexico's underestimated.

The receptor techniques suffer from collinear transport, which increases the standard errors and biases due to generally more accurate transport from near source regions compared to far source regions. In chapter 7 it was shown that the FMBR analysis generally overestimated the contributions from nearby source regions in Mexico and Texas and underestimated the contributions from the more distant eastern and western U.S. source regions. In addition, the FMBR technique did not account for contributions from outside of the simulated source regions. Sensitivity testing using the REMSAD synthetic sulfate concentrations and source attributions found that the contribution from REMSAD boundary conditions were primarily placed in the Mexican and Texas source areas.

### 9.2 Reconciled Source Attribution Results – Fusion of Available Information

Several techniques were developed to account for and correct the identifiable biases. For the source-oriented modeling, the synthesis inversion technique was used. In this technique, the modeled source attribution results were regressed against the measured particulate sulfur data at Big Bend. The resulting regression coefficients then accounted for the biases in the original source attribution results. This is a technique similar to FMBR and TrMB, but in addition to the transport information which was incorporated into FMBR and TrMB, the source emission rates, atmospheric chemistry, and removal processes were incorporated into the analysis since these processes were directly incorporated into the REMSAD and CMAQ models.

The biases in the FMBR and TrMB analyses were quantified by applying these techniques to synthetic data derived from the REMSAD model. In this case, the predicted sulfate concentrations at Big Bend from REMSAD were used and the receptor techniques attempted to reproduce the REMSAD source contributions to Big Bend's predicted sulfate. As discussed in section 8.2.3.2, the ratio of the REMSAD attribution to the FMBR or TrMB attribution accounted for the bias. This technique is only applicable to the FMBR and TrMB attributions using the MM5 wind fields because these wind fields were used in the evaluation.

The biases in the Eulerian and receptor models are in opposite direction of each other for the same source regions. Therefore the ranges in the source attribution results in Table 9-2 appear to bound the correct source attribution results. Table 9-3 and Figure 9-1 present the reconciled source attribution results where the original model results are corrected for their respective biases. As shown, the differences between the techniques have narrowed with all source attribution estimates within 5 percentage points of the mean. Mexico is the largest average contributor to Big Bend's sulfate at about 40%, of which ~22% is attributed to the Carbón facilities. The U.S. is responsible for about 55% of the sulfate, of which the eastern U.S. accounts for 28% and Texas accounts for 17%. The western U.S. and contributions from sources outside of the domain are each responsible for less than 10% of the sulfate.

**Table 9-3. Reconciled relative source attributions of Big Bend's sulfate concentrations. The source attribution results by the various air quality and receptor modeling techniques have been adjusted to correct for identifiable biases. The relative contributions are the ratio of the average source attributions to the average predicted concentration at Big Bend during the entire BRAVO period.**

| **Source Region** | **Synthesized CMAQ (%)** | **Synthesized REMSAD (%)** | **Scaled FMBR – MM5 (%)** | **Scaled TrMB – MM5 (%)** | | **Range (%)** | **Average (%)** |
|---|---|---|---|---|---|---|---|
| Carbón | - | 23 ± 1.8 | 22 | 23 | | 22–23 | 22 |
| Mexico | 38 ± 1.7 | 39 ± 2.3 | 43 | 34 | | 34–43 | 39 |
| Texas | 17 ± 1.3 | 16 ± 1.2 | 17 | 16 | | 16–17 | 17 |
| Eastern U.S. | 30 ± 1.2 | 32 ± 0.5 | 24 | 25 | | 24–32 | 28 |
| Western U.S. | 8.5 ± 1 | 6 ± 1 | | 11 | | 6–11 | 9 |
| Outside of domain | 6.4 ± 1 | 7 ± 1 | | | | 6–7 | 7 |
| ***Unaccounted Mass** | | | 17 | 14 | | | |

* The unaccounted mass is the difference between the scaled source attribution results and 100%. For the scaled FMBR this is primarily due to contributions from the boundary conditions and the western U.S., and for TrMB this is primarily due to contributions from the boundary conditions.

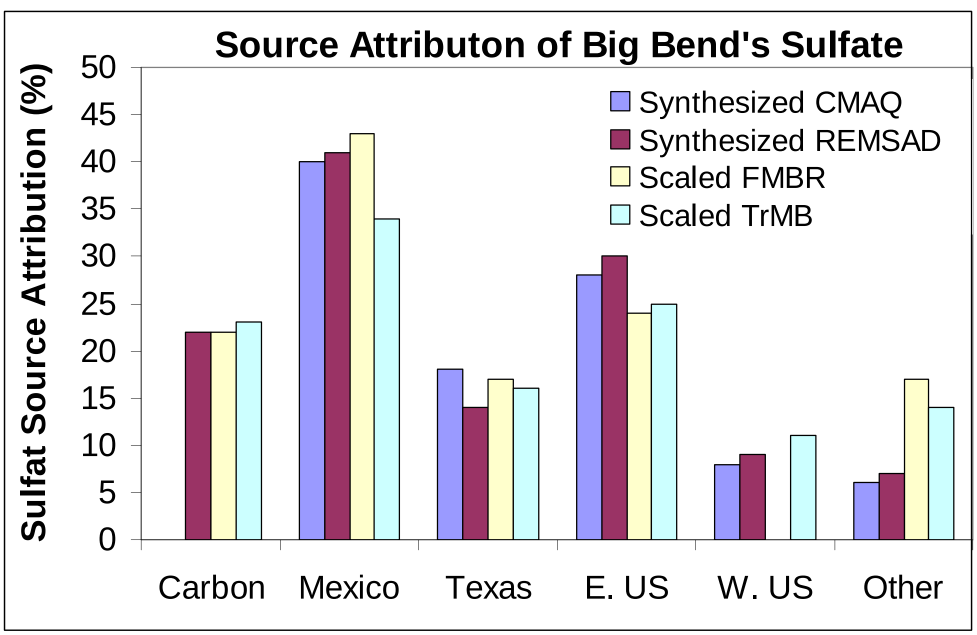



**Figure 9-1. The average relative contribution of sulfate from major source regions to Big Bend's sulfate during the BRAVO study using various methods. The relative contributions are the ratio of the average source attribution to the average predicted Big Bend sulfate.**

The close agreement between the reconciled values is encouraging. However, the four reconciled source attribution estimates are not independent attributions, and their close agreement does not insure that they are correct. All four techniques incorporated the measured particulate sulfate data at Big Bend into the analyses, and they all used the same MM5 wind fields in calculating airmass transport from the source to the receptor. The CMAQ and REMSAD simulations are closely linked since CMAQ used the REMSAD predicted sulfur concentrations at its boundary. Last, the scaling factors for the receptor modeling technique were derived using the REMSAD modeling results. Therefore these final values should be viewed as the result of the fusion of the best available information from all techniques applied in this study.

The only truly independent analysis was the TAGIT analysis conducted by *Green et al.* [2003]. In this analysis they used $SO_2$ concentrations measured in and around Big Bend as a tracer for the plume from the Carbón facilities. They estimated that Carbón was responsible for about 15% of Big Bend's sulfate during the BRAVO study. This is about 30% smaller than the reconciled values.

### 9.2.1 Daily Attribution of Big Bend's Sulfate Concentrations

The synthesized CMAQ and REMSAD results are applicable to shorter time periods, but the receptor techniques are not. On average, the two synthesis inversion results are nearly identical. However, there were some differences on a higher time resolution, particularly during the Big Bend sulfur episodes (see section 8.2.2). While neither result is truly correct, the synthesized CMAQ source attribution results are likely more trustworthy for the same source regions. The original CMAQ runs attempted to reduce known biases that occurred in REMSAD, and CMAQ compared better to the measured data. In addition, the synthesized CMAQ had smaller bias correction factors indicating the initial model results were less biased than REMSAD.

The following presents the source attributions results for everyday, and high and low sulfur days, using the synthesized CMAQ results. CMAQ did not apportion the contributions from Carbón. To estimate the Carbón contribution, the synthesized REMSAD results were used to calculate the ratio of Carbón's to Mexico's contribution and this ratio was applied to the synthesized CMAQ results.

The absolute and relative contributions on a daily basis are presented in Figure 9-2 smoothed by a three-day moving average. Each source region's contribution has unique trends over the four-month period. Mexico's contributions dominate the predicted sulfate concentrations in July and August, contributing from 0.5 to 1.5 $\mu g/m^3$ of sulfate every day and occasionally exceeding 2 $\mu g/m^3$. In September and October, the contribution from Mexico to Big Bend's sulfate decreased typically less than 1 $\mu g/m^3$ per day. The Texas sources contribute little in July and have their largest contribution in October. The Texas source contributions are episodic with their largest absolute contributions (> 1 $\mu g/m^3$) during the highest sulfate days. The eastern U.S. contributions also tend to occur during Big Bend sulfate episodes and exceeded 4 $\mu g/m^3$ during the September 1 episode. The coincidence of the Texas and Mexican contributions to Big Bend are corroborated by the airmass history analyses which showed that airmass transport from the eastern U.S. to Big Bend typically passed through Texas.

Mexico is the largest contributor to Big Bend's sulfate during the days with the lowest sulfate concentrations. This is particularly so from July–September where Mexico was often responsible for more than 80% of Big Bend's sulfate concentrations when the overall concentrations were below 2 µg/m³. In October, the western U.S. and boundary conditions were also large contributors to the lowest sulfur days, together accounting for 60–80% of the sulfur from October 19–24.

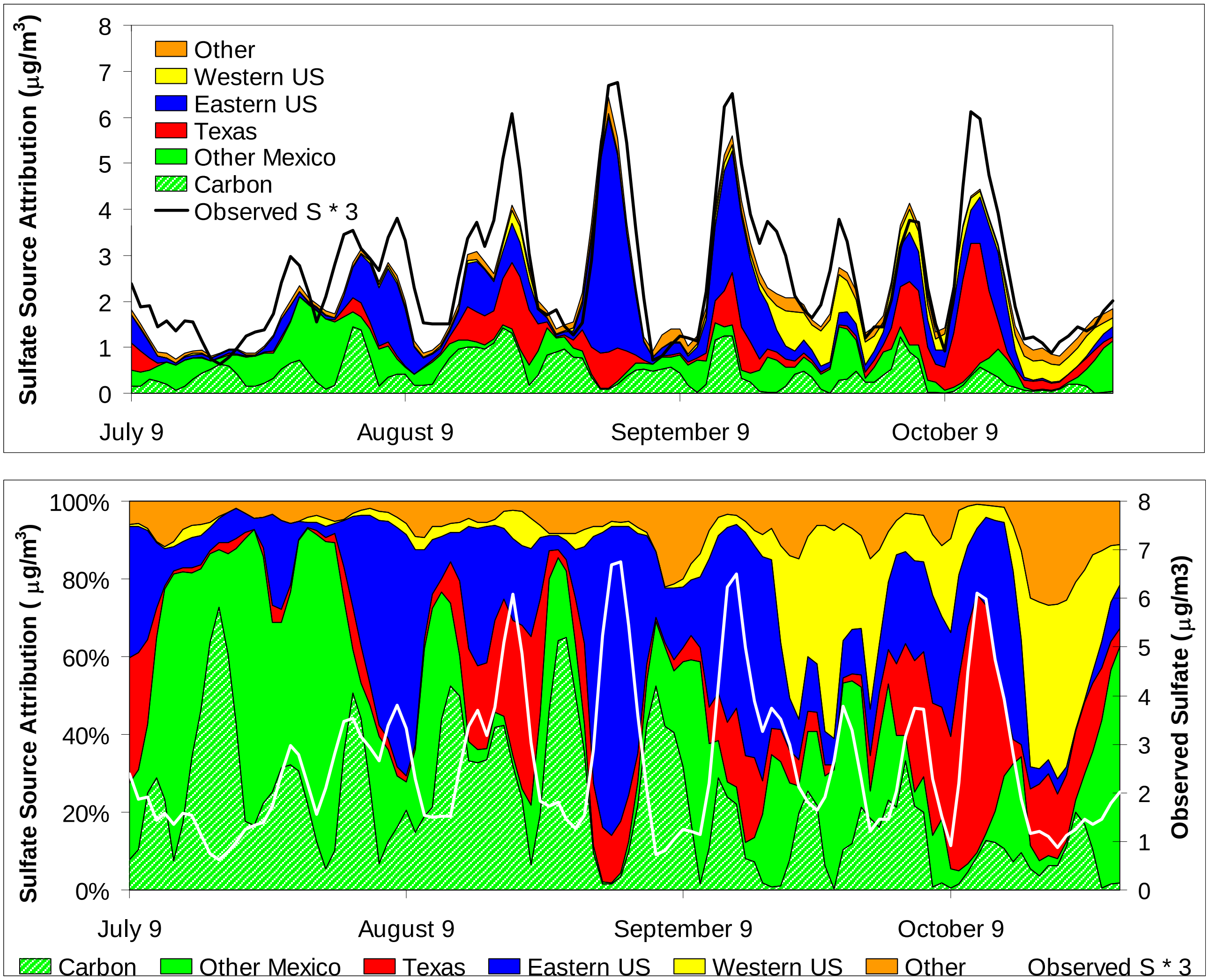



**Figure 9-2. Synthesized CMAQ daily absolute and relative source attributions to the predicted sulfate at Big Bend. The daily source attributions were smoothed using a three-day moving average. The Carbón attributions are derived from synthesized REMSAD.**

Figure 9-3 presents the Big Bend sulfate attribution for days with observed sulfate concentrations above the 80$^{th}$ percentile (high sulfate days) and below the 20$^{th}$ percentile (low sulfate days) during BRAVO. As shown, when Big Bend's sulfate concentrations were high, the United States was responsible for almost 70% of the sulfate, 43% from the eastern U.S. and 25% from Texas. Mexico was responsible for about 23% of the sulfate with the Carbón facilities accounting for 16%. However, on the low sulfur days, the United States' contribution decreased to about 40% with Texas and the western and eastern U.S. each contributing 11–15%. Mexico's contribution increased to about 48% with Carbón contributing 24%.

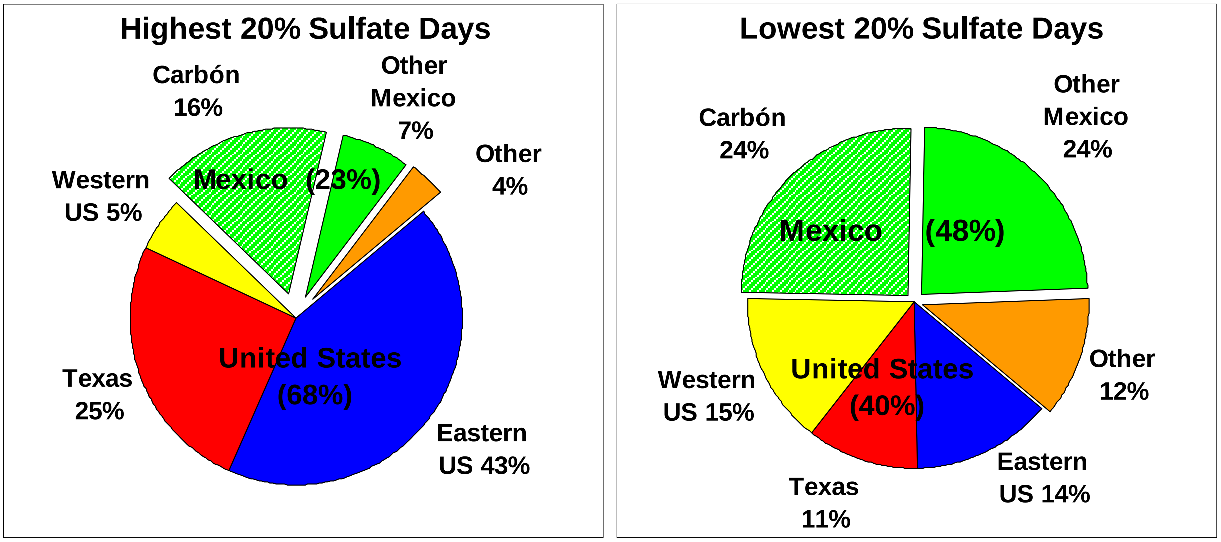



**Figure 9-3. Big Bend's average sulfate attribution for days with the highest and lowest 20% measured sulfate concentration during BRAVO.**

A detailed look at seven multi-day sulfate episodes (Figure 9-4) showed that Mexico, the eastern U.S., and Texas all contributed 30% or more of the predicted sulfate concentrations in two or more episodes. Also, usually more than one of the major source regions had significant contributions to the sulfate, the exception being for the September 1 episode, in which the eastern U.S. accounted for more than 70% of the sulfate.

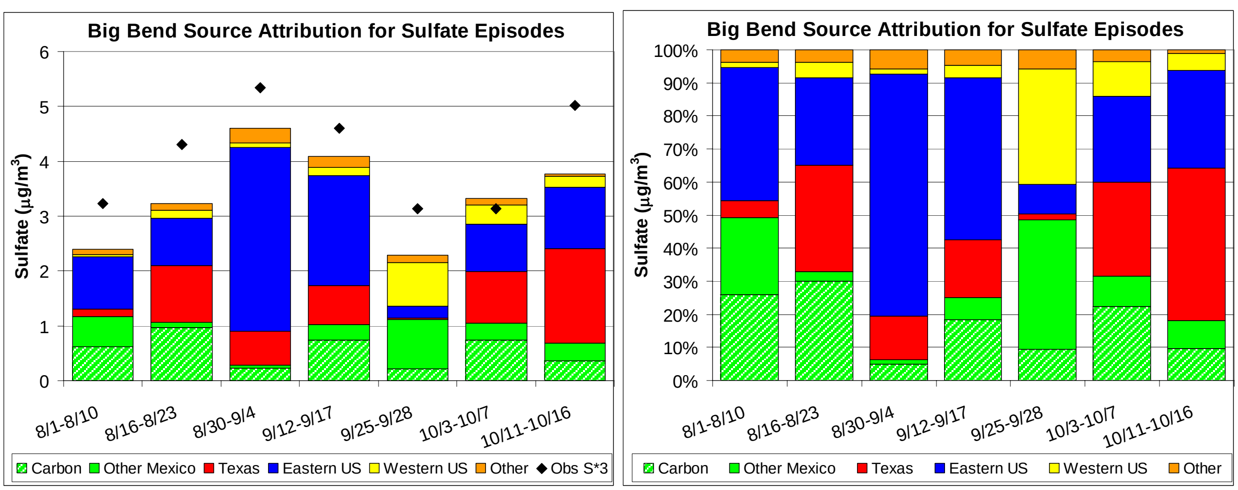



**Figure 9-4. The synthesized CMAQ average source attribution to Big Bend's sulfate for each Big Bend sulfate episode. The Carbón contribution was derived from the synthesized REMSAD results.**

The source attributions for six multi-day low sulfate periods are presented in Figure 9-5. In July and August the Mexican sources are responsible for more than 70% of the sulfate. This contribution decreases in September and October when contributions from the western U.S. and "other", i.e., the modeling boundary conditions, increase. During the October 17–24 period the western U.S. was responsible for about 40% of the sulfate and the boundary conditions about 25%. Sources in the eastern U.S. and Texas generally were responsible for less than 20% of the sulfate at Big Bend during these low sulfate periods.

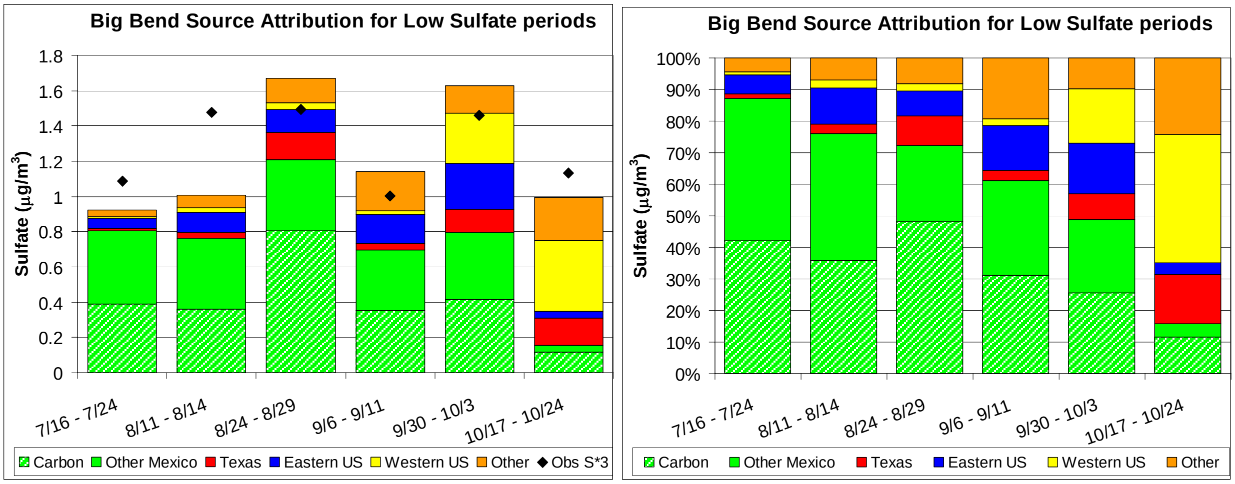



**Figure 9-5. The synthesized CMAQ average source attribution to Big Bend's sulfate for each low sulfate periods at Big Bend. The Carbón contribution was derived from the synthesized REMSAD results.**

### 9.2.2 The Relative Errors in the BRAVO Source Attribution Estimates

The synthesis inversion technique used to generate the final BRAVO source attribution results estimates a standard error for each three-day average source attribution. In order to perform the synthesis inversion, an estimated error, the prior error in the original CMAQ source attribution results of 100%, and an error of 200% for REMSAD were used. A higher prior error was chosen to prevent erroneously constraining the synthesis inversion technique. However, the resulting standard errors are sensitive to the chosen CMAQ and REMSAD prior errors and larger prior errors result in larger standard errors.

Each set of daily source attribution scaling coefficients resulting from the synthesis inversion was derived using data from multiple receptor sites for three consecutive days and then applied to the daily Big Bend source attribution results. If the assumption of linearity and independence in the regression analysis are met, then the scaling coefficients and standard errors are reasonable estimates for Big Bend. The source attribution biases were shown to be approximately constant over the monitoring sites used in the analysis, so the linear assumption should be valid for most days. There are known collinearities in the system, for example, transport from the eastern U.S. to Big Bend usually traverses Texas, but collinearities are reflected in the inversion procedure though increased standard errors. Therefore the standard errors resulting from the synthesis inversion are believed to be applicable to Big Bend's source attribution results.

The average daily relative standard error and the error in the average source attribution estimates during BRAVO are presented in Table 9.4. Note that these errors are relative to each source's average contribution to Big Bend's predicted sulfate. Previously, the errors were reported relative to the total predicted sulfate concentration, so these errors are larger. As shown, the standard errors in Big Bend's daily sulfate source attribution estimates are on average between 50 and 70% for the U.S. and Mexican source regions. The errors decrease with increased averaging. The sulfate episodes and low period lasted for 4 to 9 days and over these time periods the average errors would decrease about 2 to 3 times resulting in errors typically less than 25%. The averaging over the BRAVO period decreased the errors further resulting in errors from 4 to 10% (Table 9-4).

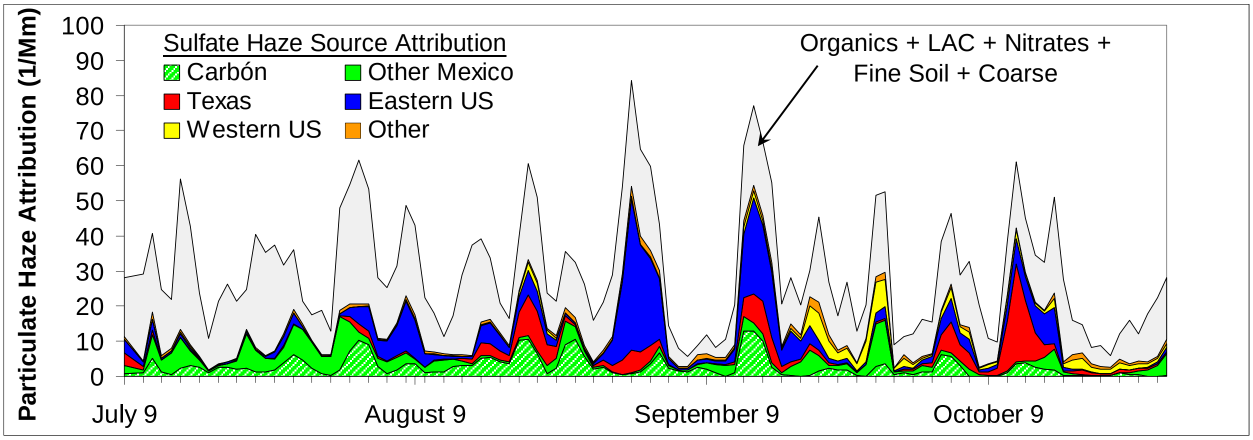
**Table 9-4. The average of the three-day average relative standard errors and their standard deviation for the BRAVO estimated source contributions of Big Bend's sulfate and the error of the four-month average source contributions. In the aggregation of the errors, values were used only if the source contributed 5% or more to Big Bend's estimated sulfate concentration.**

| | **Relative BRAVO Source Attribution Errors** | | | |
|---|---|---|---|---|
| **Source Region** | **Average Daily Error ± Standard Deviation** | **Average BRAVO Day** | **Worst 20% haze days** | **Best 20% Haze Days** |
| Carbon | 67% ± 29% | 7% | 12% | 12% |
| Mexico | 49% ± 33% | 5% | 9% | 22% |
| Eastern US | 53% ± 40% | 4% | 6% | 21% |
| Texas | 68% ± 39% | 7% | 7% | 26% |
| Western US | 69% ± 20% | 9% | 16% | 27% |
| Outside of Domain | 85% ± 15% | 10% | 14% | 23% |

### 9.3 The Sulfate Contribution to Haze from each Source Region.

The direct attribution of haze to its sources was not conducted in this study. However, the sulfate source attribution results can be related to haze by scaling the sulfate portion of the Big Bend haze budget (see chapter 3) by the relative sulfate source attribution results. Figure 9-6 presents the daily absolute and percent fractional contribution by sulfur source regions to Big Bend particulate haze. Particulate haze refers to the non-Rayleigh portion of the light extinction that is the result of both man-made and naturally occurring particles in the atmosphere. Rayleigh scattering, light scattered by particle-free air, was not included in these figures, since Rayleigh scattering is a natural and fixed consequence of the earth's atmosphere.

Sources in Mexico generally contribute a moderate amount of 5 $Mm^{-1}$ to 15 $Mm^{-1}$ of the light extinction on most days during the study period. However, during some of the smaller haze episodes in July and August their relative contributions are 30% to 40% of the average light extinction. Sources in Texas contributed less than 5 $Mm^{-1}$ on most days of the study period, but during one of the periods of higher contribution Texas was responsible for nearly 30 $Mm^{-1}$, or about 40% of the light extinction on the haziest day in October. Sources in the eastern U.S. also contributed less than 5 $Mm^{-1}$ on most days of the study period, but during the two haziest episodes its sources contributed about 50 $Mm^{-1}$ and about 30 $Mm^{-1}$, respectively, corresponding to about 50% and 30% of the total light extinction.



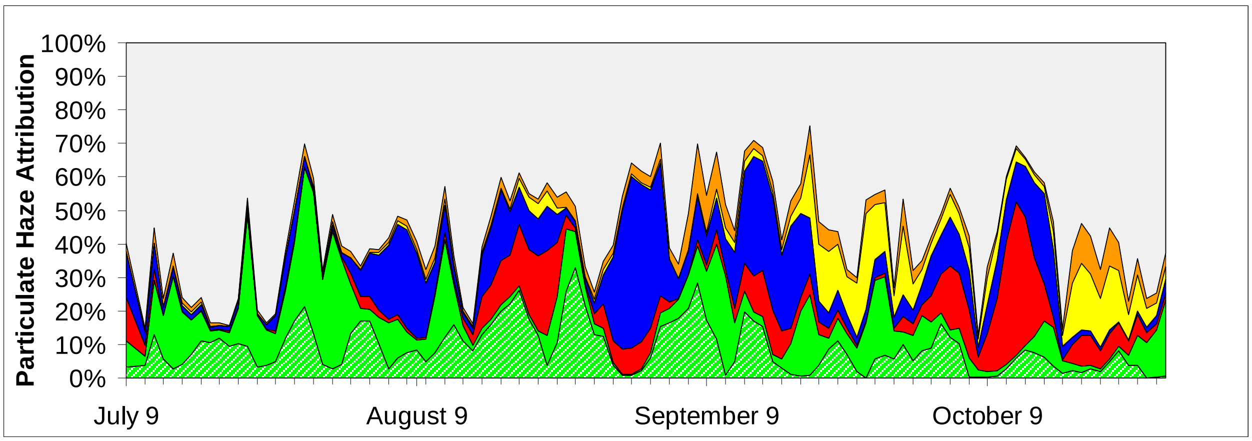




**Figure 9-6. Estimated contributions to particulate haze by various particulate sulfate source regions. The top plot shows the absolute haze contributions by the various particulate sulfate sources as well as the total particulate haze level (black line). The bottom plot shows the fractional contribution to haze by the various sources.**

Figure 9-7 presents pie diagrams illustrating the differences in particulate sulfate contributions by various source regions to particulate haze for the study period's 20% haziest days compared to the study period's 20% least hazy days. The numbers of 20% haziest days for each month of the BRAVO study from July 9th through October are 1, 8, 10, and 4, while the numbers per month for the 20% least hazy days are 3, 1, 10, and 9, respectively.

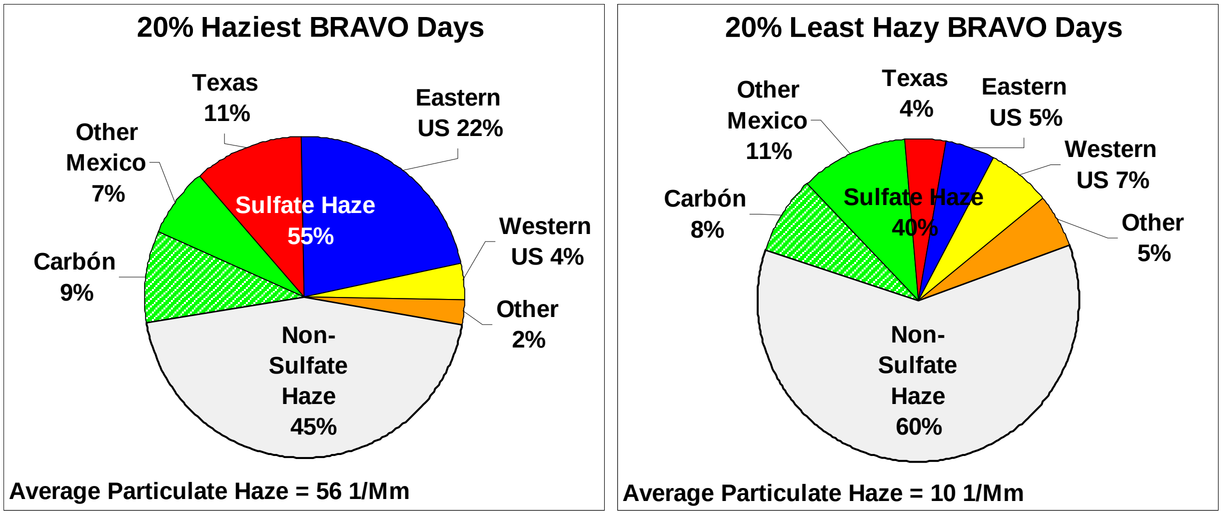



**Figure 9-7. Estimated contributions by particulate sulfate source regions to Big Bend particulate haze levels for the 20% haziest days and the 20% least hazy days of the BRAVO study period.**

As shown, particulate sulfate contributions to light extinction are higher on the haziest days compared to the least hazy days (55% compared to 40%). The non-sulfate haze is primarily due to dust (fine soil and coarse particles) and carbonaceous (organic and carbon) compounds. Compared to the least hazy days, the haziest days have a higher relative contribution to light extinction by dust (25% compared to 19%) and a lower relative contribution by carbonaceous particles (19% compared to 39%).

The increase in the sulfate contribution on the haziest days compared to the least hazy days is driven by increased relative contributions from the eastern U.S. and Texas. The relative contribution of sulfate on the haziest days from Texas increased by about a factor of 3 (4% to 11%), and from the eastern U.S. increased by about a factor of 4 (5% to 22%) compared to the least hazy days. In contrast, the relative contributions for the Carbón I & II power plants remained about the same at 8 to 9% and the contribution of other sources in Mexico decreased from 11% on the least hazy days to 7% on the haziest days. The relative contribution of sulfate sources in the western U.S. to Big Bend's sulfate haze also decreased from 7% on the least hazy days to 4% on the haziest days. These results are consistent with the observation that the Texas and eastern U.S. sources had their largest sulfate contributions during the highest sulfate episodes.

The errors for the haze attribution results in Figure 9-7 are presented in Table 9-4. As shown, for the 20% haziest BRAVO days, the errors are from 6–12% for the Mexican, Texas, and eastern U.S. source regions, but increase to about 15% for the western U.S. and other regions. The relative errors are larger on the 20% least hazy BRAVO days with errors between 22–26% for all source regions except Carbon which has an error of only 12%.

### 9.4 Application of the Source Attribution Results to other Months and Years

In order to assess the applicability of haze attribution results for the BRAVO study to other years or other times of year, it is necessary to compare the four-month study period with the same months in other years and with other months of the year. Emissions and meteorology are the two most important factors that influence haze levels. Between 1999 and the present the annual emissions responsible for particulate sulfate concentrations in North America have not appreciably changed (U.S. emissions have decreased about 15%, but less is known about emission trends in Mexico). Seasonal variations in $SO_2$ emissions and in the $SO_2$ to particulate sulfate oxidation rate make extrapolations of the BRAVO study results to other months of the year prone to additional uncertainty. One of the most influential meteorological processes affecting the haze at Big Bend is the airflow patterns that determine which potential source regions are upwind of Big Bend. In spite of the uncertainties inherent in such a simple approach, comparisons of the meteorological flow patterns from the residence time analysis (section 8.1.3) were used alone in an attempt to assess the applicability of BRAVO study results to other years and times of year.

Residence time plots convey information about both the frequency of transport over potential source regions and its duration over the regions. In section 8.1.3 it was shown that the residence time transport patterns are driven by the variations in transport frequency over regions as opposed to duration variations. Consequently, a change that doubles the residence time over a source region for a specific month can be thought of as doubling the probability of influence of that source region during that month. In this example the monthly average contribution would likely double because the numbers of impacting periods would about double, but the amount of the peak impact is not expected to change much.

As shown in section 8.1.3.5, during the BRAVO study period airflow to Big Bend was mostly similar to the airflow conditions during the five-year period. However, in September 1999 there was typically less flow over the eastern U.S. than for the five-year average, implying that the BRAVO results may underestimate the average haze contributions by that region's sources. In addition, during October 1999 there was typically more flow over Texas and less flow over Mexico, implying that the average October BRAVO haze contributions may be overestimated for Texas and underestimated for Mexico compared to the five-year average. While the estimated average contributions by these source regions may change, the peak contributions are likely not affected by the atypical frequency of flow.

Comparing the airflow patterns for the BRAVO study period to that of the other months of the year (section 8.1.3.5), it is evident that $SO_2$ sources in Mexico are likely to contribute less in November through March. This is because airflow across Mexico is less in general and is over lower emission density regions of Mexico to the west of Big Bend. $SO_2$ sources in Mexico are likely to be contributing to the particulate sulfate portion of the Big Bend haze during the months of April through June comparable to their contributions during the BRAVO study for July and August months. Sources in Texas are likely to contribute little to the particulate sulfate portion of the Big Bend haze for the months from November through June since the airflow is not frequently over the high emissions regions of east Texas, similar to July 1999. Eastern U.S. sources are unlikely to contribute to Big Bend haze during the months from November through March since airflow to Big Bend is rarely over that region during those months. During the months from April to June, the eastern U.S. sources may contribute a modest amount to sulfate haze, comparable to that estimated for July and early August.

### 9.5 Implications.

There is no simple answer to the question of what sources are responsible for the haze at Big Bend National Park; sources in both the U.S. and Mexico are responsible. Mexican $SO_2$ emissions contribute to the sulfate haze more frequently, but to generate the haziest events that occur in the late summer and fall, contributions from Texas and the eastern U.S. must occur.

The greatest contributors to haze are the Carbón I & II power plants in northern Mexico. Substantial changes in those facilities' emissions would likely result in small but noticeable changes in haze levels on many days, but it would not make much difference to the worst haze episodes during late summer and early fall. To substantially affect all of the haze episodes during the late summer and fall where U.S. contributions are large at Big Bend will require $SO_2$ emission changes in both Texas and the eastern U.S. Because of the high frequency of air transported to Big Bend from the southeast along a corridor on both sides of the Rio Grande River, emission changes there have a potential to affect haze levels at Big Bend, especially during June through September when transport from this region is most frequent.

The clearest days at Big Bend also had low sulfate concentrations. The visual scene on a clear day is more sensitive to small changes in haze than a hazy or moderately hazy day, and small changes in sulfate concentrations can lead to perceptible changes in haze. On these clear days, the Carbón I & II power plants and other sources in northeast Mexico were the largest contributors to Big Bend's sulfate. Reduction in emissions from Carbón would likely result in creating more clear days. On the other hand, growth along this border region will likely further reduce the number of clear days.