7) Receptor Model Evaluation

A test of the accuracy of any source attribution method is to determine how well it can reproduce known attributions. There are at least two known source attributions with which to test receptor models during BRAVO. The first is the attribution of the measured perfluorocarbon tracer concentrations to the four tracer release sites. In that case only transport and dispersion are tested since the tracers are non-reacting and essentially non-depositing. Both MM5 and EDAS/FNL wind fields can be tested using the tracers.

A similar test involving chemistry and deposition in addition to dispersion is to determine how accurately the receptor models can reproduce the sulfate source attributions simulated by the REMSAD model. In this case we know the concentrations and source attributions of the 24-hour average sulfate predicted by REMSAD. For purposes of the test, there is no assertion that the REMSAD-simulated sulfate source attributions are accurate in the real world, only that in the simulated REMSAD world we have known emissions, chemistry, and meteorology and so if we model these using receptor techniques, we would expect to reproduce the simulated source attributions. In this case only the MM5 meteorological data are used since this is what was used by REMSAD and so is therefore the correct wind field for transport of the simulated sulfate. The tests of REMSAD sulfate attribution also allow testing of the receptor models with source areas beyond the state of Texas, whereas the perfluorocarbon tracers were all emitted within the state and therefore cannot be used to test longer range transport. All three back trajectory models, ATAD, HYSPLIT, and CMC, were used in both these tracer and REMSAD sulfur tests. See chapter 5 for another evaluation for the forward transport models using the tracer data.

7.1 Source Apportionment of Tracer Data

7.1.1 Trajectory Mass Balance

To test the accuracy of the Trajectory Mass Balance (TrMB) model, it was exercised using the total perfluorocarbon tracer concentration at Big Bend as the dependent variable and back trajectory endpoints from the four tracer release sites as the independent variables.

7.1.1.1 Tracer Concentrations

The daily total tracer concentrations for attribution testing by TrMB were calculated by summing the 24-hour average concentrations of the four perfluorocarbon tracers for each day during the second half of the study when the four tracers were released from four different sites. The fraction of the total concentration due to each tracer is the fraction arriving from each tracer's corresponding release site. The four tracer release sites and the TrMB source areas used to represent each are shown in section 2.3.2.5.

A problem for use of the tracer concentrations in TrMB is how to handle the many negative concentrations. Negative concentrations result when the mean background is subtracted from the analyzed value. The most severe problem is with iPPCH, the northeast Texas or Big Brown tracer, which has a negative mean concentration at K-Bar and would therefore require a negative regression coefficient in order to reproduce the mean concentration accurately. Four options were explored: 1) leaving negative concentrations as negative, 2) setting all negative concentrations to zero, 3) adding a constant concentration to all days to raise the minimum concentration of each tracer to zero, or 4) removing all days with negative concentrations. Option 4 proved unreasonable since too many days would be removed. All of the remaining options were tried. There were only minimal differences in the results and model performance for these three options, though in general options 2 and 3 performed slightly better. It was decided to continue using option 2, setting all negative 24-hour concentrations of the individual tracers to zero before summing them to calculate the total tracer concentration for each day and further, to proceed without using iPPCH, because on average, the concentration of this tracer was so low as to be mostly undetectable at K-Bar, and in fact, was zero plus or minus measurement error.

Another option for the total tracer concentration was to use the mean of the measured tracers at K-Bar, Persimmon Gap, and San Vicente. These three sites are all within or near the boundaries of Big Bend National Park. Again, the model results and model performance did not change substantially when data from one or both of the additional sites were averaged with the K-Bar data. For simplicity, the experiment was continued using data from K-Bar only. Figure 7-1 shows the resulting total tracer concentrations at K-Bar for each day with the bar colors showing the contribution of each of the three remaining tracers.

Several different begin and end dates were also tried, and these did affect model results and performance because different dates resulted in different regression coefficients. This is expected due to changes in the mean meteorology and emissions when different time periods are used. The final time period chosen was September 17–October 28. Releases of PDCB and PTCH were halted at Eagle Pass on September 13 and begun at San Antonio and Houston, respectively, on September 17, so this was a reasonable beginning date. Tracer release of PTCH from Houston was stopped on October 25, though emissions for the other three sites continued until November 1. After October 28, release of ocPDCH from Eagle Pass became erratic with much higher releases than average on the 29th and lower than average on the 30th, so an end date of October 28 was chosen. Tracer emissions during the second half of BRAVO are shown in Figures 2-3 through 2-6. A constant release of tracers throughout the study would be ideal for TrMB analysis. This did not occur, with each of the tracers having periods of higher than average, lower than average, and no emissions.

7.1.1.2 Results and Discussion of TrMB Tracer Test

Table 7-1 is a summary of the TrMB model results for total tracer at K-Bar during the second half of the BRAVO study. Several model and data combinations are able to reproduce the known attributions of all three tracers to within the error in the measured concentration and the standard errors of the regression coefficients. These include HYSPLIT with EDAS/FNL input, CAPITA MC with MM5, ATAD with MM5, and ATAD with raw sounding data. In general, the choice of 5-, 7-, or 10-day back trajectories made little difference and the results were usually the same within the standard error of the regression coefficients, no matter what the trajectory length. This is most likely because the tracer release sites are all within 5 days transport of K-Bar.

Worst model performance is by HYSPLIT with MM5 input which dramatically over-predicts Eagle Pass and under-predicts San Antonio. Eagle Pass is approximately 250 km from K-Bar, while San Antonio is almost twice as distant at approximately 400 km. This may be an indication that HYSPLIT with MM5 has too many back trajectory endpoints close to the receptor at the expense of too few farther away. Other problem combinations were ATAD with EDAS/FNL input and CAPITA MC with EDAS/FNL input, which both overestimate tracer arriving from Houston and underestimate San Antonio. Houston is the most distant of the three modeled release sites at approximately 700 km from K-Bar, so these latter combinations are overestimating the most distant source area.

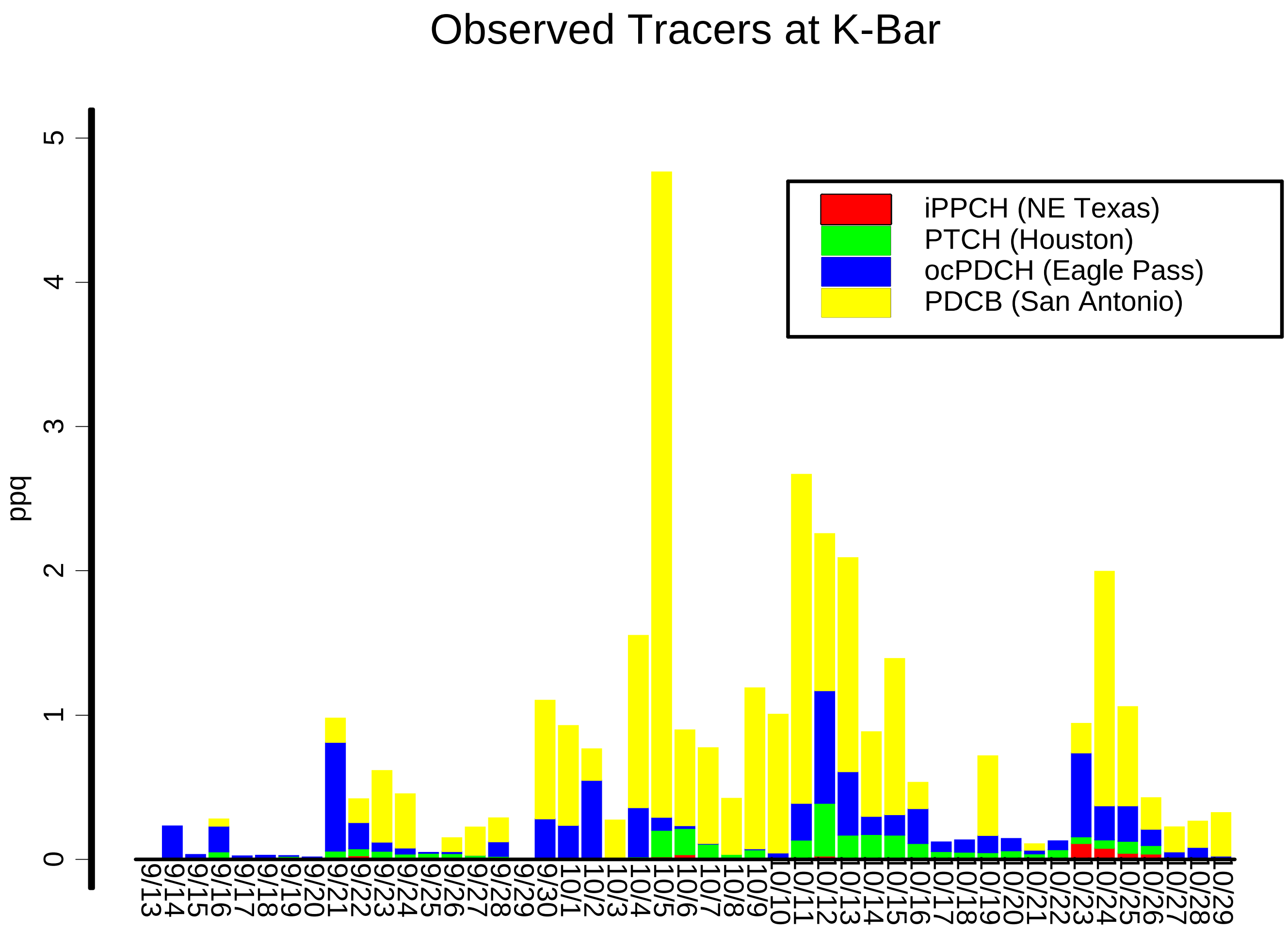

Figure 7-1. Observed perfluorocarbon tracer concentrations (ppq) at K-Bar during the second half of the BRAVO study after negative concentrations were set to zero.

The problems with the under-performing combinations seem to be unrelated to collinearities between the source areas. In all cases the variance inflation factors (VIF) (see section 2.3.2.1) were less than 3 for all sources. A high VIF indicates that the endpoints in a single source region could be nearly explained by a linear combination of the endpoints in other source regions. A VIF greater than 10 is considered strong collinearity. Correlations between source area endpoints were as high as 0.675, but the amount of correlation between source areas was unrelated to how well the model/wind field combination predicted the correct attribution.

The remaining two possibilities for error are either the tracer concentrations or the back trajectory endpoints. Since several trajectory model and wind field combinations were able to reproduce the tracer attributions, we assume that the concentrations are reasonably accurate to within their reported uncertainties, or at least that we do not have errors so large that the mean percentages due to the three release sites are grossly wrong. This leaves the placement of the back trajectory endpoints within Texas as the only possibility for error. Influential days in the regression are October 5, 11–13, and 23–24. However, examination of the relationships between tracer concentrations and endpoints and between the endpoints from different model/wind field combinations did not reveal any obvious problems with the back trajectories on these individual days.

Results of this test then suggest that source attribution results generated by TrMB using back trajectories from the HYSPLIT model with MM5 input wind fields should not be used for source attribution of sulfate. TrMB modeling using back trajectories from either CAPITA MC or ATAD models with EDAS/FNL input are also suspect. The best model/wind field combination for tracer attributions, and thus for accurate back trajectory placement within south Texas, was HYSPLIT with EDAS/FNL input. The remaining combinations of CAPITA MC or ATAD with MM5 input and ATAD with raw sounding input also were able to re-create the known tracer attributions.

Table 7-1. Results of TrMB modeling of three BRAVO tracers at K-Bar for 9/17–10/28 (42 days). The first three rows show the tracer name, the release site, the mean measured concentrations, and the percent of the total measured concentration due to each tracer. Negative concentrations were set to zero before summing. The remaining rows give the TrMB modeled percent attributions for several combinations of back trajectory models and input meteorological data and trajectory lengths. Attributions that are accurate to within the uncertainty of the measurement and standard error of the regression coefficients are shown in bold and a larger font for easy identification.

	ocPDCH **Eagle Pass**	**PTCH** **San Antonio**	**PDCB** **Houston**	**R^2**
Mean Concentration (ppq)	0.155±0.024	0.559±0.081	0.062±0.008	NA
Mean Percent (%)	**20±4**	**72±13**	**8±1**	NA
ATAD Raw 5-day	**35±12**	**65±14**	**0±9**	0.495
ATAD EDAS/FNL 5-day	**16±8**	33±9	51±9	0.708
HYSPLIT EDAS/FNL 5-day	**28±12**	**67±13**	**5±9**	0.640
HYSPLIT EDAS/FNL 7-day	**29±13**	**68±15**	**3±11**	0.603
HYSPLIT EDAS/FNL 10-day	**27±13**	**73±16**	**0±11**	0.612
CAPITAMC EDAS/FNL 5-day	**30±9**	43±10	27±8	0.721
CAPITAMC EDAS/FNL 7-day	**33±10**	**50±10**	18±8	0.643
CAPITAMC EDAS/FNL 10-day	**30±9**	39±10	31±8	0.689

ATAD MM5 5-day	**34±12**	**60±13**	**6±9**	0.564
HYSPLIT MM5 5-day	82±18	18±18	**0±11**	0.484
HYSPLIT MM5 7-day	81±18	19±19	**0±11**	0.489
HYSPLIT MM5 10-day	81±18	19±18	**0±12**	0.502
CAPITAMC MM5 5-day	**23±16**	**77±19**	**0±12**	0.643
CAPITAMC MM5 7-day	**26±17**	**74±21**	**0±13**	0.616
CAPITAMC MM5 10-day	**24±17**	**76±21**	**0±14**	0.626

7.1.2 Forward Mass Balance Regression

The forward mass balance regression technique is a quantitative source apportionment technique that merges measured receptor data with forward airmass transport simulations. The technique is based upon the inversion of the source receptor technique to retrieve a source/sink term and is described in section 2.3.2.2. The basic equation is:

$$c_{ik} = \sum_j T_{i,k|j} \left(KE\right)_j + error_{ik} \qquad (2\text{-}26)$$

where c_{ik} are the concentration values at the receptor *i* and time k, $(KE)_j$ are the source/sink term for each source region j, and $T_{i,k|j}$ is the transit probability or the probability that mass emitted from source j will impact the receptor i at time k. Given c and T, equation 2-26 is inverted to retrieve (KE) which in turn can be multiplied by T to estimate the source contribution to the receptor concentrations, i.e. $c_{ikj} = T_{i,k|j}\left(KE\right)_j$.

As part of the BRAVO study four unique perfluorocarbon tracers were released to estimate dispersion from four industrial or urban sites (see section 2.1.1.2). The tracers were continually released at near constant rates from Eagle Pass and the Big Brown power plant in northeast Texas from about July 5–October 30 and from San Antonio and the Parish power plant in Houston from about September 17–October 30. The tracers are conservative species, so the source/sink term is equal to the tracer release rate and equation 2-26 describes the dispersion of the tracer plume. These data were used to test and validate the FMBR technique by inverting equation 2-26 to estimate individual tracer release points and locations. In addition, the tracer concentrations were added together, creating integrated concentrations, and the FMBR was used to reconstruct all four tracer release rates and source attributions to the combined tracer concentration. This last test is more synonymous with using FMBR to estimate the source attribution of Big Bend's sulfate concentrations.

7.1.2.1 Transit Probabilities

The transit probabilities were estimated using the CAPITA Monte Carlo (CMC) model. The model was used to generate forward 10-day plumes every two hours from ~670 sources evenly distributed over Mexico and most of North America about 100 km apart (Figure 7-2). A plume identifies the downwind three-dimensional location of particles that were previously released from the source and can be viewed as a direct simulation of the dispersion of a source's emissions. The plumes were calculated by releasing five tracer particles from each source every two hours and tracking their movement in space and time. Daily transit probabilities were calculated through simple particle accounting, where the particles, from each source at a 100 km grid cell at Big Bend within the first 1 km of the surface, were summed together over each day. These particle counts were then divided by the total number of particles comprising a source's plume, e.g., 7 day plume * 12 releases/day * 5 particles/releases * 12 time increments /day= 5040 particles/day. The transit probabilities were calculated using both the 36 km MM5 meteorological data and the combined EDAS/FNL meteorological data. Both wind fields produced similar results so only the MM5 results are presented. In addition, as discussed in the previous section, the TrMB approach with MM5 data proved to be superior to the EDAS/FNL winds when tested against the tracer data.

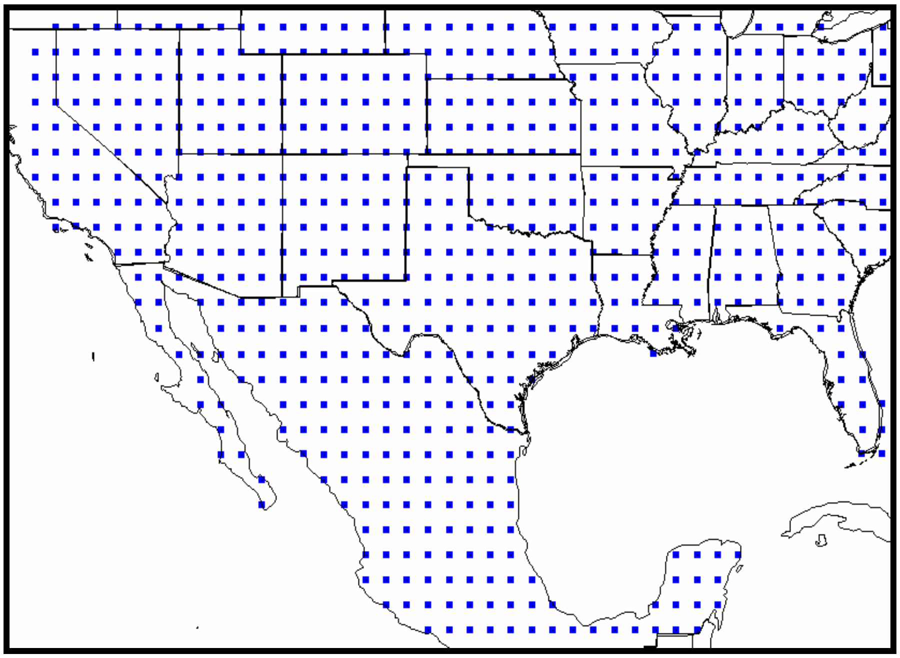

Figure 7-2. The source regions from which 7-day forward plumes were simulated using the CAPITA Monte Carlo model.

The Big Bend transit probabilities are presented in Figure 7-3 for the BRAVO time period and July and October. As shown, over the BRAVO time period transport to Big Bend primarily occurs from the Gulf of Mexico up along the Texas/Mexican border. Airmass transport also occurs from the north and northeast of Big Bend from Texas and the eastern U.S. There is a monthly pattern to the airmass transport with transport during July coming almost exclusively from the southwest of Big Bend along the Texas/Mexican border, and with more transport from Texas and the eastern U.S. during October. These transport patterns are similar to those derived from the residence time analysis in section 8.1.3.3. This is expected since these are similar transport assessments, but the transit probability identifies potential transport of airmasses from the surface to Big Bend, while the residence time analysis examines transport throughout the column.

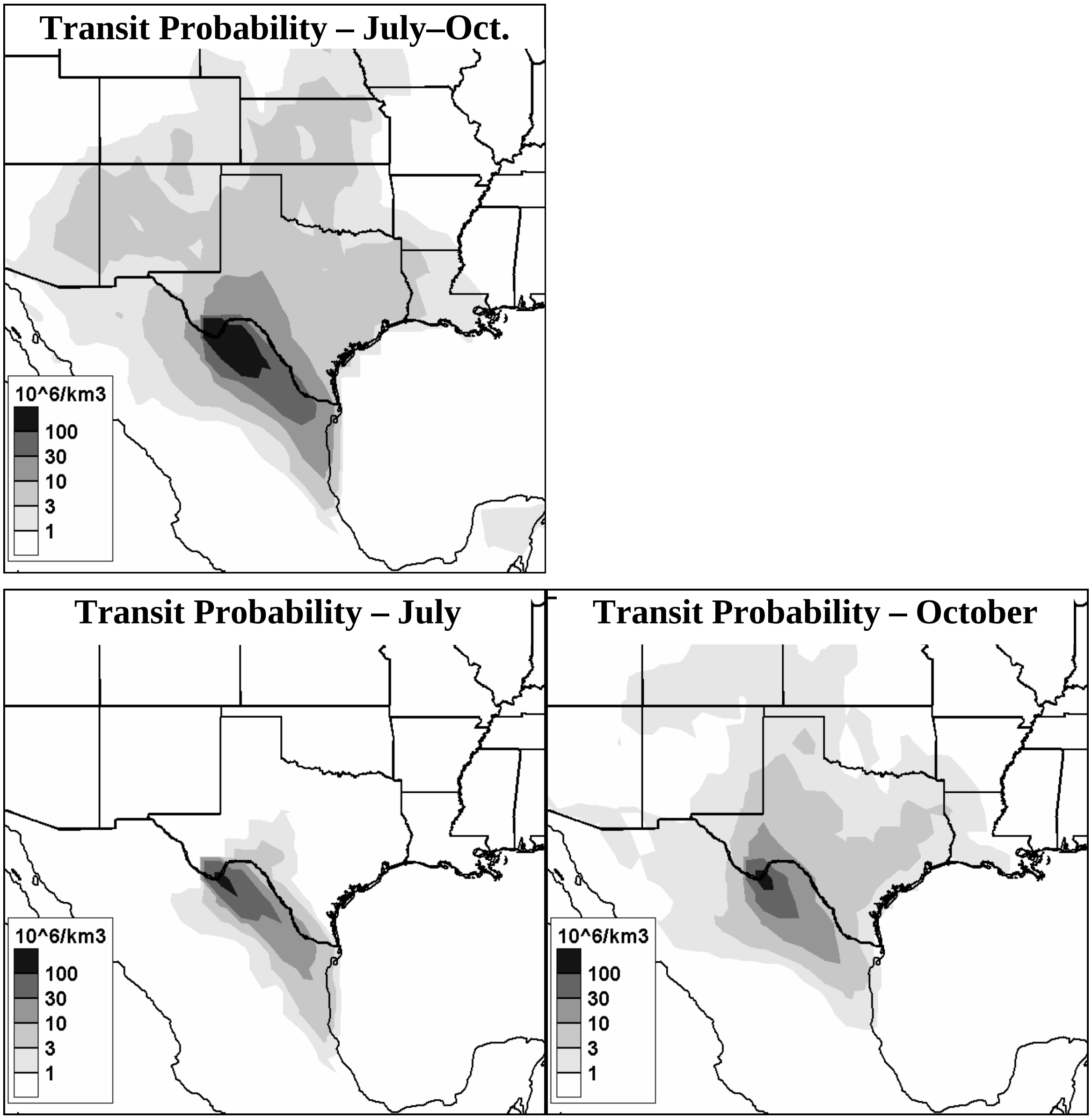

Figure 7-3. Big Bend transit probabilities for the July–October BRAVO time period and the months of July and October.

7.1.2.2 Retrieval of Tracer Emission Rates

The inversion technique was applied to the ocPDCH tracer continuously released from Eagle Pass from July 5–October 30 and to the iPPCH tracer continuously released from northeast Texas from July 9–October 30 in order to test the ability of the inversion technique to properly locate a source's location and emission rate. The tracer concentrations are the measured concentration minus an assumed background. A number of these delta concentrations are negative, reflecting the uncertainty in the measurement and background concentration. However, for the northeast Texas tracer there may have been a systematic bias in the concentrations; for example, the July average iPPCH tracer at Big Bend was negative. All negative values were included in the analysis. The transit probabilities from all ~670 source region plumes were also used in the analysis. The inversion was conducted using singular value decomposition (SVD) which can invert an under-determined system and dampen instabilities that result in least square regressions of ill-conditioned systems, such as the source receptor relationship (see section 2.3.2.2).

The reconstruction of the Eagle Pass and northeast Texas tracer release sites and rates using all measured concentrations values are presented in Figure 7-4 and Table 7-2. This was an over-determined system with about 800 data points and 670 unknown potential source regions, and there were monitoring sites in the vicinity and partially surrounding the tracer release sites. As shown, in both cases the retrieval was able to properly identify the tracer release site. For Eagle Pass, the reconstructed total tracer release rate of 4.2 kg/day is close to the actual rate of 3.7 kg/day. However, the release rate for the northeast Texas tracer was underestimated by about a factor of 6. In both cases the average predicted tracer concentrations resulting from the inversion were within 10% of the measured concentrations and the correlation coefficient was about 0.6.

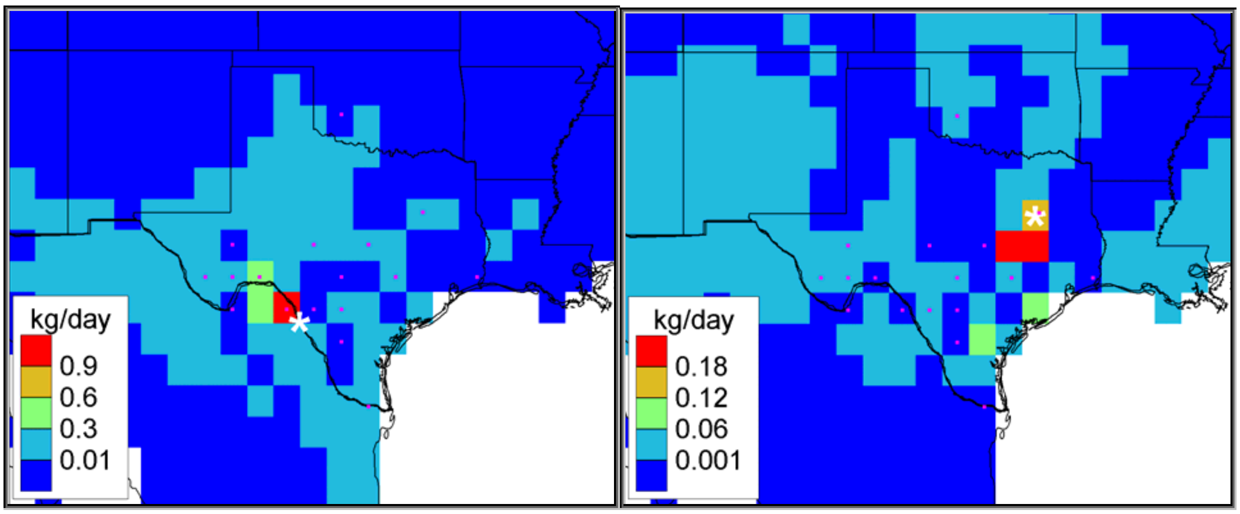

Figure 7-4. Reconstruction of the Eagle Pass and Big Brown tracer release sites and rates using tracer concentration measured all monitoring sites from July–October. The tracer release sites are identified by the stars and the monitoring site locations by the squares.

Table 7-2. The retrieved Eagle Pass and northeast Texas tracer release rates and inversion performance statistics.

			Average (ppq)			Tracer Release Rate (kg/day)	
Tracer	**Receptors**	**# Obs**	**Obs**	**Pred**	**r**	**Actual**	**Retrieved**
Eagle Pass (ocPDCH)	All	798	0.4	0.38	0.6	3.7	4.2
Eagle Pass (ocPDCH)	Big Bend	118	0.148	0.144	0.61	3.7	2
N.E. Texas (i PPCH)	All	798	0.03	0.027	0.59	2	0.34
N.E. Texas (i PPCH)	Big Bend	118	0.007	0.006	0.35	2	0.016

The perfluorocarbon tracers are an inert species not subjected to chemical and physical removal processes and receptor sites had the same source/sink term. However, this study is concerned with the attribution of sulfate, and sulfate is primarily a secondary species generated by the transformation of sulfur dioxide. Both sulfur dioxide and sulfate are removed from the atmosphere by wet and dry deposition. These chemical and physical processes vary in space, and it is not appropriate to derive a single source sink term for distant receptor sites. Therefore application of the FMBR technique to Big Bend's sulfate source attribution can only use measured concentration data from the Big Bend region.

To test the inversion using this limited set of observations, FMBR was applied to only the Big Bend tracer concentrations. In this case, the inversion is a highly under-determined system with only 118 concentration values to resolve the impact from 670 potential sources. In order to get stable results, the SVD solution added more weight to the solution stability at the expense of the model fit and resolution. Therefore the retrieved release rates were spread out over a larger domain. This is seen in the spatial patterns of the retrieved tracer release rates in Figure 7-5 where the retrieved tracer emissions extend from the Big Bend receptor to the actual release sites and beyond.

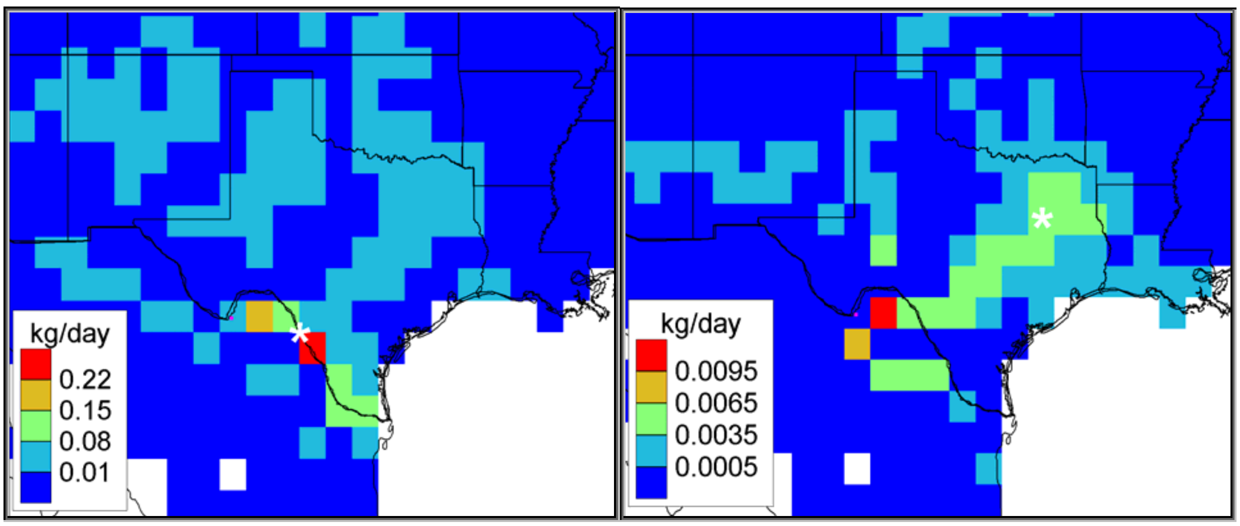

Figure 7-5. Reconstruction of the Eagle Pass and Big Brown tracer release sites and rates using tracer concentration from only the Big Bend monitoring site from July–October. The tracer release sites are identified by the stars and the monitoring site locations by the squares.

The retrieved Eagle Pass tracer still identifies the largest release rates around the true Eagle Pass release site, but now the release rates have been spread out from Big Bend to the Gulf of Mexico, along the Texas-Mexican border. In addition, the total retrieved release rate is about a factor of two lower than the actual rate, 2 kg/day compared to 3.7 kg/day. The retrieved release rates for the northeast Texas tracer cover a large domain with the highest release rates near Big Bend and a region of near constant release rates extending from Big Bend to the northeast Texas release sites. The total release rate severely underestimates the actual rate by over a factor of a hundred (Table 7-2). Also, the correlation of the observed values and predicted concentrations is only 0.35. The underestimation is likely due to a bias in the northeast tracer delta concentrations. The average northeast tracer at Big Bend was only 0.006 ppq and 70% of the delta concentrations were below zero. This demonstrates the fact that the FMBR technique will compensate for biases in both the model and observations.

The spatial patterns in the retrieved tracer release rates are similar to the transport patterns to the Big Bend receptor site for the highest 20% of the tracer concentrations explored in section 8.1.3. Therefore, without any addition information, FMBR when used with the Big Bend receptor data is best suited for identifying the transport pathways. It is possible that if a longer time period were used, then the FMBR could better isolate the tracer release site.

7.1.2.3 Source Apportionment of Combined Tracer Concentrations

The previous section illustrated the difficulty of clearly identifying the tracer release sites and rates using only the Big Bend observed tracer concentrations. In this section, a different question is addressed: given the location of a small set of source regions, can FMBR properly retrieve the contribution of each source region to the ambient concentration at Big Bend? To investigate this question, the four tracer concentrations at Big Bend were added together, creating an integrated tracer concentration time series. Transit probabilities from four source regions encompassing the tracer release sites were estimated from the database of source plumes. The contribution of each individual tracer to the total was then estimated using FMBR. Least square regression was used to invert the relationship and obtain the source attributions. SVD was not used since the system was highly over determined with only four unknowns and 40 known concentration values.

Tracers were only released from the Houston and San Antonio sites from September 17 onward. This analysis included only data from September 19–October 30 and aggregated the values from San Vicente and K-Bar since they fell into the same 100 x 100 km cell around Big Bend. The combined tracer time series is presented in Figure 7-6, where it is evident that the San Antonio tracer dominated the total. The four source regions are presented in Figure 7-7. The sizes of the source regions were dictated by the modeled source regions as shown in Figure 7-2. In addition, the Eagle Pass source region was made smaller than the others to account for the fact that errors in transport increase with distance.

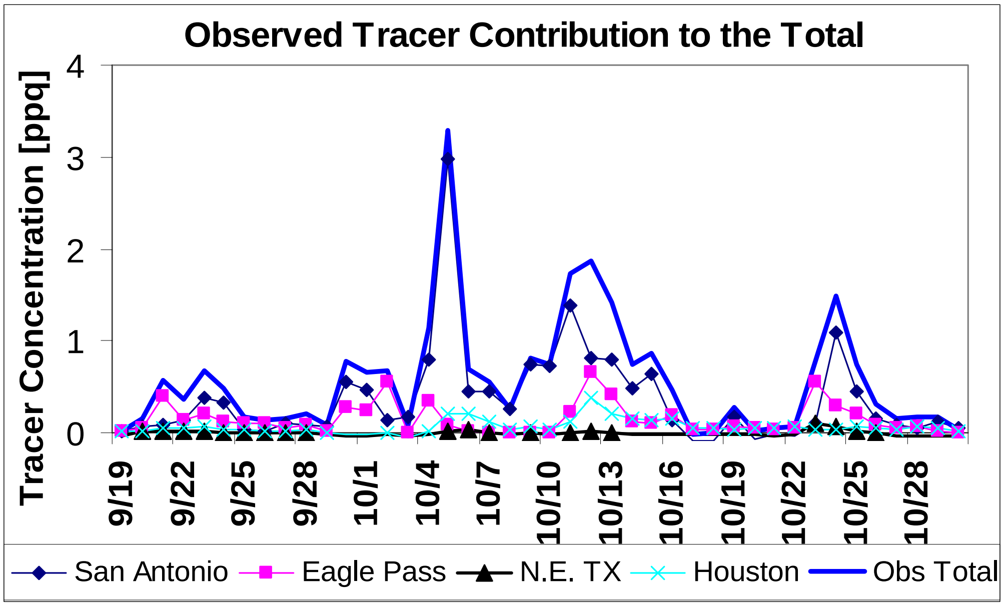

Figure 7-6. The combined tracer time series at Big Bend (K-Bar) monitoring site and the contribution of each tracer to the total.

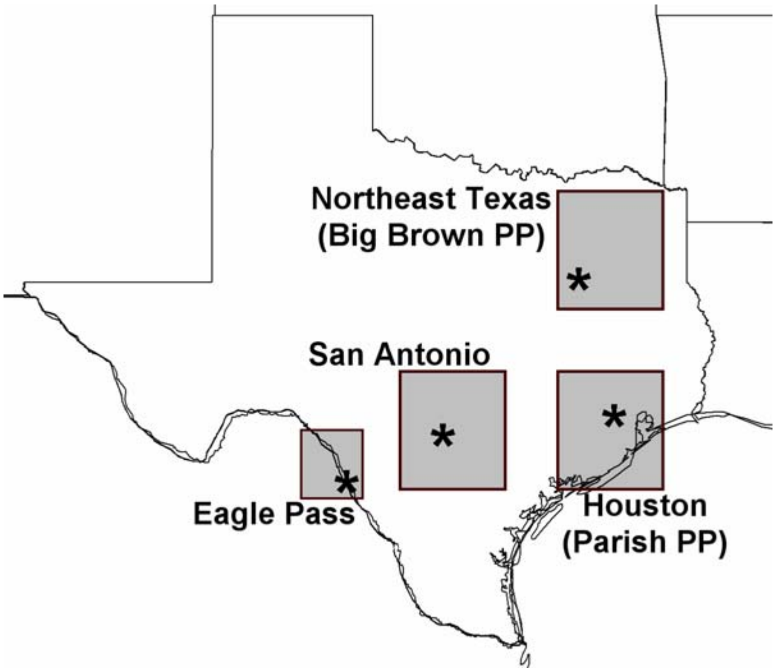

Figure 7-7. The four tracer source regions used in the FMBR analysis.

The results of the FMBR are presented in Table 7-3 and Figure 7-8. As shown, on average the retrieved source attributions properly identify the San Antonio tracer as the largest contributor and northeast Texas as the smallest. In addition, the average contributions are within the observed tracer contributions and the standard error for Eagle Pass, Houston, and northeast Texas. The retrieved San Antonio tracer plus the standard error is about equal to the observed contribution.

Table 7-3. Average observed and retrieved tracer emission rates and their contribution to the combined Big Bend trace concentrations.

Tracer Release Site	**Tracer Emission Rates [kg/day]**		**% source Attribution**	
	Actual	**Retrieved**	**Observed**	**Retrieved**
Eagle Pass	3.7	1.3 ± 2.2	26	28 ± 8
N.E. Texas	1.8	1.3 ± 3.5	-0.6	5 ± 9
San Antonio	9.3	9 ± 2.6	63	51 ± 11
Houston	2.3	4.9 ± 3	12	17 ± 7
Total	17	16.4 ± 5.7		

Performance Statistics

r^2	0.72
Mean Obs (ppq)	0.62
Mean Pred (ppq)	0.61
RMS Error (%)	54

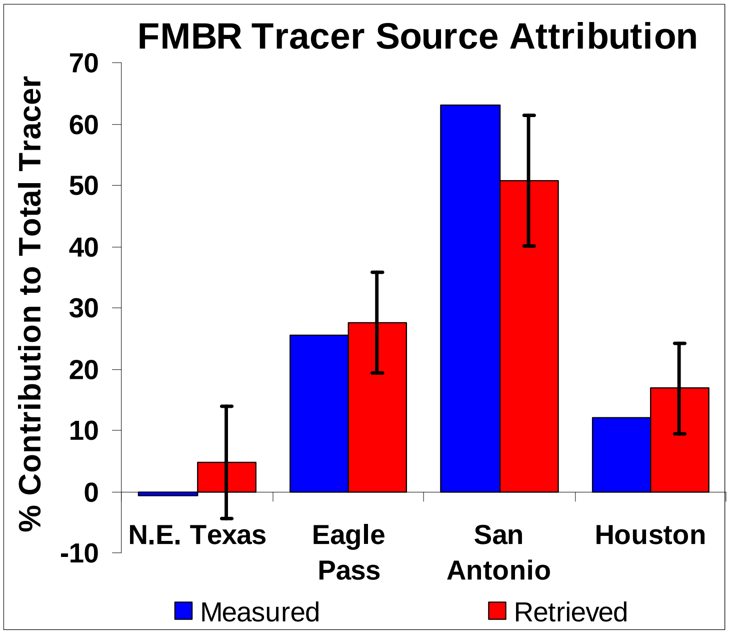

Figure 7-8. The tracer average source attribution and standard errors.

The kinetic probabilities for tracers are 1 and release rates were near constant. Therefore the predicted tracer concentrations and daily source attributions are valid. As shown in Figure 7-9 and Table 7-3, the predicted concentrations reproduce the observed total tracer concentrations well with an $r^2 = 0.72$ and no bias. Similar to the measured results, the retrieved San Antonio tracer is the largest contributor on a daily basis; however, from October 4–16 the San Antonio contribution is smaller than the average and is responsible for the retrieved San Antonio average contribution to be underestimated.

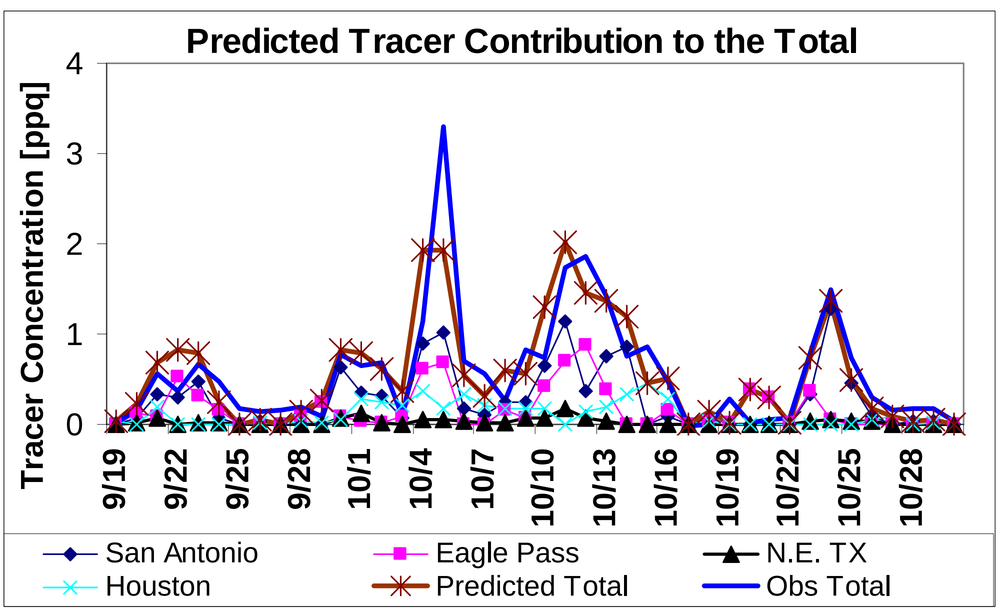

Figure 7-9. The observed and predicted total tracer and the predicted source attribution of each tracer to the total.

7.1.2.4 Discussion of FMBR Tests

These results illustrate that, provided enough data and sufficiently accurate transport model, FMBR can properly identify the tracer release sites and rates with no *a priori* information on the tracer release site locations. However, using only the Big Bend data, the resolution of the results diminish and it identifies the common transport pathways associated with the highest tracer concentrations at the Big Bend monitoring site. For the Eagle Pass tracer site, which frequently impacted the Big Bend monitor with concentrations several times the background concentrations, the FMBR was able to isolate the tracer release site from within the transport pathway. However, for the northeast Texas tracer, which was farther away, with Big Bend tracer concentration and usually near the background concentration FMBR could not isolate the tracer release site.

When the tracer release sites were added to the FMBR, the technique was able to decompose the integrated tracer time series into its contributions from the four individual tracers. The decomposition worked better for an on-average than on a day-by-day basis. Also, the average tracer release rate for each tracer was properly retrieved. Consequently, the FMBR is a valid technique for retrieving average source contributions of tracers. When using data from only Big Bend it is necessary to predefine a small set of source regions.

7.2 Source Apportionment of REMSAD Base Case Sulfate using Receptor Models

The previous section validated the TrMB and FMBR source attribution technique using inert tracer concentrations. However, sulfate source attribution has the complicating factor that the emitted sulfur undergoes transformation and removal processes during transport from the source to the receptor. To validate this technique for a reactive species, these analyses were applied to the REMSAD-predicted sulfate at Big Bend and compared to the REMSAD source attribution results. In addition to TrMB and FMBR techniques, synthesized REMSAD was applied to these artificial data.

7.2.1 Trajectory Mass Balance

7.2.1.1 Results

There are known concentrations of the 24-hour average sulfate predicted at K-Bar by the REMSAD model and known attributions of this simulated sulfate which are the REMSAD attributions of the predicted sulfate to the BRAVO source areas. For purposes of this test, there is no assertion that the REMSAD-simulated sulfate source attributions are accurate in the real world, only in the simulated REMSAD world. Only the MM5 meteorological data are used because this is what was used by REMSAD.

Table 7-4 summarizes the attributions of REMSAD sulfate to each of the four large BRAVO source regions. The attributions to the large areas were generated by aggregating the attributions of the 27 smaller source areas described in section 2.3.2.5. The top line of the table gives the REMSAD attributions both with and without the boundary conditions and non-linear fractions being redistributed proportionally. Values within 10 percentage points of the correct answer (defined as the REMSAD attributions with the redistributed boundary conditions and non-linearity) are shown in bold and in a larger font for easy identification.

Table 7-4. Columns 2–5 are the percent attributions of predicted REMSAD sulfate by TrMB, all using MM5 winds. For comparison, the top row shows the corresponding REMSAD attributions. REMSAD has boundary conditions = 7% and non-linear = 2%. Values in parentheses are the REMSAD attributions if these are proportionally redistributed. TrMB attributions within 10 percentage points of the REMSAD attributions are shown in larger font bold type. The last three columns are statistics comparing the TrMB predicted concentrations to "observed" (REMSAD predicted) sulfate concentrations. All values are for July 6–October 28, 1999 (115 days).

Model	Texas	Mexico	Eastern U.S.	Western U.S.	R^2	Mean Over-pred. SO_4 (ng/m^3)	Mean % Over-pred.
REMSAD ("Correct")	16 (18)	23 (25)	42 (46)	9 (10)	1.000	0	0
CAPITA 5-day	**19**	**31**	**39**	**11**	.778	-23	-1%
CAPITA 7-day	**20**	**24**	**36**	**20**	.798	-12	-1%
CAPITA 10-day	**21**	**21**	**37**	21	.775	-13	-1%
HYSPLIT 5-day	43	**25**	16	**17**	.768	11	+1%
HYSPLIT 7-day	43	**23**	16	**18**	.820	-26	-1%
HYSPLIT 10-day	46	**21**	19	**13**	.801	-14	-1%
ATAD 5-day	**25**	**33**	**36**	**8**	.735	-24	-1%

The best reproduction of the REMSAD sulfate attributions was with the CAPITA Monte Carlo model using 5-day back trajectories, although the 7-day and 10-day trajectories were nearly as good. The ATAD model with MM5 input also attributed the sulfate correctly to all four source regions within 10 percentage points of the correct values. The HYSPLIT model with MM5 input was able to reproduce the correct source attributions for Mexico and the western U.S., but was unable to correctly apportion sulfate from Texas and the eastern U.S., attributing much more to Texas and much less to the eastern U.S. than REMSAD.

Use of 5-, 7-, or 10-day trajectory lengths from the CAPITA Monte Carlo model made little difference in the predicted attributions except for the western U.S., which is predicted to be the source of twice as much sulfate with 7- or 10-day trajectories as with 5-day lengths. This is intuitively reasonable since airmasses rarely arrived at Big Bend directly from the western U.S. but more often traversed across the eastern U.S. and/or Texas prior to arrival. Thus, on average, the travel time from the western U.S. was longer than from the other large source areas and so longer trajectories attributed more to this region.

Why is HYSPLIT with MM5 input not accurately reproducing the REMSAD sulfate attributions when it uses essentially the same input meteorological data as CAPITA MC and ATAD? Figures 7-10 and 7-11 show time lines of the observed and predicted REMSAD sulfate and the number of back trajectory endpoints in each of the four large source areas. Figure 7-10 is for 5-day back trajectories using the CAPITA MC while Figure 7-11 is the same for the HYSPLIT model. A large difference between CAPITA MC and HYSPLIT occurs on September 1 (day 244). This is also the day of the highest measured sulfate concentration at Big Bend and so an influential point in the multiple linear regression. Notice that while the CAPITA MC model has most of the endpoints in the eastern U.S. on this day, HYSPLIT puts most of them in Texas. ATAD, which is not shown, also has most of the endpoints in the eastern U.S. on this day.

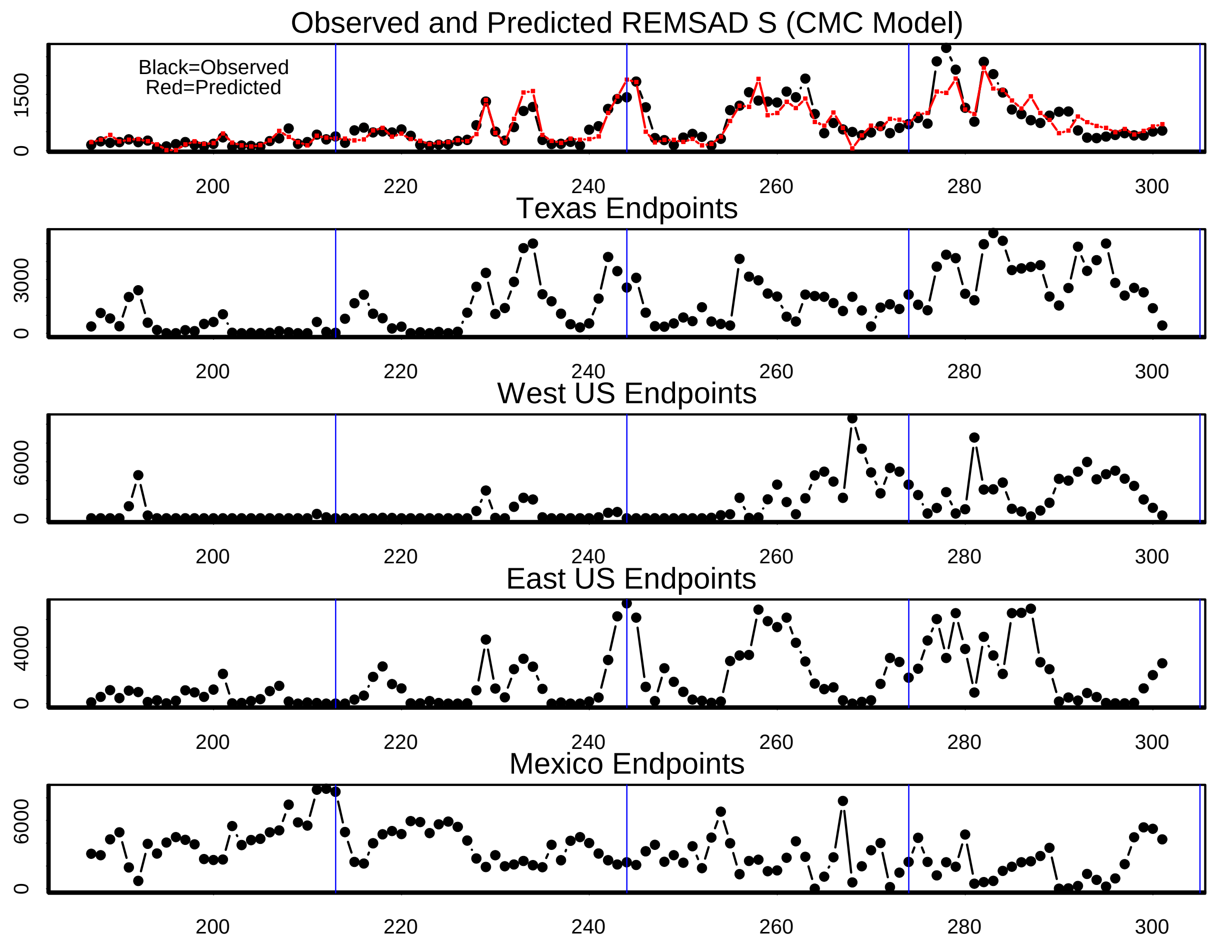

Figure 7-10. Time lines of the observed (black) and predicted (red) REMSAD sulfate in ng/m^3 (top) and number of back trajectory endpoints in each of the source areas using the CAPITA MC model and 5-day back trajectories. The blue vertical lines are the first days of August, September, October, and November.

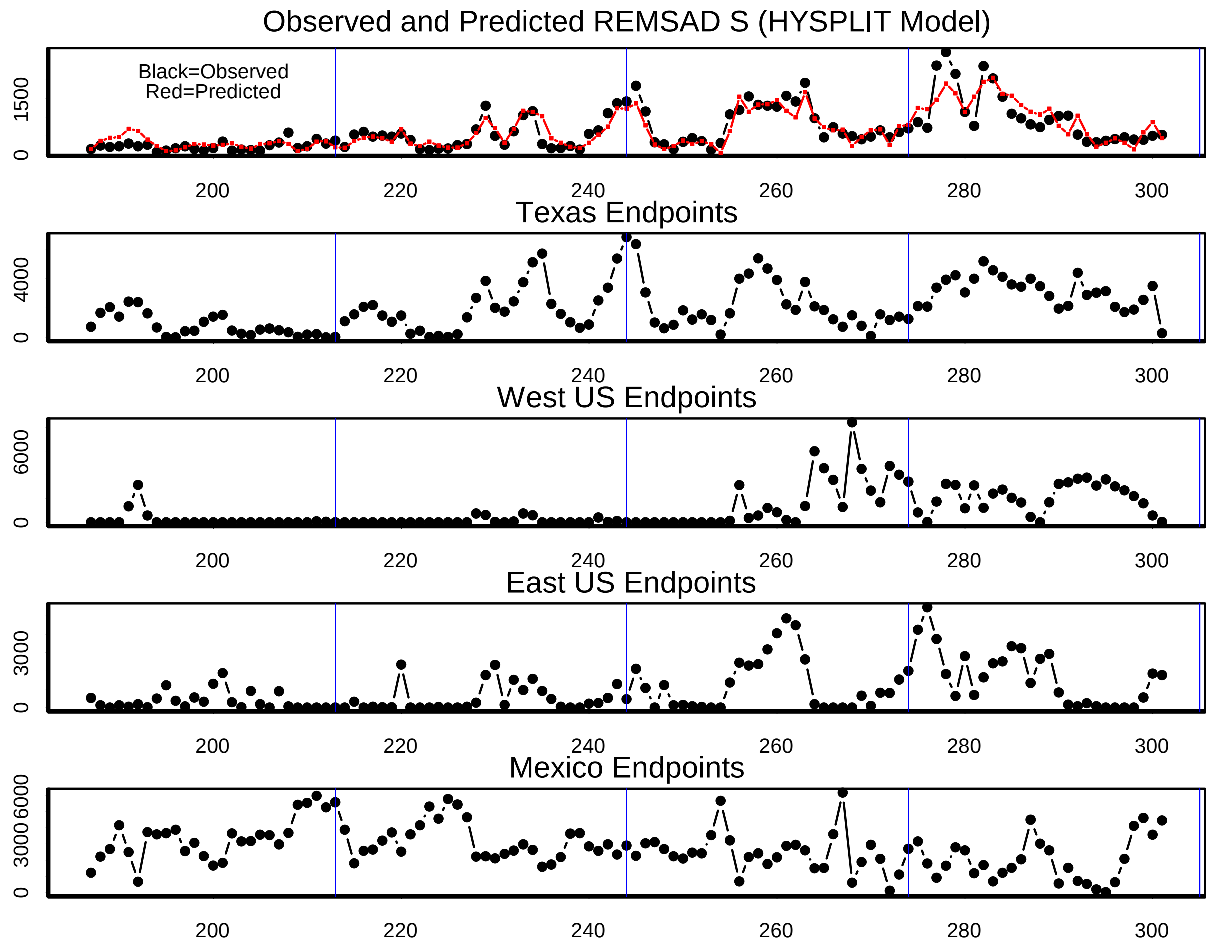

Figure 7-11. Time lines of the observed (black) and predicted (red) REMSAD sulfate (top) and number of back trajectory endpoints in each of the source areas using the HYSPLIT model and 5-day back trajectories. The blue vertical lines are the first days of August, September, October, and November.

Figure 7-12 shows the back trajectories generated by each of the three models for September 1. While they all have the same general direction, the HYSPLIT trajectories are much lower in height and in fact are essentially on the ground. This is true even for trajectories with a start height of 1000 m. Due to the lower height they also have much lower wind speeds and so remain in Texas while the CAPITA MC and ATAD trajectories extend into the eastern U.S. Because this was an influential day, the differences in trajectory heights on this day alone may explain why HYSPLIT was unable to reproduce the REMSAD source attributions.

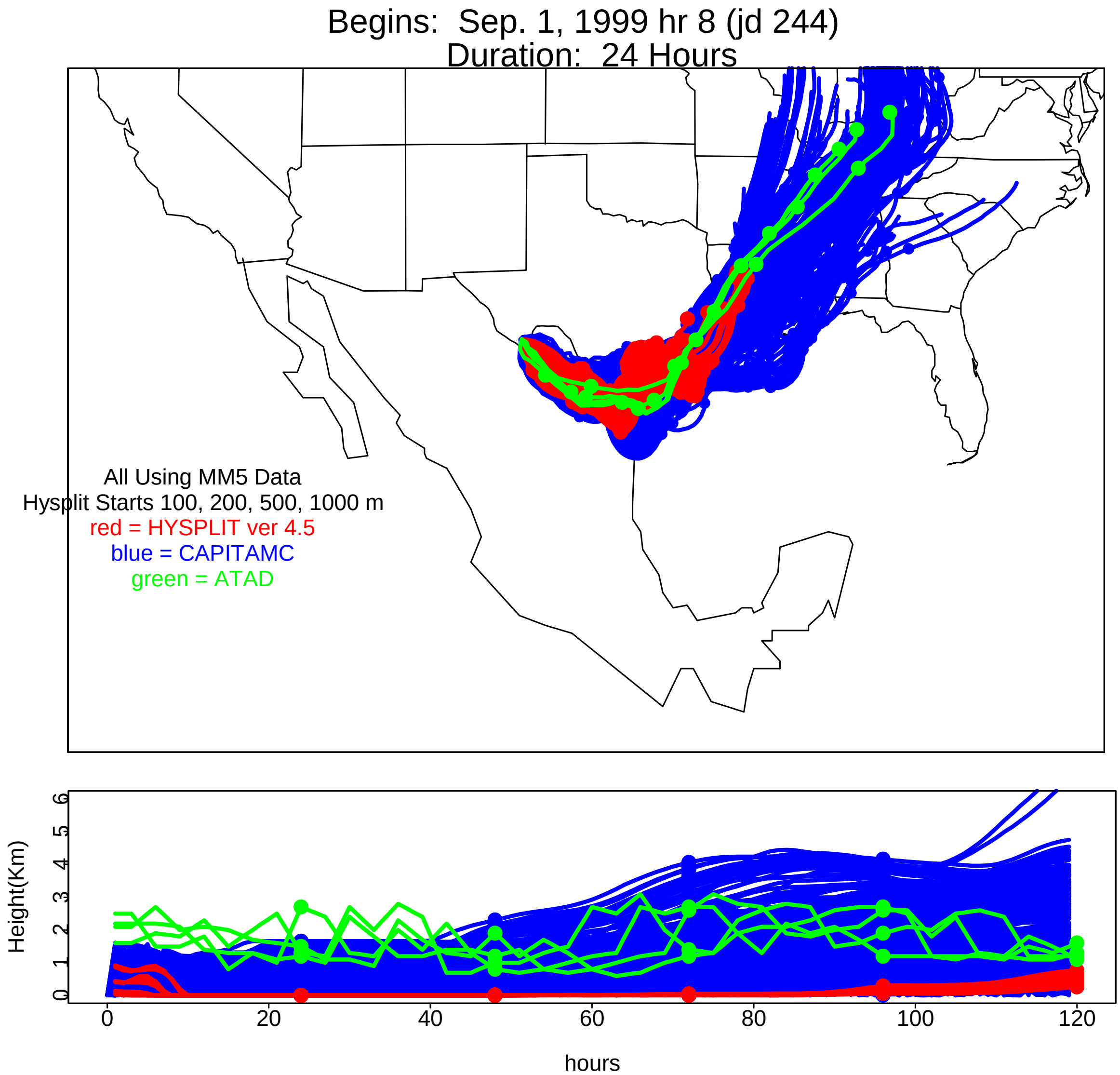

Figure 7-12. 5-day duration back trajectories arriving at Big Bend during the 24-hours beginning 9/1/99 at 8:00 a.m. Blue are CAPITA MC trajectories (20 every 2 hours), red are HYSPLIT trajectories (1 per hour for each of 4 start heights) and green are ATAD trajectories (4 per day).

In the case of the simulated REMSAD sulfate where we know the concentration of sulfate attributed to each source area, it's possible to examine the linearity of the relationship between endpoints and sulfate concentrations. Figures 7-13 and 7-14 show scatter plots of the daily number of endpoints in each of the four large areas vs. the sulfate attributed to the area for each day by REMSAD. The endpoints in Figures 7-13 and 7-14 were generated using the CAPITA MC model and the HYSPLIT model, respectively. The solid blue lines shown in each graph are the mean ratio of REMSAD sulfate from each region to the number of endpoints in the region. This is the "correct" relationship that the TrMB model would have to re-create in order to give the same attributions as REMSAD. Similarly, the dashed red line is a representation of the TrMB predicted relationship. If only these four source areas had been used in the regression, the slope of the dashed red line would be the regression coefficient. However, since each of the large areas is a composite of several smaller areas, the dashed red line was estimated by dividing the mean TrMB predicted concentration for the composite large area by the mean number of endpoints in that area. The scatter about the solid blue lines gives some idea of the linearity of the true relationship between endpoints and concentrations and how far the attribution for an individual day could deviate from the mean attribution. The angle between the dashed red and solid blue lines shows how well the TrMB model was able to reproduce the mean REMSAD attribution of sulfate.

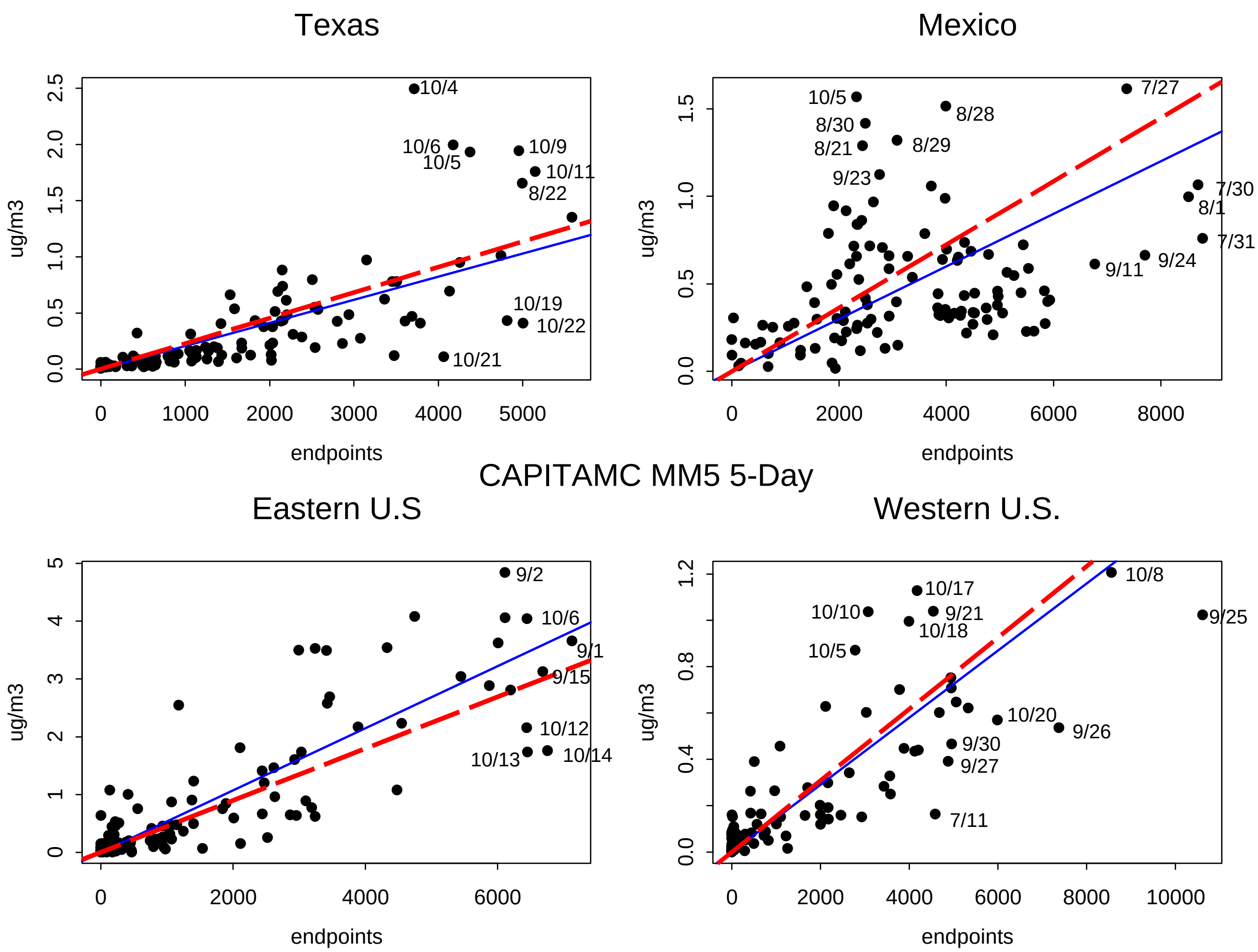

Figure 7-13. Scatter plots of REMSAD sulfate attributions vs. number of endpoints for each of four large source areas using 5-day back trajectories from the CAPITA MC model and MM5 winds. The solid blue lines are the "true" slopes; the dashed red lines are the TrMB modeled slopes.

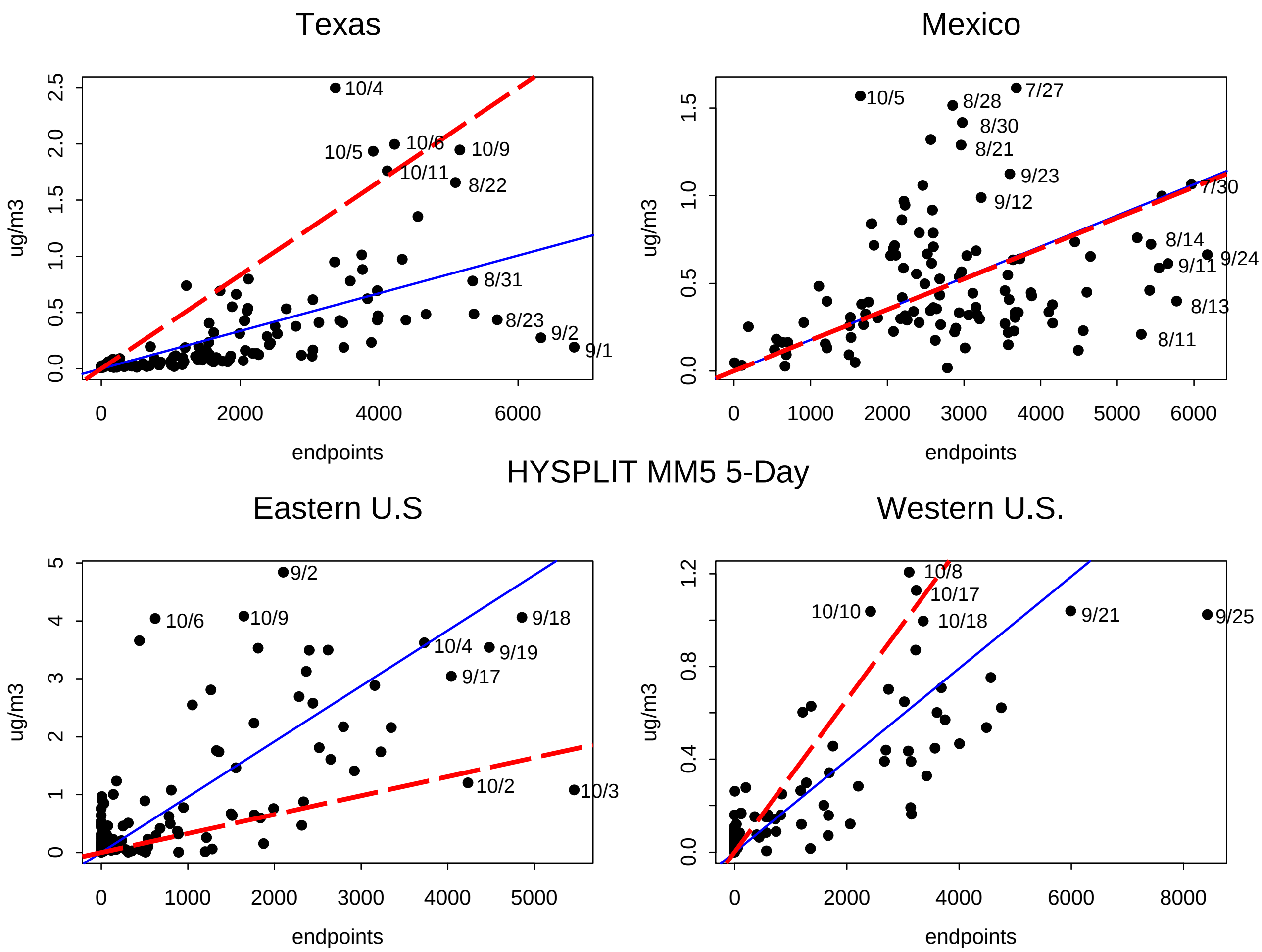

Figure 7-14. Scatter plots of REMSAD sulfate attributions vs. number of endpoints for each of four large source areas using 5-day back trajectories from the HYSPLIT model and MM5 winds. The solid blue lines are the "true" slopes; the dashed red lines are the TrMB modeled slopes.

When using endpoints generated by the CAPITA MC model, the assumption of linearity seems to be fairly good, though certainly some large deviations from the mean relationship exist. For example, for the Texas source area in Figure 7-13, all except a few days in October when REMSAD gives large attributions to Texas seem to fall close to the lines. These days probably had unusual meteorological or chemical characteristics that caused the relationship between sulfate at Big Bend and transport from areas within Texas to be different than average. In the case of October 4, it appears that the sulfate attribution to Texas on this individual day could be underestimated by TrMB by nearly a factor of 5, even though on average the mean attribution to Texas was within one percentage point of being correct. So, as expected, the errors in the attributions predicted by the TrMB model are larger for individual days than for the mean.

7.2.1.2 Discussion

Both the CAPITA MC and ATAD models, when run with the MM5 wind fields used by REMSAD, can reproduce the REMSAD sulfate attributions for four large source areas to within ten percentage points for all areas. HYSPLIT can similarly reproduce the attributions to Mexico and the western U.S., but is significantly overestimating the attribution to Texas and underestimating the eastern U.S. This may be due to trajectory heights that are too low on a single highly influential day, thus giving too much influence to Texas and not enough to the eastern U.S. Similarly, HYSPLIT with MM5 input also failed the tracer attribution test. Use of EDAS and FNL wind fields could not be tested by this simulated sulfur attribution test since REMSAD was run only with the MM5 winds as input and these are the correct winds for this simulated world.

Examination of the relationship between sulfate attributions and endpoints indicates that caution should be exercised when using TrMB to examine source attributions on individual days. The model is, in general, much more accurate for the four-month average. Even if the mean source attributions were reproduced with total accuracy, the degree of accuracy of the model on any one individual day depends on how much the meteorological and chemical characteristics on that day deviated from the mean. In general, this will not be known.

7.2.2 Forward Mass Balance Regressions

The transit probabilities were generated for each source region using the plumes generated from the CAPITA Monte Carlo model driven by the MM5 wind fields (Figure 7-3). All plumes originating from virtual sources within a REMSAD source region were aggregated together. Transit probabilities from the two eastern U.S. source regions, LA/MISS & MO/IL/AR, were highly collinear so they were combined into one source region. Boundary conditions were not included in the FMBR since the simulation of the transport from the REMSAD boundary was not conducted, but they were incorporated in the percent source contributions so the FMBR source attributions could be compared to the REMSAD source attributions. The analysis was conducted using different plumes lengths of 5, 7, and 10 days to test the sensitivity of the FMBR on plume length. This is important since if the plumes are too short then contributions from distant source regions will not be represented, and if the plumes are too long then impacts from “old” particles will be too large due to the single average kinetic factors that are derived as part of the source/sink term.

Results are presented in Table 7-5 for the three different lengths of the simulated source plumes. As shown in the FMBR performance statistic, the predicted sulfate fits the REMSAD sulfate well, with $r^2 \sim 0.8$, an error of 40%, and bias of 4%. Consequently, the transport from 10 source regions, some 1000’s of km^2 in size, multiplied by a constant explains 80% of the variance in the REMSAD sulfate daily time series.

The FMBR performance statistics are independent of the plume lengths. However, as the plume lengths increased, the number of source regions with contributions within one standard error of the REMSAD results increased from 4 of 9 for the 5-day plumes and to 8 of 9 using the 10-day plumes.

Using the 10-day plumes, all but the northeast Texas source region is within the standard error of the REMSAD results. Aggregating over the large source regions, the western U.S., the eastern U.S., Texas, and Mexico, the Texas source contribution is overestimated by about 9 percentage points and the western U.S. contribution is zero or underestimated by about 9 percentage points. Eastern U.S. and Mexican source regions properly reproduce the REMSAD result. Therefore it appears that the FMBR technique is unable to resolve the western U.S. source contributions and erroneously attributes them to the Texas source region.

Table 7-5. Source attribution results from the application of the FMBR technique to the REMSAD predicted sulfate concentrations. The REMSAD source attribution results are also provided. Bolded source attributions are within 1 standard error of the REMSAD results. Propagation of errors was used to estimate standard errors for aggregated regions.

	% Contribution ± Standard Error			
Source Regions	**5 day**	**7 day**	**10 day**	**REMSAD Model***
Carbón	**16.1 ± 4**	**16.5 ± 4**	**15.4 ± 4**	14.1
Rest of Mexico	**13.6 ± 10**	**12.2 ± 10**	**11.6 ± 11**	10.0
NE Texas	**7.8 ± 3**	9.5 ± 3	10.2 ± 3	5.4
SE Texas	14. ± 4	**11.9 ± 4**	**11.8 ± 5**	8.8
Rest of Texas	**5. ± 4**	**4. ± 5**	**3.7 ± 5**	2.3
LA/MISS & MO/IL/AR	33.8 ± 6	**28.7 ± 8**	**27.6 ± 8**	22.3
E Central	0.6 ± 2	7.4 ± 4	**8.8 ± 5**	13.6
Rest of E U.S.	2.1 ± 3	2.7 ± 4	**4. ± 5**	7.4
W U.S.	0. ± 8	0. ± 9	**0. ± 9**	9.3
GOCART BC	7	7	7	7.2
Mexico (all)	**29.6 ± 10**	**28.8 ± 11**	**27. ± 12**	24.0
Texas (all)	26.8 ± 6	25.4 ± 7	25.7 ± 7	16.5
Eastern U.S. (all)	36.5 ± 7	**38.8 ± 10**	**40.4 ± 11**	43.2
Western U.S. (all)	0. ± 8	0. ± 9	**0. ± 9**	9.3
GOCART BC	7	7	7	7.2

FMBR Performance Statistics				
Base Case Avg (μg/m^3)	1.81	1.81	1.81	
Predicted Avg (μg/m^3)	1.73	1.72	1.73	
Bias (%)	-4.4	-4.5	-4.1	
RMS Error (%)	42	41	41	
r^2:	0.79	0.80	0.80	
# Data Values	115	115	115	

***REMSAD source attributions were scaled by 2.7% to account for missing mass due to nonlinearities in the model simulations.**

To investigate the Texas and western U.S. bias, the FMBR technique was used with each source region in the western U.S. fixed at a constant *a priori* source/sink term so their total contribution to Big Bend's sulfate was about 9%. The results are presented in Table 7-6. Fixing the western U.S. contribution to 9% had little impact on the regression performance statistics, with r^2 dropping from 0.8 to 0.74 and the bias improving somewhat from -4% to -3%. The western U.S. was divided into two source regions, the central plains north of Texas and west of Texas. The central plains source contribution accounted for 3.8 % of the 9% western contribution. The increases in the contributions from western U.S. source regions were primarily compensated for by decreases in nearby source regions, with the largest changes occurring for Texas where the source contribution decreased from 25.7 to 21.2%. The airmasses from the western U.S. source regions traversed these nearby source regions en route to Big Bend. Therefore, for source regions with collinear transport, FMBR tended to increase the source attributions of the closer source regions at the expense of the more distant source regions.

Table 7-6. Source attribution results from the application of the FMBR technique to the REMSAD predicted sulfate concentrations. The REMSAD source attribution results are also provided. The FMBR used the 10-day plumes and was run under three different conditions defined below. Propagation of errors was used to estimate standard errors for aggregated regions.

	% Contribution ± Standard Error				% contribution Bias
Source Regions	**Run 1**	**Run 2**	**Run 3**	**REMSAD Model***	**Run3/REMSAD**
Carbón	15.4 ± 4	15.3 ± 4	16.7 ± 4	14.1	1.19
Rest of Mexico	11.6 ± 11	9.4 ± 11	13.8 ± 11	10.0	1.39
NE Texas	10.2 ± 3	9. ± 3	10.6 ± 3	5.4	1.96
SE Texas	11.8 ± 5	12.1 ± 5	12.9 ± 5	8.8	1.46
Rest of Texas	3.7 ± 5	0. ± 5	4.4 ± 5	2.3	1.93
LA/MISS & MO/IL/AR	27.6 ± 8	27.5 ± 8	27.9 ± 8	22.3	1.25
E Central	8.8 ± 5	8.4 ± 5	8.8 ± 5	13.6	0.65
Rest of E U.S.	3.9 ± 5	2. ± 5	4.9 ± 5	7.4	0.66
Western U.S.				9.3	
Central Plains	0. ± 3	3.8 ± 3	0. ± 3		
West of TX	0. ± 9	5.3 ± 9	0. ± 9		
GOCART BC	7	7		7.2	
Mexico (all)	27. ± 12	24.7 ± 12	30.5 ± 12	24.0	1.27
Texas (all)	25.7 ± 7	21.2 ± 7	27.9 ± 7	16.5	1.69
Eastern U.S. (all)	40.3 ± 11	38. ± 11	41.6 ± 11	43.2	0.96
Western U.S. (all)	0. ± 9	9.2 ± 9	0. ± 9	9.3	0.00
GOCART BC	7	7		7.2	

FMBR Performance Statistics			
	Run 1	**Run 2**	**Run 3**
Base Case Avg (μg/m^3)	1.81	1.81	1.94
Predicted Avg (μg/m^3)	1.73	1.75	1.85
Bias (%)	-4.1	-3	-4.7
RMS Error (%)	41	47	40
r^2:	0.8	0.74	0.79
# Data Values	115	115	115

***REMSAD source attributions were scaled by 2.7% to account for missing mass due to nonlinearities in the model simulations.**

Run 1: REMSAD sulfate minus the contribution from the boundary conditions, same run as in Table 7-5.
Run 2: REMSAD sulfate minus the contribution from the boundary conditions and the western U.S. and central plains source contributions fixed to a total contribution of 9%.
Run 3: Total REMSAD sulfate (includes the contribution from the boundary conditions).

This bias can also be seen by determining in which source regions the contributions from the REMSAD boundary conditions are placed. Ideally, this mass would be attributed to source regions at the edge of the domain. The FMBR technique was applied to the total REMSAD sulfate concentration; that is, the contribution from the boundary conditions were included. These results are presented in Table 7-6 as Run 3. The boundary condition accounted for about 7% of the total REMSAD simulated sulfate at Big Bend. As shown, more than 80% of this mass was assigned to the closer-by Texas and Mexico (Carbón and Monterey) source regions, while the distant source regions in the western U.S., Mexico, and eastern U.S. had little change. This is consistent with the previous finding that the FMBR technique tended to overestimate the contribution from the closer source regions at the expense of more distant source regions with collinear transport.

The magnitude of these systematic biases was estimated by taking the ratio of the relative attribution of the FMBR in run 3 and the actual relative REMSAD source attribution results and are presented in Table 7-6. By taking the ratio of the relative contributions as opposed to absolute concentrations, the factors account for the differences in the distribution of the contributions from the various source regions as opposed to the absolute values of these contributions. As shown, Texas and Mexican sulfate contributions were biased high by 27% and 69%, respectively. The western U.S. was biased low by 100% and the eastern U.S. was biased low by only 4%.

In conclusion, the FMBR technique was successfully validated against the synthetic sulfate data generated by REMSAD and the 10-day plumes produced the best results. The contributions from the eastern U.S., Mexican, and Carbón source regions could be independently resolved. However, it appears that nearly all of the contributions from the western U.S. and boundary conditions were attributed to Texas and Mexican sources. Based upon these results, FMBR is applied to the measured fine particle sulfur at Big Bend National Park (see chapter 8) using the 10-day plumes. When interpreting those results, one needs to be mindful of the potential biases and relationship between the predicted western U.S. and boundary conditions and the Texas and Mexican source attributions.

7.2.3 Synthesized REMSAD

The apportionment of Big Bend's sulfate was also conducted using a synthesis inversion technique. In this method the REMSAD or CMAQ-MADRID source apportionment results are regressed against the observed sulfate concentrations throughout Texas. The same simulated attribution results are then scaled by the regression coefficients to derive alternative source attribution estimates that, when summed together, better fit the measured data. This approach compensates for some of the systematic errors present in the two simulations of the air quality models and thus should provide more accurate attributions of Big Bend aerosol.

The synthesis inversion method is based upon the inversion of the conservation of mass equation and is fully described in section 2.3.2.3.1. However, in brief, it can be shown that a discrete form of the conservation of mass equation can be derived from the Green function solution of a linear differential conservation of mass equation:

$$c_i = \sum_j G_{ij} s_j + \varepsilon_i = m_i + \varepsilon_i \qquad (2\text{-}33)$$

where c_i are the observed concentration values for each receptor site/time pair i, m_i are the modeled concentration values, ε_i are the errors in c_i,, s_j are the source attribution scaling coefficients for each source time pair $_j$, and G_{ij} are the absolute source contributions of source/time pair j to observation i. In this application, c_i are the observed fine particle sulfur data scaled to sulfate and G_{ij} are calculated from the REMSAD model. Equation 2-33 is solved for s_j using constrained linear regression. If REMSAD is a perfect model, in the least square sense, then the source attribution scaling coefficients (s_j) will be equal to 1.

In order to test the synthesis inversion technique, it was applied to the synthetic REMSAD sulfate source apportionment results. To account for measurement error, a 10% normally distributed random error was added to all predicted observations:

$$(m_i + \varepsilon_i) = \sum_j G_{ij} s_j \tag{7-1}$$

where $(m_i + \varepsilon_i)$ is the model prediction plus the 10% error. A valid model would result in regression coefficients of $s_j = 1$. Coefficients other than one would be due to highly collinear source attribution estimates between source regions and the regression could not resolve the source region contribution due to the error ε_i and non-linearities in the REMSAD model. Also, a source region's contribution may be smaller than the error in the system and the regression technique would be unable to properly resolve this source's contribution to the receptors.

The inversion process was conducted using a number of different configurations including using only the predicted Big Bend sulfate data for all four months, the predicted sulfate data from all BRAVO monitoring sites for all four months, and the predicted sulfate data from all BRAVO monitoring sites for different five-day periods. In most cases the source attribution scaling coefficients were within 15% of 1. For example, Table 7-7 presents the results from two analyses, one using the predicted daily sulfate concentrations from only the Big Bend monitoring site and the other using data from all BRAVO monitoring sites and times. As shown, when only the Big Bend data are used, the scaling coefficients are within 10% of 1, except for northeast Texas and then the r^2 is essentially one. When the predicted sulfate from all BRAVO monitoring sites are used, the regression coefficients are generally within 5% of 1 with most standard errors less than 1%.

Three conclusions can be drawn from this analysis. First, the technique is valid, provided sufficiently accurate data and model are used. Second, there is sufficient variability between the source regions with large contributions to Big Bend and Texas that the system can adequately resolve them. Last, the non-linearities in the REMSAD chemistry module are sufficiently small that the source regions' contributions from each other can be resolved with this linear technique. This testing does not account for errors in the REMSAD model itself. As shown in chapter 6, the REMSAD modeling error is on the order of a factor of 2. This is much larger than the 10% error added to the predicted sulfate and the error introduced by the non-linearities in REMSAD's chemistry module. Therefore this does not conclusively validate the system for application to observed data.

Table 7-7. The source attribution scaling coefficients and performance statistics resulting from regressing the REMSAD source attribution results for all modeled source regions against the REMSAD-predicted sulfate concentrations at Big Bend and at all BRAVO monitoring sites for the entire BRAVO time period. The predicted sulfate concentrations had a 10% normally distributed error added to them.

	Scaling Coefficients (s_i)	
Source Regions	**Big Bend Data**	**All Data**
Carbon	**1 ± 0.01**	**1.01 ± 0.002**
Rest of Mexico	**1 ± 0.01**	**1 ± 0.003**
NE Texas	**1.13 ± 0.02**	**1.02 ± 0.001**
SE Texas	**0.95 ± 0.02**	**1 ± 0.001**
Rest of Texas	**1.08 ± 0.04**	**1.06 ± 0.005**
LA/MISS	**1.07 ± 0.02**	**1.01 ± 0.001**
E Central	**0.99 ± 0.01**	**1 ± 0.001**
MO/IL/AR	**0.97 ± 0.01**	**1 ± 0.001**
Rest of E US	**1.03 ± 0.02**	**1.01 ± 0.01**
W US	**1.03 ± 0.02**	**1.05 ± 0.02**
GOCART BC	**0.94 ± 0.03**	**0.95 ± 0.03**

Regression Performance Statistics

	Big Bend Data	**All Data**
r^2	**0.9999**	**0.9999**
Basecase Avg ($\mu g/m^3$)	**1.97**	**3.3**
Predicted Avg ($\mu g/m^3$)	**1.97**	**3.3**
RMS error (%)	**2.4**	**3**
# Data Values	**112**	**3628**

7.3 Discussion and Conclusions of Test Results

Several tests of the abilities of trajectory-based receptor techniques to reproduce two known source attributions were conducted. The purposes of these tests were to 1) determine the accuracy of the techniques themselves, 2) to determine which wind field MM5 or EDAS/FNL is better for use in further source attribution of sulfate during BRAVO and 3) to determine the optimum trajectory length.

7.3.1 Model Accuracy

On the first point the tests were successful and demonstrated that both the TrMB and FRMB techniques could reproduce the known mean attributions of both the total tracer concentrations and the REMSAD-simulated sulfate concentrations within the standard errors of the method. There are caveats to this conclusion for both models. TrMB was successful only with back trajectories from some trajectory model and input wind field combinations. Since those sets of combinations that worked well were consistent for both tests, there is confidence in using this subset of back trajectories for attribution of measured sulfate. FMBR can identify the tracer release sites and rates. However, when using only one receptor site, the resolution of the results diminish and it identifies only the common transport pathways associated with the highest tracer concentrations. FMBR was also able to isolate the tracer release sites when the concentrations were sufficiently high. When the release sites were identified, FMBR was able to decompose the integrated tracer into its contributions from the four individual sites and the mean release rate for each tracer was properly retrieved. As expected, both TrMB and FMBR give more accurate results for the four-month average source attribution than for individual days.

7.3.2 Wind Fields

On the second goal of determining which wind field is superior, these tests were not conclusive. Only the MM5 wind field could be tested with REMSAD-simulated sulfate concentrations so no insight regarding wind fields was gained by that test. For the attribution of the tracers, both wind fields did equally well in FMBR. For TrMB, MM5 input resulted in accurate tracer attributions when trajectories were generated by the CAPITA MC or ATAD model, but EDAS/FNL input was superior in the HYSPLIT model.

7.3.3 Trajectory Lengths

Trajectory lengths of 5, 7, and 10 days all performed similarly and within the uncertainties of the regressions in TrMB for the tracer. Insensitivity to trajectory length is not surprising for the tracers, since mean transport time to Big Bend for all of the tracer release sites is probably less than 5 days on average. Thus, longer trajectories do not change the relative fractions of endpoints in the source regions and so do not change the results of the regressions. For REMSAD sulfate, the western U.S. was better predicted by TrMB when using the CMC/MM5 5-day trajectories, while 7- and 10-day lengths over-predicted the contribution from this region. FMBR with REMSAD sulfate was able to reproduce the REMSAD sulfate attributions for all nine tested source regions within the standard error of the regression coefficients only with 10-day trajectories, while FMBR with 5-day and 7-day lengths could reproduce the results for 4 and 8 of the regions, respectively.

A significant difference between the forward and backward techniques is particle or endpoint height. In the forward technique (FMBR), particles were released from source areas at heights of 1 km or less. However, in the reverse technique (TrMB),all endpoints over a source region, regardless of height, were included in the regression. Many of the endpoints over the western U.S. between 7 and 10 days prior to arrival at Big Bend were at relatively high heights, > 3 km, and thus less likely to be in contact with emissions released in this region. It is possible that had the TrMB tests using trajectories from the CAPITA MC model limited the modeled endpoints to those below the mixing height, longer trajectories would have performed better since the information would not have been contaminated by the noise of endpoints above the mixed layer. Other analyses (see chapter 4) have shown that HYSPLIT trajectories are on average much lower than those generated using the CAPITA MC model, and HYSPLIT trajectories generated with EDAS/FNL input, which performed well in the tracer test, could not be tested with the REMSAD sulfate.

These tests therefore give us indications that: 1) for the tracers, which are less than 5 days away, trajectory length is not important, and 2) for sulfate, longer trajectories are probably better able to accurately reproduce the attributions as long as the endpoint or particle heights are limited to those below the mixed layer.