ICFA Analysis of Los Angeles OC Data – June 2012

1 Introduction

I have run dozens of ICFA runs (each taking a few hours to run) and have found that for these data, the outputs are dependent on the rigidity of the bounds placed on each element of the profile matrix. The volatility for the source apportionment (SA) results based on these organic measurements seems to greatly outweigh the volatility seen in SA results obtained from PM2.5 data. For most of the runs, we use bounds of the following form. For "known" elements of the mobile profiles (which were also used in CMB), the bounds on the elements were estimate $\pm$ $S \times$ uncertainty. For the unknown elements of the 3 CMB-based profiles, the estimates for these elements were initially unbounded (see discussion of starting values below). For elements of the 4 PMF-based profiles (wood, biomass, SOA1, and SOA2), the bounds on the elements were $(1/M \times \text{PMFestimate}, M \times \text{PMFestimate})$. For these roughly $(1/M \times \text{PMFestimate}, M \times \text{PMFestimate})$. For the majority of ICFA runs, I used S between 2 and 5 and M between 1.5 and 1000, with $M \geq 20$ indicating that the PMF estimates are allowed to evolve as much as 20-fold. A few lessons learned:

- The inclusion of WSOC among the apportioned species is problematic, but probably necessary. When included, it often leaches from the expected sources (wood, biomass, SOA1, SOA2) into diesel or gas profiles in sizable quantities. A further complication is that the WSOC's move into the mobile profiles differs for different specified tightness/looseness of bounds. That is, for the same data, when bounds are adjusted, "excess" WSOC sometimes moves into the diesel profile and sometimes into the gas profile.

  However when WSOC is removed from the list of apportioned species, the pre-specified profiles for several of the sources become less well-differentiated. For example, the nature of the two SOA sources from PMF are difficult to distinguish. So, when ICFA is run without WSOC, one of the SOA sources takes on a dramatically different structural interpretation, one involving a large amount of EC. Thus, it seems best to include WSOC when apportioning these mostly OC-based species.

- Placing bounds that are too tight yields computational problems. When $S = 2$ and $M = 3$, we had computational instability, with bounds so tight that the estimates were unable to move away from a local maximum.

- Placing bounds that are too loose on the CMB-related profiles yielded estimated profiles that failed to retain the diesel/gas/smoker interpretation, thus eliminating the original purpose of the exercise: to identify components of mobile emissions in the presence of other major sources (wood, biomass, SOA1, and SOA2).

- WSOC apportionment seemed more reasonable when starting values were given (as opposed to leaving unspecified the WSOC elements of the diesel/gas/smoker profiles and leaving the possible estimates unbounded except for constraining nonnegativity). The other estimated elements of the d/g/s profiles that were not a part of the CMB-related profile estimates also behaved more reasonably when reasonable starting values were given. Consequently, I obtained estimates of WSOC and other elements that were not available in the CMB-related profiles by substituting in the corresponding elements from the PMF/Mobile profile, which acts as an average mobile (d/g/s) profile. For these initial estimates, I then used values of $10 \times M$ in place of M in the parameter boundary equation and allowed WSOC to be virtually unbounded in these profiles.

2 ICFA Estimates

2.1 Optimal Estimates: Using WSOC

Based on the above observations, the estimates that were chosen to be optimal were based on $S = 5$ and $M = 10$ when using all 80 elements (including WSOC). This set of bounds yielded profile estimates that generally agreed with the PMF profiles for wood, biogenic, SOA1, and SOA2 (see Figure 1. The roughly 25 elements of the d/g/s profiles from the CMB-based analysis also remained fairly stable, with the other 55 elements initialized by the PMF mobile profile changing fairly dramatically (as one might expect). The main source of instability was the WSOC abundance. The magnitude of the contribution of the d/g/s sources was heavily dependent on the WSOC element of the source profiles. But, as noted above, retaining WSOC in the set of species was central to the identification and interpretation of the sources.

For this specific analysis, the WSOC fraction in the smoker profile went to zero while the WSOC fraction in the diesel profile was quite large. In fact, the ICFA analysis (which uses WSOC) has diesel contributing much more (average of 0.80) than either gas (0.09) or smoker (0.04). (Figure 2 illustrates contributions for each of the 7 ICFA sources.) In contrast, the CMB analysis (which did not use WSOC) had d/g/s averages of 0.25, 0.36, 0.58, respectively.

It is interesting to note that despite differences in the d/g/s split between ICFA and CMB, the sum of d/g/s for ICFA, the sum of d/g/s for CMB, and the PMF combined mobile source are all very similar (see Figure 3).

2.2 Tightly-Bounded Estimates: Omitting WSOC

In order to obtain reasonably comparable estimates of source contributions *without* the inclusion of WSOC, we need to put quite restrictive bounds on the assumed profiles. For these analyses we use $S = 2$ and $M = 1.5$. Comparable to Figures 1 through 3, Figures 4 through 6 give illustrations of the profile, contributions, and mobile contributions for this analysis that excludes WSOC from the list of species and uses the tightly-bounded profiles.

2.3 Tightly-Bounded Estimates: Using WSOC

For comparison, the same constraints were fit as in the previous section ($S = 2$ and $M = 1.5$) but now including WSOC. Comparable to Figures 1 through 3 and Figures 4 through 6, Figures 7 through 9 give illustrations of the profile, contributions, and mobile contributions for this analysis when including WSOC from the list of species and using the tightly-bounded profiles.

2.4 Comparison of Three Analyses

Table 1 shows the estimated contribution of each source when using PMF, CMB, or ICFA (with various settings for S, M, and the inclusion of WSOC among modeled species. Note that although the total amount of mobile emissions is reasonably consistent, the diesel/gas/smoker breakdown changes quite dramatically as the nature of the profile constraints are subtly changed. While the CMB analysis indicates that the smokers are the largest part of the mobile contributions, the 3 ICFA analyses point to Diesel, Gas, and Diesel as the largest mobile contributors. Thus, ICFA can be used, but for these data, it appears that the data may be too sensitive to adequately identify the d/g/s split.

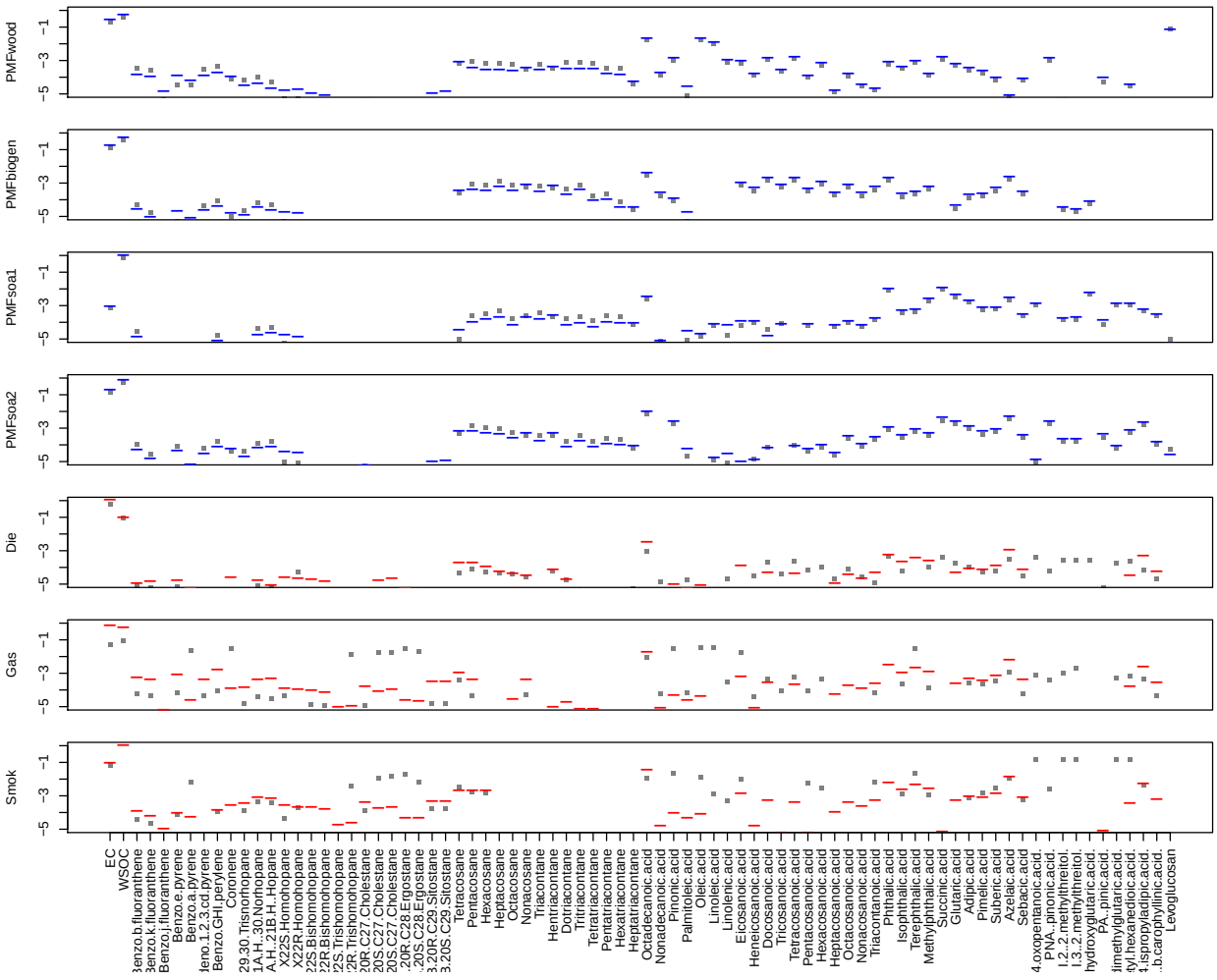

Figure 1: Profiles with $S = 5$ and $M = 10$.

Table 1: Comparison of estimated mean source contributions when using PMF, CMB, or ICFA. Numbers in parentheses are "total mobile" after summing diesel/gas/smokers.

			ICFA		
			$S = 5$, $M = 10$	$S = 2$, $M = 1.5$	$S = 2$, $M = 1.5$
	PMF	CMB	(inc. WSOC)	(no WSOC)	(inc. WSOC)
Mobile	1.11	(1.18)	(.93)	(.67)	(.97)
Diesel	-	.25	.80	.17	.67
Gas	-	.36	.09	.34	.03
Smoker	-	.58	.04	.16	.27
Wood	.38	.46	.66	.85	.66
Biogenic	.86	-	.04	1.14	.54
SOA1	.67	-	1.02	.43	.73
SOA2	.82	-	1.21	.60	.96
VegDet	-	.10	-	-	-

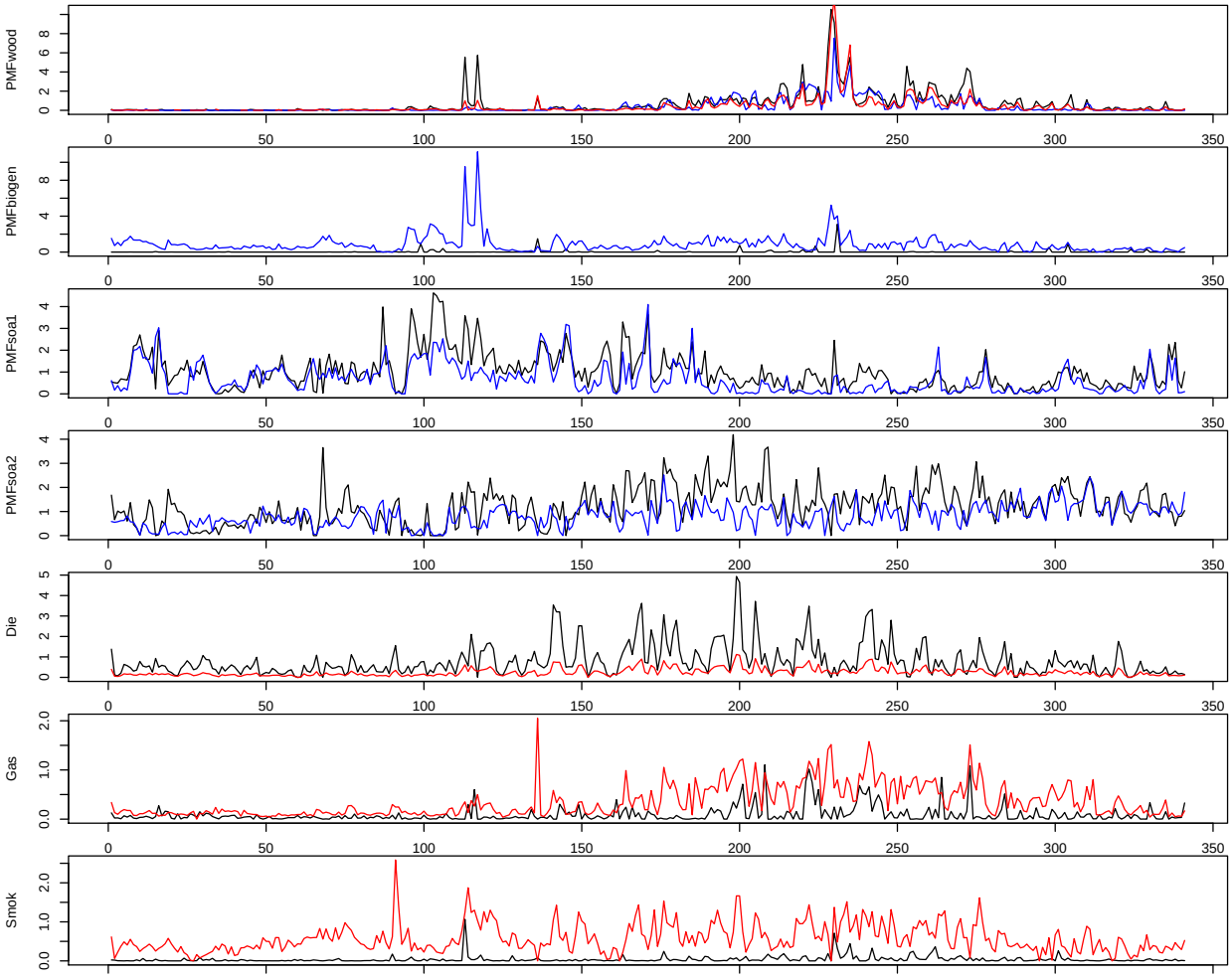

Figure 2: Contributions with $S = 5$ and $M = 10$.

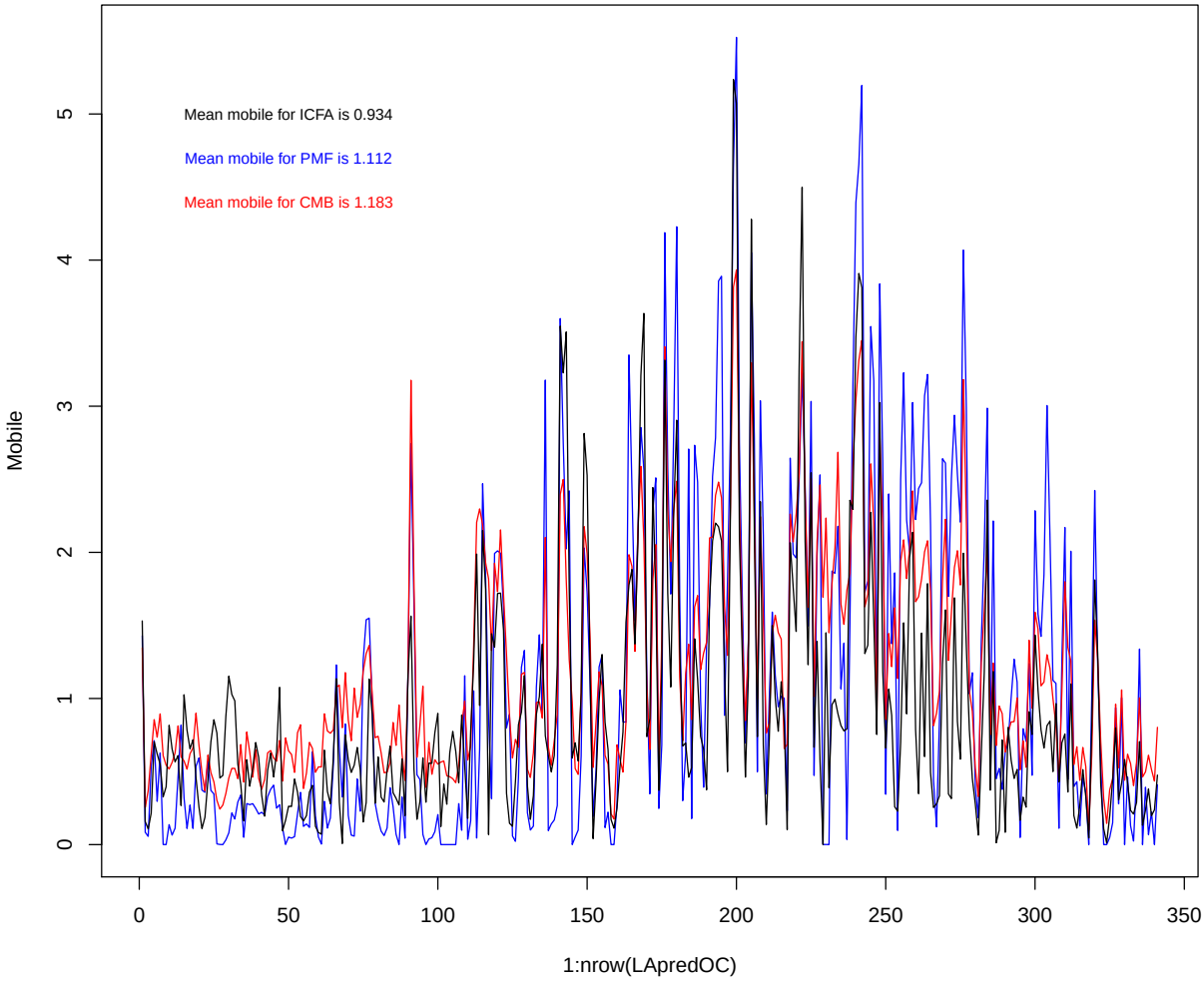

Figure 3: Combined Diesel/Gas/Smoker Contributions with $S = 5$ and $M = 10$.

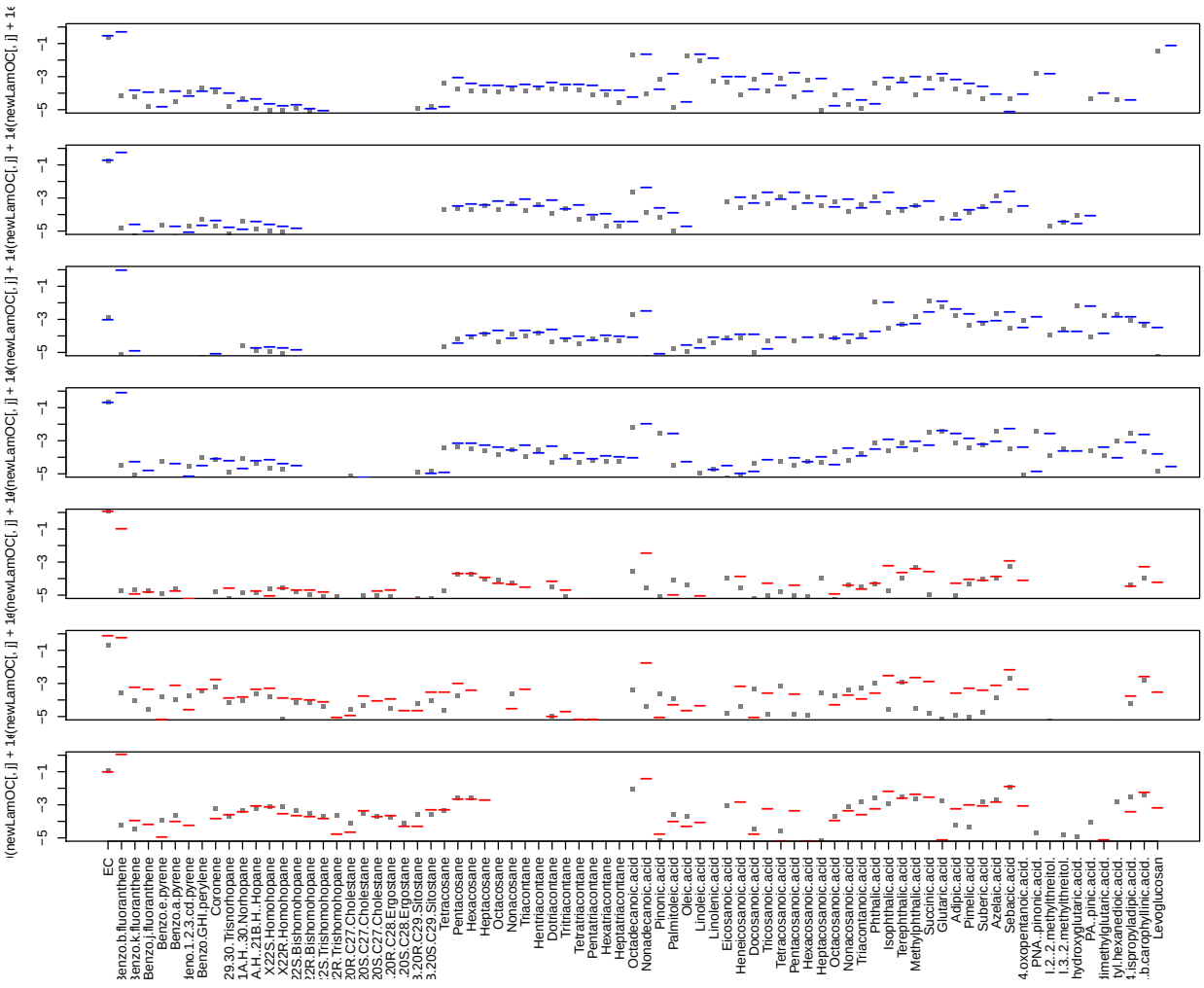

Figure 4: Profiles with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.

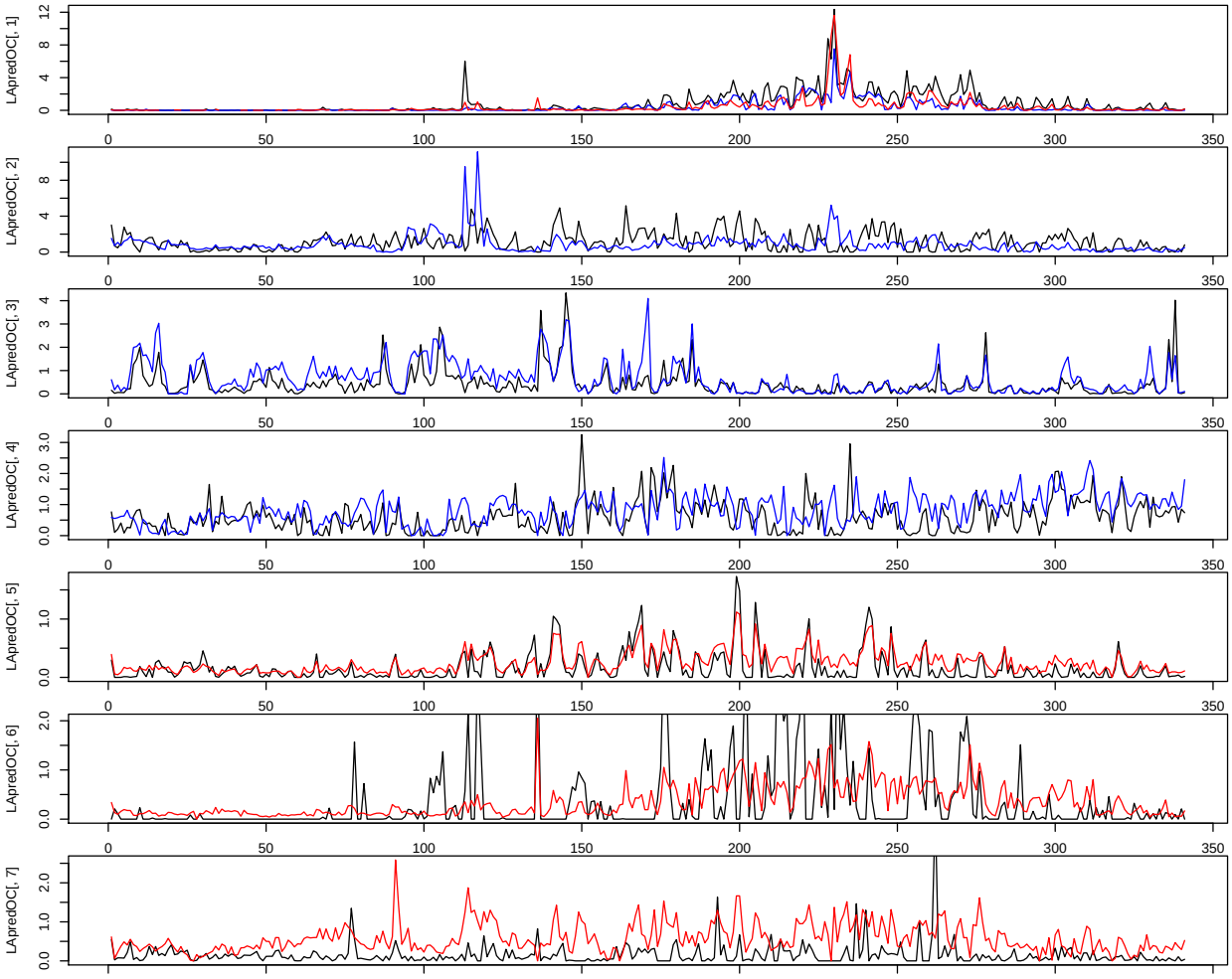

Figure 5: Contributions with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.

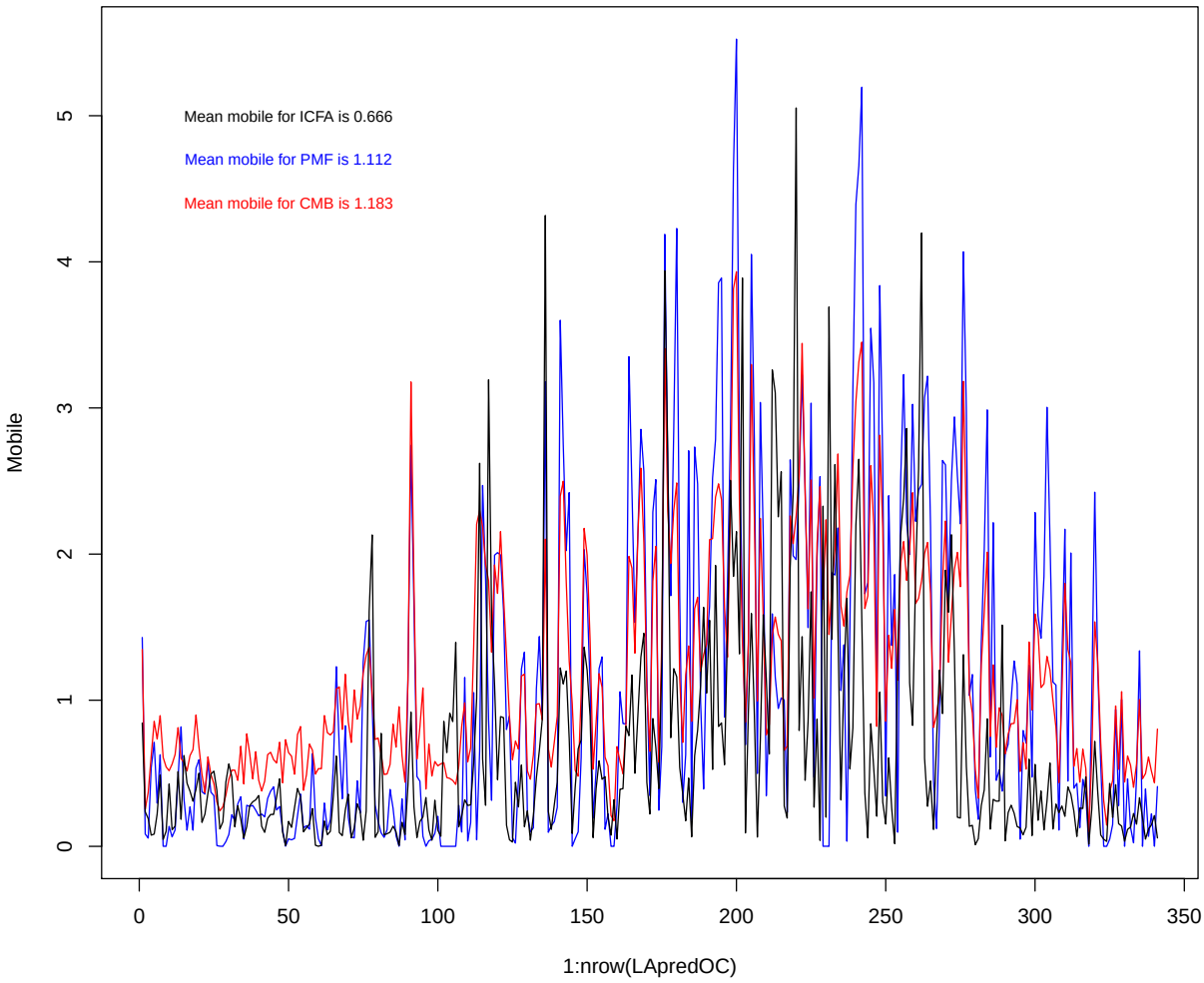

Figure 6: Combined Diesel/Gas/Smoker Contributions with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.

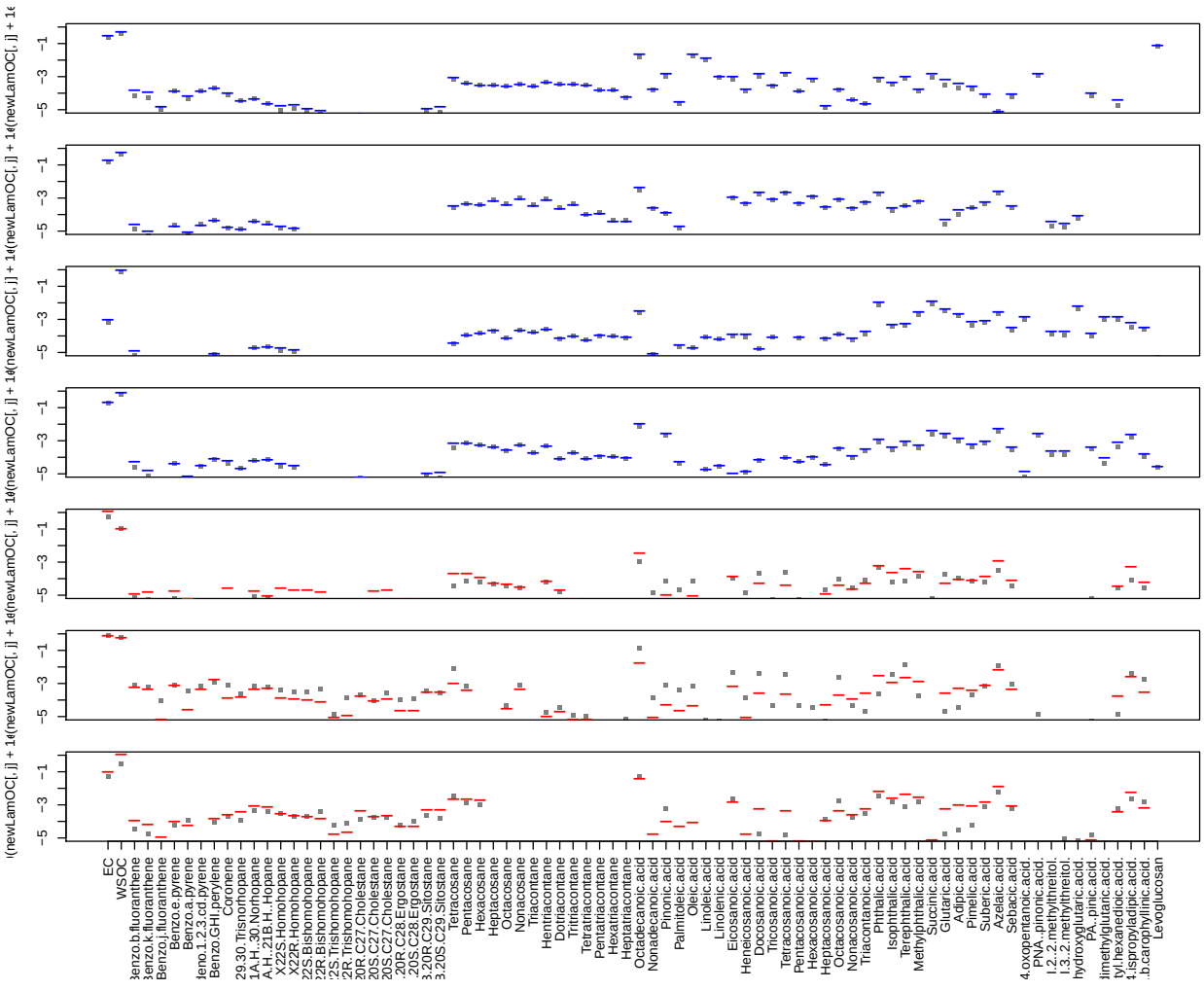

Figure 7: Profiles with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.

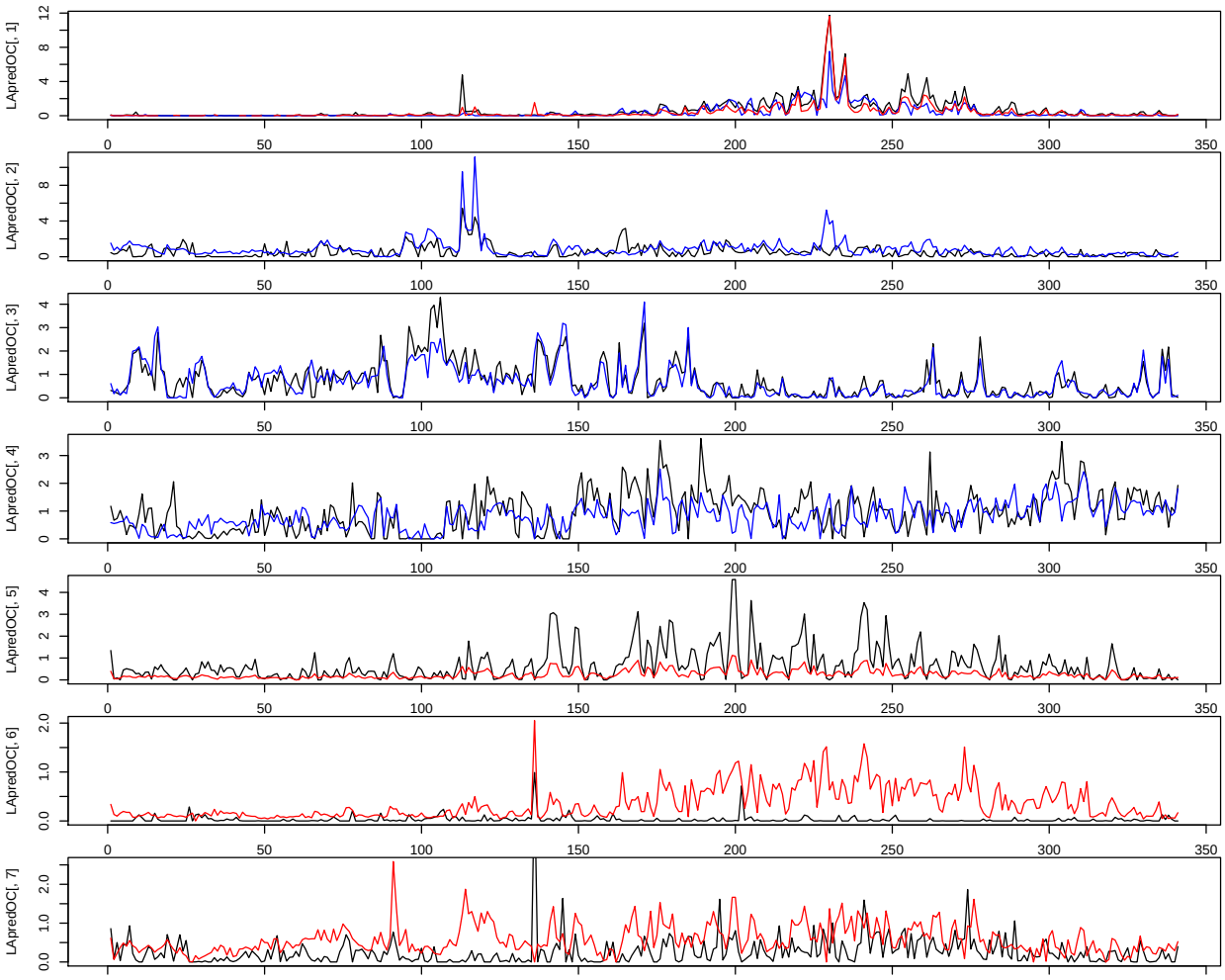

Figure 8: Contributions with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.

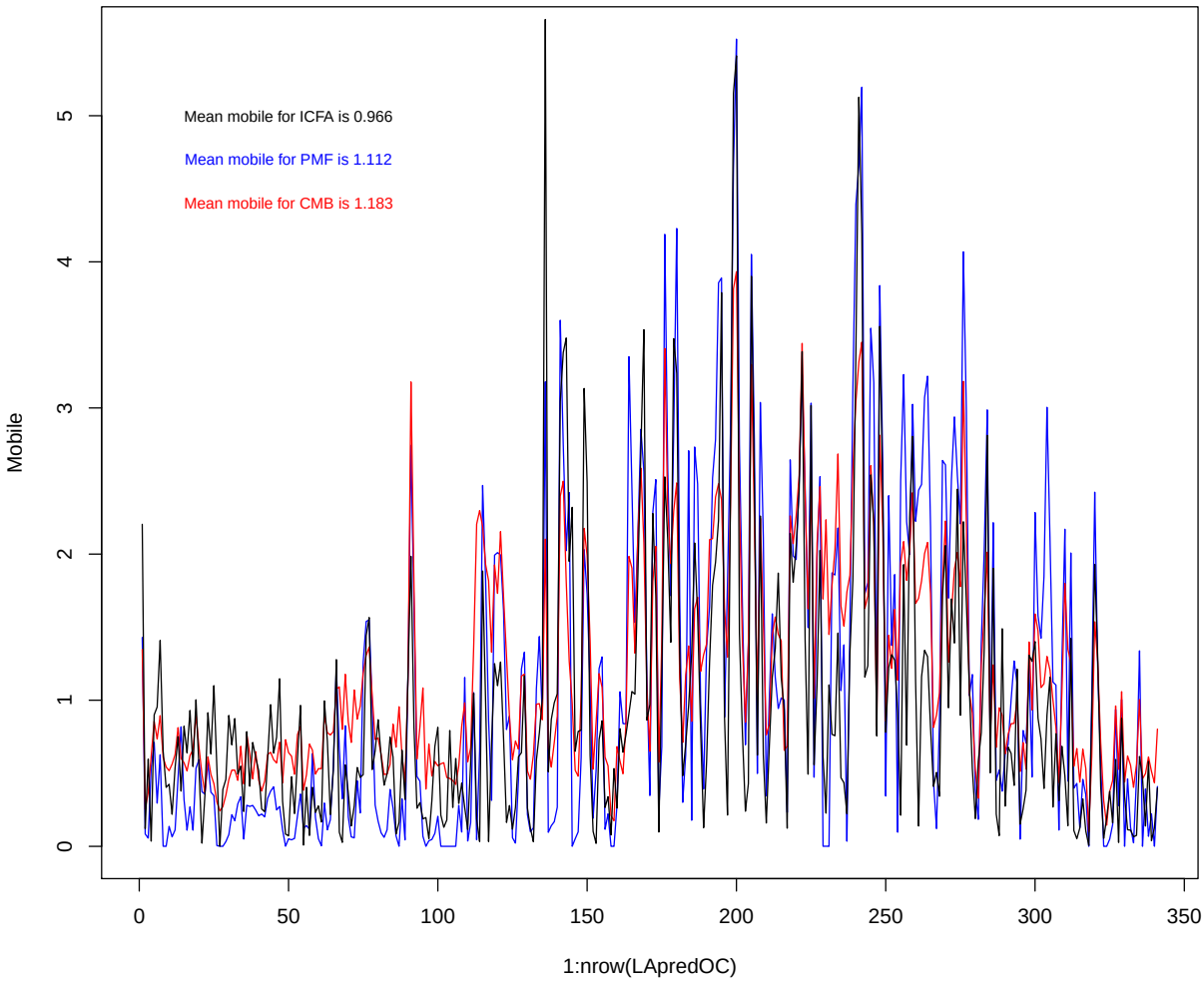

Figure 9: Combined Diesel/Gas/Smoker Contributions with $S = 2$ and $M = 1.5$, excluding WSOC from analyses.