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Source apportionment by Positive Matrix Factorization at Houston North Wayside

Report, Version 2

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Executive summary:

Sampling of fine particulate matter ($PM_{2.5}$) and subsequent chemical speciation was accomplished for the Houston North Wayside site from for 128 days between Jul 2022 and Jun 2023. The chemical composition for 116 of those 128 days was used to estimate source contributions to the local $PM_{2.5}$ at North Wayside. Twelve of the sample days were removed as outliers as they had extreme concentrations of select compounds, which could not be modeled. For the remaining 116 days, a source contribution model, the US Environmental Protection Agency (US EPA) Positive Matrix Factorization (PMF) model version 5 was used (https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses). This model reports source factors which are not necessarily representing specific emission sources but do represent which chemical species vary together at the site. These results can help to determine which type of sources impact the site.

An 8-factor solution was identified with the largest source factor is Urban BB mix (34.8%), which is a mixed urban factor that includes biomass burning (BB) and traffic emissions. The two crustal factors combine to contribute 26.7% of the total mass. The two sulfate factors combine to contribute 28% of the total mass, while the magnesium chloride and nitrate factors have very minor contributions at around 2%. The zinc metal contribution stands at nearly 6% over the year but does have one extreme day.

There are additional trace metals that were not included in the model as they primarily contributed to a few peak days and were not consistently present in the results, so the model could not affectively deal with them. Additional analysis may be needed to better understand the metals sources in the area.

PMF methodology and data evaluation:

Model input:

The datasets were received via direct communication from TCEQ (email and attachment from Erik Gribbon). These included chemical speciation of filter based $PM_{2.5}$ collected at the North Wayside site in the Houston metropolitan area, including concentration, uncertainty and minimum detection limit (MDL) as reported in the received datasets. The ambient concentration (µg m^{-3}) and the accompanying reported uncertainty of these chemical species was used as the input for PMF model version 5.0 from the US EPA (https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses). The per sample uncertainty was used for the model input. As the uncertainty reported with the ambient concentration was the analytical uncertainty associated with the chemical methods, an extra 10% modeling uncertainty was added to the input species to account for additional uncertainty associated with the data that may not be represented by the analytical uncertainty such as variation of source profiles and chemical transformations in the atmosphere (Norris, Duvall et al. 2014). This prevents the model from being too rigid with the fit on each sample point.

The PMF model does not accept blank values for any species. For this study the data was first filtered to remove species that had values for less than 50% of the sample days. The next step of data preparation was to evaluate the species concentration with respect to the reported MDL. For species concentrations below the MDL, a value of one half the MDL was used for model input. For example, roughly 50% of the potassium ion data was below the detection limit, so a value of one half the MDL was used for those days. The fraction of days above MDL, the uniqueness of the species as a tracer, and the signal to noise (S/N; indicates whether the variability is noise or real) of the species concentration were all used to evaluate the potential strength of the species in the model. The reported S/N in the table below was calculated within the EPA PMF 5.0 model (Norris, Duvall et al. 2014). The signal is calculated as below where concentration (x_i) and uncertainty (s_i) is used as the signal.

$$d_{ij} = \left[\frac{x_{ij} - s_{ij}}{s_{ij}}\right]$$

The S/N is then calculated as below.

$$\left[\frac{S}{N}\right]_j = {}^{1}/_{n} \sum_{i=1}^{n} d_{ij}$$

The EPA PMF 5 guidebook suggests that S/N using compounds with at least a S/N > 1, while compounds that have ambient concentration less than the uncertainty are assigned a S/N = 0. The model uses compounds that are rated good and downweights importance of compounds rated weak. Compounds rated bad are not included. Table 1 is a summary of compound classification for the input compounds.

Table 1 Input species for the PMF model including categorization of the compounds and species statistics.

Species	Category	S/N	Min	25th	Median	75th	Max
Mass	Weak	10.0	2.48062	8.2340	11.8295	17.9281	34.4578
Aluminum	Strong	3.9	0.00489	0.0234	0.0617	0.1115	0.7336
Bromine	Weak	2.9	0.00064	0.0022	0.0036	0.0054	0.0296
Calcium	Strong	9.2	0.00758	0.1543	0.2603	0.6034	2.7689
Chlorine	Bad	8.9	0.00011	0.0054	0.0319	0.1700	1.1792
Chromium	Bad	1.4	0.00011	0.0010	0.0023	0.0054	0.1088
Copper	Weak	1.5	0.00057	0.0015	0.0040	0.0076	0.0286
Iron	Weak	8.2	0.01453	0.0987	0.1953	0.3020	1.0574
Lead	Bad	2.2	0.00099	0.0014	0.0037	0.0071	0.0375
Magnesium	Weak	0.5	0.00275	0.0028	0.0135	0.0418	0.2151
Manganese	Bad	3.4	0.00038	0.0022	0.0047	0.0087	0.3268
Molybdenum	Bad	1.1	0.00031	0.0006	0.0012	0.0027	0.0485
Nickel	Bad	5.9	0.00009	0.0011	0.0027	0.0052	0.1125
Potassium	Bad	10.0	0.00840	0.0584	0.0820	0.1401	0.2882
Silicon	Strong	9.5	0.00109	0.0937	0.2098	0.3203	1.5481
Strontium	Bad	1.1	0.00024	0.0006	0.0016	0.0025	0.0053
Titanium	Weak	3.0	0.00023	0.0041	0.0070	0.0110	0.0596
Zinc	Weak	9.7	0.00281	0.0186	0.0391	0.0693	0.8235
Ammonium	Strong	9.9	0.01268	0.2370	0.3726	0.6741	1.8544
Chloride	Strong	8.1	0.00500	0.0170	0.0354	0.1643	0.9894
ECTOR	Bad	7.6	0.26420	0.6682	0.9504	1.4691	5.1470
ECTOT	Strong	8.0	0.09854	0.5115	0.7303	1.1210	3.6337
Nitrate	Weak	9.4	0.00500	0.1320	0.3404	0.5637	3.4886
OCTOR	Bad	8.4	1.20804	2.5180	3.7802	5.6038	13.0847
OCTOT	Strong	8.9	1.38558	2.5985	3.9538	5.9103	14.5980

Potassium Ion	Weak	7.9	0.00430	0.0150	0.0150	0.0431	0.1459
Sodium Ion	Weak	9.6	0.00500	0.0308	0.0947	0.3623	1.4667
Sulfate	Strong	9.8	0.27040	0.9318	1.4673	2.2774	6.1067

The categorization included evaluation of species times series, which are depicted in graph form below (Figures 1-3). The time series also provide a means of finding outlier days for each compound. Inputs to the PMF model typically have outlier events removed as the model cannot replicate outlier event concentrations. Figure 1 includes the key species which proved to be crucial in determining the factors in the PMF model output. From the time series in Figure 1, the zinc in the top panel can be seen to have a high event during the fall of 2022 but otherwise has low concentration. Calcium in panel 2 has high concentrations focused in Sept/Oct 2022. Nitrate in panel 3 is rather consistent in the cooler months and peaks in December, while ammonium (panel 4), organic carbon (panel 5), and sulfate (panel 9) are present year-round with seasonal peaks. Chloride (panel 6) has intermittent influence at the site, while Silicon (panel 7) peaks in late summer (Jul-Aug) and again in Sept/Oct. The $PM_{2.5}$ mass in panel 9 is present year-round.

Figure 2 includes an additional set of measured species for North Wayside. Of the species in Figure 2, only Strontium was not included in the PMF input. Strontium (panel 1) can be a crustal metal, however, for North Wayside the concentrations were generally quite low except a couple event concentrations that coincide with increases in Magnesium (panel 2). The strontium, magnesium, and potassium ion in panels 1, 2, and 3 in Figure 2, respectively have a very high point on 1/1/2023 and lesser on 7/4/2022. These dates were removed from the PMF input as outliers. These peaks likely represent a firework signal, but as it is only 2 days out of 128, the PMF model would not be able to model this source accurately. The magnesium (panel 2), titanium (panel 3) and aluminum (panel 8) show similar peak times in Jul – Aug. Bromine (panel 4) and EC TOT (elemental carbon by thermal optical transmission) are present year-round (panel 7).

Figure 3 includes the trace metal species from North Wayside that were present in more than 50% of all sampled days, however, not all these species were included in the final PMF model. Iron was chosen because of the high S/N (Table 1), while copper was chosen because it had a good model to observation correlation in the preliminary model runs and the S/N was greater than 1. Also, as mentioned above, all outlier days had to be removed as preliminary models could not replicate the observations on those days. The preliminary model runs did include many of the dates in Table 2, however, the predicted concentrations for the trace metals did not match the observed peaks and no factor was calculated to match these trace metal peaks. Therefore, these were deemed outlier days and were removed from the model to improve the estimation of the more routine sources and improve the model goodness of fit parameters (as discussed in the next section). Similarly, the trace metal species that were present in > 50% of all sampled days, but did not have good predicted vs observed relationships were also not included in the model. The PMF user guide specifically discusses cases of trace metals which have brief and infrequent peaks in concentration, which cannot be resolved by the PMF model due to insufficient information, e.g. not enough samples or species to define the factor (Norris, Duvall et al. 2014).

However, although these metals were not all included in the model runs, the time series does provide valuable information about potential local influences. There are many key days with overlapping peak concentrations in these metals. For example, nickel (panel 1) and molybdenum (panel 2) share many key dates, while lead (panel 4), iron (panel 5), copper (panel 6) and chromium (panel 7) also share some, but not all peak dates. Additional analysis would help to better define these potential sources; suggested additional analysis would include evaluation of elemental ratios and comparison to known sources, evaluation of wind direction during peak events, and additional sampling with chemical speciation to better constrain the frequency of source impact in this area (see Future Work).

Based on these time series, twelve samples were removed from the batch of 128 sample days. The excluded dates are defined in Table 2. The minimal number of samples for PMF analysis is typically 100 and the sample set here, with outliers excluded, is 116. This is at the lower end of sample numbers. In the EPA PMF 5 user guide (https://www.epa.gov/sites/default/files/2015-03/epa_pmf_5.0_setup.exe), the example dataset for chemical speciation of filter samples at a single site has an n>400 samples. The number of samples impacts how well the model can define a factor, with a higher number of samples often increasing the accuracy of that factor replication in the model.

Table 2 List of excluded dates from PMF input.

07/04/22 00:00
11/03/22 00:00
11/07/22 00:00
11/09/22 00:00
11/10/22 00:00
11/28/22 00:00
12/25/22 00:00
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02/23/23 00:00
02/27/23 00:00

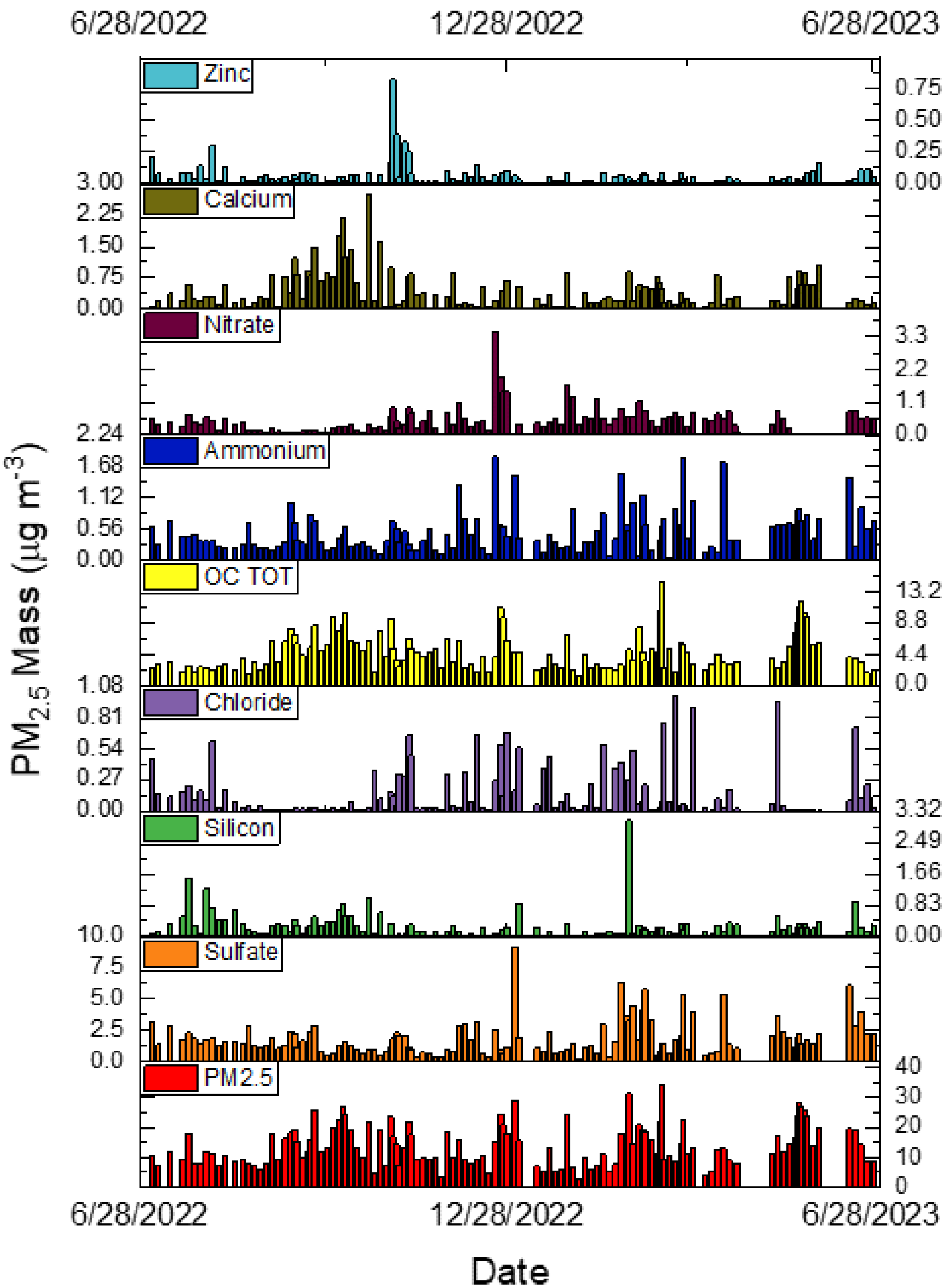

Figure 1 Time series of select key species presented as ambient concentration. These species are colored in the same as the output source factor for which they are key species. OC TOT is organic carbon by thermal optical transmission.

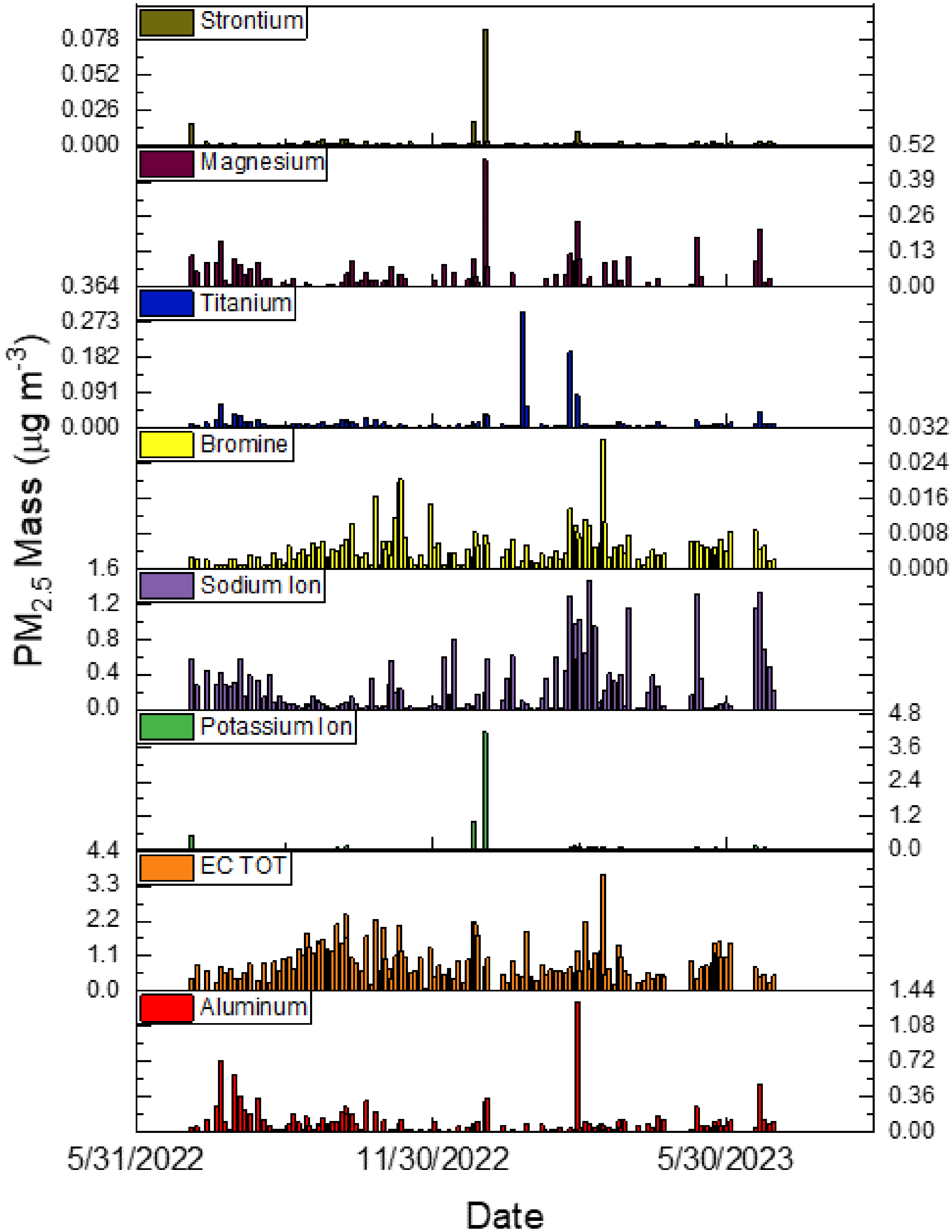

Figure 2 Time series of additional species presented as ambient concentration. EC TOT is elemental carbon by thermal optical transmission.

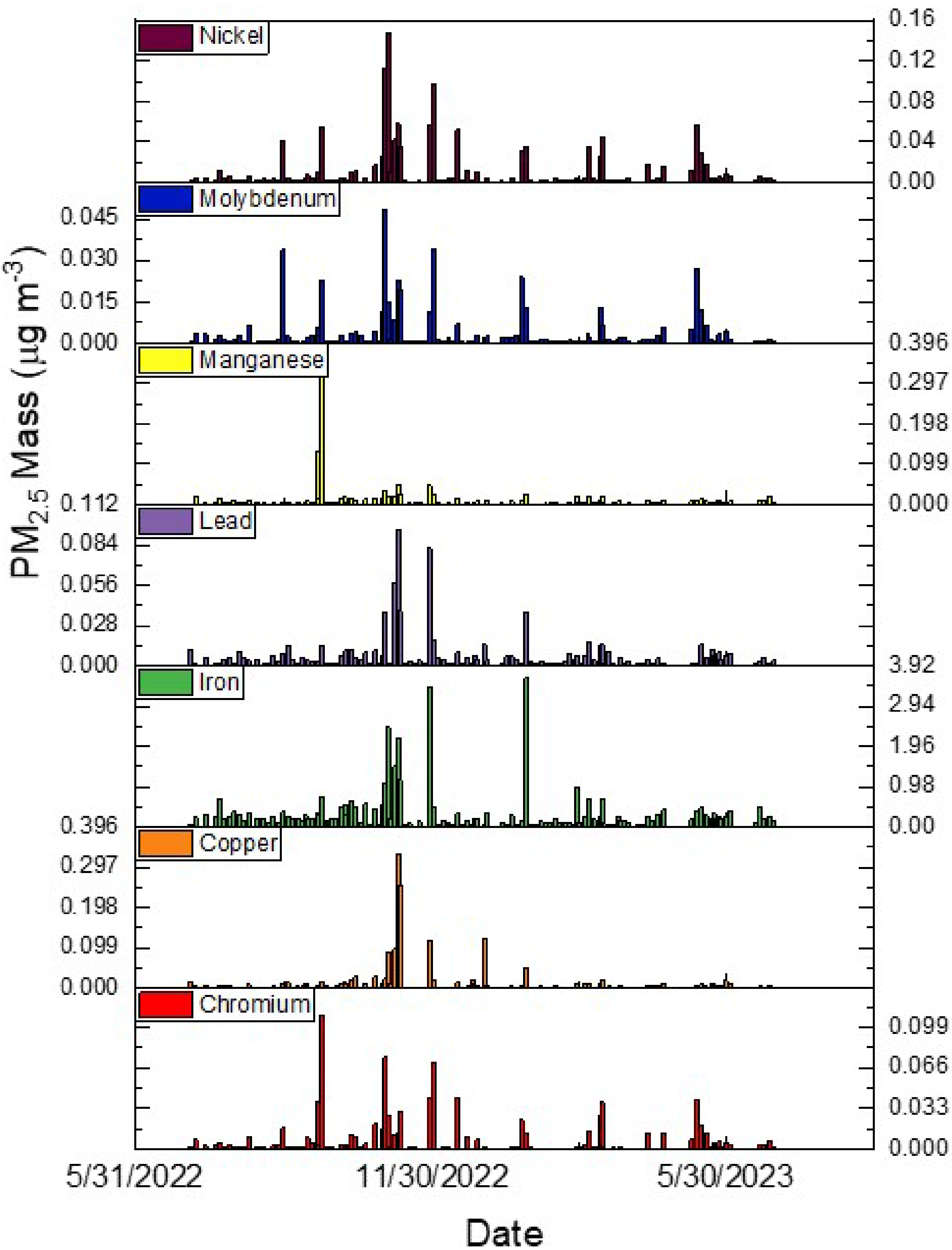

Figure 3 Time series of select metals species presented as ambient concentration. Only copper and iron were included as input species in the PMF model.

Model runs:

As suggested in the user guide, the initial PMF runs were for 6 factors and 20 runs. The model was then rerun for 7, 8, and 9 factors, each with 20 runs. The PMF model reports Q-values in the base run landing page for each run. The Q-value is a goodness-of-fit parameter and helps one determine how well the model fits the input data. For this PMF analysis, the largest drop in the Q-value occurred between the 7 and 8-factor runs, with only an incremental drop in Q-value with the addition of a 9th factor. The rotational analysis highlighted that the 8-factor solution had the lowest change in Q-value, indicating that this was the preferred solution, statistically. Additional analysis of the source profiles and time series confirmed that the 8-factor solution was the best fit for this sample set at North Wayside.

The residual analysis is another means of determining the best run conditions (e.g. number of factors, species categorization, uncertainty, removal of outliers). The residuals help determine how well the model fits each species. Residuals (model not fitting with observation) should be minimized and the residual analysis should result in a normal distribution, centered on zero, between -3 and 3. The residual analysis for the PM mass is included in Figure 4 below.

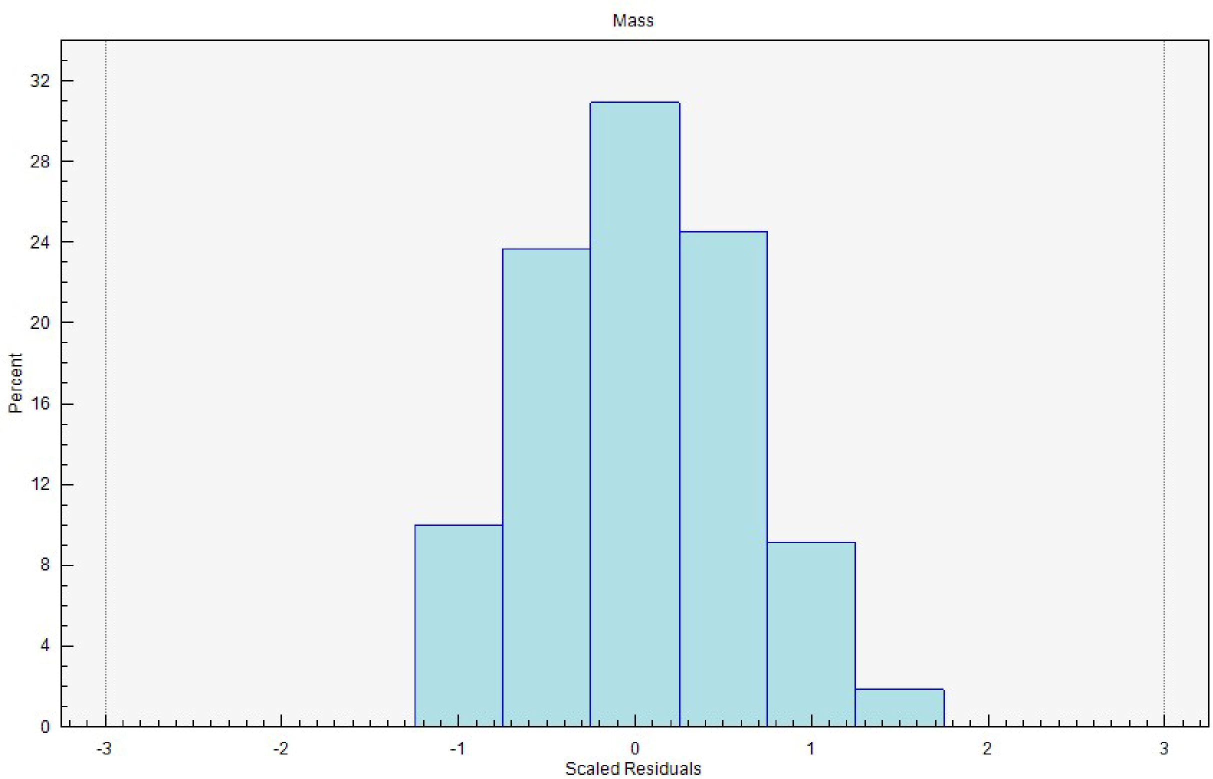

Figure 4 Residual analysis for PM mass for the 8-factor run in the final analysis.

The comparison between input observations and modeled results are also used to evaluate the model runs. The correlation plot in Figure 5 shows that the regression is close to the one-to-one line with no major outliers. In initial runs with the metals in Figure 3, there was poor correlation between observed and predicted and many outliers (see Figure 6). The predicted and observed time series also helps identify any outliers that the model cannot predict (Figure 7 and 8).

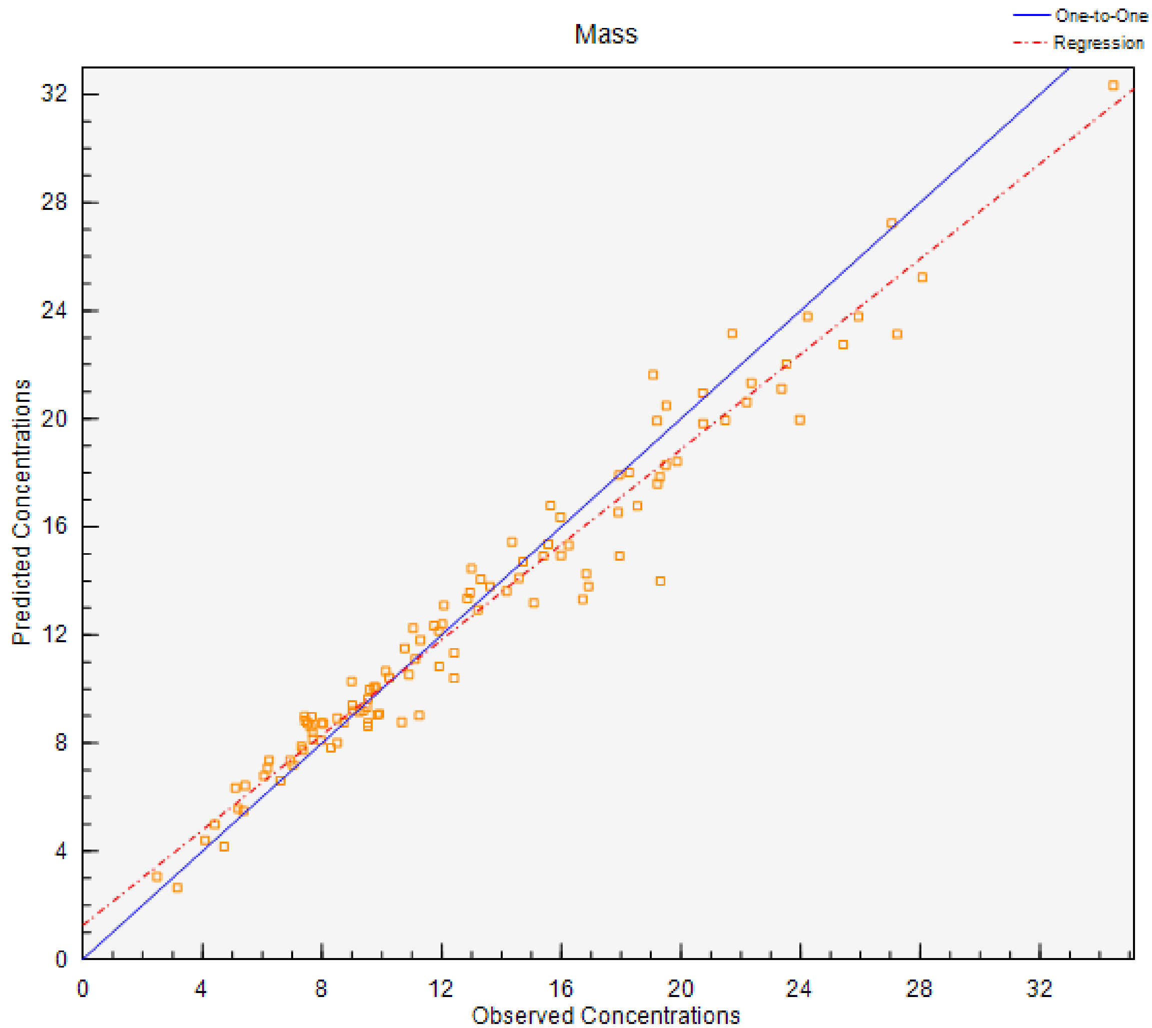

Figure 5 Correlation plot of predicted or modeled PM mass versus observed PM mass for the 8-factor solution.

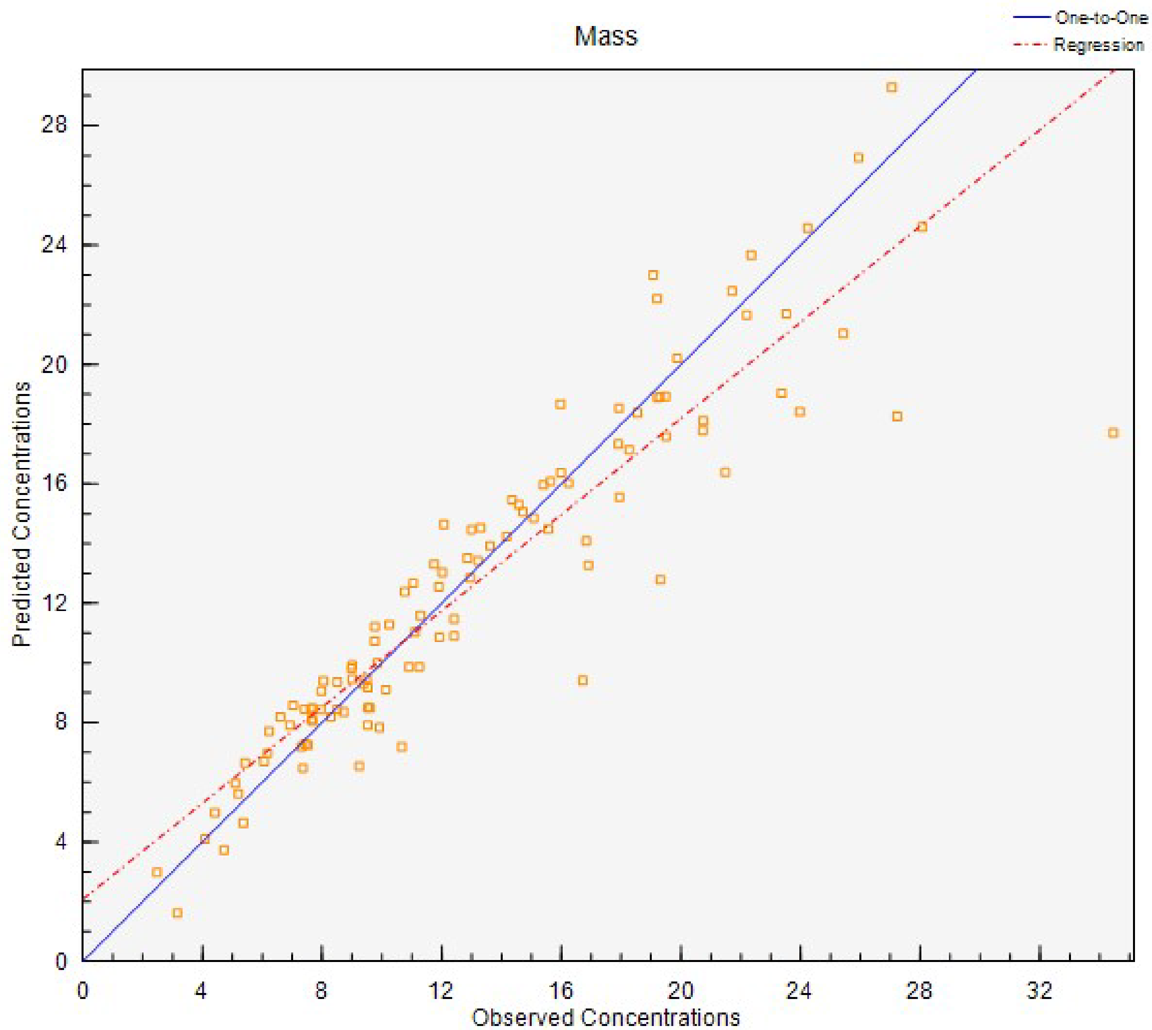

Figure 6 The 7-factor solution showing dates that were not well modeled and a weaker correlation.

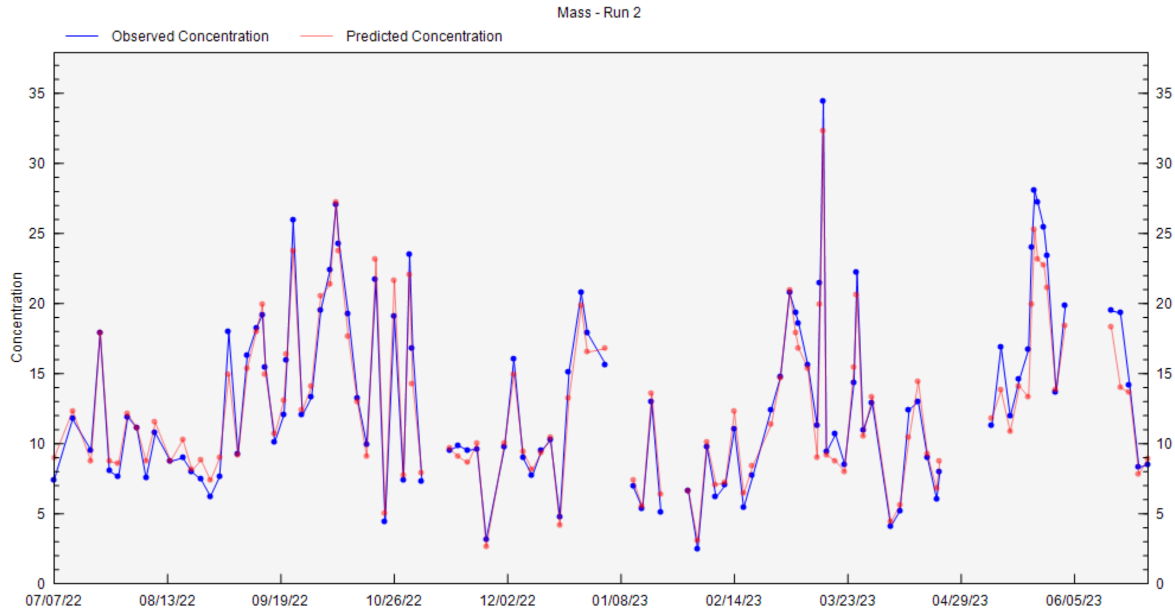

Figure 7 Time series of predicted and observed PM mass for the 8-factor solution.

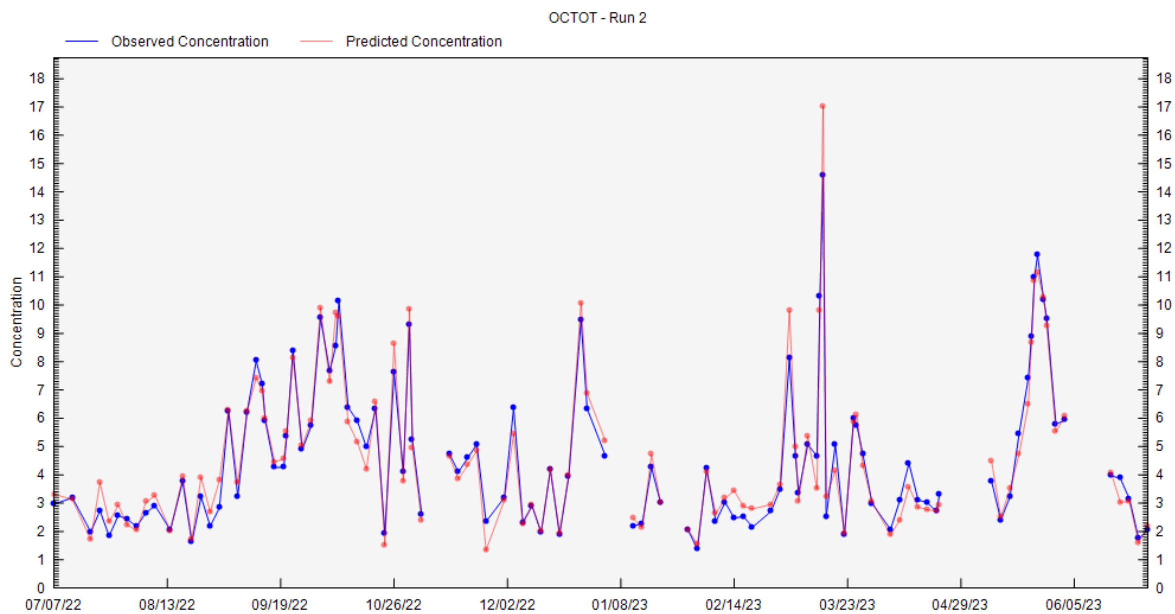

Figure 8 Time series of the organic carbon predicted and observed concentrations for the 8-factor solution.

There are three different types of additional error estimation analyses that can be run in the PMF 5 program: base model displacement method, base model bootstrap method, and the combined base model bootstrap displacement method. These were run on a 50 run version of the 8-factor solution. The displacement analysis did not reveal factor swaps at any dQmax level. The bootstrap displacement analysis showed a maximum decrease in Q that was less than 5, which is not a large value. There were swaps in factors 2 (Crustal 1) and 8 (zinc metals); which may indicate that these are not completely resolved from each other. The combined results are discussed below for each source factor to give an indication of which species is the most certain and most definitive for that source factor. This helps to define which species can be used to assess potential sources contributions to that factor and help to identify when a species is not clearly defined within this PMF analysis. For each factor, the range in the bar depicts the error associated with the inclusion of the species within the factor. The species with the shortest range in concentration, regardless of the actual concentration, are the most definitive for that factor. The figures are all included in Appendix 1. These species are included in Table 3.

Table 3 The error estimate for species within each factor with strong certainty marked + and weak marked – and no inclusion in the factor marked 0.

Species	Na Mg SO4	crustal 1	Mg Cl	Urban BB mix	NH4 SO4 aged	nitrate	crustal 2 urban	zinc metals
Mass	+	+	+	+	+	0	+	+
Aluminum	0	+	0	0	0	-	-	0
Bromine	-	0	-	+	+	0	+	0
Calcium	0	0	-	0	0	0	+	0
Copper	0	0	0	-	0	0	+	+
Iron	-	+	0	-	-	0	+	+
Magnesium	+	+	+	0	0	0	0	0
Silicon	0	+	0	-	+	0	+	0
Titanium	0	+	-	-	-	0	-	0
Zinc	0	0	0	0	0	0	0	+
Ammonium	0	0	0	0	+	-	0	0
Chloride	0	0	+	0	0	0	0	-
ECTOT	-	0	0	+	0	0	+	+
Nitrate	0	0	0	0	0	+	0	0
OCTOT	0	-	0	+	-	0	-	-
Potassium Ion	-	0	-	+	0	0	0	0
Sodium Ion	+	-	-	0	0	0	0	0
Sulfate	+	0	0	0	+	0	0	0

The 8-factor solution

The 8-factor solution can be presented in multiple formats. We will look first at the factor profiles. It should be noted here that factors are not emission sources. Factors indicate species that trend together in time during the sample period, which can indicate co-emission from the same source but may also indicate co-transport within an urban area or co-production from precursor species. Therefore, these will be called source factors, but not called emission sources. These profiles include both the concentration (left axis) and the % of the total species (right axis) as well as the species list (bottom axis). The 8-factor solution is displayed in Figure 9 as output directly from the PMF 5 model with the factor names assigned based on prior knowledge of tracer species and known source profiles. In this particular case, many of the inorganic ions found their own factors, while the remaining elements and metals grouped into 2 crustal factors and an additional zinc metal factor. The organic carbon was primarily present in a single factor with potassium ion, elemental carbon, bromine, and copper. The organic carbon had a more uncertain and lower inclusion in the ammonium sulfate factor. However, it is well-known that organic carbon has many sources within urban areas (e.g. traffic emissions, biomass burning, food cooking, secondary production in the atmosphere from gas-phase precursors, and biogenic sources). These results indicate that the model can only give a general urban mix for the organic carbon sources, as the potassium ion would indicate biomass burning and the copper might indicate traffic emissions or road dust.

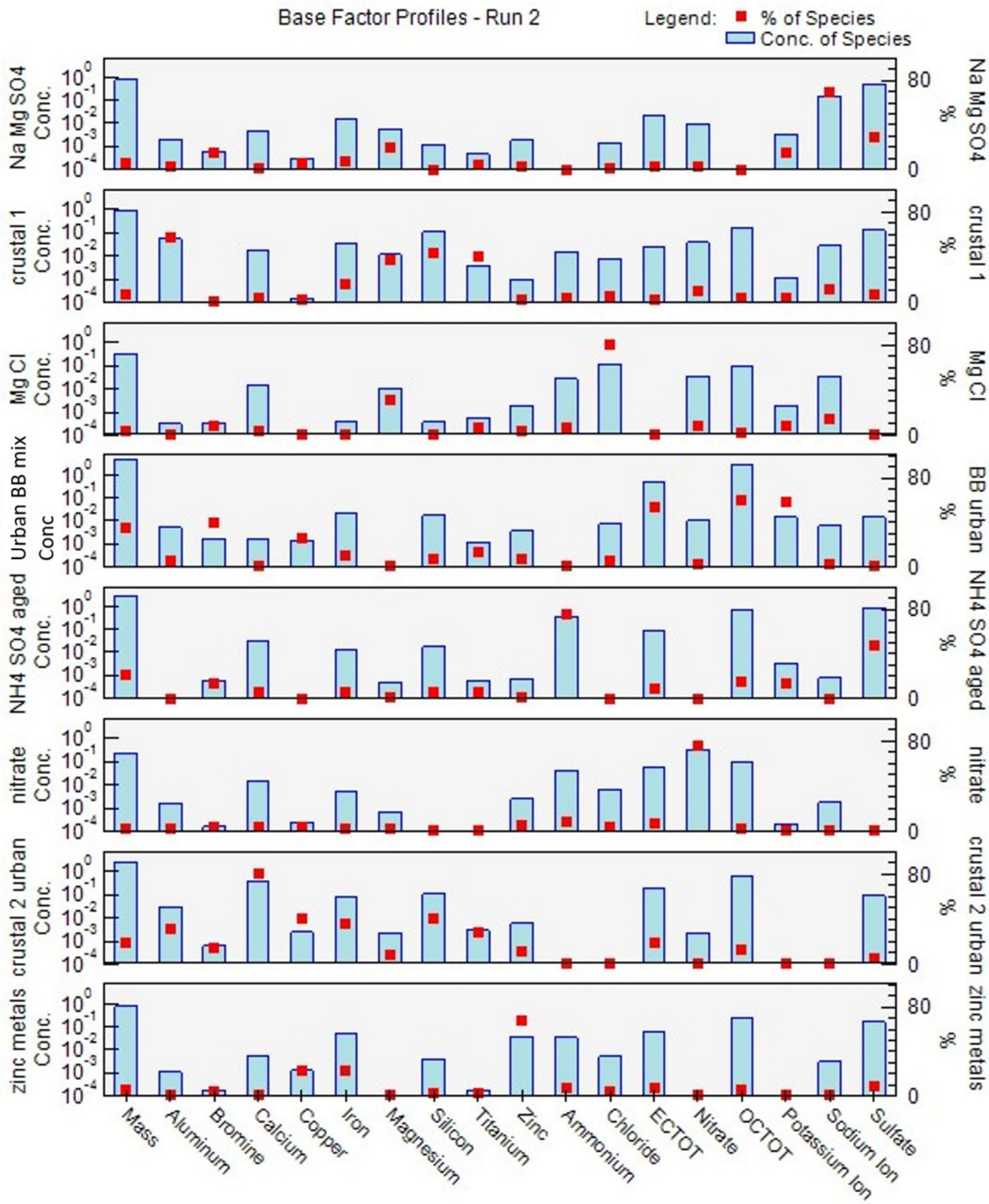

Figure 9 Source profiles for the 8-factor solution.

Each individual factor can be further plotted as a time series of contribution to the PM mass (Figure 10). This time series, combined with the factor profile in Figure 9 is useful in distinguishing between the sources for the two crustal factors. Crustal 1 factor is present predominantly in Jul-Aug and includes aluminum, iron, magnesium, silicon, and titanium, which are all naturally occurring in crustal materials. It is known that Saharan dust often impacts the Houston area during this time frame and it likely contributes to this source factor. The Crustal 2 factor is dominated by calcium, includes an enhancement of copper and elemental carbon, and is present after the end of the Saharan dust season; this likely indicates more local contribution (e.g. road dust) included within this crustal factor as copper and elemental carbon are both present in traffic emissions. As silicon, iron and titanium are present in both crustal factors, this supports the designation of these are including crustal or dust material.

The Zinc metal factor includes one high event which is captured well in the predicted time series. The composition of this factor only includes metal species (iron, copper and zinc), likely indicating a local metal industry source. Additional wind direction and emission inventory analysis would help identify the source. Additional filter sampling and chemical analysis would confirm the frequency of contribution for this source factor.

The ammonium sulfate factor includes the "aged" designation (Figure 9 and 10) as it may include organic carbon (the contribution is uncertain) and ammonium sulfate is produced in the atmosphere from separate emissions of sulfur dioxide and ammonia. These do not have to come from the same emission source. This factor is present through the calendar year.

The Urban BB mix factor is the dominant organic carbon factor, as discussed above. It is present year-round but cannot be directly attributed to only one emission source. Comparing to BC2 data for specific fire events would help to confirm this. The urban BB mix factor also included elemental carbon, bromine, and copper. Organic and elemental carbon with metals (e.g., copper) and non-metal materials (e.g., bromine) within a single factor may provide insight into possible sources. Urban structures bring many elements of this factor in close proximity including the building material and the consumer products within. Urban fires that destroy these structures and their consumer and industrial contents (e.g., plastics, electronic, textiles, wood, and metal and non-metal materials) could potentially support the fire-induced mobilization (thru, combustion, thermal degradation or dehalogenation) and subsequent release of aerosols that contain both OC and EC as well as metals and non-metals. This has been reported indirectly through the characterization of burning urban waste (Kumar, et al. 2015), for example incinerators, and directly through the characterization of urban fire plumes for metals and non-metals (Li et al 2023). While these types of studies are extremely rare, they may shed light into the different components of the urban mix factor seen in this PMF analysis. For example, Li et al 2023, reported the presences of both bromine and toxic metals in ambient PM from urban fires in Hong Kong (2021). During these urban fires both bromine and copper atmospheric concentrations (along with zine, lead, and chlorine) increased. There are some differences between this single study and the composition of the urban BB mix factor including the presence of chlorine and zinc. These differences may be due to several factors including atmospheric processing and half-lives, building materials, and consumer products. Note: this PMF study did not directly sample urban fire plumes but did utilize similar sampling and analytical techniques as Li et al 2023. In addition to urban fire plumes, another potential urban source of bromine may be gasoline exhaust, which has been implicated in other Houston PMF studies (Sadeghi, Choi et al. 2020).

The magnesium chloride factor (Figure 10) is present intermittently throughout the year and may be include marine emissions. Likewise, the sodium and magnesium sulfate factor is present intermittently throughout the year but also potentially contains marine contributions. Additional wind direction analysis would help to confirm this.

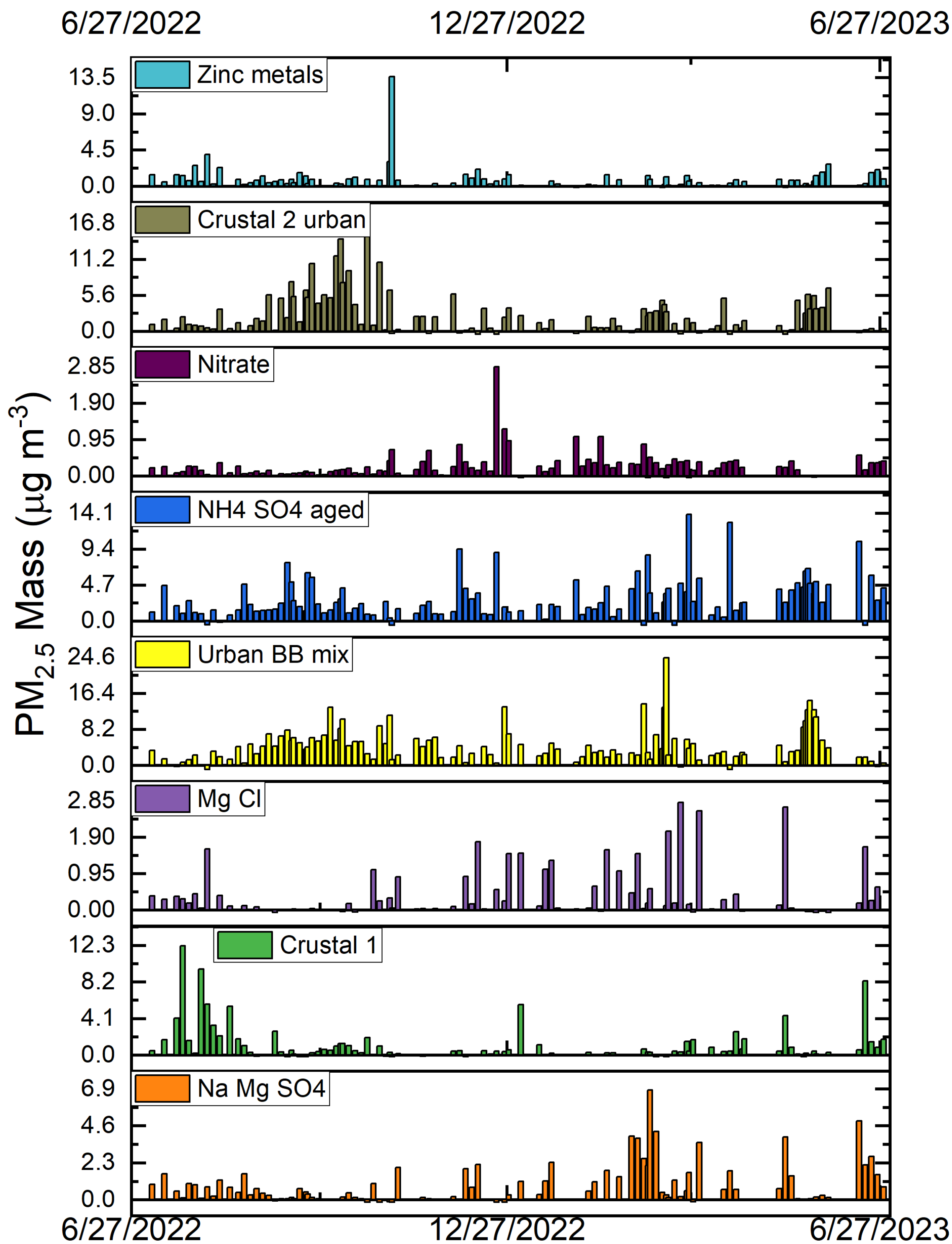

Figure 10 Time series of factor contributions to PM mass at North Wayside.

The time series of all factors can be viewed as 3-dimensional plots and stacked bar plots (Figure 11 and 12). These highlight the magnitude of impact of different source factors on the PM mass and seasonality of the source factors. Figure 11 highlights the mass contribution differences among the factors by season. The Crustal 1 source factor has a very high mass contribution, but is very limited in the season of impact. While Figure 12, the stacked bar time series of source factor contribution to PM mass, highlights that the Sept – Oct time period experiences peaks in two major factors: Urban BB mix and Crustal 2 urban, and the confluence of these factors is also evident in the consistently elevated PM in this season. The Zinc metal source factor has low contributions throughout the year, but the model does capture one event day (Nov 11, 2022) with a very high impact from this factor.

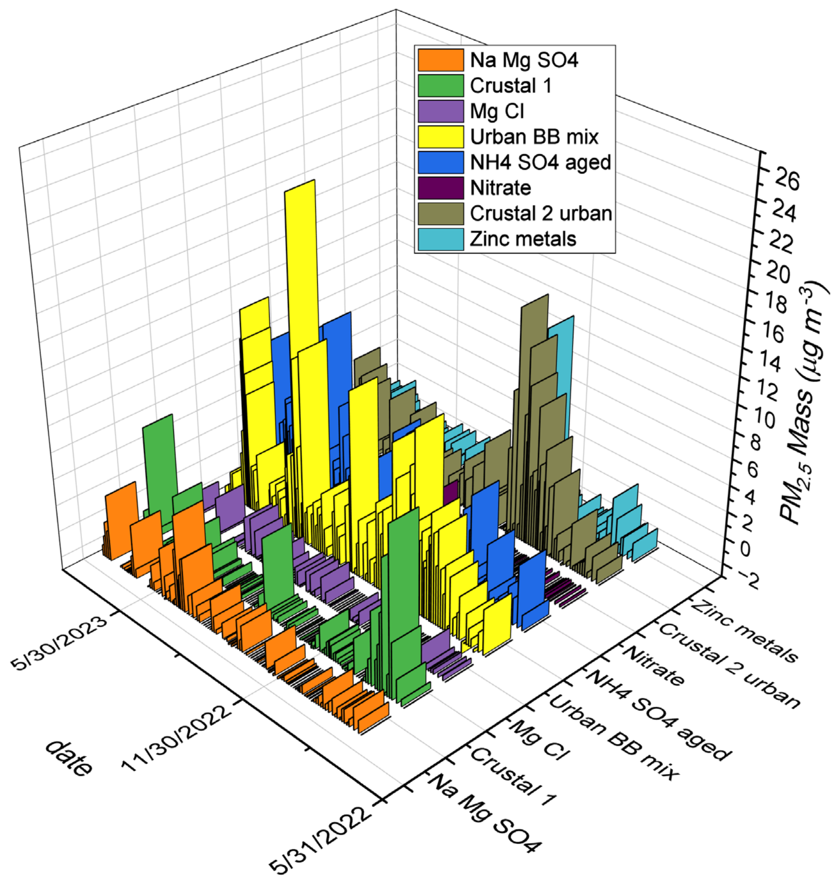

Figure 11 Ambient contributions by source factor and date depicted in three dimensions for $PM_{2.5}$ ($\mu g\ m^{-3}$) at the TCEQ site, North Wayside.

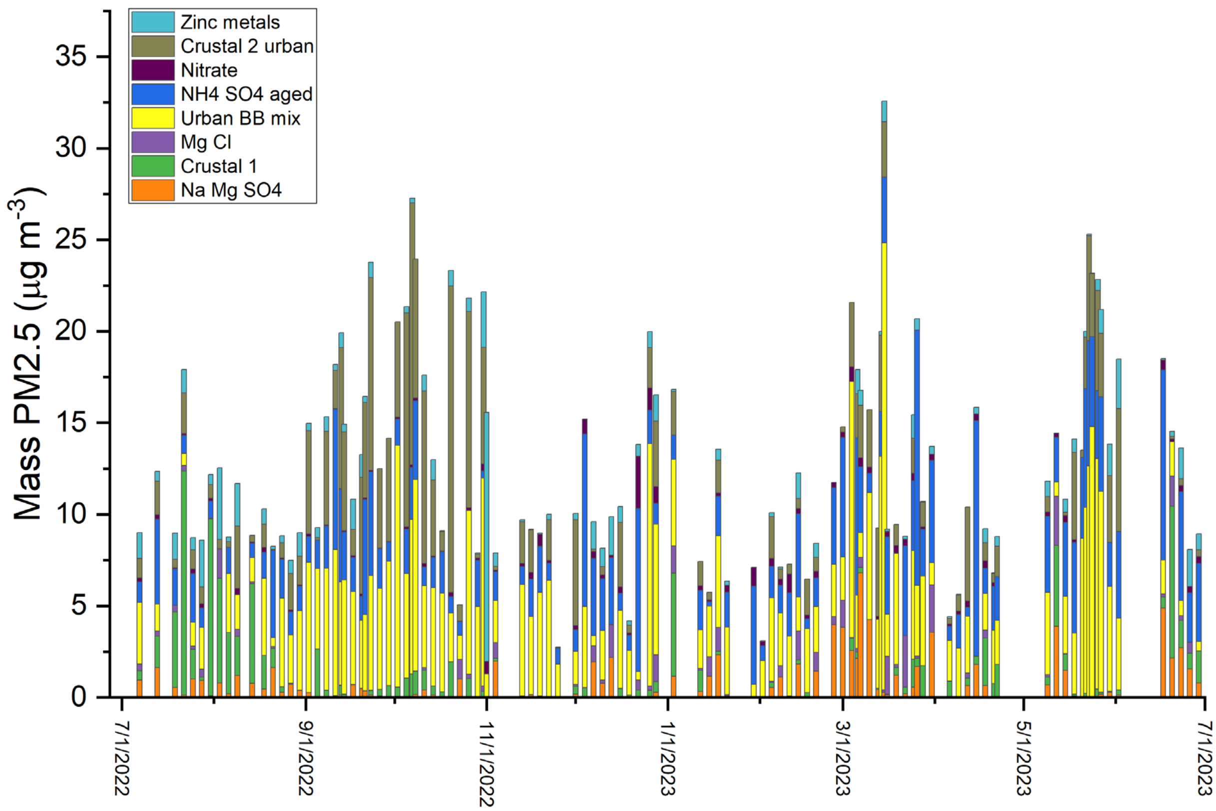

Figure 12 Time series of source contributions in stacked bar of combined PM mass by date at the TCEQ site, North Wayside.

A pie chart of the total mass for the entire sample set depicts the contribution by source factor to the year of samples (Figure 13). The largest source factor is Urban BB mix (34.8%), which is a mixed urban factor that includes biomass burning and traffic emissions. The two crustal factors combine to contribute 26.7% of the total mass. The two sulfate factors combine to contribute 28% of the total mass, while the magnesium chloride and nitrate factors have very minor contributions at around 2%. The zinc metal contribution stands at nearly 6% over the year, but does have one extreme day.

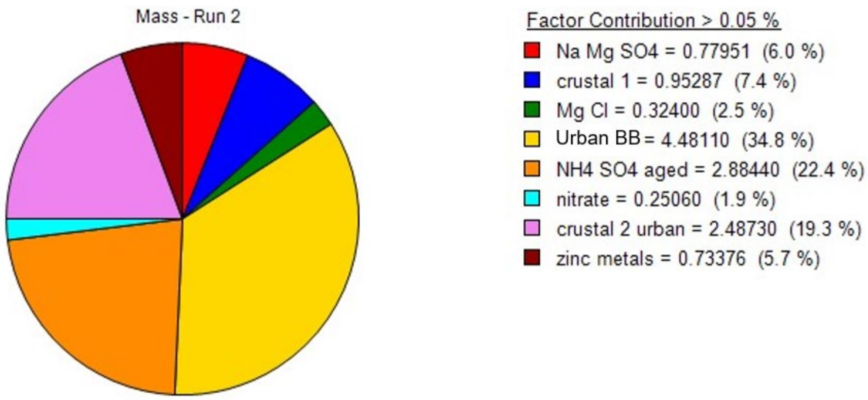

Figure 13 Pie chart of source contributions to the total PM mass.

Future work

- As mentioned previously, this source modeling was completed using a year's worth of data at roughly a one-in-three day sampling routine. Completing additional filter sampling and speciation to add to this data set would likely lower the error and may enable improved characterization of source factors.
- The trace metal source contributions were not complete for all the species in the input files. Chromium, lead, nickel, and molybdenum were removed from the model due to poor predicted vs observed response in the model and peak days for iron, copper and manganese were removed as outliers. The PMF model cannot produce factors for elements that have insufficient information (e.g. few peaks and few samples). Wind direction and elemental ratio analysis could be combined with emission inventory information to further assess potential sources in the local area. Additional sampling would be required to accrue enough sample dates with metals sources for the PMF model to be able to apportion these metals accurately to a source factor. However, the zinc – copper-iron source factor was clearly depicted in the model and wind direction analysis could be used to identify potential local sources.
- Organic carbon was primarily included in the Urban BB mix factor, with some potential inclusion in the ammonium sulfate aged factor. Separation of organic carbon sources would require additional speciation of the organic components (e.g. oxidized vs hydrocarbon-like, or individual organic compounds). For example, the inclusion of levoglucosan might enable the separation of an independent biomass burning factor. The potassium ion data was only 50% above the MDL, which may have impacted the ability to cleanly separate the BB source factor from the general urban emissions.
- Compare these results to other TCEQ data at nearby sites, including filter based PM speciation as well as aerosol optical for dust and biomass burning at BC2 sites.

Appendix 1.

Figures of combined error estimate analysis by factor.

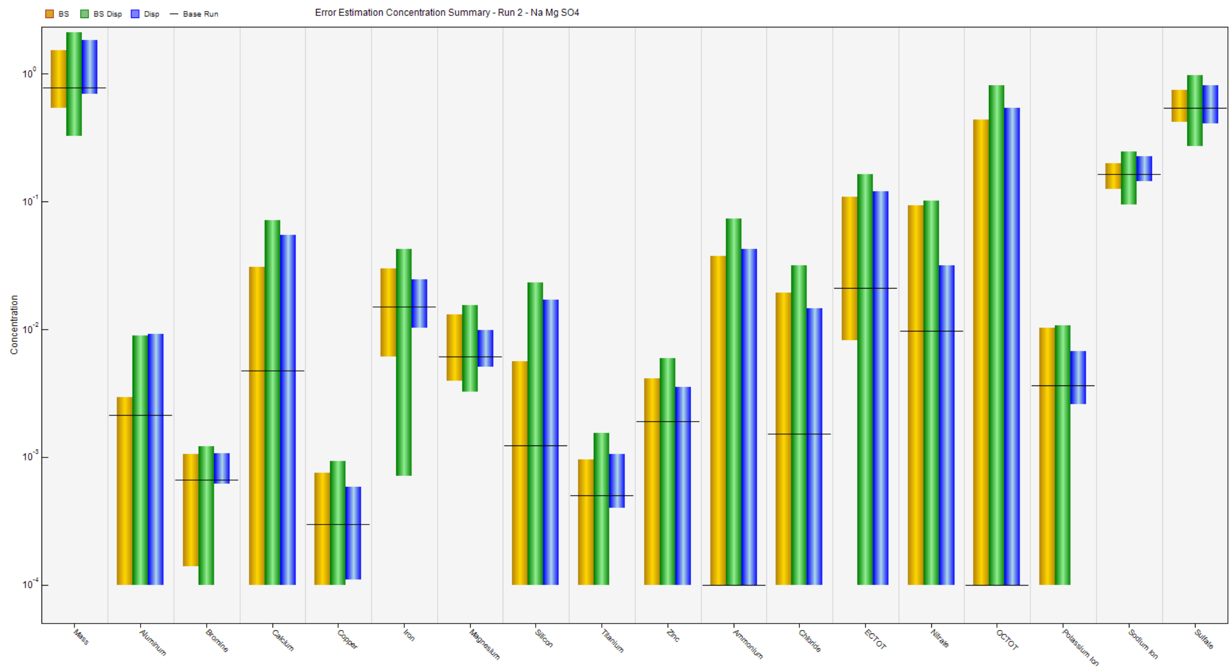

Figure A 1 Combined error estimation for Source Factor 1: sodium magnesium sulfate (Na Mg SO4).

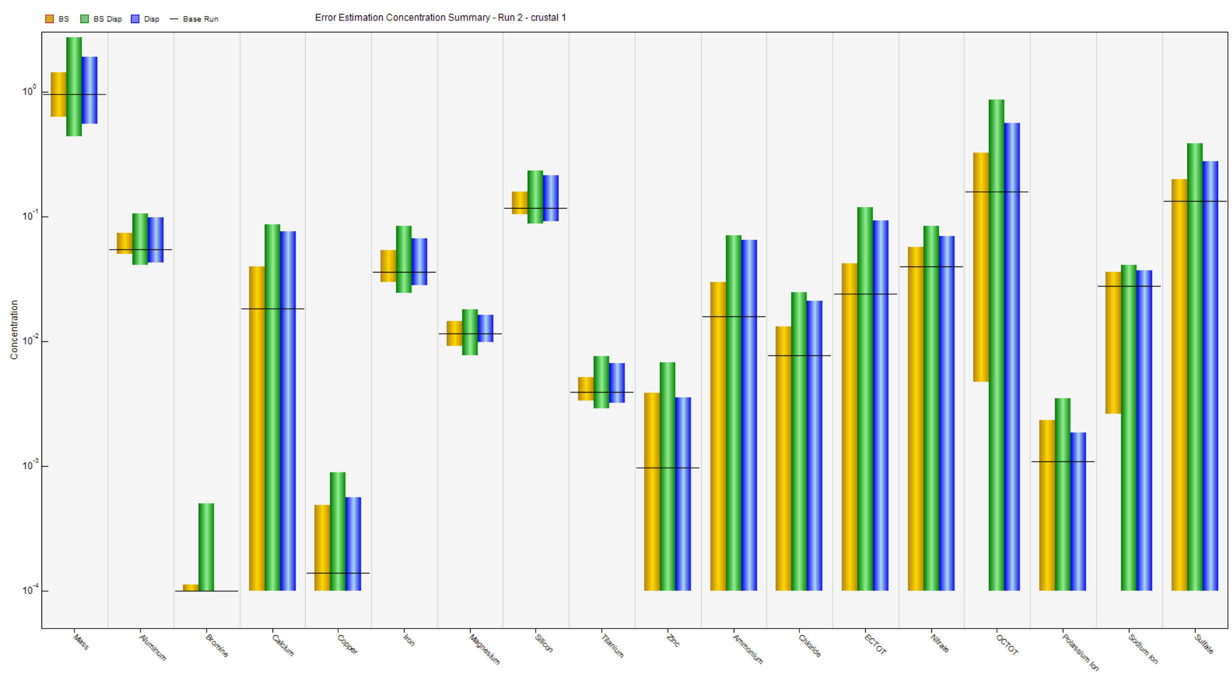

Figure A 2 Combined error estimation for Source Factor 2: Crustal 1.

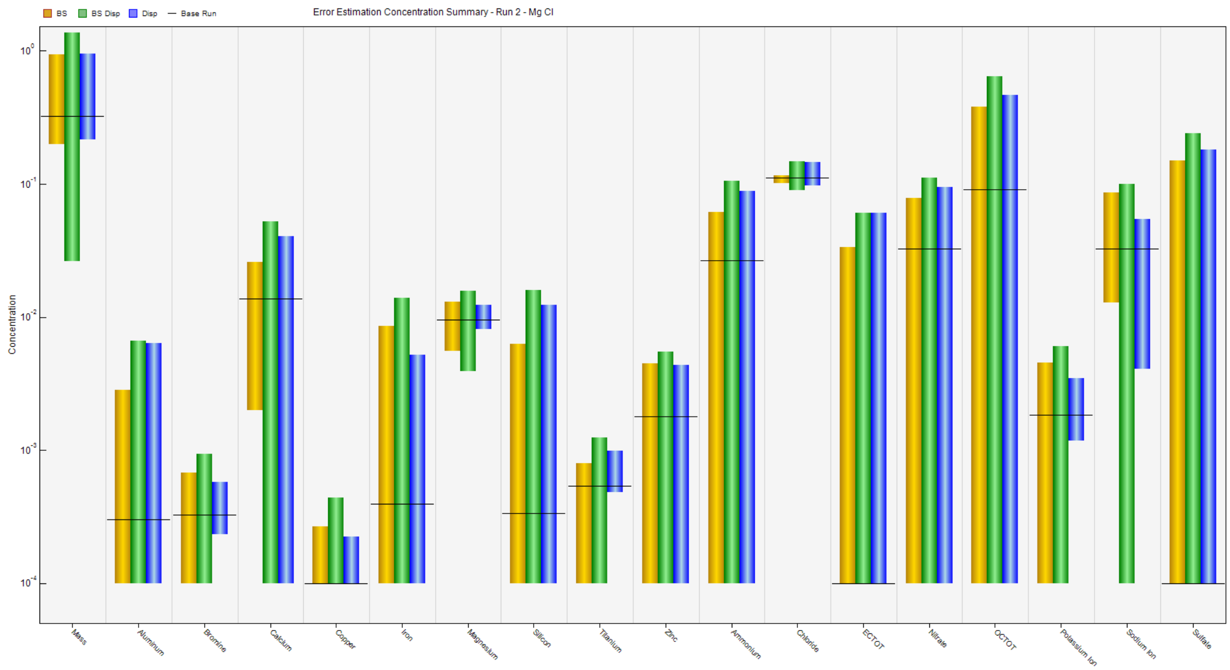

Figure A 3 Combined error estimation for Source Factor 3: magnesium chloride (Mg Cl).

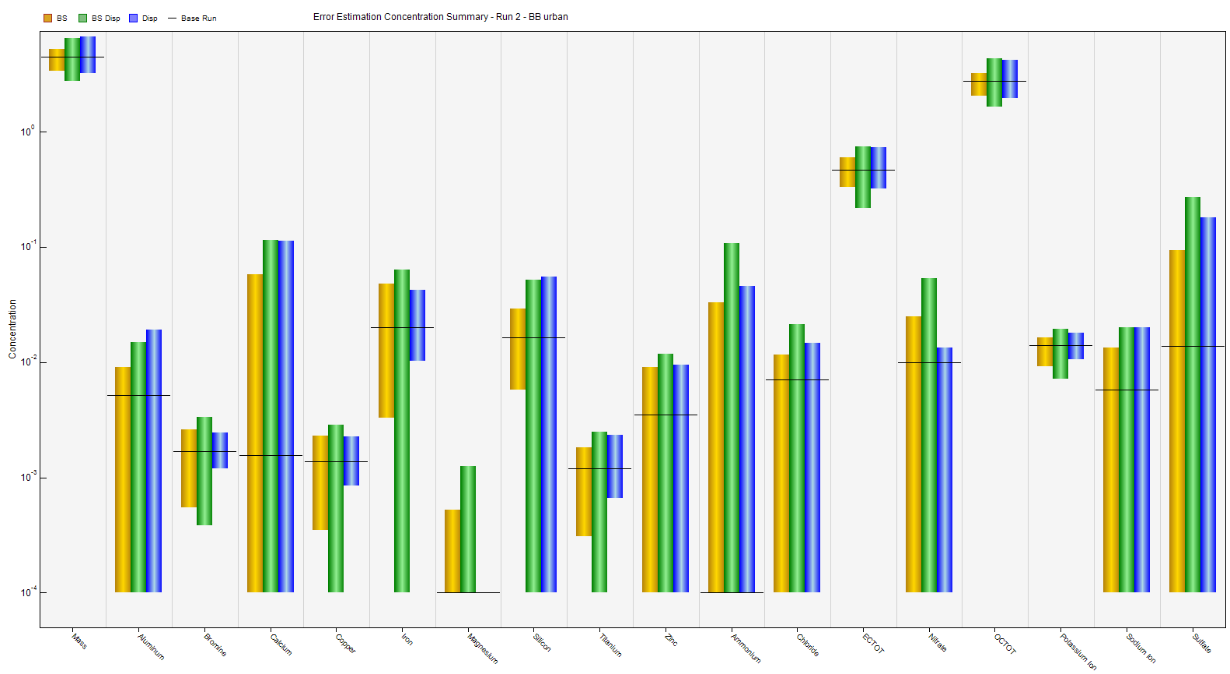

Figure A 4 Combined error estimation for Source Factor 4: urban biomass burning mixture (Urban BB mix).

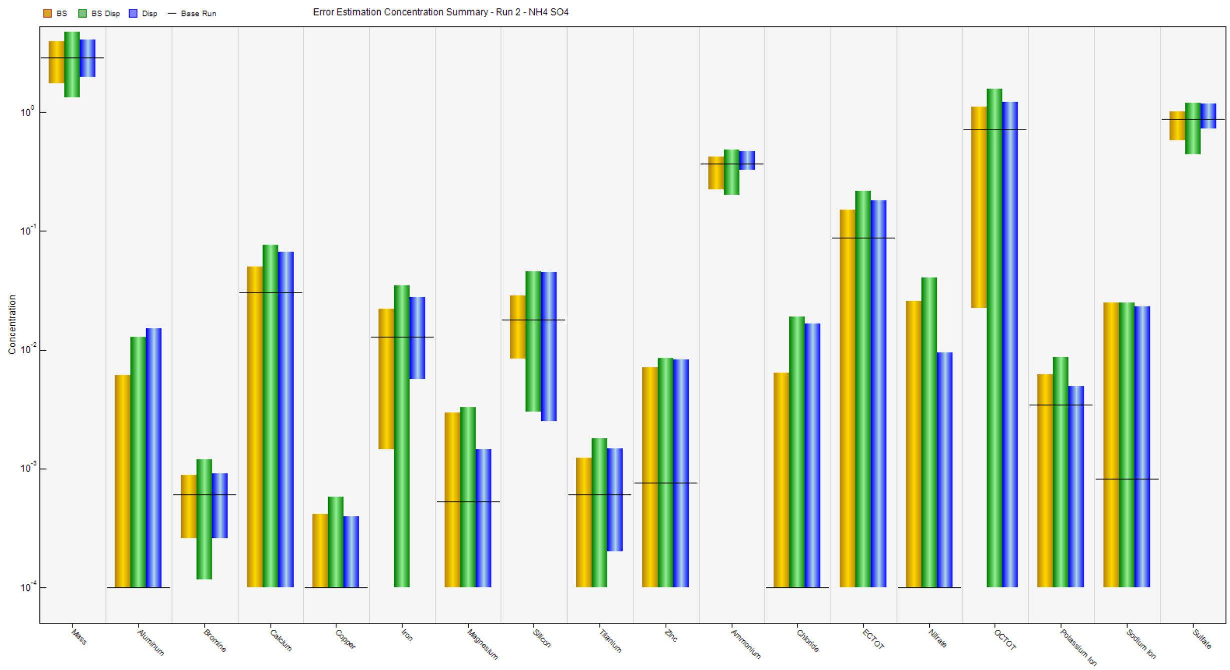

Figure A 5 Combined error estimation for Source Factor 5: ammonium sulfate aged (NH4 SO4 aged).

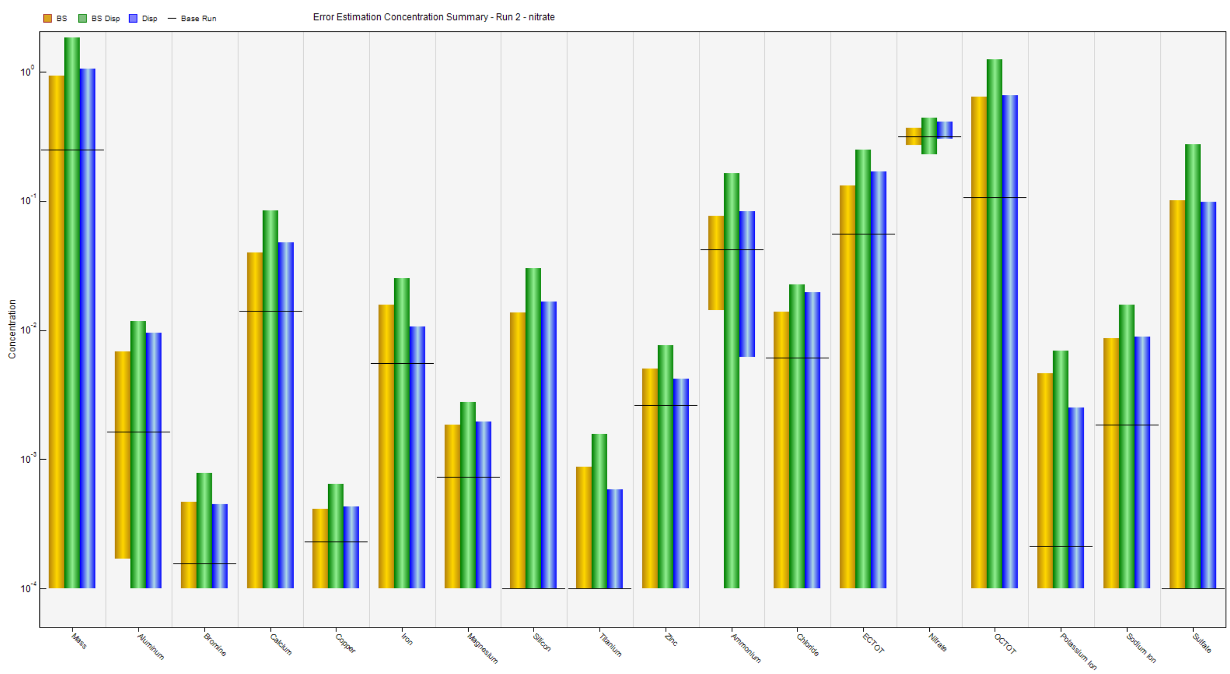

Figure A 6 Combined error estimation for Source Factor 6: nitrate.

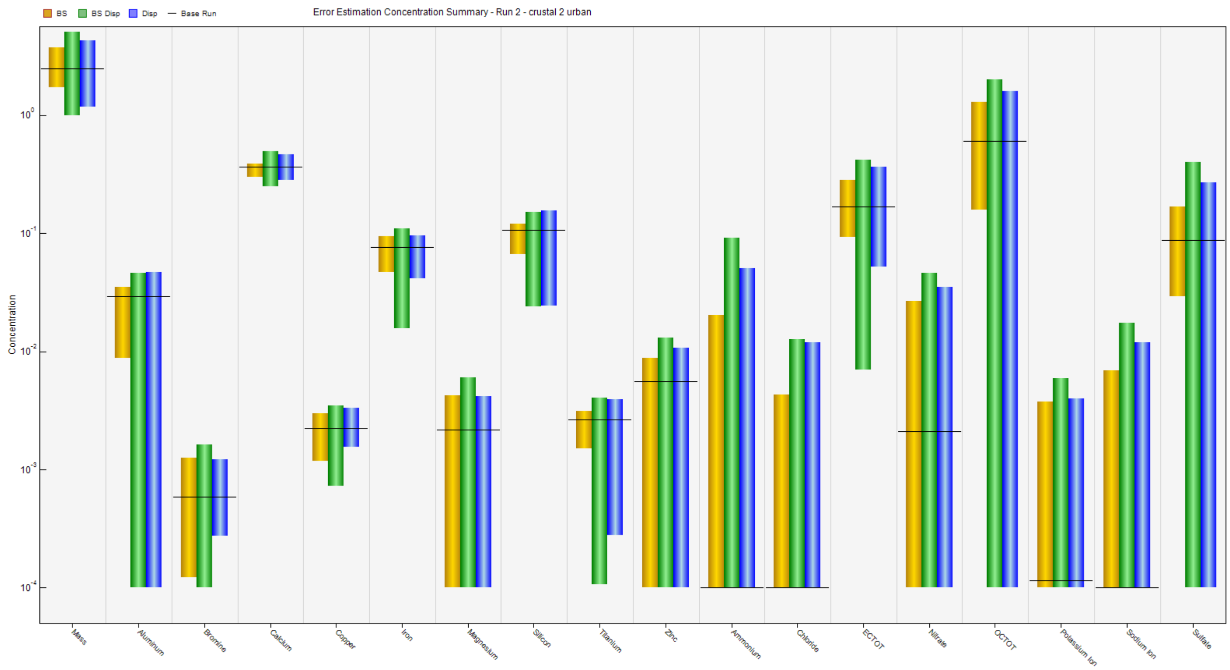

Figure A 7 Combined error estimation for Source Factor 7: Crustal 2 urban.

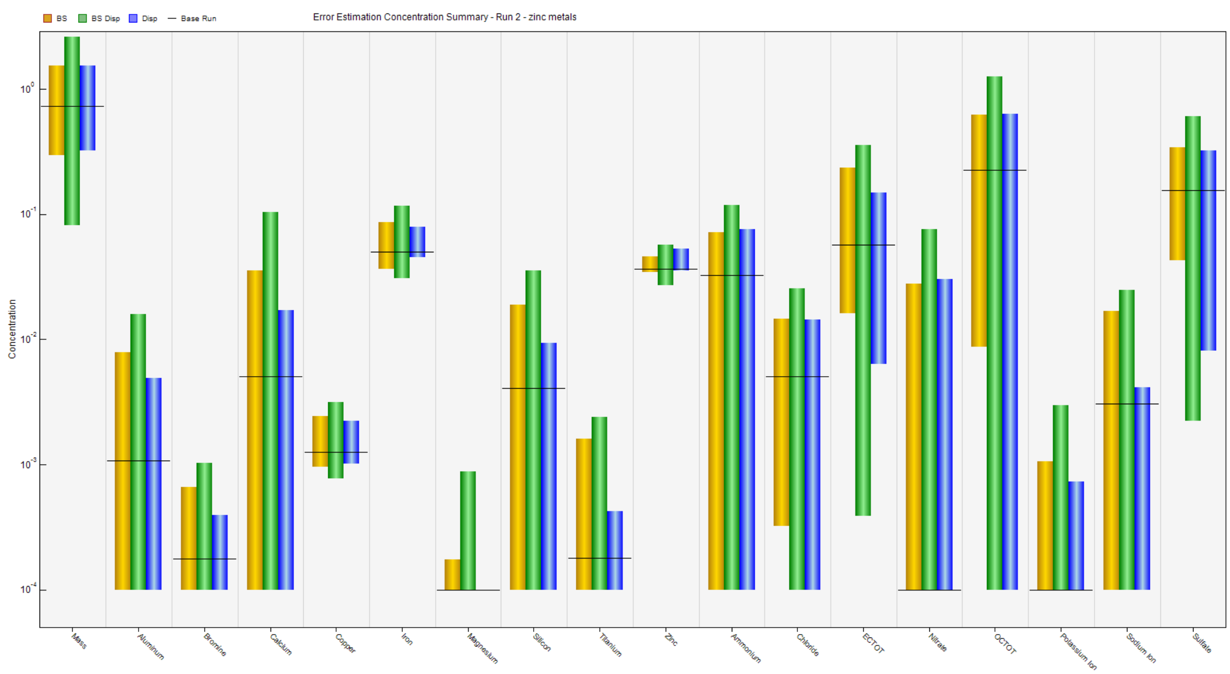

Figure A 8 Combined error estimation for Source Factor 8: Zinc metals.

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Appendix 2:

PMF source contribution dataset by factor and date by mass (µg m^{-3}). All -999 are sample dates that were removed due to the presence of outliers.

	Na Mg SO4	crustal 1	Mg Cl	Urban BB mix	NH4 SO4 aged	nitrate	crustal 2 urban	zinc metals
7/4/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
7/7/2022	0.94	0.50	0.37	3.36	1.15	0.21	1.05	1.41
7/13/2022	1.62	1.73	0.27	1.47	4.63	0.24	1.83	0.53
7/19/2022	0.53	4.14	0.36	-0.23	1.98	0.07	0.45	1.43
7/22/2022	0.12	12.25	0.30	0.65	0.99	0.11	2.21	1.29
7/25/2022	1.02	1.61	0.18	1.30	2.66	0.25	1.04	0.67
7/28/2022	0.93	0.18	0.42	2.29	1.07	0.24	0.90	2.55
7/31/2022	0.10	9.65	0.05	-0.03	0.94	0.15	0.75	0.54
8/3/2022	0.79	5.72	1.60	-0.90	-0.53	0.03	0.49	3.91
8/6/2022	0.21	3.33	-0.01	3.22	1.43	-0.01	0.32	0.25
8/9/2022	1.21	2.12	0.38	1.91	-0.17	0.34	3.41	2.31
8/14/2022	0.77	5.44	0.10	1.33	0.78	0.07	0.35	-0.09
8/18/2022	0.45	1.83	-0.01	4.22	1.42	0.26	1.28	0.83
8/21/2022	1.62	1.05	0.11	0.48	4.77	0.05	-0.16	0.18
8/24/2022	0.29	0.29	-0.01	4.84	2.12	0.07	0.82	0.40
8/27/2022	0.70	-0.12	0.08	2.63	1.26	0.12	1.97	0.73
8/30/2022	0.39	0.00	-0.02	4.35	1.34	0.06	1.58	1.27
9/2/2022	0.26	-0.04	0.00	7.13	1.40	0.14	5.63	0.41
9/5/2022	-0.06	2.66	-0.06	4.38	1.56	0.00	0.12	0.56
9/8/2022	0.05	0.35	-0.01	6.65	2.31	0.06	5.12	0.79
9/11/2022	0.07	-0.17	0.01	8.01	7.64	0.04	2.10	0.31
9/13/2022	0.13	0.52	0.01	5.65	5.06	0.03	7.71	0.81
9/14/2022	0.05	0.11	0.02	6.24	2.61	0.06	5.39	0.42

9/17/2022	0.69	-0.18	0.01	5.07	1.89	0.08	1.43	1.66
9/20/2022	0.48	-0.17	0.01	3.71	1.33	0.12	6.38	1.24
9/21/2022	0.35	-0.08	0.01	4.16	6.29	0.08	5.22	0.32
9/23/2022	0.14	0.23	0.03	6.26	5.68	0.08	10.52	0.85
9/26/2022	0.06	0.37	0.00	5.51	2.18	0.02	4.34	-0.09
9/29/2022	-0.01	0.64	-0.01	6.81	1.05	0.03	5.62	-0.07
10/2/2022	0.00	0.55	0.00	13.22	1.44	0.11	5.19	-0.01
10/5/2022	0.04	1.00	-0.01	5.72	2.41	0.14	11.67	0.34
10/7/2022	-0.02	1.29	-0.01	8.43	2.84	0.15	14.30	0.25
10/8/2022	0.16	1.28	-0.05	10.46	4.29	0.16	7.58	-0.11
10/11/2022	0.42	1.05	0.16	4.46	1.02	0.20	9.41	0.87
10/14/2022	0.14	0.49	-0.05	5.38	1.65	0.07	4.15	1.11
10/17/2022	0.06	0.24	-0.02	5.39	2.27	0.05	1.07	0.03
10/20/2022	-0.16	1.95	-0.01	2.65	0.89	0.23	16.75	0.85
10/23/2022	1.01	0.00	1.06	1.33	0.73	0.04	0.90	-0.08
10/26/2022	-0.16	1.03	0.23	8.96	-0.04	0.14	10.73	0.73
10/29/2022	0.00	-0.15	0.00	4.96	2.52	0.12	0.21	0.06
10/31/2022	-0.13	0.32	0.32	11.35	0.38	0.39	6.35	3.03
11/1/2022	-0.16	-0.19	0.05	1.24	-0.58	0.68	-0.37	13.60
11/3/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
11/4/2022	2.00	0.14	0.86	2.29	1.59	0.07	0.22	0.71
11/7/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
11/9/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
11/10/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
11/13/2022	0.04	-0.05	0.03	6.10	0.98	0.16	2.29	0.10
11/16/2022	0.13	-0.15	0.03	4.25	2.05	0.37	2.32	0.05
11/19/2022	0.06	-0.02	0.00	5.67	2.52	0.66	-0.33	0.05
11/22/2022	0.01	-0.03	0.03	6.35	0.94	0.14	2.22	0.30
11/25/2022	0.02	-0.08	0.01	1.80	0.88	0.02	-0.02	0.03
11/28/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
12/1/2022	0.18	0.42	0.10	1.82	1.18	0.24	5.77	0.34
12/4/2022	0.01	0.51	0.02	4.45	9.40	0.81	-0.26	0.00
12/7/2022	1.94	-0.19	0.87	0.56	4.23	0.37	0.15	1.47
12/10/2022	0.78	-0.02	0.16	2.72	2.83	0.20	0.44	1.01
12/13/2022	2.18	-0.05	1.78	0.02	3.64	0.14	-0.50	2.11
12/16/2022	-0.02	0.48	0.02	4.26	0.93	0.36	3.51	0.88
12/19/2022	0.04	0.08	-0.01	2.44	0.84	0.11	0.44	0.23
12/22/2022	-0.16	0.42	0.53	0.46	8.92	2.85	-0.47	0.64
12/25/2022	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
12/26/2022	-0.16	0.38	0.24	13.24	1.83	1.23	2.18	0.89
12/28/2022	0.28	0.57	1.47	7.15	1.13	0.91	3.58	1.43
1/1/2023	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
1/3/2023	1.15	5.63	1.49	4.74	1.31	-0.05	2.40	0.11
1/12/2023	0.31	1.17	0.10	2.11	2.16	0.26	1.31	-0.06
1/15/2023	1.16	-0.10	1.06	2.76	0.23	0.11	0.40	-0.14
1/18/2023	2.33	0.19	1.30	5.00	2.16	0.20	1.78	0.61
1/21/2023	0.04	0.06	0.05	3.70	1.88	0.40	-0.02	0.23
1/24/2023	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
1/27/2023	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
1/30/2023	-0.01	-0.01	0.03	0.68	5.37	1.02	-0.32	-0.15

2/2/2023	0.00	-0.03	0.02	1.99	0.80	0.25	0.04	-0.02
2/5/2023	0.53	0.29	0.05	4.56	1.74	0.43	2.27	0.21
2/8/2023	1.11	-0.06	0.62	2.88	1.51	0.34	0.55	0.11
2/11/2023	-0.01	0.06	-0.03	3.29	2.36	1.03	0.53	-0.02
2/14/2023	1.82	0.23	1.58	1.87	4.54	0.28	0.53	1.42
2/17/2023	-0.01	0.25	0.00	3.50	0.55	0.21	1.96	-0.02
2/20/2023	1.43	-0.02	1.02	2.52	1.55	0.36	0.78	0.77
2/23/2023	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
2/26/2023	3.96	-0.10	0.45	2.85	4.18	0.32	-0.20	-0.12
2/27/2023	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00	-999.00
3/1/2023	3.82	0.03	1.48	2.35	6.52	0.30	0.29	-0.08
3/4/2023	2.56	0.67	0.03	13.98	-0.58	0.83	3.50	-0.03
3/6/2023	2.13	0.35	0.18	2.92	8.59	-0.05	2.42	1.31
3/7/2023	6.81	0.29	0.56	1.33	3.62	0.48	2.85	0.83
3/10/2023	4.24	-0.19	-0.02	6.95	1.04	0.34	3.13	-0.14
3/13/2023	0.44	0.05	0.00	3.77	-0.09	0.18	4.82	-0.15
3/14/2023	0.00	-0.02	0.01	13.18	2.45	-0.02	4.15	0.21
3/15/2023	0.28	-0.19	0.12	24.44	3.57	-0.05	3.03	1.13
3/16/2023	0.13	0.05	2.06	2.31	4.22	0.29	0.03	0.11
3/19/2023	1.21	0.41	0.18	6.06	-0.55	0.43	1.16	-0.13
3/22/2023	0.19	0.36	2.81	-0.36	4.90	0.36	-0.44	0.18
3/25/2023	0.56	1.53	0.01	5.93	3.81	0.40	1.93	1.28
3/26/2023	1.69	0.44	0.14	3.85	13.93	-0.05	-0.02	0.62
3/28/2023	-0.12	1.73	-0.05	4.91	2.55	0.14	1.33	0.04
3/31/2023	3.56	-0.06	2.59	1.21	5.57	0.36	-0.29	0.41
4/6/2023	-0.02	0.88	0.03	2.21	0.78	0.13	0.29	0.11
4/9/2023	0.01	-0.05	-0.01	2.69	1.83	0.19	0.84	0.07
4/12/2023	0.64	0.41	0.27	3.13	0.49	0.34	5.11	0.03
4/15/2023	1.80	0.44	0.01	-0.90	12.86	0.37	-0.50	0.36
4/18/2023	0.65	2.60	0.41	2.00	1.39	0.41	0.99	0.76
4/21/2023	0.04	0.70	-0.03	2.92	2.39	0.22	0.49	0.05
4/22/2023	-0.06	1.79	0.00	2.41	2.40	-0.01	1.65	0.55
5/9/2023	0.69	0.43	0.13	4.49	4.16	0.24	0.83	0.84
5/12/2023	3.89	4.40	2.69	0.79	2.43	0.22	-0.48	-0.15
5/15/2023	1.48	0.87	0.04	3.13	4.02	0.38	0.18	0.72
5/18/2023	0.06	0.11	-0.01	3.35	4.95	0.15	4.77	0.72
5/21/2023	0.03	-0.17	-0.02	8.66	4.43	0.00	0.38	0.02
5/22/2023	0.01	0.22	-0.02	10.15	6.49	-0.02	2.81	0.32
5/23/2023	0.03	0.02	-0.04	12.60	6.83	-0.02	5.73	0.08
5/24/2023	0.05	-0.01	-0.04	14.74	4.91	0.00	3.44	0.04
5/26/2023	0.00	0.45	-0.05	12.61	3.69	-0.03	5.48	0.59
5/27/2023	0.17	0.12	-0.06	10.98	5.14	-0.02	3.48	1.30
5/30/2023	0.28	0.08	-0.03	5.71	2.40	0.00	3.65	1.71
6/2/2023	0.13	0.29	-0.06	3.92	4.72	0.00	6.72	2.71
6/17/2023	4.89	0.58	0.18	1.85	10.38	0.54	-0.23	0.09
6/20/2023	2.15	8.29	1.65	1.87	-0.55	0.15	0.15	0.28
6/23/2023	2.70	1.50	0.24	0.84	5.95	0.34	0.37	1.68
6/26/2023	1.56	0.85	0.60	-0.23	2.71	0.34	-0.03	2.02
6/29/2023	0.79	1.75	-0.01	0.51	4.27	0.38	0.36	0.87

References