Supplemental of Temporally-Refined Sources of Major Chemical Species in High Arctic Snow

S1 Alternative PMF Solutions

S1.1 Source Apportionment based upon Snow Concentration and Snow Flux per Day

Table S1: Factor compositions based on different snow metrics.

Metric	Snow Concentration							Snow Flux per Day						
Factor	1	2	3	4	5	6	7	1	2	3	4	5	6	7
BC	5	0	66	17	0	8	5	4	0	55	15	3	15	7
MSA	5	0	10	12	27	4	42	7	0	18	7	30	0	39
ACE	0	5	10	79	0	0	6	2	7	7	79	0	0	5
FOR	0	10	0	80	1	8	0	3	11	0	80	0	7	0
Cl^-	79	3	7	2	2	2	5	75	2	5	6	3	4	6
Br^-	33	0	0	26	23	15	4	32	0	0	29	20	15	4
NO_3^-	0	10	4	0	86	0	0	3	8	2	1	79	0	6
SO_4^{2-}	9	4	5	0	4	9	68	10	0	5	0	4	15	67
$C_2O_4^{2-}$	27	8	9	8	14	22	12	25	11	7	5	15	22	15
Na^+	79	4	0	2	0	7	9	75	2	0	7	0	8	8
NH_4^+	15	2	17	47	5	5	8	16	2	13	49	6	7	8
K^+	38	10	0	4	20	10	19	38	7	0	7	19	11	18
Mg^{2+}	43	34	1	5	0	0	17	46	30	1	8	0	0	15
Al	2	84	0	0	3	3	7	3	88	0	0	3	1	4
V	2	84	1	1	3	5	5	3	87	1	1	3	5	2
Cu	6	48	0	0	7	28	11	6	45	0	0	7	33	9
As	5	44	7	0	0	44	0	4	40	7	0	2	48	0
Se	0	81	2	1	0	3	12	0	85	2	0	0	3	9
Sb	0	0	4	18	1	60	17	0	0	4	19	2	60	15
Pb	4	25	8	8	0	53	2	3	21	8	7	0	57	3
Fit with Flux/Period	0.99	0.99	0.97	0.99	0.99	0.98	0.99	1.00	1.00	0.98	0.99	1.00	0.99	0.99

Notes: Pearson's correlation coefficients provided for fit with flux/period solution

Table S2: Factor contributions based on different snow metrics.

Metric	**Snow Concentration**							**Snow Flux per Day**						
Factor	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**1**	**2**	**3**	**4**	**5**	**6**	**7**
mm-dd	**2014**													
09-14	0.0	0.4	0.1	0.6	0.5	1.4	0.3	0.0	0.3	0.3	1.0	0.9	0.9	0.9
09-19	-0.2	-0.2	-0.2	1.1	0.6	-0.2	11.4	-0.2	0.5	-0.2	0.3	-0.2	-0.2	4.7
09-24	0.3	0.5	0.0	0.5	0.3	-0.2	5.3	0.9	-0.2	-0.1	1.6	-0.2	-0.2	16.5
09-29	0.9	-0.2	-0.1	0.8	2.8	-0.2	13.1	0.1	0.3	-0.2	0.2	0.5	-0.2	4.9
10-05	0.1	0.1	0.0	0.8	1.4	0.2	0.4	0.1	0.1	-0.1	0.6	1.2	0.2	0.5
10-07	0.1	0.5	0.4	2.3	2.6	5.0	1.5	-0.2	2.2	-0.2	5.9	7.5	13.5	4.5
10-11	0.0	0.9	-0.2	4.2	1.3	2.7	4.4	-0.1	0.4	-0.1	1.3	0.3	0.8	1.7
10-18	0.6	0.2	1.0	1.1	0.4	0.6	1.5	0.3	0.2	0.7	0.5	0.1	0.2	0.9
10-22	0.2	1.0	0.0	0.2	0.1	1.2	0.1	0.1	0.8	-0.1	0.1	0.1	0.8	0.1
10-26	0.3	0.0	-0.1	0.3	0.4	0.5	3.0	0.2	0.2	-0.2	0.2	0.1	0.3	3.1
11-01	0.2	0.4	0.0	1.3	0.7	1.1	0.3	0.2	0.8	-0.2	1.9	1.2	1.7	0.6
11-06	0.1	0.0	-0.1	0.5	0.2	0.2	0.3	0.1	0.1	-0.2	0.5	0.2	0.3	0.5
11-09	0.1	4.3	1.1	0.1	-0.2	1.7	0.1	0.2	4.5	1.1	-0.1	-0.2	1.4	0.3
11-11	1.6	3.5	2.3	0.0	-0.1	3.0	-0.2	2.3	5.6	3.6	-0.2	-0.2	3.9	-0.2
11-13	1.4	0.1	0.8	0.7	0.4	0.0	0.4	1.9	0.1	1.4	0.8	0.4	0.0	0.7
11-16	0.8	0.5	0.4	0.2	0.2	0.4	0.0	0.7	0.5	0.4	0.1	0.2	0.3	0.1
11-18	1.0	0.2	0.2	0.2	0.1	0.5	0.0	1.3	0.2	0.3	0.2	0.1	0.6	-0.1
11-23	4.4	4.6	0.9	-0.2	0.7	2.0	0.2	1.7	2.1	0.4	-0.2	0.2	0.7	0.2
12-01	2.2	0.9	0.5	0.1	0.1	0.3	0.5	1.3	0.6	0.4	0.0	0.0	0.2	0.4
12-09	0.4	1.3	3.9	0.5	2.7	1.7	0.1	0.1	0.6	1.8	0.1	1.1	0.6	0.1
12-14	-0.2	4.6	5.1	-0.2	6.3	8.7	-0.2	-0.2	4.8	4.3	-0.2	5.9	7.1	-0.2
12-16	0.1	-0.2	1.4	2.3	4.0	0.5	0.9	-0.2	-0.2	2.3	2.9	5.6	0.5	1.4
12-20	0.2	-0.2	1.4	0.5	5.3	1.5	0.4	0.1	-0.1	0.7	0.3	2.9	0.7	0.2

Table S2 (continued): Factor contributions based on different snow metrics.

Metric	Snow Concentration (continued)							Snow Flux per Day (continued)						
Factor	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**1**	**2**	**3**	**4**	**5**	**6**	**7**
mm-dd	**2015**													
01-01	9.4	3.8	-0.2	0.3	0.5	1.8	0.3	9.5	4.4	-0.2	-0.2	0.1	1.7	0.6
01-17	0.3	0.8	1.4	0.3	0.3	0.1	0.3	0.1	0.3	0.6	0.1	0.1	0.0	0.2
01-27	1.0	0.4	0.1	0.1	0.2	0.0	0.0	0.4	0.2	0.0	0.0	0.1	0.0	0.0
02-01	0.5	0.7	0.5	0.3	0.2	0.0	0.2	0.4	0.7	0.6	0.2	0.1	0.0	0.2
02-18	0.1	0.3	1.0	0.3	0.4	0.0	0.1	0.3	0.7	3.2	0.6	1.0	-0.2	0.3
02-21	0.6	0.4	2.8	0.9	2.2	0.3	0.4	0.3	0.3	1.8	0.4	1.2	0.1	0.2
02-28	0.8	0.1	2.3	0.2	0.5	0.0	0.2	0.3	0.0	0.9	0.1	0.2	0.0	0.1
03-05	0.1	0.0	0.5	0.2	0.1	0.1	0.1	0.1	0.1	0.8	0.2	0.2	0.1	0.2
03-08	0.4	0.0	1.5	0.5	0.6	-0.2	0.2	0.4	0.0	1.7	0.4	0.5	-0.2	0.2
03-10	0.1	0.0	2.1	1.3	0.4	0.5	0.4	0.1	0.0	3.8	1.7	0.5	0.6	0.6
03-12	1.9	-0.2	2.4	1.5	0.7	5.0	0.0	2.3	-0.2	3.3	1.7	0.9	5.9	-0.2
03-15	0.6	0.4	3.4	1.0	0.1	4.9	0.2	0.5	0.5	3.4	0.8	0.1	3.9	0.0
03-20	2.0	1.0	2.9	1.2	0.8	0.6	0.1	0.8	0.4	1.5	0.4	0.3	0.2	0.1
03-25	0.6	0.3	0.2	1.2	0.5	0.6	0.1	0.4	0.3	0.2	0.7	0.3	0.4	0.1
03-29	1.0	0.2	0.8	1.3	0.9	0.2	0.4	0.8	0.3	0.9	1.0	0.7	0.2	0.4
04-01	0.6	0.5	3.5	1.8	-0.2	-0.1	0.9	0.3	0.4	3.1	1.1	-0.2	-0.1	0.7
04-04	5.0	0.8	0.7	2.4	0.2	-0.2	0.6	7.1	1.3	1.9	2.8	-0.1	-0.2	1.1
04-11	2.4	0.6	0.5	0.9	2.6	1.0	-0.2	1.2	0.4	0.2	0.4	1.4	0.5	-0.1
04-14	0.8	0.3	0.5	2.7	0.0	0.6	0.2	0.9	0.5	1.2	3.5	-0.2	0.9	0.4
04-18	0.9	0.4	0.6	0.6	0.7	0.6	0.3	0.9	0.5	0.7	0.4	0.7	0.5	0.4
04-22	0.5	5.6	2.8	0.2	0.7	0.9	0.4	0.5	6.1	3.2	-0.1	0.7	0.7	0.7
05-12	0.7	0.3	-0.1	1.9	1.0	0.0	0.0	1.9	1.1	-0.1	5.3	3.4	0.0	0.3
05-13	3.5	0.2	-0.2	1.7	2.0	-0.1	-0.2	9.0	0.6	-0.2	3.7	5.3	-0.2	-0.2
05-16	-0.2	8.5	0.3	5.2	0.0	-0.2	-0.2	-0.2	5.1	0.4	2.3	-0.1	-0.1	0.2
05-26	0.3	0.1	-0.2	2.2	1.1	0.1	0.3	0.2	0.2	-0.2	2.7	1.6	0.1	0.5
Fit Flx/Per	0.82	0.88	0.60	0.69	0.69	0.74	0.82	0.87	0.90	0.83	0.65	0.75	0.82	0.60

Notes: Pearson's correlation coefficients provided for fit with flux/period solution

S1.2 Four and Six-Factor PMF Solutions

The four-factor PMF solution is described below (Figure S1). Factors 1, 2, and 3 were practically identical to their counterparts in the seven-factor solution. Factor 4 of the four-factor solution was found to be characterized by several major ions, fall/spring peaks, and local Arctic source areas. Thus, Factor 1 was identified as a sea salt source, Factor 2 as a crustal source, Factor 3 as a combined long-range transport source, and Factor 4 as a mixture of aged transported emissions and local emissions.

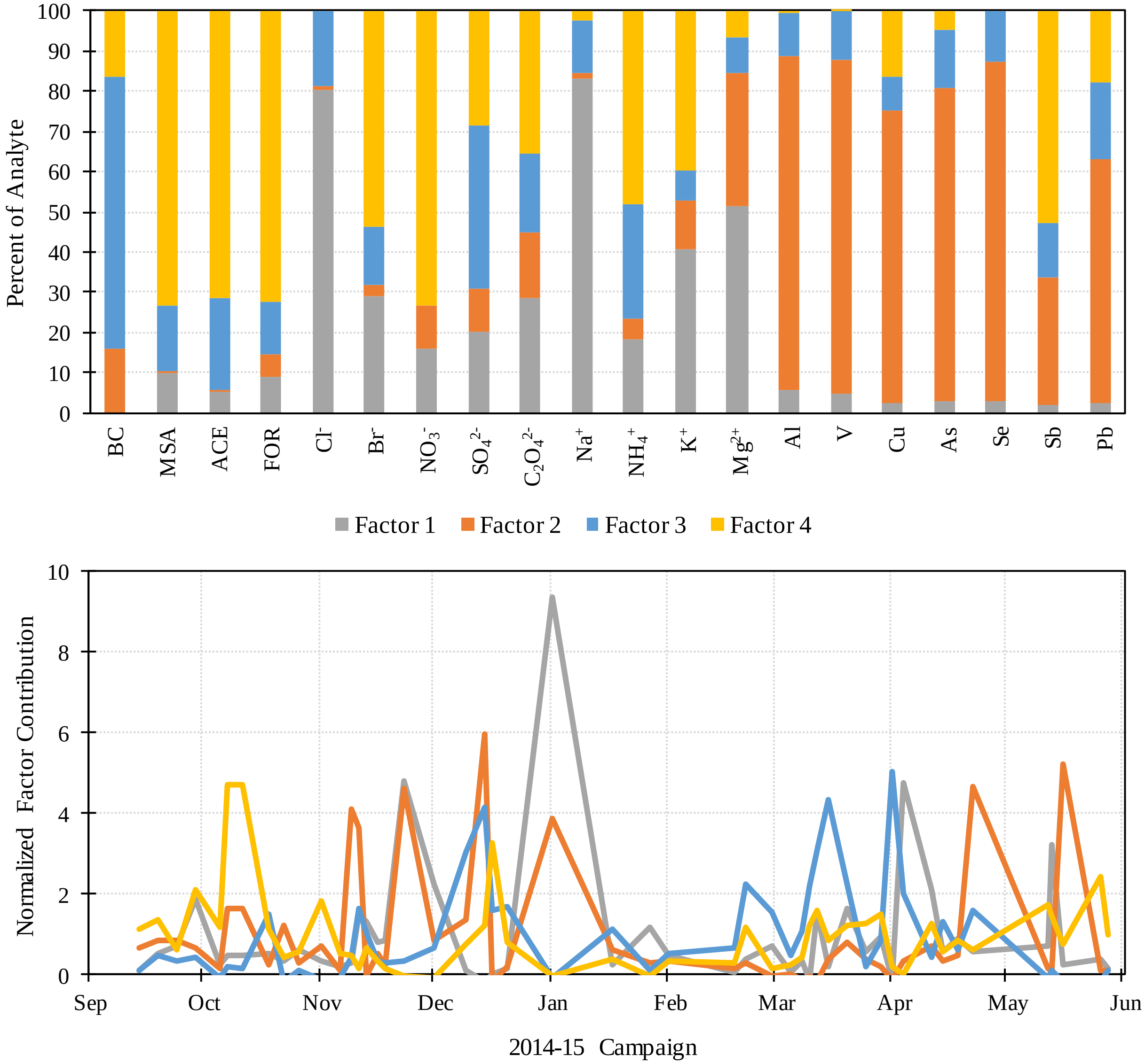

Figure S1: Four-factor PMF solution: Factor compositions and contributions.

The six-factor PMF solution is described below (Figure S2). The six-factor solution was found to be essentially identical to that of the seven-factor solution, with the exception of that Factors 6 and 7 of the seven-factor solution combined to form Factor 6 of the six-factor solution. As discussed in Chapter 5, this new Factor 6 was not as readily interpretable as the split factors. The six-factor solution however provided the largest improvement in Q values and prediction accuracy with the addition of a factor, thus was included here as a potential solution.

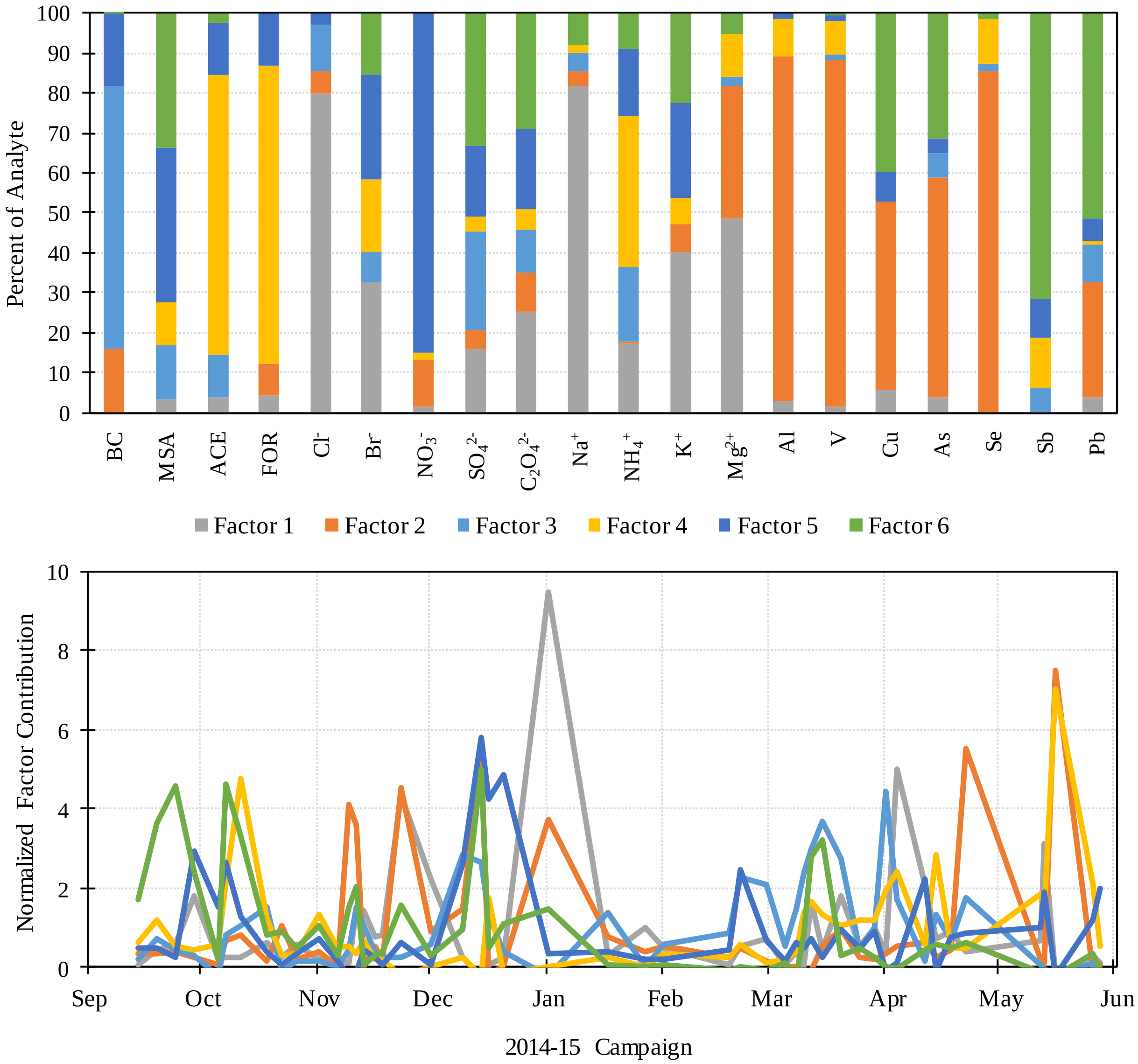

Figure S2: Six-factor PMF solution: Factor compositions and contributions.

S2 Seven-Factor Solution Parameters

S2.1 Optimal Solution Selection

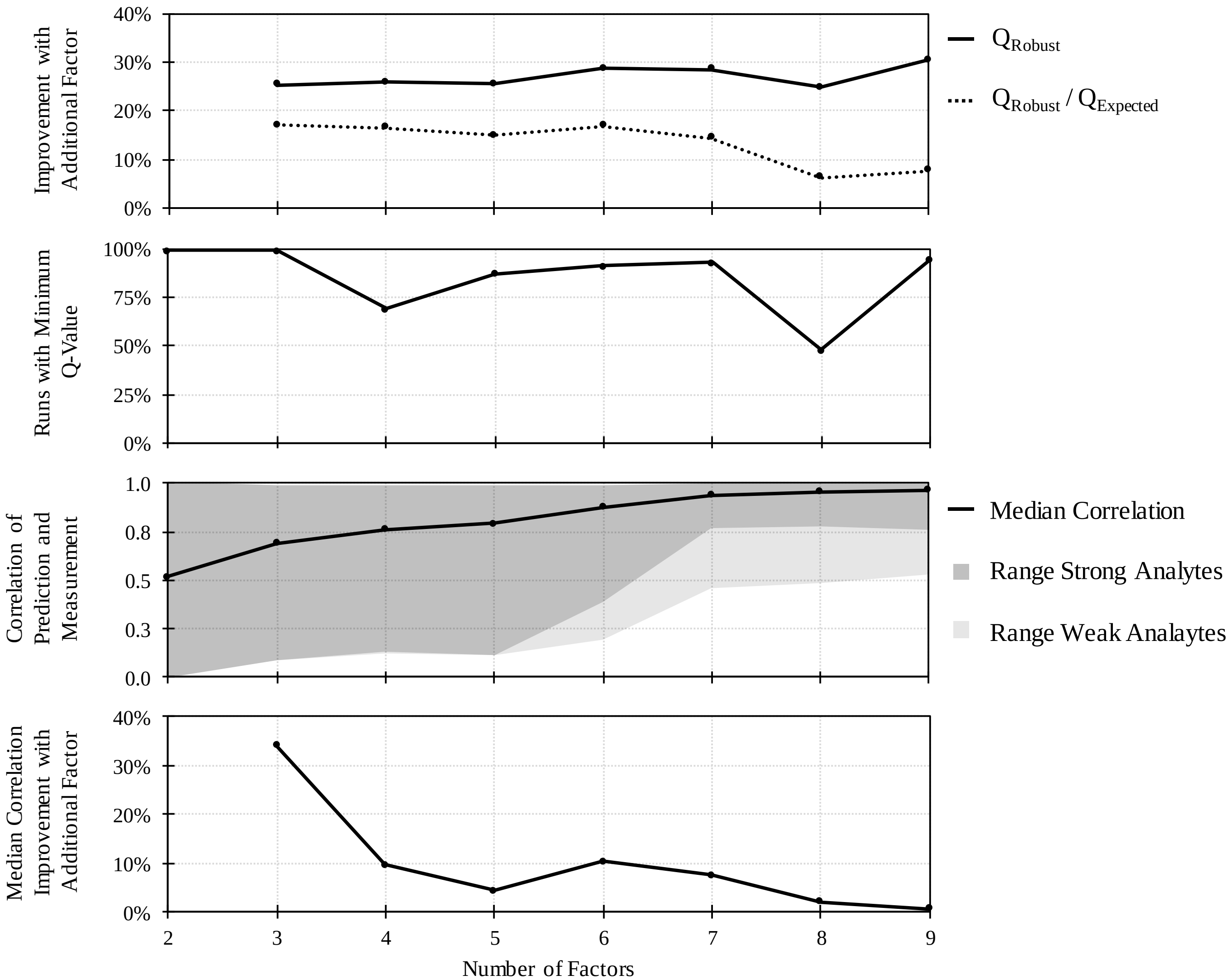

Figure S3: PMF Flux per Period Solution Parameters across the Number of Factors Used. Agreement of measurements and PMF prediction: line depicts median Pearson's correlation coefficient, dark shaded area depicts range of correlations for strong analytes and light shaded area range for weak analytes.

S2.2 Fit of PMF Seven-Factor Solution

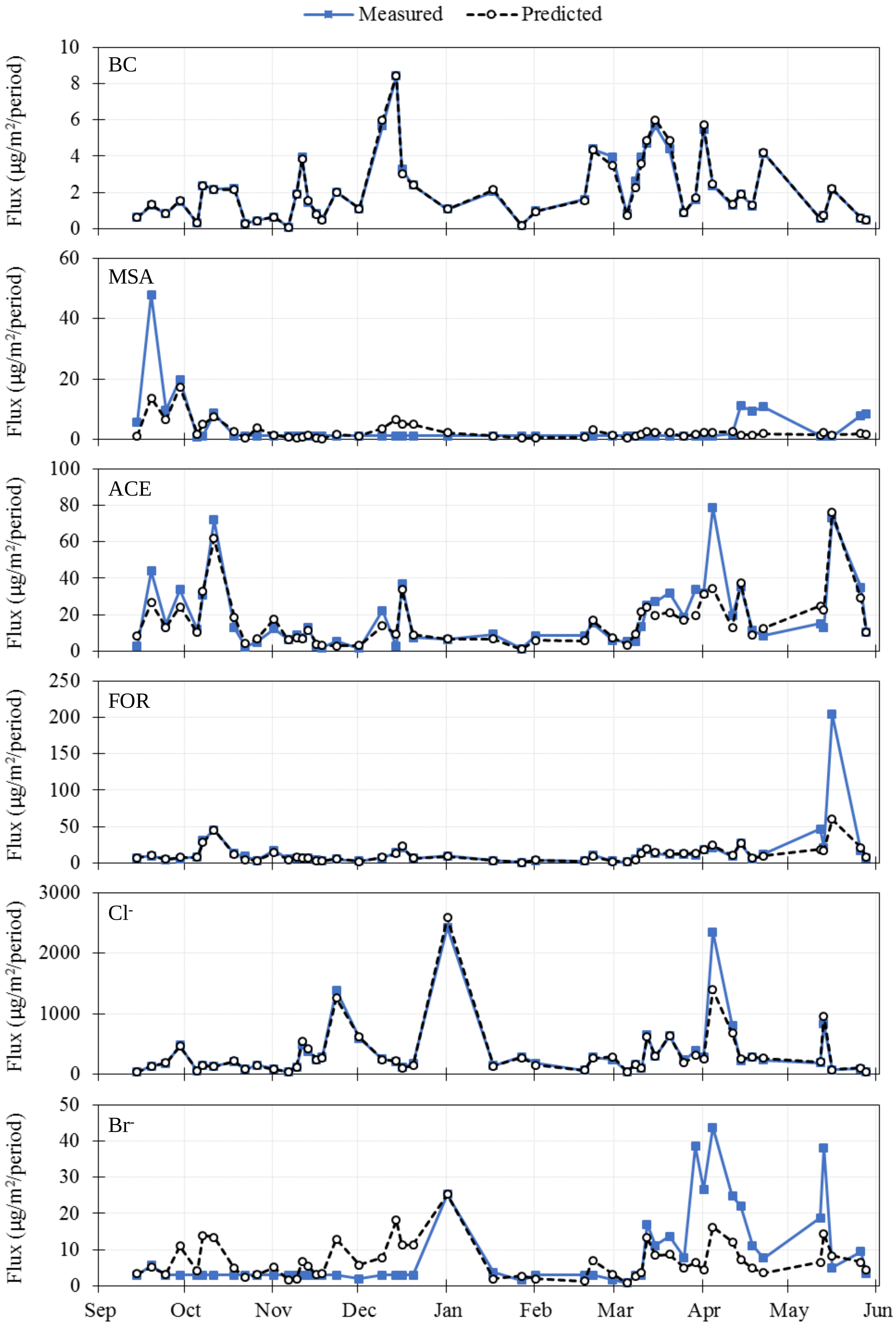

Figure S4: Measured and predicted fluxes of seven-factor PMF solution.

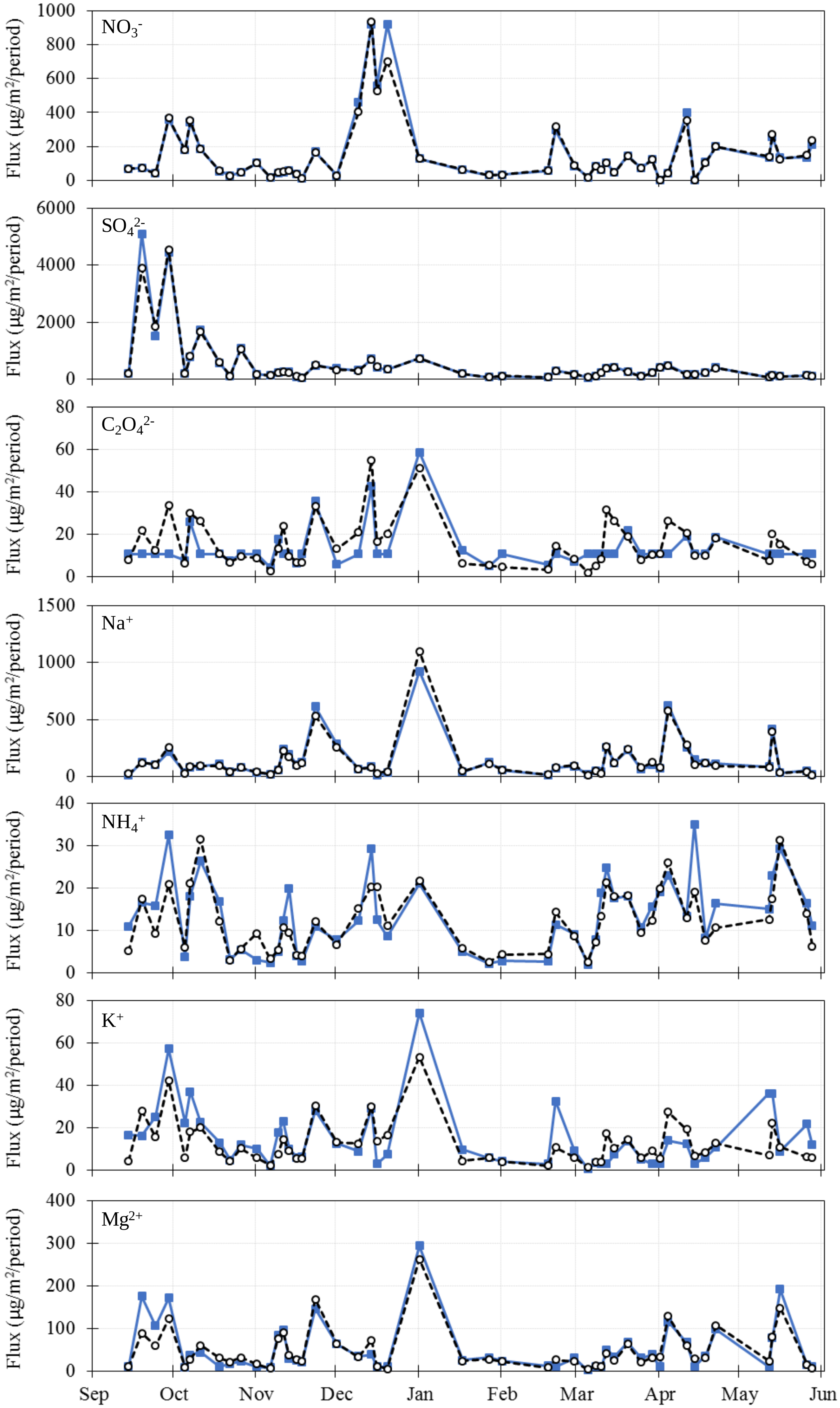

Figure S4 (continued): Measured and predicted fluxes of seven-factor PMF solution.

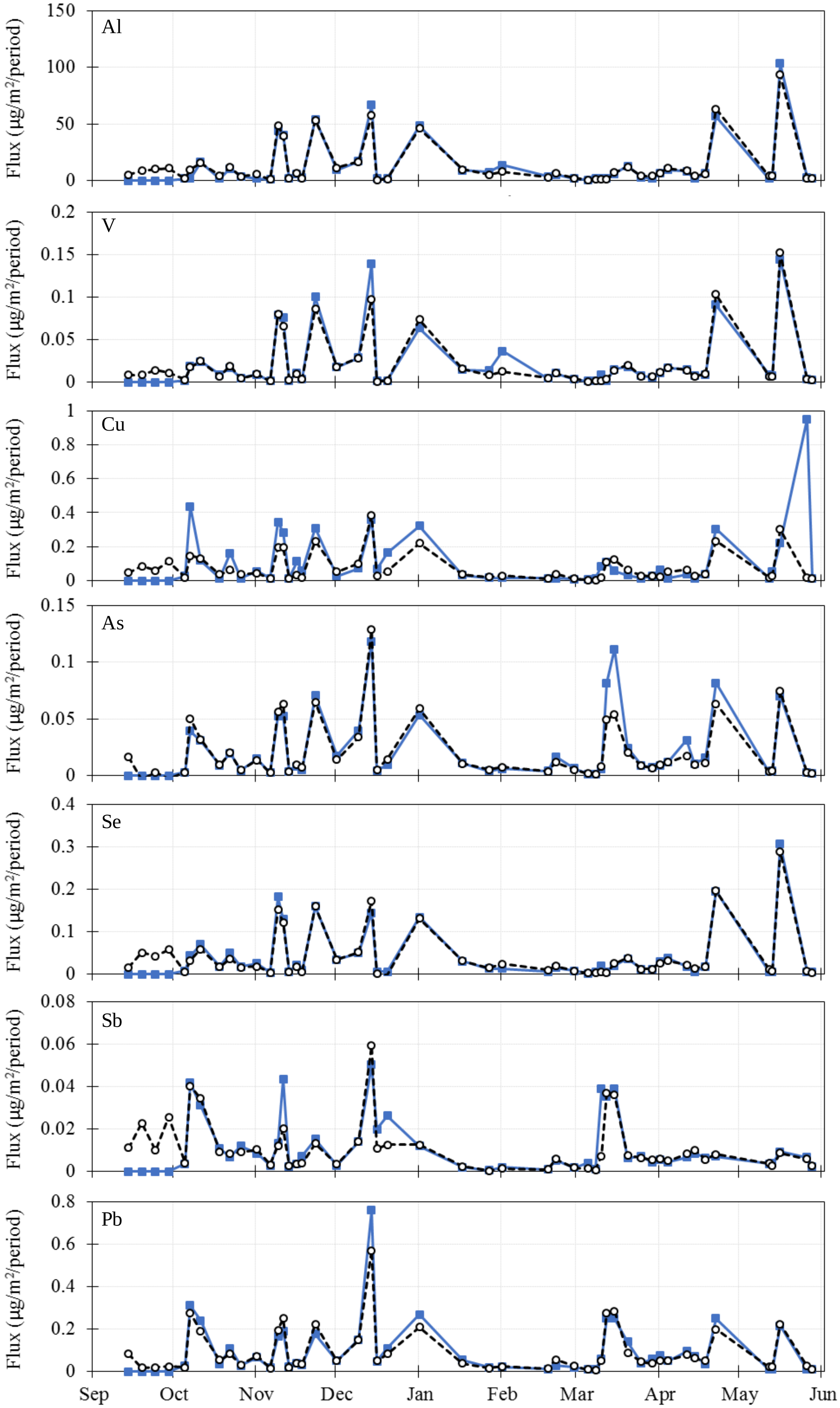

Figure S4 (continued): Measured and predicted fluxes of seven-factor PMF solution.
Notes: All missing values plotted as measured medians.

S2.3 Evolution of PMF Solution Factor Composition

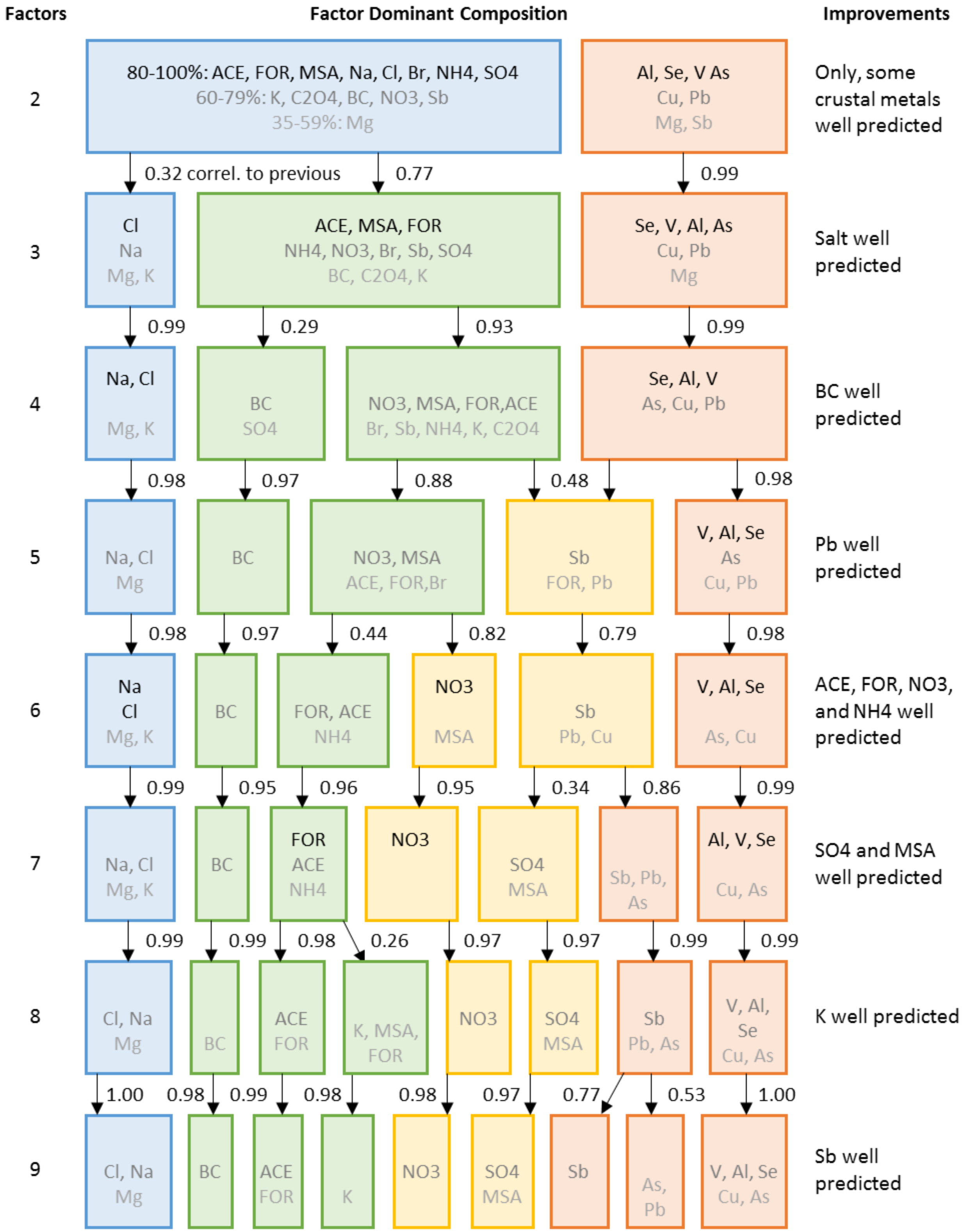

Figure S5: Evolution of factor composition of 2 to 9-factor PMF solutions.

S3 PMF Solution Sensitivity and Validation

S3.1 Solution Sensitivity to Excluded Analytes

Three analytes, which matched the conditions set in section 2.4.1, were excluded from this PMF analysis for simplicity and to increase the measurement/analyte ratio: Ca^{2+}, propionate, and H^+. While bootstrapping analysis explores the impact of removing particular measurements from the apportionment it cannot address the impact of adding additional analytes to the run. Thus, repeated PMF runs were completed including each of these analytes in turn to assess what impact they may have on the solution. All factor profiles were maintained in the augmented runs with Pearson's correlation coefficients of 0.93 or greater, with the exception of Factor 7 (Sulphate) in the H^+ run which correlated by only 0.59. Inclusion of H^+ caused MSA to no longer be loaded onto Factor 7, instead distributing MSA among the other factors. Since this H^+-augmented solution caused MSA to be loaded onto the clearly anthropogenic Factor 6 it was considered to be less interpretable than the base solution. However, this result may indicate that the H^+ associated with Factor 7 is mostly related to volcanic and/or smoking hills sources which were suggested to coincide with a marine biogenic SO_4^{2-} source. The time series of Factor 7 (Sulphate) was found to be fairly consistent even with the addition of H^+ with a correlation of 0.77. Therefore, the exclusion of these analytes was considered to be acceptable.

S3.1 Principal Component Analysis Validation

A brief principal component analysis (PCA) was completed to corroborate the PMF findings, using identical input data. The first seven principal components identified by PCA were found to explain 89% of the measured variance and agree well with the PMF factors, although a perfect correlation is not expected given the lack of non-negative constraint on the PCA results. The PCA and PMF predicted compositions agreed with Pearson's correlation coefficients of 0.39 to 0.77 and contribution time series with coefficients of 0.37 to 0.80, with the crustal and salt factors showing the best agreement. In particular, the analytes found by PMF to be dominant for each factor were generally well reflected in the PCA solution. The PCA solution found the component similar to the identified crustal and salt factors to explain the largest portion of measured variability followed by the components which resembled Factors 4 (Carboxylic Acids), 6 (Non-Crustal Metals), 3 (BC), and 5 (Nitrate), from most to least variance explained. Thus, the PCA results provided some corroboration to the PMF solution.

Table S3: PCA solution eigenvalues, eigenvectors, and contributions over campaign.

Principal Component	PC1	PC2	PC3	PC4	PC5	PC6	PC7
Eigenvalue	6.8492	3.2453	2.3119	2.0279	1.7876	0.9174	0.7215
Eigenvectors							
BC	0.19	-0.24	-0.06	-0.27	-0.28	0.33	0.01
MSA	0.01	0.21	-0.47	-0.12	0.29	0.16	-0.16
ACE	0.11	0.16	-0.39	0.15	-0.39	-0.14	-0.10
FOR	0.16	-0.06	-0.26	0.46	-0.19	-0.23	0.21
Cl-	0.18	0.38	0.30	-0.03	-0.14	0.11	-0.01
Br-	0.06	0.32	0.11	0.01	-0.48	-0.04	-0.05
NO3-	0.14	-0.17	-0.06	-0.35	-0.04	-0.33	0.63
SO4=	0.06	0.24	-0.45	-0.23	0.28	0.15	0.03
C2O4=	0.29	0.08	0.25	-0.18	0.14	-0.07	0.07
Na+	0.20	0.38	0.29	-0.05	-0.04	0.11	-0.06
NH4+	0.23	0.14	-0.29	-0.11	-0.32	-0.10	0.02
K+	0.20	0.26	0.06	-0.19	0.23	-0.35	0.18
Mg++	0.27	0.32	-0.05	0.12	0.21	0.13	0.04
IS Al	0.32	-0.11	0.02	0.31	0.13	0.08	0.12
IS V	0.33	-0.15	0.03	0.22	0.14	0.10	0.08
IS Cu	0.20	-0.09	0.04	-0.02	0.15	-0.61	-0.58
IS As	0.31	-0.19	0.03	-0.06	-0.04	0.28	-0.19
IS Se	0.31	-0.11	-0.01	0.34	0.14	0.07	0.01
IS Sb	0.20	-0.24	-0.07	-0.33	-0.13	-0.02	-0.28
IS Pb	0.31	-0.21	0.00	-0.20	-0.03	0.07	-0.02
Component Contribution							
9/14/2014	-1.777	-0.381	0.1027	0.1039	0.804	-0.159	0.0959
9/19/2014	-0.004	3.7118	-5.881	-1.286	3.0331	1.7229	-0.745
9/24/2014	-0.788	1.2373	-1.006	-0.222	1.3064	0.181	0.0384
9/29/2014	1.2476	3.7531	-3.676	-2.13	1.9661	-0.351	1.2667
10/5/2014	-2.193	-0.275	0.4071	0.1777	0.7021	-0.678	0.6511
10/7/2014	2.2723	-1.207	-0.507	-1.96	0.0395	-2.298	-0.913
10/11/2014	1.733	-0.062	-3.131	-0.257	-1.023	-0.633	-0.419
10/18/2014	-1.281	-0.188	-0.087	-0.135	-0.186	0.1674	0.0714
10/22/2014	-1.487	-1.035	0.6771	0.8351	0.936	-0.081	-0.573
10/26/2014	-1.906	-0.076	0.251	-0.012	0.9286	0.1673	0.0169
11/1/2014	-1.749	-0.737	0.3842	0.4436	0.4989	-0.195	0.0619
11/6/2014	-2.915	-0.546	0.4566	0.7499	0.5667	0.1511	-0.067
11/9/2014	2.1621	-1.657	0.8076	1.5717	1.8824	0.07	-0.924
11/11/2014	2.9388	-1.462	0.6757	-0.003	0.687	0.6053	-1.397
11/13/2014	-1.482	0.5422	0.2844	0.0877	-0.236	0.1044	0.1716
11/16/2014	-1.977	-0.407	0.8266	0.6004	0.7078	0.0604	-0.303

Table S3 (continued): PCA solution eigenvalues, eigenvectors, and contributions over campaign.

Principal Component	PC1	PC2	PC3	PC4	PC5	PC6	PC7
Component Contribution (continued)							
11/18/2014	-2.14	-0.214	1.0614	0.3649	0.6969	0.1541	-0.261
11/23/2014	4.9011	0.9812	2.7053	0.6318	1.937	0.6514	-0.359
12/1/2014	-1.071	0.5607	1.0837	0.5232	0.7537	0.7392	0.0061
12/9/2014	0.591	-1.722	-0.206	-0.971	-0.662	0.5508	0.8866
12/14/2014	8.4113	-4.899	0.1763	-3.536	0.0949	0.1842	1.1751
12/16/2014	-0.962	-1.26	-0.938	-0.967	-1.103	-0.808	1.1113
12/20/2014	-0.641	-1.883	-0.01	-2.218	-0.131	-1.548	1.8145
1/1/2015	7.3992	5.5857	4.206	-0.591	1.7431	-0.505	0.1849
1/17/2015	-1.545	-0.573	0.5883	0.3957	0.5222	0.3855	0.181
1/27/2015	-2.345	-0.138	0.9806	0.8369	0.9214	0.3837	0.0971
2/1/2015	-1.817	-0.558	0.7511	0.9225	0.743	0.4697	0.1679
2/18/2015	-2.603	-0.716	0.454	0.5469	0.3575	0.3799	0.1046
2/21/2015	-0.755	-0.418	0.1606	-0.832	-0.121	-0.19	1.1472
2/28/2015	-1.797	-0.484	0.4269	-0.143	-0.027	0.7385	0.2814
3/5/2015	-2.796	-0.738	0.6729	0.4513	0.6189	0.3021	-0.047
3/8/2015	-2.203	-0.557	0.5125	0.1383	0.0826	0.4174	0.2675
3/10/2015	-0.742	-1.469	-0.429	-0.913	-0.981	0.2232	-0.841
3/12/2015	1.4551	-0.538	-0.135	-1.555	-2.43	1.136	-1.395
3/15/2015	1.5565	-1.786	-0.363	-1.538	-1.895	1.7849	-1.537
3/20/2015	0.8284	0.579	0.3942	-0.349	-1.188	0.6161	0.2933
3/25/2015	-1.673	-0.078	0.2275	0.4618	-0.253	0.0499	0.0304
3/29/2015	-1.073	1.1995	0.1091	0.3616	-2.274	-0.089	-0.022
4/1/2015	-0.555	0.06	-0.389	0.0803	-2.365	0.664	-0.397
4/4/2015	1.6846	4.9962	0.7832	0.8217	-3.982	0.5339	-0.388
4/11/2015	0.2854	1.1339	1.0431	-0.477	-0.995	-0.165	0.8896
4/14/2015	-0.63	1.0493	-1.468	0.1646	-2.183	0.0817	-0.495
4/18/2015	-1.413	0.2557	0.1022	0.1346	0.2222	0.4202	-0.135
4/22/2015	3.7308	-1.593	-0.156	1.0841	1.2759	1.1381	-0.506
5/12/2015	-1.181	0.8304	0.1325	0.5419	-0.57	-1.295	0.9403
5/13/2015	0.0918	3.0195	1.1441	-0.173	-1.556	-1.032	0.8812
5/16/2015	6.7261	-1.775	-3.354	7.1336	-0.942	-0.646	1.2897
5/26/2015	-0.374	0.0607	-0.606	0.1098	0.4894	-4.202	-2.983
5/28/2015	-2.139	-0.125	-0.246	-0.007	0.5846	-0.359	0.5833
Comparison with PMF Results - Pearson's Correlation Coefficient							
Similar Factor	2	1	4	6	7	3	5
Composition	0.68	0.77	-0.50	-0.39	0.55	0.40	0.59
Contribution	0.80	0.67	-0.54	-0.49	0.37	0.39	0.53