Supplement to Aerosol composition and sources in the Central Arctic Ocean during ASCOS

R. Chang et al.

September 13, 2011

S1 Solutions from positive matrix factorisation

For this study, a four factor solution was determined to best represent the measured aerosol. A two factor solution separates the Ship Emissions factor from the ambient background aerosol, while a three factor solution includes the Organic factor. It is only with four factors that the Marine Biogenic factor is separated from the Continental factor. Additional factors either identify instrumental noise, or split the existing factors. Figure S1 shows the decrease in Q/Q_{exp} as additional factors are included, where Q_{exp} is the expected Q. We see that including a 5^{th} factor decreases Q/Q_{exp} by identifying instrumental noise, while additional factors only serve to capture episodic events, often coinciding with ship emissions. Even though Q/Q_{exp} decreased slightly from 3.36 for four factors down to 3.06 for 10 factors, including more factors did not contribute additional information about the measured aerosol. As such, the four factor solution was deemed to give the most information about the measured ambient aerosol.

The robustness of the solution can be explored by either varying the initial seed, which changes the set of pseudorandom values used for the initial point (Paatero, 1997), or by using bootstrapping analysis, in which the rows of $\mathbf{X}$ are randomly sampled and PMF is executed on the new dataset (as described by Reff et al., 2007). Both of these methods were used and the four factor solution at *fPeak* = -0.75 was found to be robust: 100 values for the initial seed parameter in the PMF2 program resulted in 90 of the cases giving the solution presented here, while 100 iterations of the bootstrapping analysis

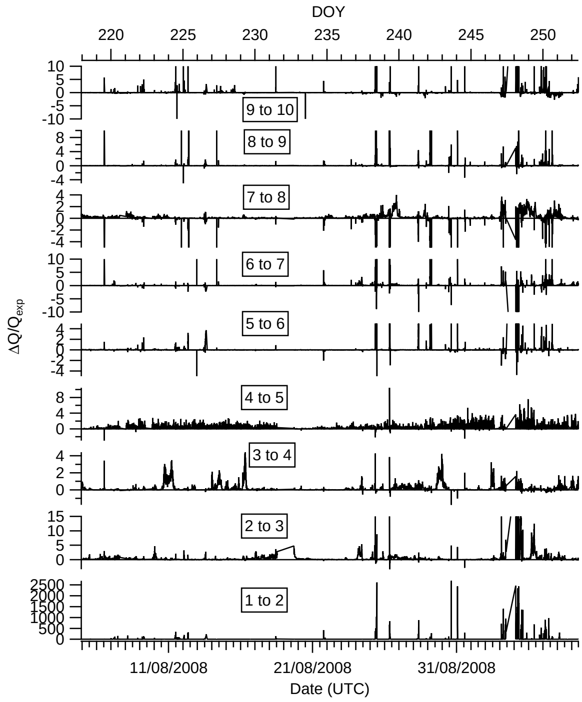
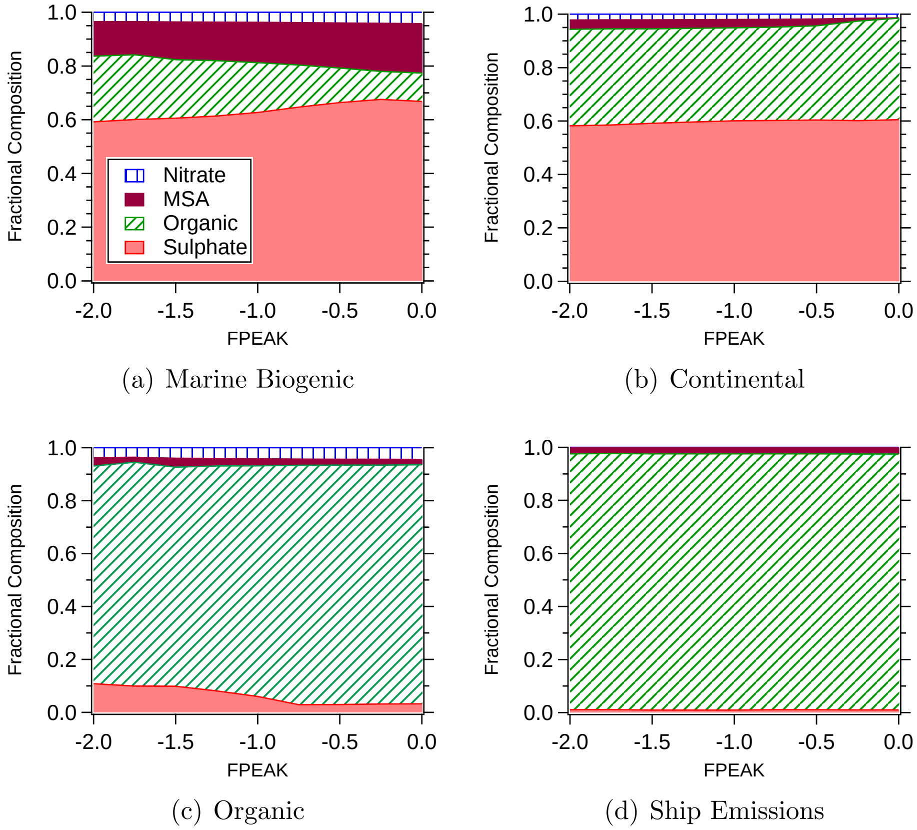


Figure S1: The decrease in Q/Q_{exp} as additional factors are included in the PMF solution.

resulted in deviations of < 0.01 (fraction of signal) in the mass spectra and < 0.015 μg m^{-3} in the time series.

Although the solution at *fPeak* = -0.75 is robust, a range of *fPeak* = -1.5 to 0 provide physically reasonable solutions. The degree to which the composition and F44 are dependent on the solution can be seen in Figs. S2 and S3.

Analyses were also performed on a data matrix calculated by adding together the mass spectra of the species of interest (i.e. nitrate, sulphate, organic and MSA) in nitrate equivalent mass with a corresponding error matrix calculated from the individual errors added in quadrature. However, results from the initial runs were similar enough to those calculated from the method described in the main text that only the latter method was pursued.



(a) Marine Biogenic (b) Continental

(c) Organic (d) Ship Emissions

Figure S2: Changes in fractional composition of the factors with varying *fPeak*.

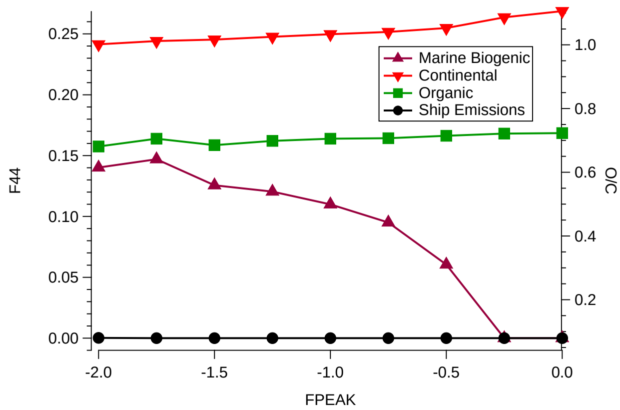


Figure S3: Changes in the F44 with varying *fPeak*.

References

Paatero, P.: Least squares formulation of robus non-negative factor analysis, Chemometr. Intell. Lab., 37, 23–35, 1997.

Reff, A., Eberly, S. I., and Bhave, P. V.: Receptor Modeling of Ambient Particulate Matter Data Using Positive Matrix Factorization: Review of Existing Methods, J. Air Waste Manage., 57, 146–154, 2007.