Supporting Information Appendix

Total Potential Source Contribution Function Analysis of Trace Elements Determined in Aerosol Samples Collected near Lake Huron

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Table 3
Average Criterion Values Used for PSCF and TPSCF Calculations

Element	Yearly Used as Criterion Value (ng/m^3)	Summer Used as Criterion Value (ng/m^3)	Winter Used as Criterion Value (ng/m^3)
Al	44.9450	57.6574	30.7587
Br	2.4106	1.7970	2.7939
Cl	41.4954	50.2319	34.3905
Fe	53.7327	63.9819	42.2952
I	0.4309	0.3349	0.5380
Na	40.7062	23.0893	61.0192
V	0.7006	0.9844	0.3666

Aluminum

The majority of the aluminum in the Burnt Island atmosphere is a result of crustal entrainment.(*24*) While re-entrained crustal particles are normally larger in size and do not have a high tendency for long-range atmospheric transport, the long-range transport of crustal materials does occur.(25,26) Therefore, we expect the majority of the aluminum on the Burnt Island atmosphere to result from distributed sources not too far

from the station. In addition to the crustal aluminum, some fine aerosol particles (< 2.5 μm) are also emitted from coal-fired power plants. Figure 7 shows the seasonal distribution of atmospheric Al concentrations and the three Al TPSCF plots. Due to the strong seasonal dependence on Al concentrations, Figure 7b illustrates the source of atmospheric Al during the months of April, May, and August when Al concentrations are at their highest. These source regions appear to be related to areas of high agricultural production. Figures 7b and 7c show the likely source regions for the Summer, and Winter, respectively. Most of the high-potential TPSCF cells for the seasonal plots represent the boundary regions of the trajectories along the northwest and south. This pattern is suggestive of local sources.

Bromine

The atmospheric Br concentrations at Burnt Island are generated from a mixture of sources. The main source is the sea salt component, but Br may also result from urban sources. The seasonal atmospheric concentrations and TPSCF plots for Br are shown in Figure 8. The summer TPSCF values, with the criterion value equal to the average Br concentration during the summer months, show the New York City area as being a strong source region. The winter Br TPSCF, with the criterion value equal to the average Br concentration during the winter months, shows source regions around the Georgia/Alabama border, the northern region of Quebec, and the southwest coast of Hudson Bay. The source regions on the Hudson Bay is particularly interesting in that it

suggests that Br may be emitted from the spring algae bloom.(*27*) The seasonal trend exhibited by Br at Burnt Island with a peak in the spring is similar to data in a less polluted site in Alaska.(*28*)

Chlorine

Chlorine predominately results from salt in the atmosphere. The salt in the aerosol at Burnt Island may originate from sea spray or may be the product of salt put on roads in the winter. Studies conclude that long-range transport of sea-salt aerosols is possible after small particles are formed from breaking of ocean waves.(*29,30*) Figure 9 shows seasonal atmospheric concentrations and the TPSCF maps for Cl at Burnt Island. Chlorine has a very strong seasonal variation. As a result, the yearly average TPSCF calculation shows the air trajectory likelihood for the winter months of the year when the Cl concentrations are high. These source regions appear to be in the southern Gulf of Mexico region and the northern regions of Canada. The summer Cl TPSCF shows source regions along Appalachian Mountains of the United States with a particular high probability around the Tennessee/North Carolina border. The winter Cl TPSCF shows that the Cl source is most likely from northern Canada during this period.

Iron

Iron is observed mostly as a crustal material at Burnt Island. However, the variance of iron is also partially explained from urban sources. Figure 10 displays the seasonal

atmospheric concentrations and TPSCF plots for Fe at Burnt Island. The yearly average TPSCF for Fe shows nearly no unique source locations. However, the summer and winter TPSCF results do reveal high probability TPSCF regions. During the summer months, Fe has high source probability in the region around Alabama, Mississippi, and Tennessee. This likely represents the steel industry in this region. In addition there is a high probability region in Manitoba, Canada. The winter Fe TPSCF shows likely source regions around Alabama and Mississippi in the United States as well as northern Quebec in Canada.

Iodine

At Burnt Island, most of the variance in atmospheric iodine concentrations is accounted by the sea salt source.(*14*) Figure 11 shows the seasonal atmospheric concentrations, yearly average, summer and winter TPSCF figures for Burnt island. The yearly average TPSCF plot shows the major high probability source region for I is in the mid-Atlantic region of the United States. This source region starts in the state of New York, goes down through Pennsylvania, and continues to run down the Appalachian Mountains to through Virginia, West Virginia, North Carolina, and Tennessee. This compares well with the results in Cheng and Hopke (*31*) where I was shown to be a marker for coal in the Washington, D.C. area. There are some other I source regions including ones in Mississippi, Kansas, Montana, Quebec, and the Northern Territories. The summer and winter TPSCF plots show subsets of the yearly average TPSCF. The summer I TPSCF shows the mid-Atlantic source region in New York and Pennsylvania as well as the

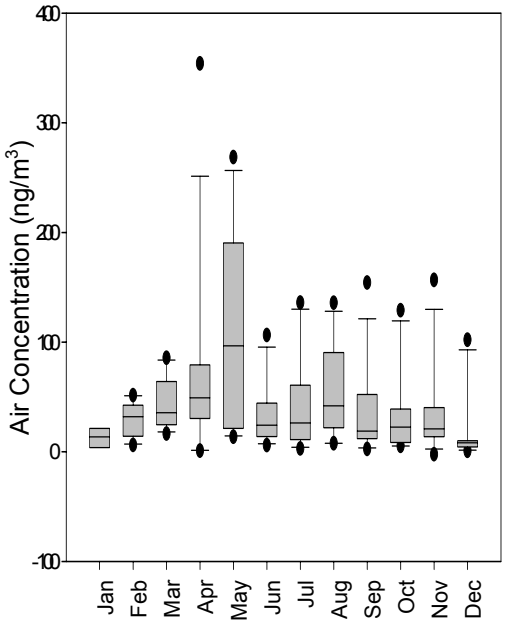
source region in Quebec. The winter I TPSCF shows source regions in the mid-Atlantic states of New York and Pennsylvania and the province of Quebec. The seasonal trend of I exhibited at Burnt Island is similar to that observed in central Alaska.(*32*)

Sodium

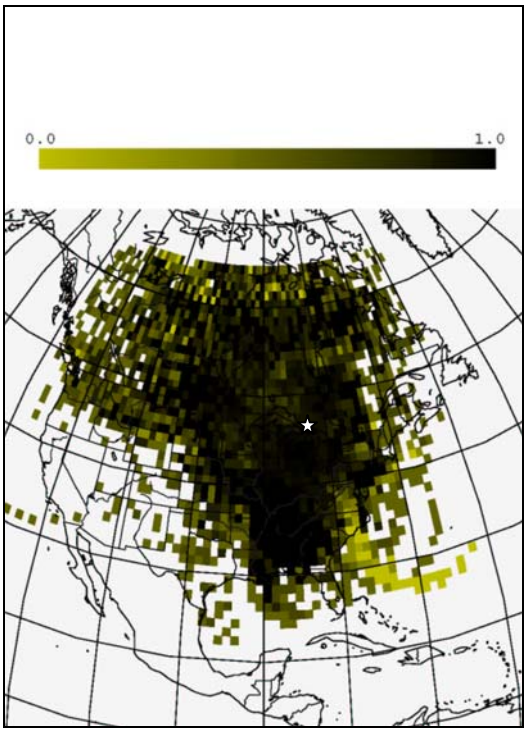
Figure 12 shows the seasonal concentrations and TPSCF figures for Na in the Burnt Island aerosol. Similar to Br, the variance of Na is explained by a salt source. The atmospheric aerosol concentrations of Na at Burnt Island are very seasonally dependent. Many of the high probability Na regions for the summer and winter TPSCF calculations are boundary regions for this calculation. However, the seasonal TPSCF plots do show that the Na is largely associated oceanic and other salt water regions (e.g., Hudson Bay).

Vanadium

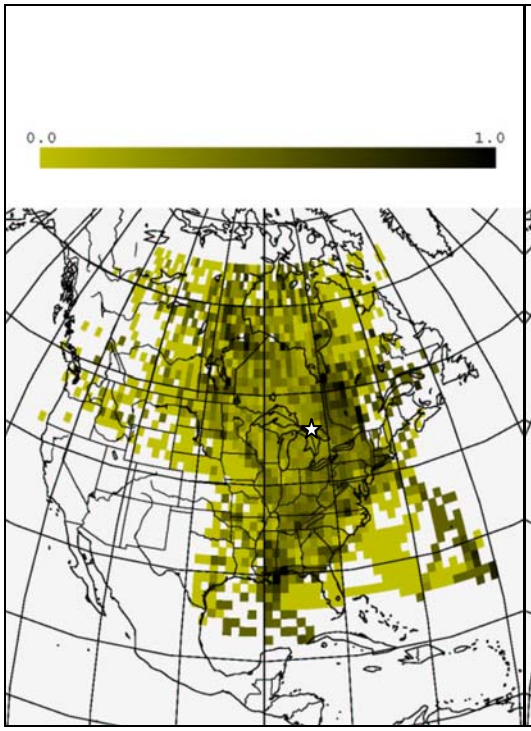
Vanadium is only moderately enriched at the Burnt Island sampling station above crustal levels. The non-crustal V is traced to the combustion of heavy fuel oils containing V-porphyrin complexes.(*33*) While the vanadium values for these TPSCF calculations were not reduced to non-crustal vanadium, the TPSCF values illustrated in figure 13 do represent the source regions expected for oil fired power plant emissions. These include the mid-Alantic coast region between Virginia and New York and the region around southern Mississippi and Louisiana. The winter V TPSCF values also show a high probability region in northern Quebec.



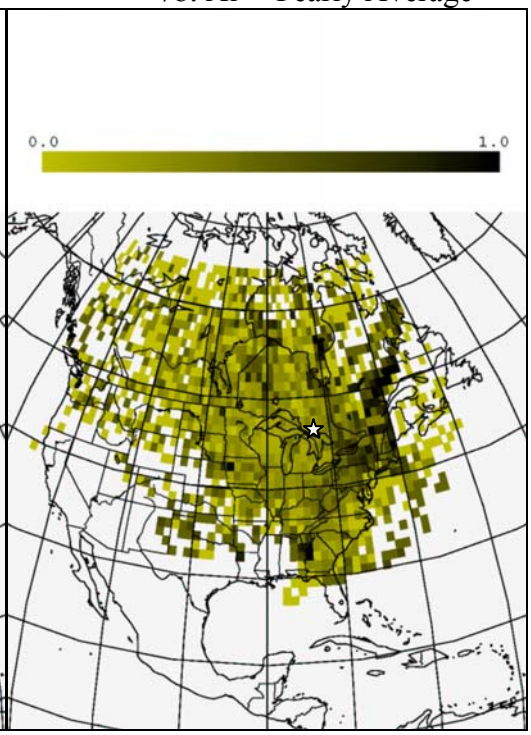
7a. Seasonal Al Concentrations



7b. Al – Yearly Average

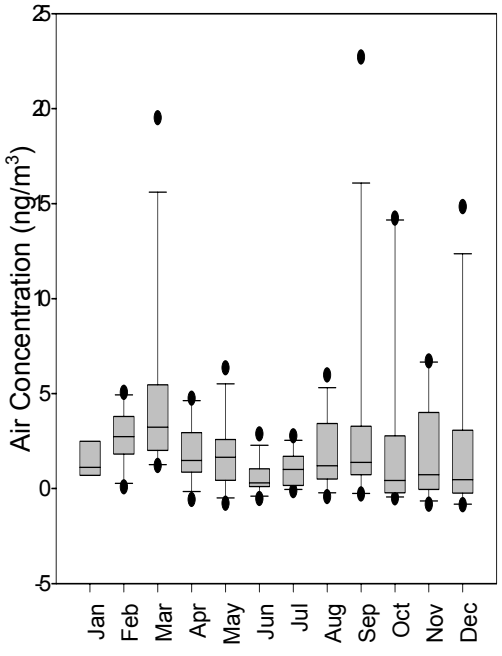


7c. Al – Summer

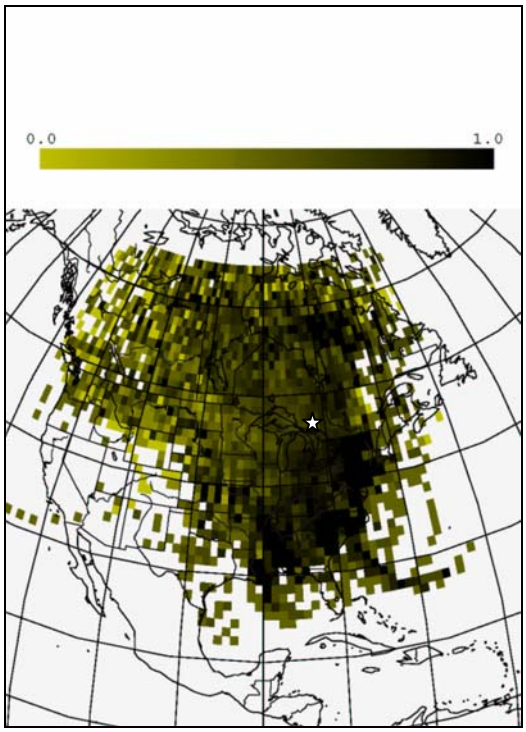


7d.Al-Winter

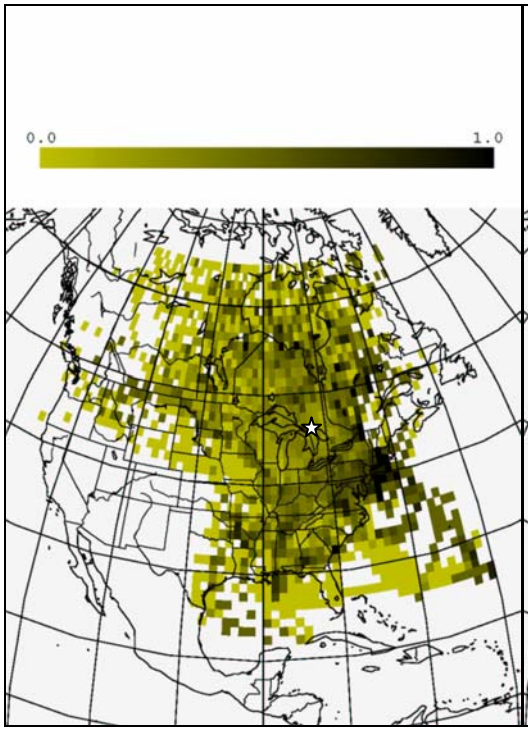
Figure 7. Aluminum TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.



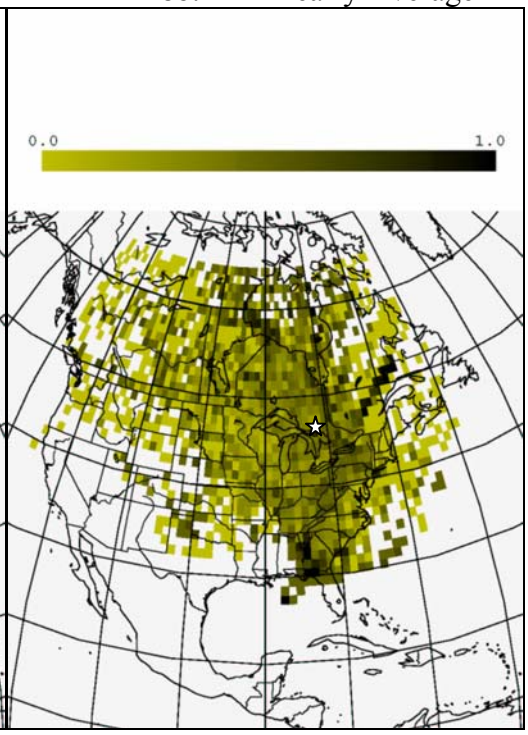
8a. Seasonal Br Concentrations



8b. Br – Yearly Average

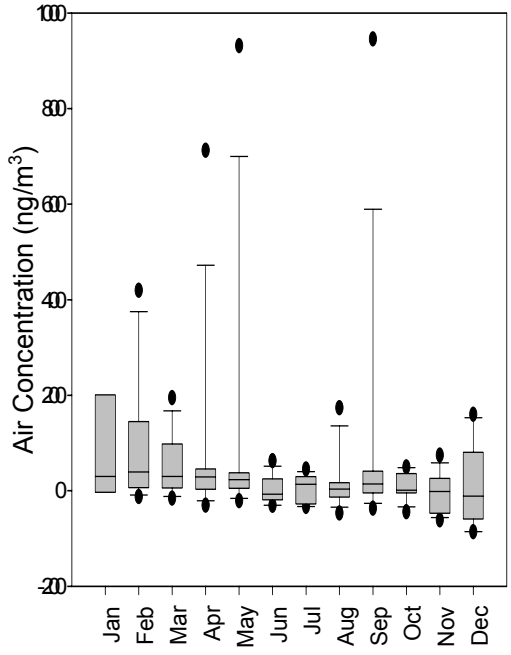


8c. Br – Summer

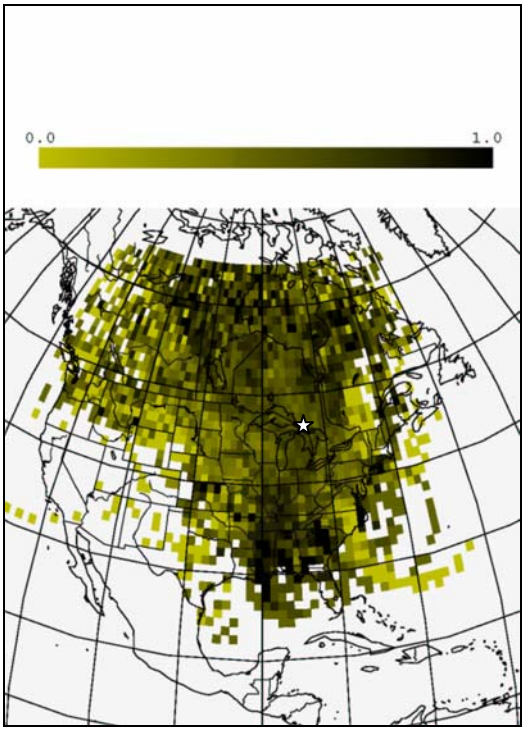


8d. Br- Winter

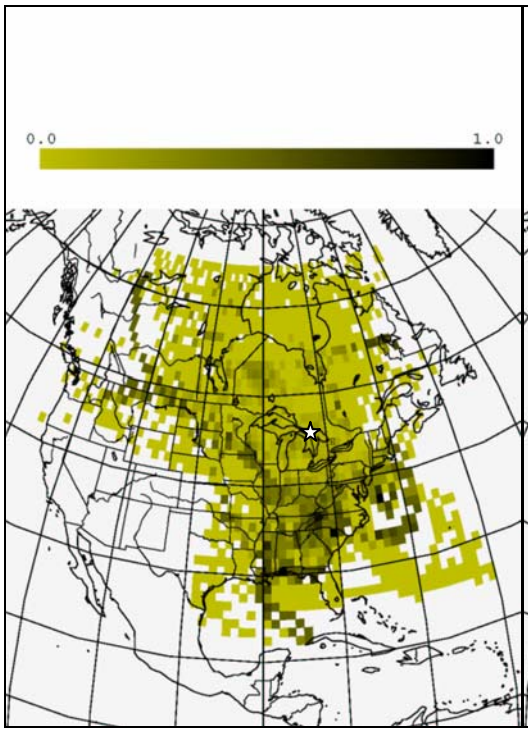
Figure 8. Bromine TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.



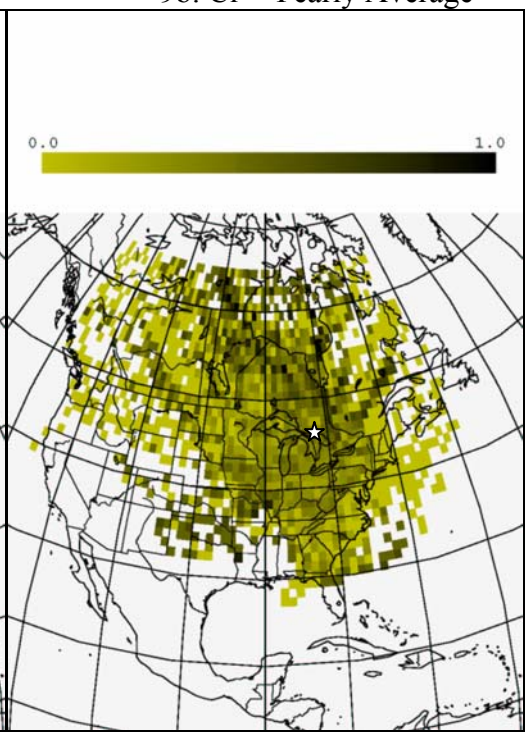
9a. Seasonal Cl Concentrations



9b. Cl – Yearly Average

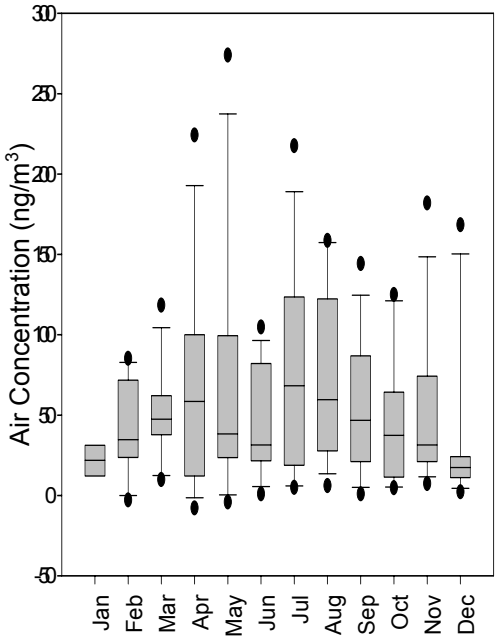


9c. Cl – Summer

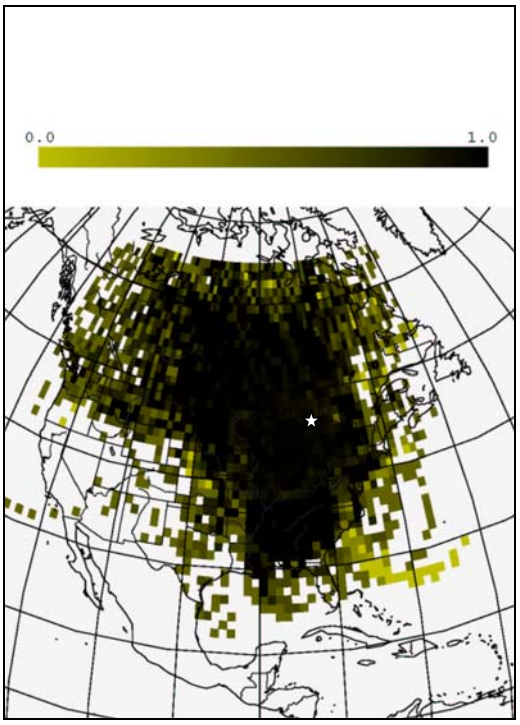


9d. Cl – Winter

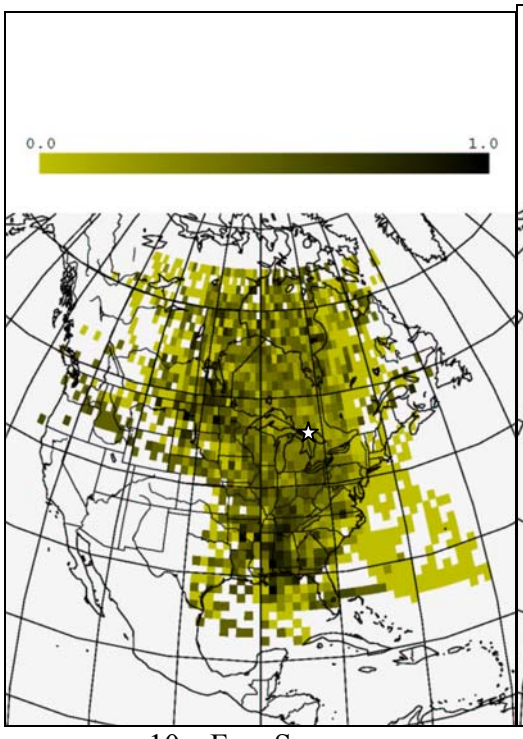
Figure 9. Chlorine TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.



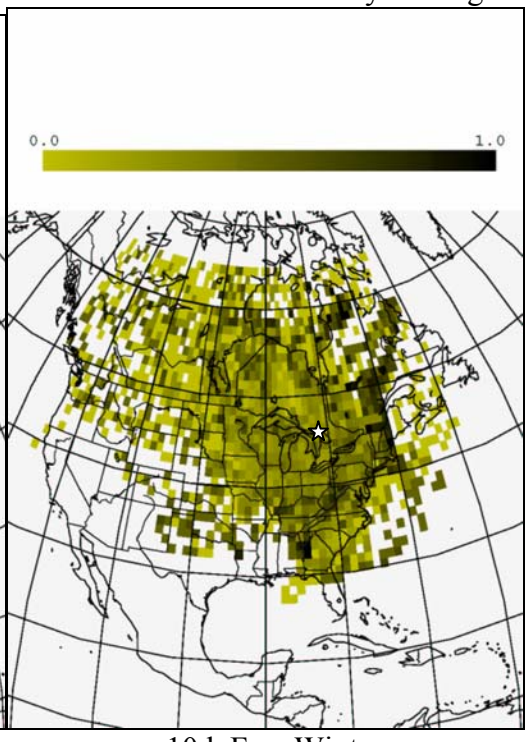
10a. Seasonal Fe Concentrations



10b. Fe – Yearly Average

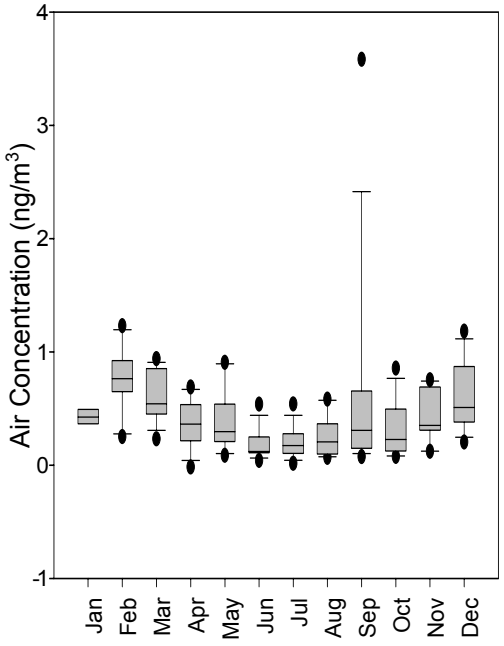


10c. Fe – Summer

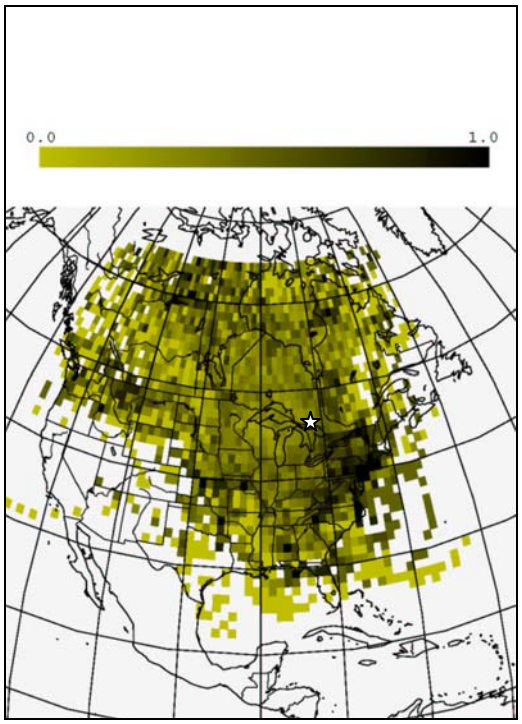


10d. Fe – Winter

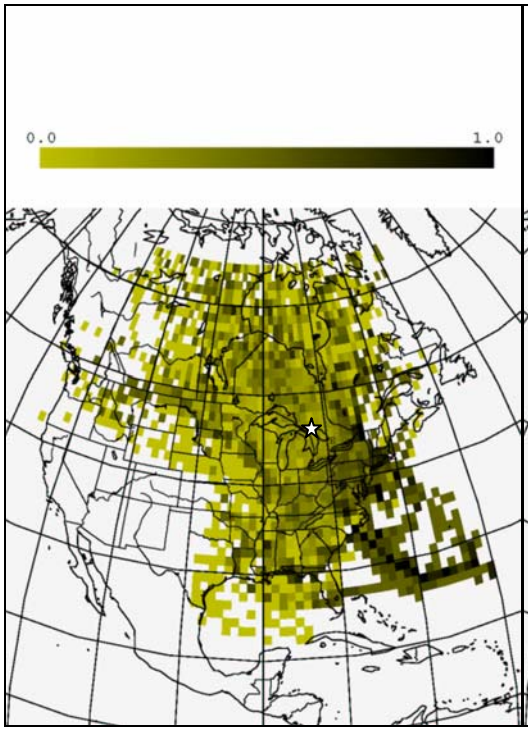
Figure 10. Iron TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.



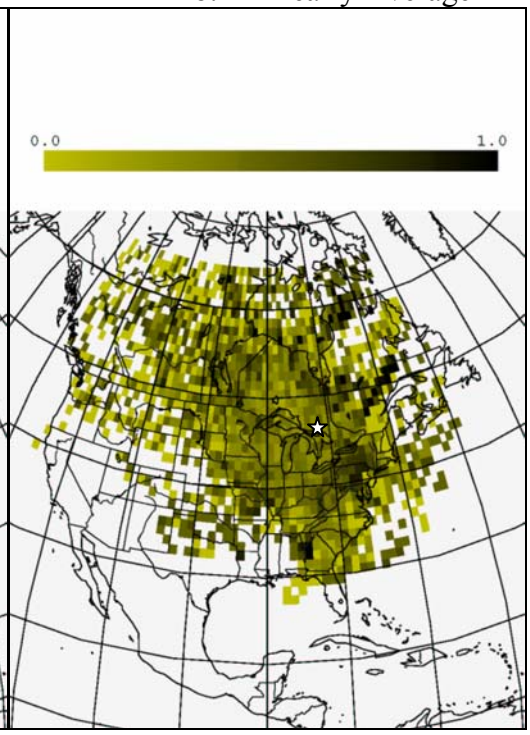
11a. Seasonal I Concentrations



11b. I – Yearly Average

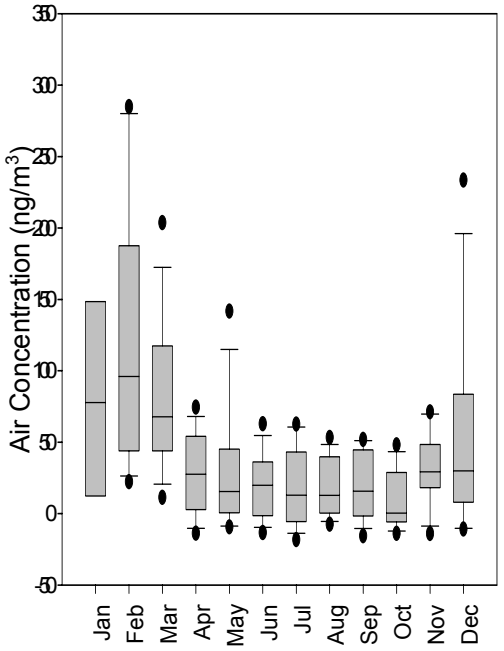


11c. I – Summer

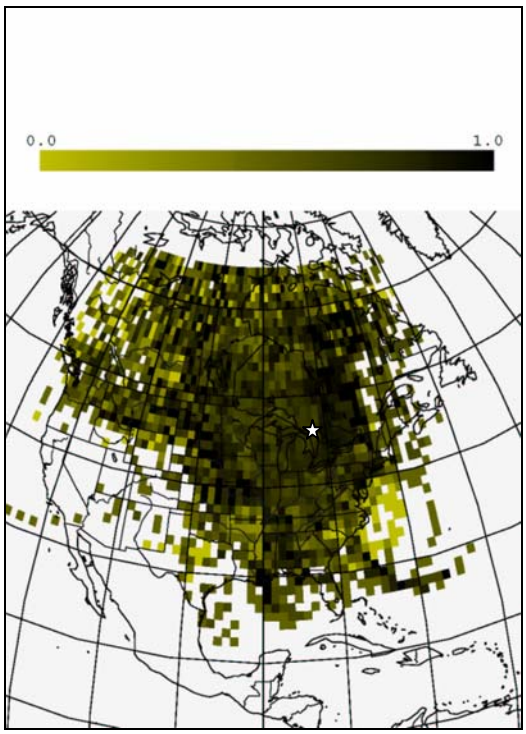


11d. I – Winter

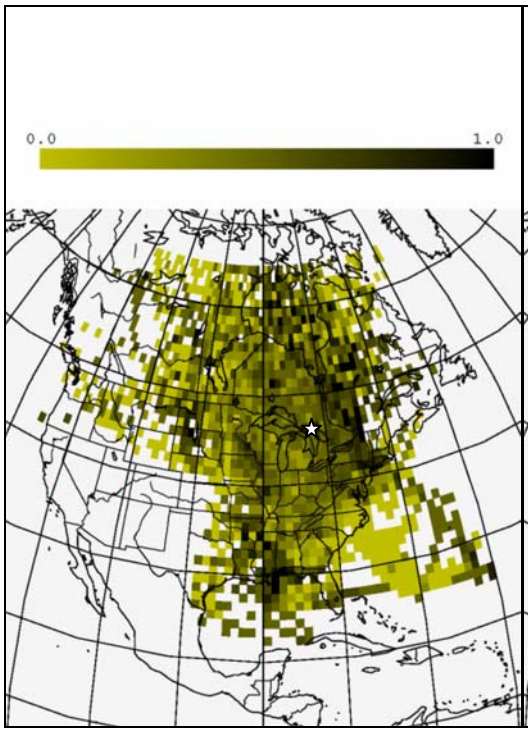
Figure 11. Iodine TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.



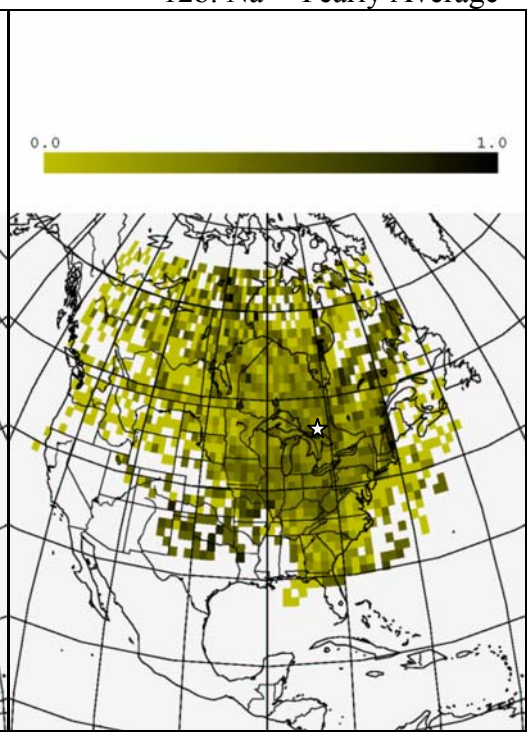
12a. Seasonal Na Concentrations



12b. Na – Yearly Average



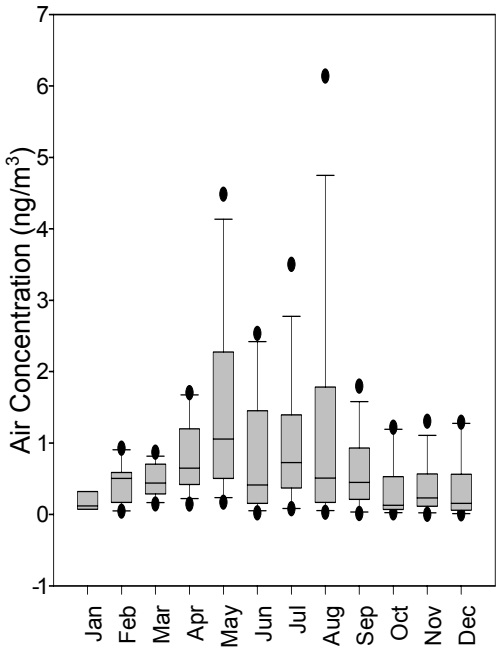
12c. Na – Summer



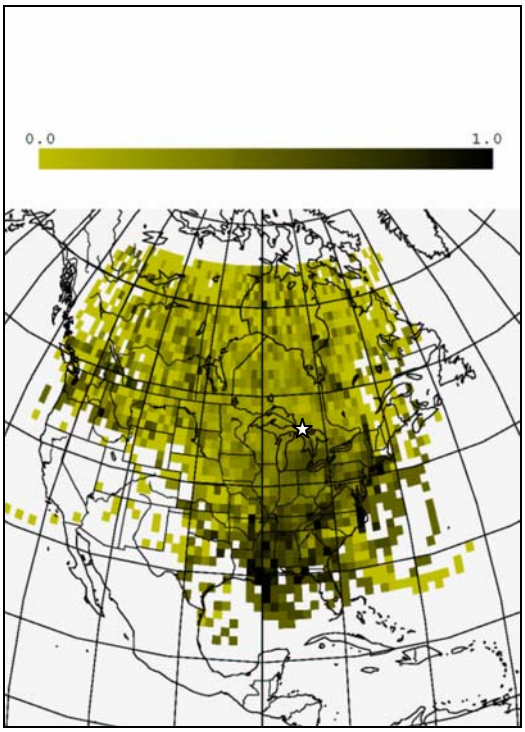
12d. Na – Winter

Figure 12. Sodium TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.

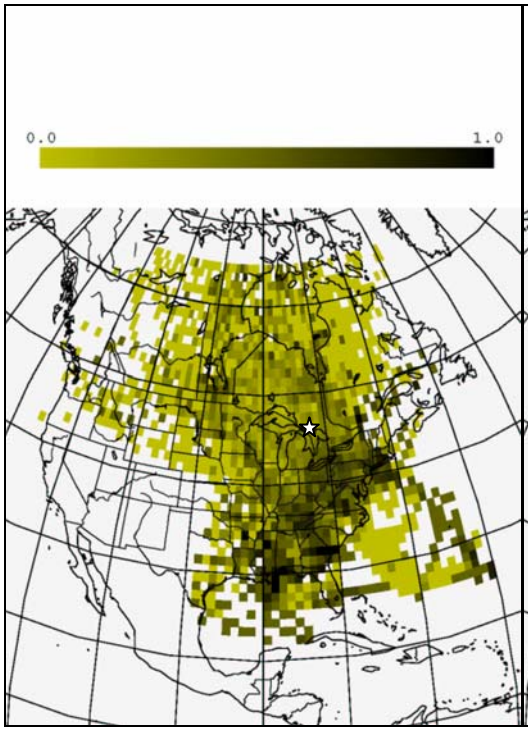
I



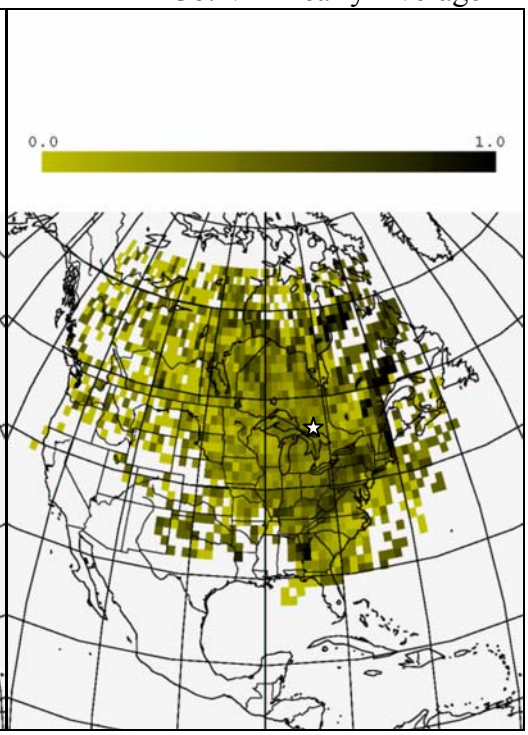
13a. Seasonal V Concentrations



13b. V – Yearly Average



13c. V – Summer



13d. V – Winter

Figure 13. Vanadium TPSCF plots for Burnt Island. The plots show the TPSCF for all the data, the summer data (April to September), and the winter data (October to March). The stars represent the location of the Burnt Island station.

References

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