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Abstract

This paper presents a quantitative comparison of the four most commonly used receptor models, namely Absolute Principal Component Scores (APCS), Pragmatic Mass Closure (PMC), Chemical Mass Balance (CMB), and Positive Matrix Factorization (PMF). The models were used to predict the contributions of a wide variety of sources to $PM_{2.5}$ mass in Halifax, Nova Scotia during the Quantifying the impact of Boreal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites (BORTAS) experiment. However, particular emphasis was placed on the capacity of the models to predict the boreal wild fire smoke contributions during the BORTAS experiment. Using PMF, a new woodsmoke enrichment factor of 52 was estimated for use in the PMC receptor model. The results indicate that the APCS and PMC receptor models were not able to accurately resolve total $PM_{2.5}$ mass concentrations below $2.0 \mu\text{g m}^{-3}$. CMB was better able to resolve these low $PM_{2.5}$ concentrations, but it could not be run on 9 of the 45 days of $PM_{2.5}$ samples. PMF was found to be the most robust of the four models since it was able to resolve $PM_{2.5}$ mass below $2.0 \mu\text{g m}^{-3}$, predict $PM_{2.5}$ mass on all 45 days, and utilized an unambiguous woodsmoke chemical marker. The median woodsmoke relative contribution to $PM_{2.5}$ estimated using PMC, APCS, CMB and PMF were found to be 0.08, 0.09, 3.59 and $0.14 \mu\text{g m}^{-3}$, respectively. The contribution predicted by the CMB model seems to be clearly too high based on other observations. The use of levoglucosan as a tracer for woodsmoke was found to be vital for identifying this source.

1 Introduction

It has been estimated that between 1990 and 2011 wildfires have consumed a median $1.7 \text{ million ha yr}^{-1}$ of Canadian boreal forest (data from Natural Resources Canada). The burning of these forests is a significant source of primary and secondary trace gases and size-resolved particulate matter (PM) to the troposphere (Drysdale, 2008).

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The tropospheric trace gases and PM generated by wildfires are transported long distances with the potential to harm health and the environment 1000 km from their source (Palmer et al., 2013; Naeher et al., 2007; Franklin et al., 2014). During July 2011, the BORTAS (Quantifying the impact of BOREal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites) experiment was conducted out of Halifax, Nova Scotia, Canada to investigate the impact of North American wildfires on the atmospheric chemistry of the troposphere (Palmer et al., 2013). Central to BORTAS-B was the operation of the UK BAe-146-301 Atmospheric Research Aircraft over Eastern Canada, which was used to characterize size-resolved particulate matter and trace gases in wildfire plumes advecting within the outflow from North America (Palmer et al., 2013). Column profile flights were also made above Halifax. In addition to the aircraft measurements there were a number of continuous and integrated surface and column observations of trace gases and size-resolved particulate matter composition made at Dalhousie University in Halifax. A description of the instrumentations and measurements made at the Dalhousie University Ground Station (DGS) are provide in Palmer et al. (2013), Gibson et al. (2013b) and Franklin et al. (2014).

This paper will explore the source attribution of boreal wildfire smoke (and other sources) to surface fine particulate matter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) during the BORTAS-B experiment using four commonly used receptor models.

There are a number of different receptor modelling approaches that can be utilized for the source apportionment of PM_{2.5}, e.g. multivariate least squares factor analysis approaches such as Positive Matrix Factorization (PMF), Pragmatic Mass Closure (PMC) methods and Chemical Mass Balance (CMB) source profile techniques (Gibson et al., 2013b, 2009; Ward et al., 2004; Gugamsetty et al., 2012; Harrison et al., 2011). The US Environmental Protection Agency's (USEPA) CMB receptor model has been used in many PM_{2.5} source apportionment studies (Subramanian et al., 2007). The CMB receptor model uses a solution to linear equations that expresses each receptor chemical concentration as a linear sum of products of source fingerprint abundances and contributions (Ward et al., 2006b; Watson et al., 1994). The advantage of CMB is that it can

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be applied to individual 24 h PM mass and chemical composition. The disadvantage is that the technique relies heavily on available source profiles being representative of regional sources impacting the receptor, which is not always the case (Hellén et al., 2008; Ward et al., 2006b). One assumption of the CMB model is that chemical species emitted from a source are conserved during sampling, and that chemical species do not react with each other (Ward et al., 2006b). CMB is well suited for apportioning local or upwind sources of primary aerosols (those emitted directly as particles). To account for secondary aerosol contributions to PM_{2.5} mass, ammonium sulfate and ammonium nitrate are normally expressed as “pure” secondary source profiles, and represented by their chemical form (Ward et al., 2006b). The USEPA CMB model has been applied to numerous urban and rural PM_{2.5} source apportionment studies in environments impacted by woodsmoke (Ward et al., 2012; Bergauff et al., 2009; Gibson et al., 2010; Ward et al., 2006b).

Pragmatic Mass Closure is a very simple method and works well for the mass closure of the major PM_{2.5} components, e.g. sea salt, secondary ions, surficial fugitive dust, organic and elemental carbon (Gibson et al., 2009). A number of studies have used PMC to apportion the major chemical species to PM mass (Yin and Harrison, 2008; Harrison et al., 2003; Gibson et al., 2009; Dabek-Zlotorzynska et al., 2011).

Another receptor model that has been used extensively in PM_{2.5} source apportionment studies is Absolute Principal Component Scores (APCS) (Song et al., 2006). APCS is a multivariate factorization based model developed by Thurston and Spengler (1985) that is still widely used for the source apportionment of particulate matter.

However, APCS can occasionally return negative mass contributions (Paatero and Tapper, 1994). In order to overcome the negative source mass contribution problem, Paatero and Hopke (2003) introduced a Positive Matrix Factorization (PMF) source apportionment method in the late 1990’s (Paatero and Tapper, 1994). PMF has since been applied widely to indoor, outdoor, urban, rural and regional PM_{2.5} source apportionment studies (Gibson et al., 2013b; Harrison et al., 2011; Larson et al., 2004).

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Chemical markers can also be important when conducting source apportionment. Both APCS and PMF rely on expert, a priori knowledge of chemical markers found within the PM_{2.5} chemical composition to identify the source of each PM_{2.5} component factor, e.g. high factor loadings of Al, Si, Ca and Fe are indicative of crustal re-entrained material (Song et al., 2006; Hopke, 1991; Gibson et al., 2013b). Many studies use levoglucosan (1,6-anhydro- β -D-glucopyranose) as an unambiguous chemical marker of wildfire and residential woodsmoke (Gibson et al., 2010; Ward et al., 2012; Simoneit et al., 1999). Levoglucosan is derived from cellulose burning at temperatures greater than 300 °C (Simoneit et al., 1999; Ward et al., 2006a). Potassium (K) is also a good tracer for woodsmoke and often used in conjunction with levoglucosan (Bergauff et al., 2010; Jeong et al., 2008; Urban et al., 2012). Other commonly used PM_{2.5} source chemical markers are described in Gibson et al. (2013b), Harrison et al. (2011) and Jeong et al. (2011). In addition, the source chemical profiles contained within SPECIATE 4.0 are another resource to aid in the identification of PM_{2.5} sources within a speciated PM_{2.5} sample (Ward et al., 2012; Jaekels et al., 2007; Gibson et al., 2013b).

This paper presents a quantitative comparison of the four most commonly used receptor models: APCS, PMC, CMB and PMF. The objective is to provide a quantitative comparison of the ability of these models to predict overall PM_{2.5} mass and the contributions of minor components. The models are compared based on their ability to apportion boreal wildfire woodsmoke (and other sources) applied to a 45 day contiguous PM_{2.5} data set sampled at the DGS in Halifax during the BORTAS-B experiment. This dataset should provide sufficient variability and contributions of minor sources to permit a comprehensive comparison of the four receptor models.

2 Measurements

A full description of the PM_{2.5} speciated sampling methods employed for this paper are described in Gibson et al. (2013b). Additional supporting instrumentation used at the DGS during BORTAS-B are described in Palmer et al. (2013) and Franklin et al. (2014),

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HP-5 MS capillary column. Splitless injection was employed. The levoglucosan and internal standard were detected by extracted ion signals at 217 and 220 m/z , respectively. Levoglucosan analysis recoveries for 100 to 2000 ng averaged $96 \pm 12\%$ ($n = 18$, ± 1 sigma). Six laboratory blanks were used to calculate an average levoglucosan blank concentration and the standard deviation and 95 % confidence interval for the blank. The limit levoglucosan of detection (LOD) is reported as the average laboratory blank and was found to be 7.7 ng m^{-3} (Bergauff et al., 2008) level plus one 95 % confidence interval for the blank. Local meteorological data at the DGS was collected using a Davis Vantage Pro II weather station (Davis Instruments Corp. Hayward, California 94545 USA). Further information on the meteorological sensors onboard the Davis Vantage Pro II and results are provided in Gibson et al. (2013b). In addition, a daily climatology review of synoptic meteorology in the greater Halifax Regional Municipality observed during the PM_{2.5} sampling is also provided in Gibson et al. (2013b).

HYSPLIT 10 day, 5 day and 2 day air mass back trajectories were used to identify the likely upwind source regions of PM_{2.5} (Gibson et al., 2013b). A plot of ensemble HYSPLIT back trajectories by source region during the sampling campaign is provided in Gibson et al. (2013). From Gibson et al. (2013b) it was observed that 40 % of the air masses entering Halifax during BORTAS-B originated from the marine sector, 16 % from the SW (NE US), 27 % from the WNW (Windsor-Quebec source region) and 16 % from the N. The SW cluster and WNW cluster appear to be mainly associated with boundary layer flow from known upwind source regions of PM_{2.5} that was mainly composed of ammonium sulfate ((NH₄)₂SO₄), ammonium nitrate (NH₄NO₃) and organic matter (up to 70 % of the total PM_{2.5} mass).

Fire hotspot maps were used to identify active burning regions of Canada. MODIS hotspot locations from NASA (see <http://earthdata.nasa.gov/data/near-real-time-data/firms>) and AVHRR hotspots from NOAA FIMMA (see <http://www.ssd.noaa.gov/PS/FIRE/Layers/FIMMA/fimma.html>) were used to generate the fire hot spot maps (Giglio et al., 2003; de Groot et al., 2013).

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method of Thurston and Spengler (1985) the relative source contributions were then determined by multiple linear regression on the measured concentrations. The developed linear regression equations could then be used to produce a time series plot and to identify the relative contributions of the various sources.

We also used the USEPA PMF v3.0 receptor model for the source apportionment of the PM_{2.5} at the DGS. In the previous manuscript by Gibson et al. (2013b), six major sources were determined and included Long-Range Transport (LRT) Pollution 1.75 $\mu\text{g m}^{-3}$ (47%), LRT Pollution Marine Mixture 1.0 $\mu\text{g m}^{-3}$ (27.9%), Vehicles 0.49 $\mu\text{g m}^{-3}$ (13.2%), Fugitive Dust 0.23 $\mu\text{g m}^{-3}$ (6.3%), Ship Emissions 0.13 $\mu\text{g m}^{-3}$ (3.4%) and Refinery 0.081 $\mu\text{g m}^{-3}$ (2.2%). The PMF model described 87% of the observed variability in total PM_{2.5} mass (bias = 0.17 and RSME = 1.5 $\mu\text{g m}^{-3}$) (Gibson et al., 2013b). The PMF factor profile used to identify woodsmoke contained 99% of the levoglucosan mass. The PMF model initialization procedure used in this paper was the same as described in Gibson et al. (2013b).

We also utilized the pragmatic mass closure (PMC) method as another alternative receptor model (Yin and Harrison, 2008). PMC offers a simple approach to estimate the source attribution or the chemical composition of size-resolved airborne particulate matter (PM) (Harrison et al., 2003). The PMC receptor modelling method is limited to major PM species only, e.g. sodium chloride, ammonium nitrate (NH₄NO₃), ammonium sulphate ((NH₄)₂SO₄), non sea salt-SO₄, sodium nitrate (NaNO₃), organic carbon (OC), elemental carbon (EC), crustal matter, trace element oxides and particle bound water (Gibson et al., 2009; Yin and Harrison, 2008; Dabek-Zlotorzynska et al., 2011). In PMC, molar weight correction factors, or enrichment factors, are applied to the individual measured PM chemical components. This then allows an estimate of the probable species that was present in the original sample, e.g. multiplying NO₃⁻ by 1.29 yields an estimate of the NH₄NO₃ concentration present in the original PM_{2.5} sample (Dabek-Zlotorzynska et al., 2011). PMC has been used to apportion contributions to urban and rural PM₁₀, PM_{2.5} and PM_{2.5-10} in Ireland (Yin et al., 2005), coastal, rural and urban PM₁₀ in Scotland (Gibson et al., 2009), urban background and roadside locations

the bias seen for APCS and PMC. However, because PMF predicts the $PM_{2.5}$ mass on all sample days, has a slope of 0.88 and the ability to predict very low $PM_{2.5}$ mass concentrations, often seen in Halifax, in these respects it is the most parsimonious of the four receptor models.

Figures 5–8 provide a time series from 7 July to 25 August 2011 of the APCS, PMC, CMB and PMF daily $PM_{2.5}$ source apportionment. Time series plots of the individual $PM_{2.5}$ chemical species (not including levoglucosan) associated with Figs. 5–8 are provided in Gibson et al. (2013b).

It can be seen from Tables 3–6 that the four receptor models identify different number and type of $PM_{2.5}$ source respectively, e.g. the APCS model identified 6 sources, PMC 10 sources, CMB 13 sources and PMF 9 sources. The reason for the different number of sources identified by each model is due to the different inherent methodology by which each model generates the source identification. In the case of PMC, a molar correction factor is applied to individual $PM_{2.5}$ species. Therefore, if the species is present and there is a corresponding molar correction factor the source will be identified and quantified. In the case of CMB receptor modelling, the sample chemical species are identified by matching with known source chemical profiles. With CMB, the number of statistically significant and logical matches determines the number of sources identified and quantified by the model, whereas APCS and PMF both use factorization and are open to identify as many sources that meet each model's inclusion criteria and would make sense being observed at the receptor. In PMC the source name is assigned from the molar factor associated with the source, in CMB the source name is assigned from the matching source profile, in APCS and PMF the source name is subjective and assigned by the user, reflecting the chemical species observed within each factor profile. It can be seen from Tables 3–6 that surface dust and woodsmoke were identified in all four models.

With reference to Fig. 6, the PMC trace metal oxide values are worthy of note. This is because Ni and V, which would otherwise identify and be used to apportion ship emissions, was included in the trace metal oxide apportioned mass. Therefore, the

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July, 1 August, 6 August and 13 August 2011. The low or absent woodsmoke days are either associated with air flow from the ocean or from Northern Canada when boreal wild fire activity was absent. These days are also associated with low PM_{2.5} mass as described in Gibson et al. (2013b). To identify upwind forest fire source regions, we use visible MODIS satellite images, MODIS fire hot spots maps, 5 day HYSPLIT air mass back trajectories (Gibson et al., 2013b), FLEXPART air mass trajectories (Stohl et al., 2005) chemical transport models (Palmer et al., 2013), Raman Lidar (Bitar et al., 2010) and aircraft measurements (Palmer et al., 2013) were used to corroborate the apportioned woodsmoke to surface PM_{2.5} for the 21 July event. Figure 10 provides an example match up of Lidar aerosol backscatter measurements at the DGS (a), GEOS-5 forecast of CO mixing ratio associated with boreal biomass burning above the DGS (b), FLEXPART vertical profile of PM_{2.5} (c) at the DGS and a plot of the aircraft profile measurements of CO, acetonitrile and aerosol backscatter obtained at midnight (d). Acetonitrile was used as it is an effective tracer for biomass fire plumes in the atmosphere (Karl et al., 2007). Figure 10a shows elevated aerosol backscatter below 2 km between 00:00 UTC 20 July and 24:00 UTC 21 July 2011. Also there is then a “V-shaped notch” of clear air located above 2 and below 5 km, followed by further aerosol backscatter between 6 and 8 km. The elevated surface aerosol backscatter measurements seen in Fig. 10a are accompanied by elevated surface PM_{2.5} concentrations as seen in Fig. 5. Since the PMF model appears to be the most parsimonious at predicting PM_{2.5} mass, and is anticipated to be the most robust at predicting woodsmoke, it was used to compare with the features contain in Fig. 10. From the PMF source apportionment timeseries plot in Fig. 8, it can be seen that the PM_{2.5} was chiefly composed of LRT (NH₄)₂SO₄ and LRT Pollution Marine Mixture (NO₃⁻, Na, NH₄NO₃), with a small spike in woodsmoke seen on 20 July 2011. Scrutiny of HYSPLIT air mass back trajectories in Gibson et al. (2013b) and HYSPLIT dispersion models in Franklin et al. (2014) show that the air flow crossed a region experiencing extensive boreal forest wildfire in Northern Ontario prior to reaching Nova Scotia. On 20 July air flow from the NE US mixed with the air flow from Northern Ontario en route to Halifax, providing a mixture of

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boreal wildfire smoke from Northern Ontario together with anthropogenic LRT aerosol from the NE US. It can be seen from Fig. 10b that GEOS-5 predicts the exact same feature for CO as the aerosol backscatter observed by the Lidar in Fig. 10a. The CO is related to both the LRT from the NE US mixed with wildfire woodsmoke from Ontario.

Evidence for the woodsmoke entrainment on 20 July 2011 in the PMF source apportionment timeseries (Fig. 8) was further corroborated by FLEXPART forward trajectory modelling from the large forest fires in Ontario that were burning on 17 July 2011. It can be seen from Fig. 10c that FLEXPART predicted the impact of woodsmoke particles at the surface in Halifax, which helps explain the small spike in levoglucosan on 20 July 2011. Finally, further proof of woodsmoke impacts at the DGS come from the aircraft spiral profiles shown in Fig. 10d. Figure 10d shows aircraft column profiles for CO, acetonitrile and aerosol backscatter. The strong agreement between CO, acetonitrile and backscatter in Fig. 10d points toward wildfire woodsmoke as the origin of these metrics in the column over Halifax. Figure 11 provides a NASA AQUA MODIS true colour satellite image that clearly shows boreal forest fire smoke from Northern Ontario advecting over Halifax, Nova Scotia on 18 July. These fires continued to impact the DGS on 20 July 2011 as shown in Figs. 9 and 10. In a similar way the largest woodsmoke spike shown in Fig. 9 on 31 July 2011 was due to boreal forest fires in Northern Quebec. This can be seen in Fig. 12 where a NOAA HYSPLIT 5 day air mass trajectory passes over the forest fires in Northern Quebec 3 days prior to arriving at the DGS. Using the same approach, it was seen that HYSPLIT 5 day air mass back trajectories together with the fire hot spot maps for 6 August showed that the elevated woodsmoke was related to wild fires in Labrador, while the woodsmoke spike on the 12 August was related to another large fire in Ontario on 8 August 2011.

Table 7 summarizes the four receptor model parameters used for predicting PM_{2.5} during the BORTAS-B experiment. Table 8 presents the woodsmoke source apportionment descriptive statistics for each receptor model. It can be seen that the estimated mean woodsmoke contribution to PM_{2.5} by APCS and PMC are almost identical, 0.32 and 0.35 $\mu\text{g m}^{-3}$. The close agreement between the woodsmoke contribution estimated

by APCS validates the new enrichment factor in this paper generated from previous PMF and PMC analyses (Gibson et al., 2013a). It can be seen that CMB estimated the mean woodsmoke contribution to be $3.23 \mu\text{g m}^{-3}$, which is an order of magnitude greater than APCS and PMC. In addition, it can be observed that PMF estimated the mean woodsmoke contribution to be $0.61 \mu\text{g m}^{-3}$, which is approximately double that estimated by APCS and PMC. However, because of the PMF model's better $\text{PM}_{2.5}$ predictive capability (especially below $2.0 \mu\text{g m}^{-3}$) and clear woodsmoke marker source identification, known statistical robustness over APCS, its results are likely the most accurate of the four models. However, boreal forest wood combustion product emissions source profiling followed by source apportionment using these four models would be needed to completely validate PMF's superiority over APCS, PMC and CMB receptor model methodologies.

5 Conclusions

Four receptor models were used to improve our understanding of the source contribution of woodsmoke, and other major sources, to $\text{PM}_{2.5}$ total mass during the BORTAS-B experiment. During the process, PMF was used to generate a new woodsmoke enrichment factor of 52. The new enrichment factor was used in the PMC model to convert levoglucosan into a woodsmoke concentration (levoglucosan multiplied by 52). Cross-referencing the woodsmoke contribution estimated by APCS helped to validate the utility of this new enrichment factor. It was found that APCS and PMC receptor models were not able to predict total $\text{PM}_{2.5}$ mass concentrations below $2.0 \mu\text{g m}^{-3}$. Further, although CMB had an improved intercept and a slope of 1, it could not be run on 9 of the 45 days of $\text{PM}_{2.5}$ samples. PMF is considered to be the most robust of the four models since it is able to predict $\text{PM}_{2.5}$ mass below $2.0 \mu\text{g m}^{-3}$, predict $\text{PM}_{2.5}$ mass on all 45 days, has a slope close to 1, has a low bias, and utilizes an unambiguous woodsmoke chemical marker (levoglucosan) within the model. The median (min : max) woodsmoke relative contribution to $\text{PM}_{2.5}$ estimated using PMF was found to be 0.14

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(0.0:4.14) $\mu\text{g m}^{-3}$. The use of a woodsmoke tracer such as levoglucosan is critical when carrying out $\text{PM}_{2.5}$ source apportionment studies of boreal forest wild fire smoke. Controlled wood combustion product sampling followed by source apportionment modeling with these four models would greatly improve our understanding of their performance for predicting woodsmoke contributions to $\text{PM}_{2.5}$ in future studies of this nature.

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Table 1. Descriptive statistics for levoglucosan.

	<i>n</i>	Mean	SD	Min	25th Pctl	Median	75th Pctl	Max
Levoglucosan (ng m ⁻³)	45	6.1	10.0	0.2	0.9	1.6	6.2	46.0

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Table 2. Principal component analysis (PCA) of the PM_{2.5} chemical species.

	Sea Salt	LRT ((NH ₄) ₂ SO ₄)	Surface Dust	Woodsmoke	Ship Emissions
BC		0.52		0.42	
Al			0.91		
Br	0.78				
Ca			0.90		
Fe			0.70		
K				0.74	
Mg	0.96				
Na	0.97				
Ni					0.95
Si			0.98		
V					0.94
Zn				0.86	
Cl	0.81				
S		0.94			
NO ₃	0.82				
SO ₄		0.97			
NH ₄		0.96			
OM		0.74		0.56	
Levogluconan				0.91	
Eigenvalue	5.72	3.65	3.11	2.03	1.72
Cumulative % var	30.1	49.3	65.6	76.3	85.4

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Table 3. Absolute principal component scores (APCS) PM_{2.5} source apportionment descriptive statistics.

Metric ($\mu\text{g m}^{-3}$)	<i>n</i>	Mean	Median	Min	Max	SD	C.I.
Observed PM _{2.5}	45	4.36	3.96	0.08	12.50	3.13	0.91
LRT pollution aged marine aerosol	45	0.75	0.61	0.16	3.42	0.61	0.18
LRT pollution (NH ₄) ₂ SO ₄	45	3.76	3.06	0.28	13.95	2.65	0.78
Surface dust	45	0.73	0.63	0.13	3.32	0.54	0.16
Woodsmoke	45	0.35	0.09	0.01	2.71	0.62	0.18
Ship emissions	43	0.14	0.09	0.00	0.76	0.15	0.04

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Table 4. Pragmatic mass closure (PMC) PM_{2.5} source apportionment descriptive statistics.

Metric ($\mu\text{g m}^{-3}$)	<i>n</i>	Mean	Median	Min	Max	SD	C.I.
Observed PM _{2.5}	45	4.36	3.96	0.08	12.50	3.13	0.91
LRT pollution NH ₄ NO ₃	45	0.12	0.09	0.01	0.83	0.13	0.04
LRT pollution (NH ₄) ₂ SO ₄	45	0.87	0.57	0.14	4.15	0.84	0.25
Organic matter	45	1.03	0.77	0.18	2.66	0.68	0.20
Black carbon	45	0.41	0.39	0.12	1.03	0.21	0.06
Surface dust	45	0.27	0.22	0.02	1.53	0.24	0.07
Trace element oxides	45	1.48	1.48	1.47	1.49	0.00	0.00
Sea salt	45	0.16	0.11	0.01	1.06	0.18	0.05
Particle bound water	45	0.29	0.20	0.05	1.33	0.27	0.08
Woodsmoke	45	0.32	0.08	0.01	2.38	0.55	0.16

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Table 5. Chemical mass balance (CMB) PM_{2.5} source apportionment descriptive statistics.

Metric ($\mu\text{g m}^{-3}$)	<i>n</i>	Mean	Median	Min	Max	SD	C.I.
Observed PM _{2.5}	45	4.57	4.04	0.08	13.73	3.39	0.98
Surface dust	2	0.81	0.81	0.39	1.24	0.6	0.83
LRT pollution (coal/industrial)	5	0.83	0.85	0.57	1.09	0.2	0.17
Woodsmoke	14	3.23	3.59	1.38	4.72	1.04	0.54
Marine aerosol	34	0.3	0.24	0.04	1.64	0.3	0.1
Ship auxiliary engines	17	1.43	1.2	0.3	3.2	0.84	0.4
LRT pollution (NH ₄) ₂ SO ₄	21	1.45	0.67	0.24	6.77	1.58	0.68
Tire wear	1	0.82	0.82	0.82	0.82	NA	NA
Diesel trucks	2	1.11	1.11	1.1	1.12	0.02	0.02
Vegetative burning	2	2.25	2.25	1.42	3.08	1.18	1.63
Small gasoline vehicles	5	2.35	2.51	0.58	5.08	1.87	1.63
LRT pollution NH ₄ NO ₃	2	0.54	0.54	0.14	0.94	0.57	0.79
SO ₄	35	1.31	0.95	0.35	5.4	1.08	0.36

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Table 6. Positive Matrix Factorization (PMF) PM_{2.5} source apportionment descriptive statistics.

Metric ($\mu\text{g m}^{-3}$)	<i>n</i>	Mean	Median	Min	Max	SD	C.I.
Observed PM _{2.5}	45	4.57	4.04	0.08	13.73	3.39	0.98
Diesel vehicles/tire wear	39	0.05	0.03	0.00	0.17	0.04	0.01
Gasoline/tire wear	30	0.14	0.02	0.00	3.43	0.62	0.22
LRT pollution (NH ₄) ₂ SO ₄	33	2.05	1.15	0.09	12.12	2.45	0.84
Ship emissions	34	0.55	0.49	0.04	1.15	0.31	0.11
LRT pollution marine mixture	38	0.88	0.44	0.02	7.00	1.31	0.42
Woodsmoke	29	0.61	0.14	0.00	4.14	1.00	0.36
LRT pollution (coal/industry)	34	0.74	0.48	0.00	2.97	0.69	0.23
Surface dust	38	0.33	0.19	0.00	2.55	0.44	0.14

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Table 7. Comparison of the four receptor model observed vs. mean predicted PM_{2.5}.

Receptor model	<i>n</i>	Mean observed (μg m ⁻³)	Mean predicted (μg m ⁻³)	Bias	RMSE (μg m ⁻³)	<i>R</i> ²
APCS	45	4.6	5.7	1.3	2.0	0.84
PMC	45	4.6	4.9	1.4	1.6	0.84
CMB	36	5.6	5.3	4.3	1.2	0.88
PMF	45	4.6	3.9	2.9	1.3	0.88

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Table 8. Boreal wildfire woodsmoke source apportionment ($\mu\text{g m}^{-3}$) descriptive statistics by receptor model.

Receptor model	<i>n</i>	Mean	Median	Min	Max	SD	C.I.
PMC	45	0.32	0.08	0.01	2.38	0.55	0.16
APCS	45	0.35	0.09	0.01	2.71	0.62	0.18
CMB	14	3.23	3.59	1.38	4.72	1.04	0.54
PMF	29	0.61	0.14	0.00	4.14	1.00	0.36

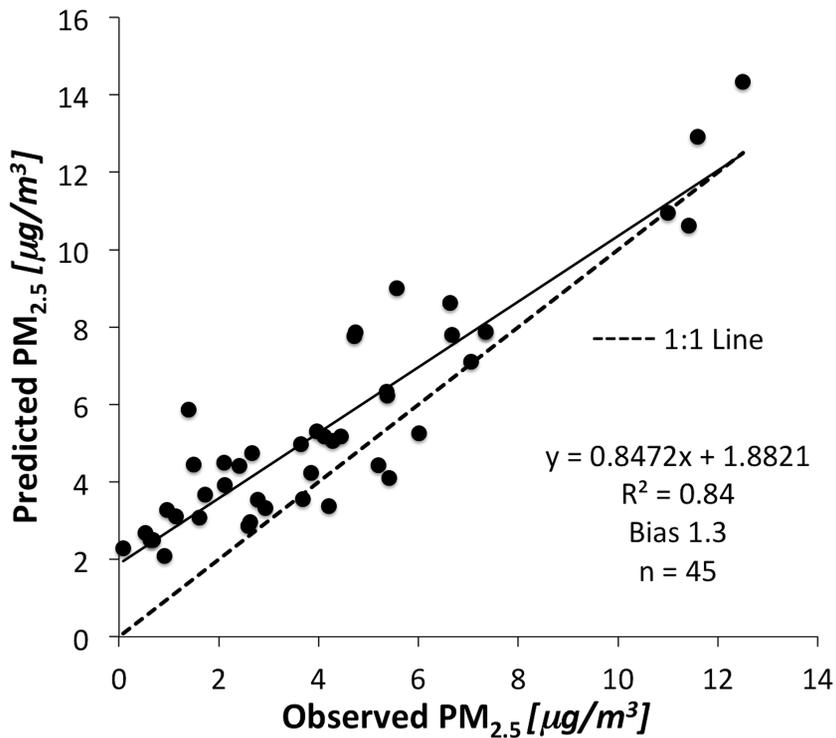


Figure 1. Absolute Principal Component Scores (APCS) predicted vs. observed $PM_{2.5}$.

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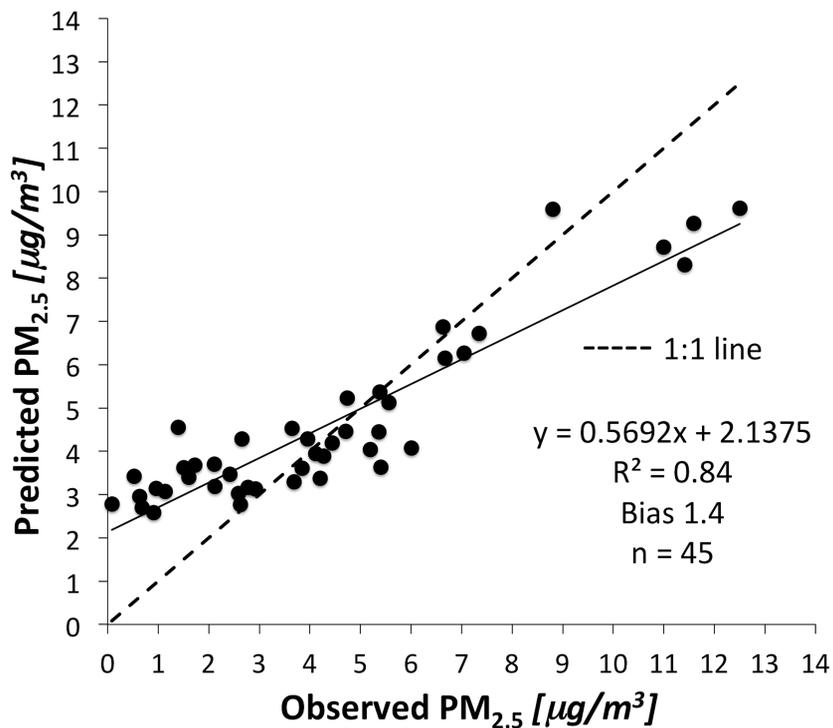


Figure 2. Pragmatic Mass Closure (PMC) predicted vs. observed PM_{2.5}.

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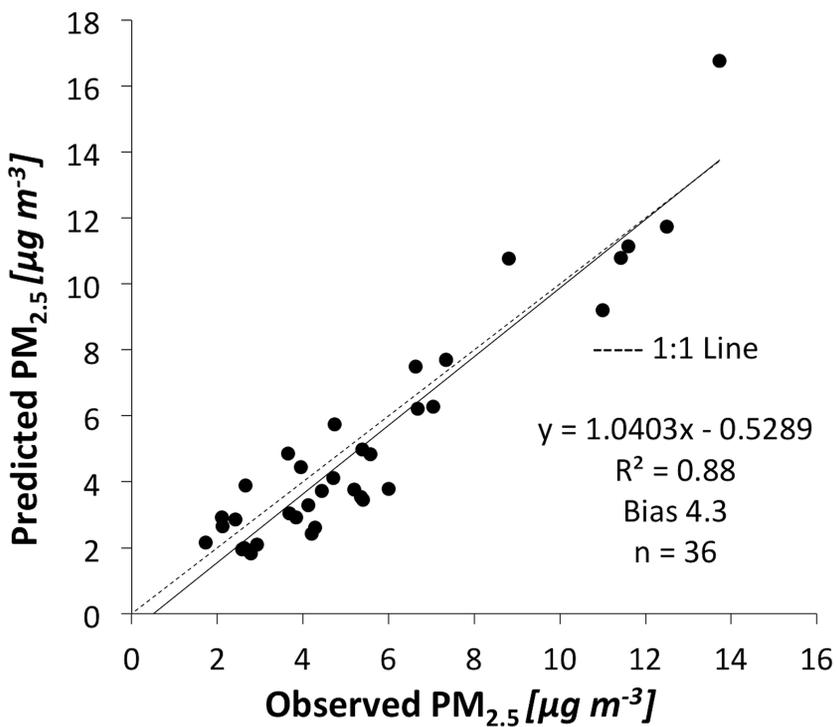


Figure 3. Chemical Mass Balance (CMB) predicted vs. observed $PM_{2.5}$.

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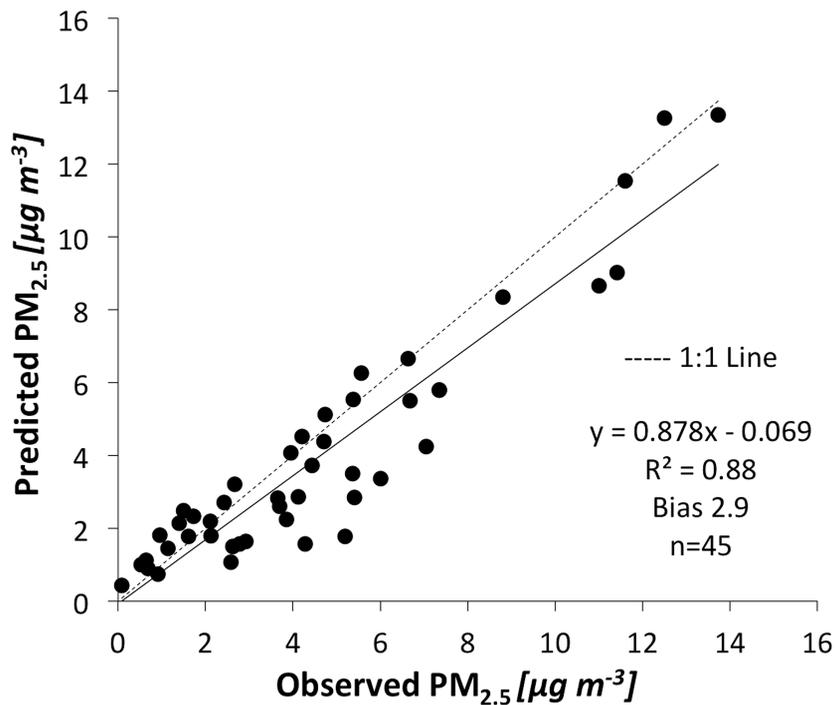


Figure 4. Positive Matrix Factorization (PMF) predicted vs. observed $PM_{2.5}$.

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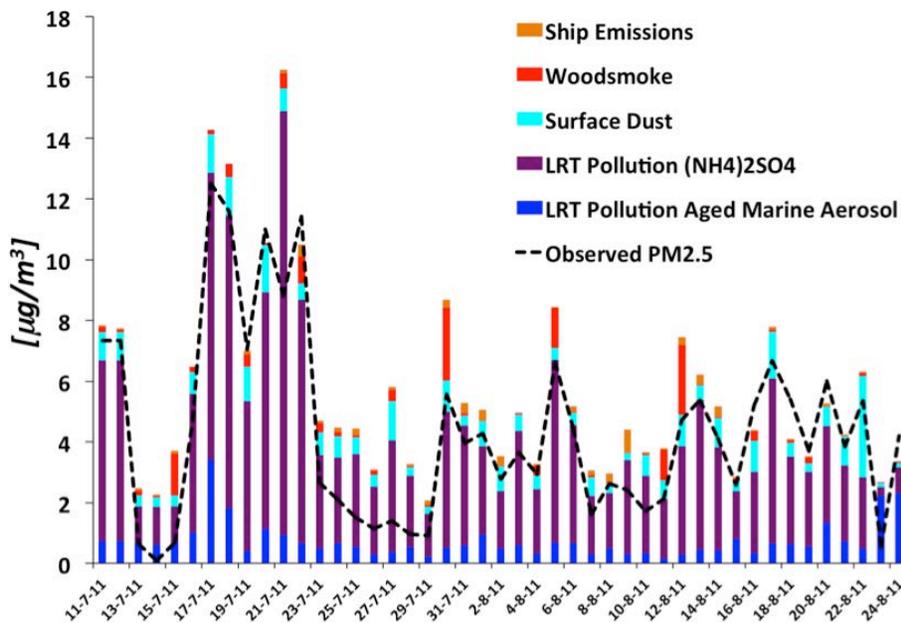


Figure 5. Time series of the relevant source contributions to PM_{2.5} estimated by Absolute Principal Component Scores (APCS).

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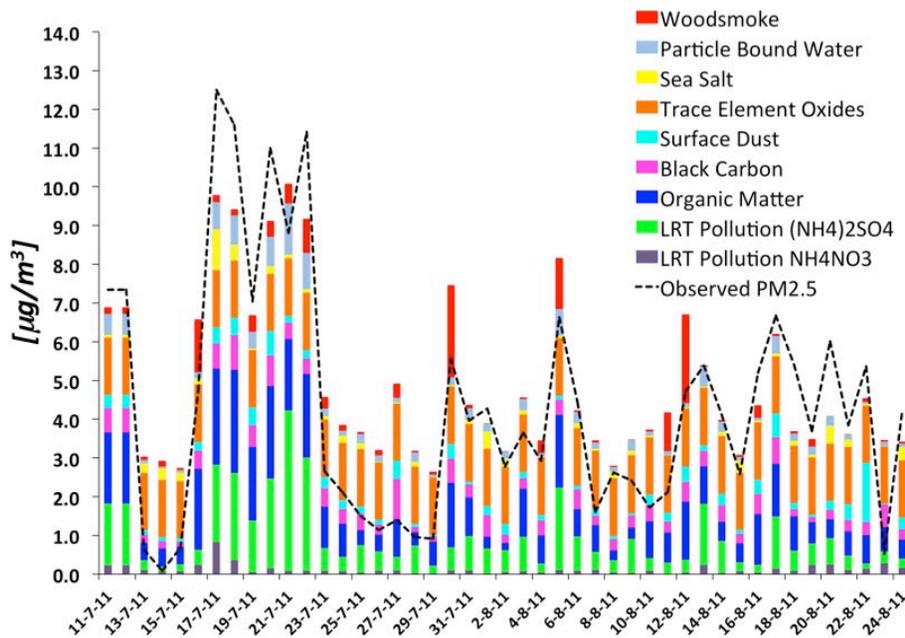


Figure 6. Time series of the relevant source contributions to PM_{2.5} estimated by Pragmatic Mass Closure (PMC) receptor model time series.

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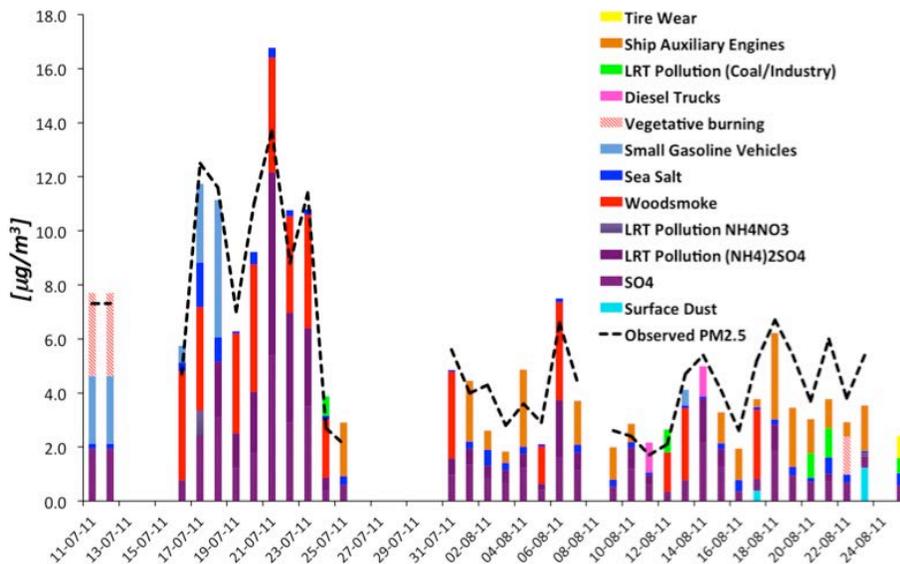


Figure 7. Time series of the relevant source contributions to PM_{2.5} estimated by Chemical Mass balance (CMB).

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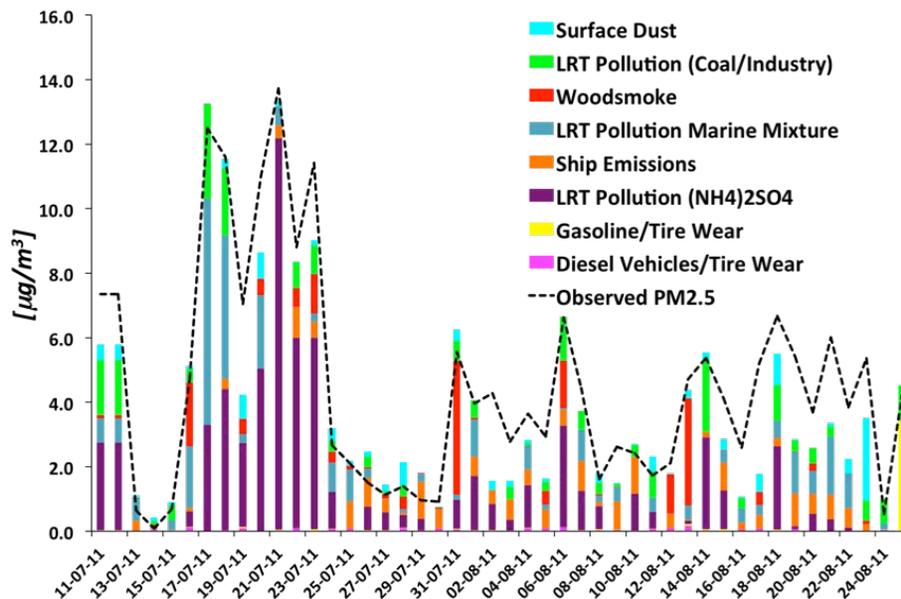


Figure 8. Time series of the relevant source contributions to PM_{2.5} estimated by Positive Matrix Factorization (PMF).

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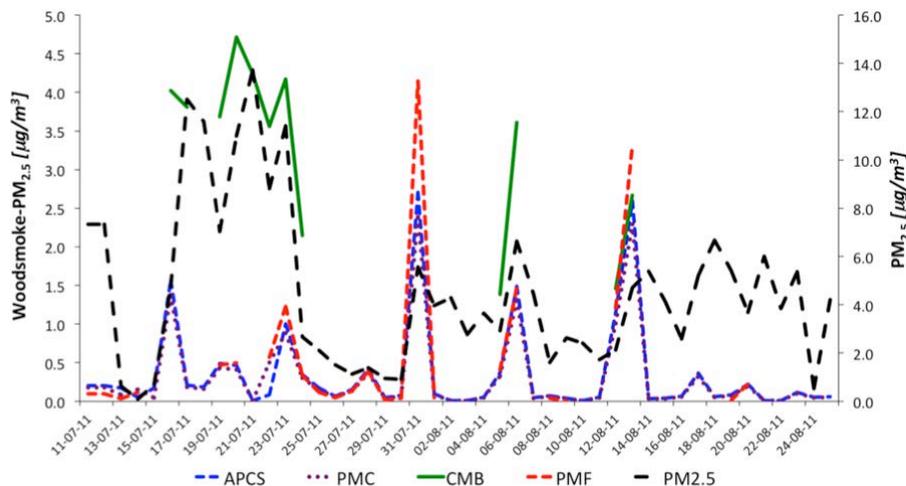
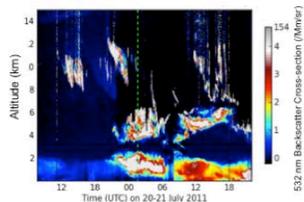


Figure 9. Time series of the woodsmoke contribution to the total $PM_{2.5}$ mass estimated from the four receptor models during BORTAS-B.

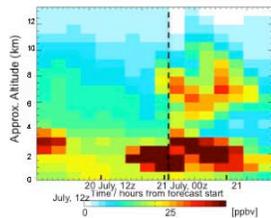
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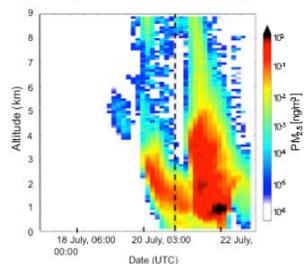
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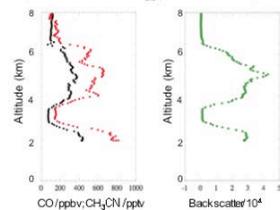
a) Lidar back scatter cross section DGS, 20/21 July 2011



b) GEOS-5 predicted CO at the DGS 20/21 July, 2011



c) FLEXPART vertical PM_{2.5} profile, DGS, 21 July 2011



d) Spiral aircraft profiles over the DGS, 21 July 2011

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Figure 10. Comparison of simultaneous observations **(a)** Lidar backscatter cross section DGS, 20/21 July 2011 **(b)** GEOS-5 CO forecast at the DGS 20/21 July 2011 **(c)** FLEXPART vertical PM_{2.5} profile, DGS, 21 July 2011 **(d)** Spiral aircraft profiles over the DGS, 21 July 2011. Vertical dashed lines in **(a)**, **(b)** and **(c)** indicate the time of the spiral aircraft profiles in **(d)**.

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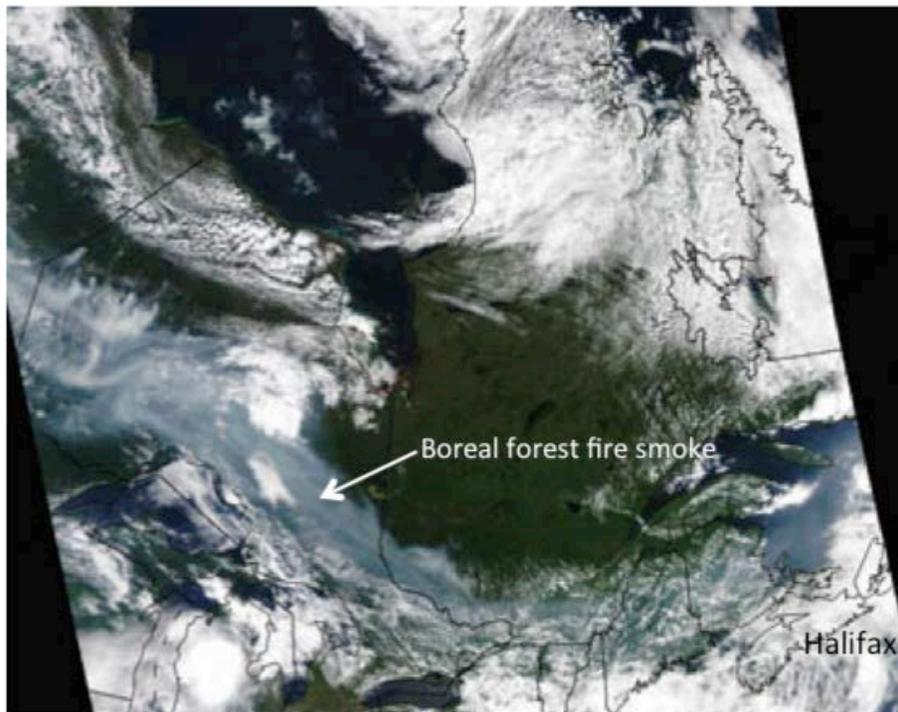


Figure 11. NASA AQUA MODIS true colour satellite image at 18:00 UTC on 18 July 2011 clearly showing boreal forest fire smoke from Northern Ontario advecting over Halifax, Nova Scotia.

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Figure 12. 5 day HYSPLIT air mass back trajectory arriving at 12:00 UTC overlaying the fire hot spot map for 28 July 2011.