



Impacts of forest fires on ambient near–real–time PM_{2.5} in Ontario, Canada: Meteorological analyses and source apportionment of the July 2011–2013 episodes

Uwayemi Sofowote, Frank Dempsey

Air Quality Monitoring and Assessment Unit, Environmental Monitoring and Reporting Branch, Ontario Ministry of the Environment and Climate Change, Etobicoke ON, M9P 3V6, Canada

ABSTRACT

The complexity of analyzing and predicting smoke plumes that originate from forest fire events and impact populated regions of southern Ontario motivates the innovative application of analytical techniques including trajectory–based receptor modeling for spatial source apportionment of the observed near–real–time particulate matter (PM) impacts. PM_{2.5} was selected as an indicator of a pollutant emitted by fires that could be transported over long distances (when entrained into the transport layer above the planetary boundary layer (PBL), and subject to sink and transformation processes) and be monitored using the existing air quality monitoring network. The source term modeling technique of simplified Quantitative Transport Bias Analysis (sQTBA) was applied to several summertime forest fire events to identify the locations of sources affecting air quality in Ontario during these events. Complementary techniques that helped to understand the movement of smoke plumes included satellite remote sensing of carbon monoxide and aerosols. All of these techniques, along with meteorological analysis, jointly provide a means of identifying the forest fire events that resulted in noticeably higher pollutant levels in Ontario. Specifically, three forest fire events in July of 2011, 2012 and 2013 were analyzed, and source regions of near–real–time PM_{2.5} concentrations were revealed to be both within Ontario and across northern Canada from Quebec to Yukon. The sQTBA was found to successfully identify the relative importance of various source regions contributing plumes from forest fires and non–wildfire related sources that caused higher pollutant levels that were measured in Ontario. The use of near–real–time PM_{2.5} data in this study facilitates the identification of the exact periods with high pollution impacts across multiple receptor sites, thus improving the overall quality of the analyses. This work shows how trajectory–based receptor models can be integrated with meteorological analyses for thorough source apportionment of wildfire–related pollution events.

Keywords: PM_{2.5}, remote sensing, near–real–time, sQTBA, long–range atmospheric transport

doi: 10.5094/APR.2015.001



Corresponding Author:

Uwayemi Sofowote

☎ : +1-416-235-6186

☎ : +1-416-235-6037

✉ : uwayemi.sofowote@ontario.ca

Article History:

Received: 21 February 2014

Revised: 19 June 2014

Accepted: 21 June 2014

1. Introduction

Forest fires are large, episodic natural sources of atmospheric pollutants released in amounts that may not be manageable by regular emissions control methods. This poses a challenge in air quality modeling and forecasting for populated regions where forest fire impacts could be significant. Episodic crests in ground–based concentrations of atmospheric pollutants such as particulate matter during summertime in relatively pristine locations may be indicative of the potential impacts of these forest fires. Meteorological analyses of geopotential height and pressure patterns, along with satellite–based remote sensing of some of the products of wildfire burning, provide some means to forecast the potential pathways and regions of impacts of observable plumes arising from known forest fire episodes.

Fires that develop in the boreal forests of Canada in summer may adversely affect ambient air quality in the more populated regions of southern Ontario during periods of northerly flow, when contributions to air pollution from industrial regions to the south and west of Ontario are minimized. Forest fires in northern Canada have been found to impact air quality as far south as Philadelphia, PA (Begum et al., 2005), New York (Wang et al., 2010) and some locations in the southeastern US (Wotawa and Trainer, 2000). Potentially negative health effects may be associated with fine

particulate matter (Pope et al., 2004) and various toxic substances including organic, inorganic as well as metallic contaminant species associated with particulates (Harrison and Yin, 2000; Neuberger et al., 2004; Ondov et al., 2006).

Meteorological forecast models and products are used to help assess the potential for forest fire events to impact the air quality at locations within the region downwind of the geographical sources of the fires. However, in cases involving concurrent multiple forest fires and other sources, absolute and relative contributions of source regions to the pollutant levels at the receptor sites are not typically known (Hecobian et al., 2011). Thus, there is a clear need for source apportionment in which the observed concentration data of a suitable tracer monitored at the receptor site can be used to resolve the potential contributory source regions. For real–time source apportionments, the challenge in reliably detecting biomass combustion has been in identifying practical unique tracer species.

Many organic and inorganic pollutants produced in forest fires (in addition to CO₂ and CO) are generally not unique to this source type. Tracers of biomass combustion have historically included high levels of certain organic species such as lignins, lignans, cellulose and their oxidation and pyrolysis products (Simoneit, 2002), specific polycyclic aromatic hydrocarbons (PAH) such as

retene (Ramdahl, 1983), and other substituted phenanthrenes (specifically, elevated ratios of dimethylphenanthrenes i.e., 1,7-DMP to 2,6-DMP) that have been suggested as tracers for coniferous (soft) wood combustion (Benner et al., 1995; Yunker et al., 2002). More universally, levoglucosan, a pyrolysis product of cellulose, can be used to trace grass, soft wood and hard wood burning (Simoneit, 2002; Jordan et al., 2006).

A good tracer species should be largely conserved during transport to remove the possibility of complicating chemical transformations and some of these pollutants have been used to track plume chemistry and ageing. Jaffe and Wigder (2012) showed that the ozone emission ratio (ER) i.e., $\Delta O_3/\Delta CO$ generally tended to increase with age of the plume and this may be important for long range advection of biomass burning emissions. Hecobian et al. (2011) commented on the ability to determine HCN in air masses in the Arctic that have undergone long-range transport. A clear increase in the ER of HCN (i.e., $\Delta HCN/\Delta CO$) could be observed for air masses that had originated in Siberia and Asia against those from Europe and North America (which included boreal biomass emissions from Canada and local wildfires/urban emissions from California, USA). Akagi et al. (2013) have also recently determined a fairly consistent ER for HCN from plumes originating from pine forest fires.

A more complete review of biomass burning markers has been presented by Simoneit (2002) but use of these organic tracers may require fairly rigorous laboratory preparation and in some cases copious amounts of the species have to be sampled so as to be detectable (Jordan et al., 2006). Laboratory methods generally lead to more infrequent sampling, thereby leaving gaps in monitoring records which will affect the ability to make timely population exposure/health based decisions. Thus, the utility of some of these tracers is negated by the need for near-real-time pollution data in large ambient air quality monitoring networks. For real time apportionments, concomitant elevation of carbon monoxide (CO), volatile organic species (e.g. CH_4), carbonaceous aerosols (e.g., OC and EC), potassium (K) and mercury (Hg) may provide a better estimation of wildfire impacts. More recently, elevated levels of real-time Delta C (the difference in black carbon concentrations measured at 370 nm and 880 nm) have been shown to be a reliable indicator for wood combustion (Wang et al., 2011; Wang et al., 2012). However, these real-time tracers may not be routinely monitored since they require specifically-designed instrumentation that is financially and logistically prohibitive to deploy across an entire ambient air quality monitoring network. Concentrations of $PM_{2.5}$ may increase during forest fires but it is not a selective tracer for these natural events due to the potential for release from multiple sources. Thus, in this work, periods with increased summertime $PM_{2.5}$ levels have been analyzed to monitor

the impacts of forest fires under favorable meteorological conditions (which provide unambiguous directions of transport for a wildfire plume that injects smoke above the planetary boundary layer (PBL), lack of significant precipitation, and subsidence toward the ground near the surface).

The objective of this work was to reveal regional locations and quantitatively determine the contributions of the probable sources of $PM_{2.5}$ impacting receptor sites using the simplified quantitative transport bias analyses (sQTBA) receptor model (Brook et al., 2004; Zhou et al., 2004; Zhao et al., 2007). This was done for three significant forest fire events with varying complexity due to multiple potential source regions during 2011, 2012 and 2013 when detectable smoke plumes correlated with known forest fire sources. July was selected because it is the month with the largest number of forest fires, as well as greatest area burned each year in Canada between 1990–2013 (NFD, 2013). The sQTBA model was used in conjunction with air mass back trajectories, meteorological analysis techniques, and satellite remote sensing products.

2. Experimental Design

2.1. Sampling locations

Ontario Ministry of the Environment (OMOE) operates a province-wide ambient air monitoring network that hosts instrumentation and technologies allowing real-time ambient monitoring of specific contaminant species that impact the air quality in the province (OMOE, 2013). Thus, $PM_{2.5}$ is monitored at 40 air quality stations and one research site in the province on a semi-continuous hourly basis. Other species that are monitored on a real-time basis in Ontario's ambient air quality monitoring network include O_3 (all stations) and NO_x (most stations) while CO and SO_2 are routinely monitored at only four and ten stations respectively (OMOE, 2013). This study focuses on the eight ambient air quality stations located in central and northern Ontario listed in Table 1. The OMOE monitoring station shelters are temperature- and humidity-controlled for the proper operation of the monitoring instrumentation. A few stations used in this study are also road-side monitoring stations which in our ambient air quality network are defined as stations that are within 100 m of a major roadway that experiences daily traffic volumes greater than 10 000 vehicles per day, (OMOE, 2013). While it is impossible to rule out contributions to $PM_{2.5}$ from these roadways during the periods of interest, we include in this work time-series plots of hourly $PM_{2.5}$ concentrations spanning a thirty-day period so that the background levels can be seen to be completely insignificant in comparison with the spikes in concentration that coincided with the arrival of the smoke plumes.

Table 1. Locations of the air quality monitoring stations used in this study

Station Location	Station Type ^a	Latitude	Longitude	Population/ Population Density (/km ²) ^b	Geographical Unit ^b	Elevation (m AMSL)	Air Intake Height (m AGL)
Thunder Bay	A/RS/C/N	48.3794	-89.2902	121 596/47.6	CMA	207	15
Sault Ste. Marie	A//N	46.5332	-84.3099	79 800/99.1	CA	252	8
Sudbury	A/C/N	46.4757	-80.9630	160 770/47.1	CMA	259	3
North Bay	A/RS/N	46.3232	-79.4493	64 043/81.2	CA	223	4
Ottawa Downtown	A/C/N	45.4343	-75.6760	1 236 324/196.6	CMA	72	4
Parry Sound	A/N	45.3379	-80.0382	6 191/463.3	Town	181	5
Cornwall	A/N	45.0180	-74.7352	58 957/115.8	CA	59	4
Morrisburg	A/N	44.8998	-75.1899	2 756/429.9	Centre	69	5

^a Station type denotes the network(s) associated with the station: A=ambient, RS=road-side, C=Canada-wide Standard (CWS), N=National Air Pollution Surveillance (NAPS). Descriptions of these networks can be found in greater detail in Ontario's annual air quality report (OMOE, 2013)

^b See Ontario data in Focus on Geography Series, 2011 Census (Statistics Canada, 2013). CMA=Census metropolitan area, CA=Census agglomeration. Centre=Population centre

2.2. Instrumentation

In Ontario, continuous real-time (hourly) ambient $PM_{2.5}$ data monitored between 2002 and 2012 were obtained with the Tapered Element Oscillating Microbalance (TEOM) developed by Patashnick and Rupprecht (1991), equipped with the Sample Equilibration System (TEOM–SES Series 1400AB; Thermo Fisher Scientific, Massachusetts, USA) within the province's air quality monitoring network. The TEOM–SES has previously been described in detail (Meyer et al., 2000). Briefly, the TEOM–SES uses a tapered element oscillating microbalance that converts changes in frequency of a particulate matter–impacted vibrating glass tube to mass loading and concentration on a continuous near–real-time basis. The sample equilibration system is a Nafion dryer configured to remove particulate–bound moisture at 30 °C which ideally should result in a smaller loss of semi–volatile particulate matter components.

As of 2013, $PM_{2.5}$ data are collected every hour at the monitoring stations with a Thermo Synchronized Hybrid Ambient Real-time Particulate (SHARP 5030, Thermo Fisher Scientific, Massachusetts, USA) monitor which is a USEPA Class III Federal Equivalent Method (FEM) analyzer (U.S. EPA, 2011). The SHARP exploits the ability of aerosols to attenuate beta radiation and scatter light (nephelometry). For real-time monitoring purposes in the province's air quality index network, $PM_{2.5}$ concentrations are computed and reported continuously as hourly averages.

TEOM and SHARP instruments are all fitted with $PM_{2.5}$ size inlets consisting of PM_{10} impactors coupled to $PM_{2.5}$ sharp cut cyclones and inlets operated at an air volumetric flow rate of 1 m³/h.

2.3. Data sets and pre-treatment

Hourly TEOM–SES or SHARP 5030 $PM_{2.5}$ data collected at the stations of interest were polled from the data loggers and archived in Ontario's Air Quality Information System (AQIS) and are publicly available on the Ontario Ministry of the Environment website (OMOE, 2010). The datasets and stations used in this study were comprised of $PM_{2.5}$ collected in July 2011 (Thunder Bay, Sault Ste. Marie, Sudbury, North Bay, and Ottawa), July 2012 (Thunder Bay, Sault Ste. Marie, Sudbury and Parry Sound) and July 2013 (Thunder Bay, Sault Ste. Marie, Sudbury, Ottawa, Morrisburg, and Cornwall).

The $PM_{2.5}$ monitors underwent regular inspection by station operators and were calibrated according to the network operational procedures. The concentration data were subject to strict quality assurance and quality control (QA/QC) review by the station operators on a daily basis and then by the network administrators on a quarterly basis (note that data for July 2013 used in this work has not been subjected to the quarterly review). No attempt was made to correct the TEOM data used in this work to the new FEM. Diagnostic parameters, monitored and stored in the data loggers, were inspected during the validation of the data in order to ensure that the samplers performed properly and could be used to report $PM_{2.5}$ concentrations. In some cases, data were missing due to instrumental failures and technical challenges while others were flagged and removed before further analyses due to operational requisites (e.g., data associated with error codes, site inspections, instrument calibrations etc). In all, >95 percent of expected hourly samples were available at all sites during the sampling periods.

3. Methods

3.1. Meteorological forecast/models

Analysis of the meteorological pattern details relevant to transport of wildfire smoke includes consideration of geopotential

height fields, winds at various levels, transport and dispersion factors, stability, convection and precipitation.

Geopotential heights at middle and upper levels of the atmosphere (which are largely unaffected by surface frictional effects) indicate the positions of large-scale ridges and troughs in the pressure pattern (Lackmann, 2011). Geopotential heights of the 700 hPa level coincide approximately with altitudes about 3 km above sea level, and heights of the 850 hPa level coincide approximately with altitudes about 1.5 km above sea level. Increasing gradients in the geopotential height indicate strengthening winds at these altitudes, and streamlines along the height contours show wind directions in the velocity field (Stull, 2000). In Ontario and the Great Lakes region, streamlines at or above the 850 hPa layer adequately represent transport directions in the lower free troposphere. While convection and other diurnal and seasonal effects may affect the stability and potential dilution of pollutants in the 850–700 hPa layer, winds at the 850 hPa level lie above the yearly median PBL height of 700–800 m for midday, globally averaged and derived using the parcel method as recommended by Seidel et al. (2010) for air quality modeling. For the wildfire plume injection median height of about 850 m above ground found by Val Martin et al. (2010) for boreal biomes, the choice of 850 hPa level winds in this study should reasonably represent transport directions above the PBL.

In this study, National Centers for Environmental Prediction (NCEP) reanalysis data (Kalnay et al., 1996) were used to plot the charts of geopotential heights. Conditions favorable for transport of smoke plumes include positions of air masses and resulting circulation patterns that result in advection of pollutant species from regions of potential wildfires to receptor locations in Ontario. As well as horizontal advection, subsidence is also required to vertically transport smoke from the transport levels above the PBL downward to the surface (Goody, 1995), and regions of subsidence are generally located west of low pressure troughs and east of high pressure ridges. Favorable conditions also require the subsidence to occur during an appropriate period, generally during the daytime, when convective mixing is most active and for a sufficient duration, typically several hours or longer, to most likely result in the smoke plume causing a measureable effect at the surface receptors.

Some remote sensing and other related products may also provide evidence of the presence in the atmosphere of products of wildfires and indicate location, depth and movement of aerosol plumes. Satellite remote sensing of total column CO may provide a useful indicator of the location and movement of smoke, since CO produced from the combustion products of a wildfire is not highly reactive and has a sufficiently long lifetime to persist and act as a tracer of smoke plumes (Seinfeld and Pandis, 1998; Wotawa and Trainer, 2000). Total column amount of CO was measured by the AIRS (Atmospheric Infrared Sounder) instrument on the polar-orbiting Aqua satellite and is available from NASA Giovanni Data and Information Services Center (NASA, 2013) but unavailable during June 2013. Another useful tool is the GOES (Geostationary Operational Environmental Satellite) product GOES Aerosol/Smoke Product (GASP), which reveals the locations of enhanced aerosol concentrations and aerosol plumes (NOAA, 2013a). GASP images display aerosol optical depth (AOD) which is a vertical column measurement and includes aerosols both near the surface and aloft. AOD is, however, limited to cloudless regions. Time-series plots of $PM_{2.5}$ concentrations may be used to illustrate the west-to-east progression of the smoke plumes (such as in the 2011 and 2012 events in this study, but not the 2013 event in which smoke came from sources northeast of the receptor sites). Comparison of the other monitored pollutants, such as O_3 and NO_2 (but not CO which is not monitored at the receptor sites in this study), might be examined to look for influence of a wildfire plume on O_3 concentrations measured at the monitoring sites. Charts have been included in the supplement for the 2012 events. Other analysis

products and techniques are also available to help in the diagnosis and prediction of wildfire plumes with the goal of anticipating potential effects on ground-level pollutant concentrations.

3.2. Trajectory computations

Five-day back trajectories (segmented into hourly endpoints) for the receptor sites were computed using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2010) with winds from the Global Data Assimilation System (GDAS) database for the periods of sampling (i.e., July 2011, July 2012 and July 2013). Errors with the HYSPLIT model are 15–30% of the total travel distance (NOAA ARL, 2008). In general, the use of ensemble methods such as the one in this work is recommended (Hopke et al., 2005) provided there are sufficient data since they average out the random errors associated with trajectories.

Relevant trajectory outputs for this study included latitude–longitude coordinates and time segments of endpoints. In the current work, the HYSPLIT back trajectories were computed for 500 m AGL arrival height to fairly minimize surface interactions before air masses reach the receptor (Begum et al., 2005; Gibson et al., 2013) and initiated every two hours each day. The use of back trajectories arriving at 500 m AGL and the resulting lack of resolution of coastal circulations near lakes and rivers, and other low-level circulations, could however be a limitation in trajectory-based receptor modeling where potential local sources may be important.

3.3. Simplified quantitative transport bias analysis (sQTBA)

QTBA (Keeler and Samson, 1989) has been described in detail by other researchers (Brook et al., 2004; Zhou et al., 2004; Brereton and Johnson, 2012; Sofowote et al., 2014). The technique of QTBA uses ensembles of back trajectory analyses from multiple receptor locations, combined with observed concentrations of air pollutants, such as $PM_{2.5}$ in this study, as well as estimates of the transition probability density function over long distances to produce spatial patterns of mean concentration associated with the air flow from upwind regions for each site. Source regions may be near or distant from the receptors, and the technique computes the Gaussian probabilities that source regions contributed air masses that could arrive at the receptor sites. This is called the natural transport potential which when overlaid on a system of grid cells (such as that defined by longitudes and latitudes) yields a potential field. Scaling the natural transport potential by the concentration measured when the trajectories arrived at the receptor produces the concentration-weighted transport field which can in turn be normalized by each grid cell's natural transport potential to yield a QTBA concentration value for that given grid cell. Higher values of QTBA in any given grid cell indicate concentration values at the receptor attributable to the source region based on the natural transport potential field of trajectories upwind of the receptor site. They are not the absolute concentrations of the species of interest at the source region (which are expected to be much higher). Thus, the QTBA results presented in this work are intended to show the source regions most often associated with higher $PM_{2.5}$ concentrations in the receptor locations. The purpose is to determine the geographic regions that systematically contribute to greater concentrations of $PM_{2.5}$ at the receptors. Simplification of the original model involved the non-inclusion of terms for chemical transformations, wet and dry deposition as well as the inclusion of a simple weighting scheme to increase confidence in model results (Zhao et al., 2007).

More details on the model can be found in the supplementary information section. In this application, sQTBA was used to reveal the potential source regions of $PM_{2.5}$ measured during periods of elevated PM levels when meteorological conditions suggested a potential for transport of smoke plumes from forest fires. MetCor

(Rastogi, 2013), a software package that computes trajectory ensemble models (TEM) (Sofowote et al., 2011; Venier et al., 2012; Sofowote et al., 2014), was used to obtain sQTBA results in $0.75^\circ \times 1^\circ$ (LAT–LON) grid cells which at latitudes of $\approx 40^\circ$ gives an approximate $80 \text{ km} \times 80 \text{ km}$ grid cell. Visualization of the sQTBA outputs was done using ArcGIS 9.3 (ESRI, California, USA).

4. Results and Discussion

4.1. The July 2013 event

The forest fire event of July 02, 2013, being the most recent of the selected cases, is first to be discussed below. Three ambient monitoring stations within about 120 km of each other in the eastern portion of the province measured sharp spikes in their $PM_{2.5}$ concentrations on the dawn of July 02 that were sustained for 12–18 hours (maximum values at Ottawa: $50 \mu\text{g}/\text{m}^3$, Cornwall: $70 \mu\text{g}/\text{m}^3$ and Morrisburg: $90 \mu\text{g}/\text{m}^3$). For context, the 24-h Canada-wide Standard (CWS) used in Ontario is $30 \mu\text{g}/\text{m}^3$ (OMOE, 2013) while the metric used for calculating the threshold for the poor category of AQI (air quality index) due to the $PM_{2.5}$ sub-index is a three-hour rolling average value of $45 \mu\text{g}/\text{m}^3$. A smaller-scale increase in $PM_{2.5}$ had previously been recorded at Thunder Bay and Ottawa on the evening of June 30 and by the evening of July 03, Sudbury had experienced levels of $PM_{2.5}$ concentrations exceeding $30 \mu\text{g}/\text{m}^3$ while Sault Ste. Marie had also experienced relatively high levels of $PM_{2.5}$ (Figure 1a). These high $PM_{2.5}$ levels could not be explained by the typical diurnal and seasonal variations and were not accompanied by significant changes in the levels of other criteria pollutants; thereby suggesting that long-range advection of the particulate matter species may have played a more important role than local emissions and processes related to photochemical production (see the discussion for the July 2012 event below for a clear example). Relevant meteorological details include an upper trough that moved slowly across northeastern Ontario during the period June 27 to July 02 while an upper ridge was quasi-stationary over western Canada, resulting in northerly flow (on the west side of the trough) across northern Ontario toward the Great Lakes region. During the afternoon of July 01, trajectories at the 850 and 700 hPa levels originating from a region of wildfires in northwestern Quebec, east of James Bay, were directed southward to eastern and northeastern Ontario. At the surface, a high pressure ridge moved eastward over northern Ontario during July 01 and subsidence ahead of the ridge transported smoke aloft downward to the surface. The chart in Figure 1b shows the upper-level (700 hPa level) ridge over northwestern Ontario and the trough east of Ontario during July 01, and wind flags at the 850 hPa level (NOAA ARL READY, 2013).

A plume of aerosols, as measured by GOES satellite remote sensing, can be seen in Figure 1c distinctly spreading southwestward toward central and southern Ontario during the morning of June 30 (before cloudiness obstructed the visibility of the aerosols). The red pixels in the AOD chart indicating maximum optical depth of the aerosol plume across northeastern Ontario correlate particularly well with the flow southwestward from the active fires in northwestern Quebec toward northeastern Ontario. The southward progression of the aerosol plume from Ottawa to the southeastern Ontario stations of Cornwall and Morrisburg can also be discerned in Figure 1a.

The most significant aspect of the synoptic-scale pattern during this event was the nearly stationary western ridge–eastern trough configuration which provided prolonged conditions suitable for smoke from forest fires in northern Quebec to advect southward to southern Canada.

Individual sQTBA analyses for these six stations were carried out and joint results were obtained by averaging individual sQTBA results per grid cell (i, j) after setting the minimum number of receptor sites to obtain a valid joint sQTBA result to five. The

sQTBA analysis was performed on a 10-day period from June 25 – July 05 that bracketed the intense $PM_{2.5}$ period thereby reducing the potential for other $PM_{2.5}$ sources to be identified, e.g. the Lac Megantic rail fires that occurred on July 06, 2013 (Transportation Safety Board of Canada, 2013). Figure 1d reveals the sQTBA-derived potential $PM_{2.5}$ source regions overlaid on the fire hotspot map for July 01, 2013 sourced from the Canadian Wildland Fire Information System CWFIS (Natural Resources Canada, 2013). This map clearly indicates that the most important potential sources during this time are located around Hudson Bay, specifically around James Bay and Foxe Channel/Basin areas in northern Quebec and in Nunavut, Canada's northernmost territory.

At this time, large wildfires around the border regions of eastern Quebec and Labrador were also burning and can be observed in the fire hotspot map for July 01, 2013 (Figure 1d). Fire

events in Ontario and adjoining provinces can be seen clearly, but coverage of the northernmost parts of Quebec as well as Nunavut are completely absent from these maps, possibly due to the limitations of satellite-derived hotspot identification, such as cloud cover, smoke, and other factors that affect the algorithms used for deriving hotspots from remote sensing imagery. Although other sources of $PM_{2.5}$ cannot be completely ruled out even with the restricted data set, the sQTBA analysis strongly suggests that the fires of northern Manitoba and eastern Quebec did not make a significant contribution to the impacts observed in Ontario when compared with more northerly regions of Quebec. This example shows how the sQTBA ensemble method distinguishes the relative importance of source regions with fires occurring simultaneously and complements the source regions derived by meteorological analysis as well as the evidence provided by satellite remote sensing.

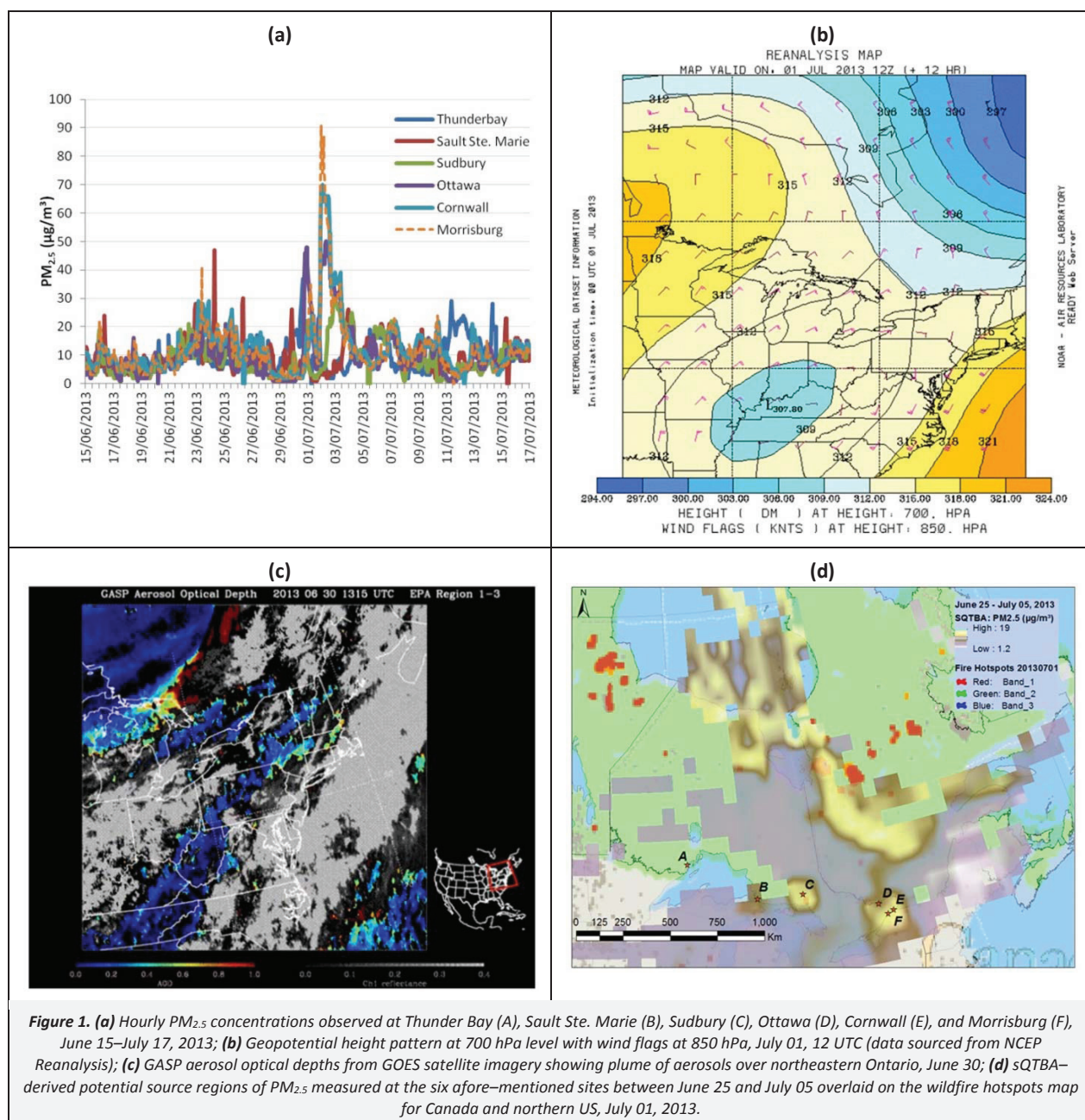


Figure 1. (a) Hourly $PM_{2.5}$ concentrations observed at Thunder Bay (A), Sault Ste. Marie (B), Sudbury (C), Ottawa (D), Cornwall (E), and Morrisburg (F), June 15–July 17, 2013; (b) Geopotential height pattern at 700 hPa level with wind flags at 850 hPa, July 01, 12 UTC (data sourced from NCEP Reanalysis); (c) GASP aerosol optical depths from GOES satellite imagery showing plume of aerosols over northeastern Ontario, June 30; (d) sQTBA-derived potential source regions of $PM_{2.5}$ measured at the six afore-mentioned sites between June 25 and July 05 overlaid on the wildfire hotspots map for Canada and northern US, July 01, 2013.

4.2. The July 2012 event

Higher than normal $PM_{2.5}$ levels were recorded briefly at Thunder Bay in the morning of July 21, 2012. A few hours later, $PM_{2.5}$ levels greater than $45 \mu\text{g}/\text{m}^3$ and sustained for about 8 hours were measured east of Thunder Bay, first at Sault Ste. Marie and subsequently at Parry Sound and Sudbury by July 22 (see Figure 2a). Peak $PM_{2.5}$ concentrations exceeding $80 \mu\text{g}/\text{m}^3$ were observed at Parry Sound in the morning of July 22 and did not return to levels lower than $20 \mu\text{g}/\text{m}^3$ at this station and at Sault Ste. Marie and Sudbury until the morning of July 23. A high pressure ridge was south of Ontario during July 21, a trough was moving eastward over northern Ontario, and a cold front moved eastward across the northern Great Lakes region. Winds became northwesterly over the northern Great Lakes area, followed by post-frontal afternoon convection. Upper level winds were northwesterly, and subsidence following the cold front allowed smoke aloft to descend to the surface. The chart in Figure 2b shows the northwesterly flow in the pattern of geopotential heights at the 700 hPa level plotted with wind direction flags at the 850 hPa level. The Hazard Mapping System (HMS) Fire and Smoke analysis (not shown in this work) produced by NOAA (NOAA, 2013b) for July 21, reveals many active

fires plotted in northwestern Ontario, and a large plume of smoke spreading eastward over northern Ontario. Some evidence of the combustion products of the wildfires can be seen in the plot of total column CO from AIRS, coinciding with the plume trajectory, during the period July 20–23 from AIRS in Figure 2c.

The sQTBA analysis run for a 10-day period spanning July 15–25 at these four sampling locations is overlaid on the forest fire hotspots (Natural Resources Canada, 2013) as of July 14, 2012 in Figure 2d. The sQTBA map shows relatively strong potential sources of $PM_{2.5}$ in northwestern Canada during this period that coincide with fire hotspots in Nunavut, Yukon and northern Alberta, and the CO remote sensing map (Figure 2c). This northwestern region contains the most intense potential sources, while western Ontario, Manitoba and Minnesota and their border regions are also indicated as strong potential source regions in Figure 2d. A possible source region appears to be lake surfaces and this is a result of back trajectories passing over lakes (particularly south of Thunder Bay and Sault Ste. Marie) onto the land. This pattern of partial stagnation and on-shore lake breeze results from the weak local high pressure cells that form over the cool lake surfaces during summer days.

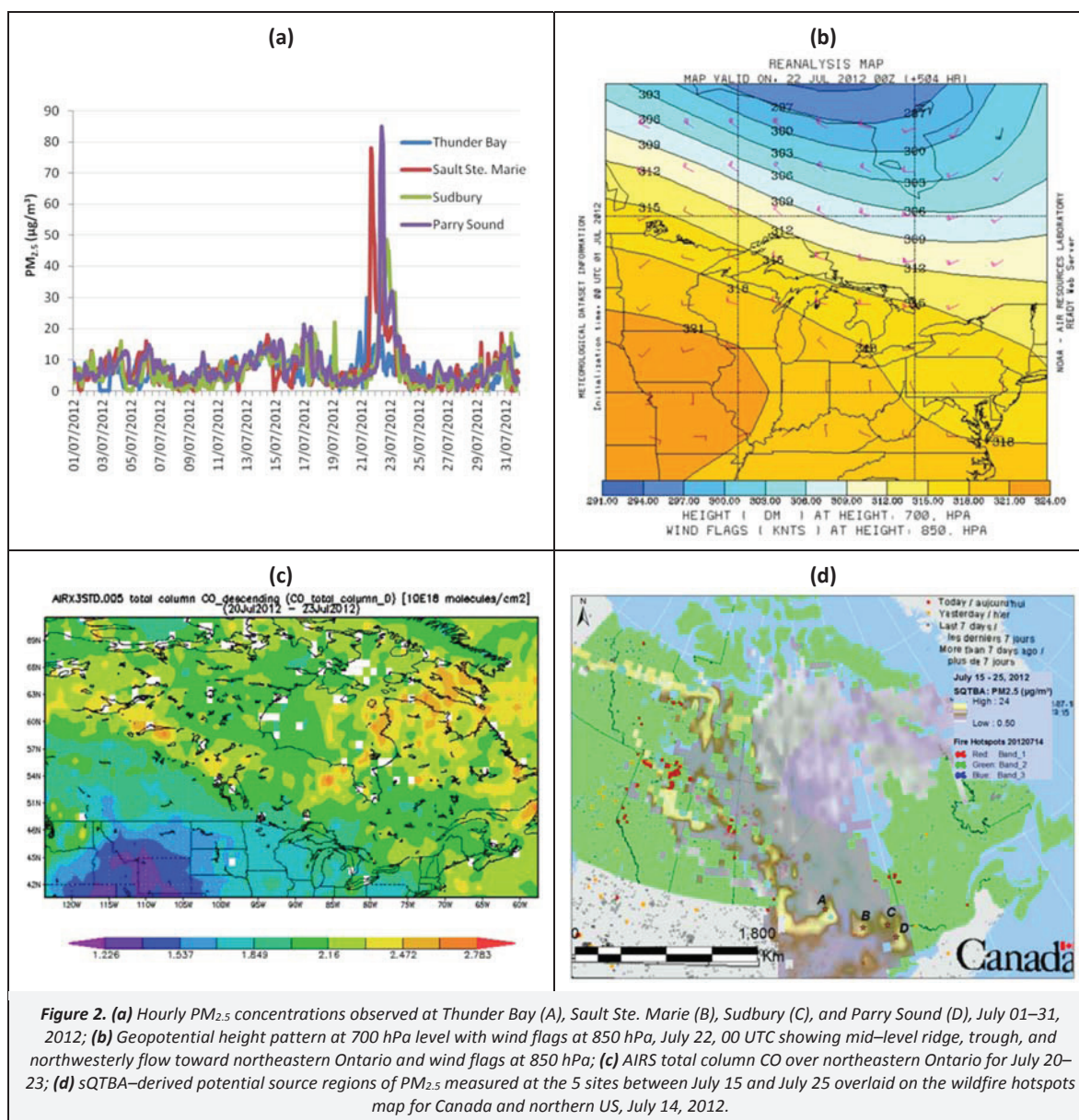


Figure 2. (a) Hourly $PM_{2.5}$ concentrations observed at Thunder Bay (A), Sault Ste. Marie (B), Sudbury (C), and Parry Sound (D), July 01–31, 2012; (b) Geopotential height pattern at 700 hPa level with wind flags at 850 hPa, July 22, 00 UTC showing mid-level ridge, trough, and northwesterly flow toward northeastern Ontario and wind flags at 850 hPa; (c) AIRS total column CO over northeastern Ontario for July 20–23; (d) sQTBA-derived potential source regions of $PM_{2.5}$ measured at the 5 sites between July 15 and July 25 overlaid on the wildfire hotspots map for Canada and northern US, July 14, 2012.

Other regions indicated in the sQTBA map may have arisen from other sources of PM_{2.5} not related to the forest fires during this period, but the strength of the sQTBA analysis in this event is the identification of the most important potential source regions.

Additionally, the west-to-east progression of the smoke plume across northeastern Ontario may be seen in the chart plotting PM_{2.5} hourly concentrations (Figure S1a in the Supporting Material, SM). The timing of the concentration peaks is consistent with transport eastward from Thunder Bay (on the west end of Lake Superior) to Parry Sound and Sudbury (near the east end of Lake Huron) the following day and the daytime peaks are consistent with daytime diurnal convection helping the plume to mix downward through the boundary layer to the ground. The air quality monitoring sites also monitor O₃ and NO₂ (except at Sudbury) and these pollutants may be plotted during the period of the PM_{2.5} concentration peaks. The charts for PM_{2.5}, O₃ and NO₂ for each of the stations are shown in Figure S2 (see the SM) but no distinctive effect appears in the concentrations of O₃ and NO₂ that could be confidently associated with a smoke plume. Distinct effects on O₃ concentrations at Ontario's air quality monitoring stations from wildfire plumes are recognizable in some events (Dempsey, 2013) but the O₃ profiles during the July 20–23, 2012 event were entirely consistent with typical diurnal trends and the trends across southern, central and eastern Ontario. Only the PM_{2.5} concentrations increased sharply to anomalously high values and could be confidently correlated with the evidence from the active forest fire sources, transport conditions for smoke plumes, remote sensing CO data, and the meteorological conditions favorable for vertical mixing to the ground.

4.3. The July, 2011 event

PM_{2.5} concentrations were observed to approach 85 µg/m³ by noon of July 19, 2011 and remain higher than 30 µg/m³ for the next 48 hours at the Thunder Bay air monitoring station (Figure 3a). Some other monitoring stations to the east of Thunder Bay (specifically, Sudbury, North Bay and Ottawa) were later impacted by high PM_{2.5} episodes (~40 µg/m³) over the next two days. The Sault Ste. Marie air monitoring station, however, experienced a sharp but brief (lasting three to four hours) spike in PM_{2.5} concentrations on the evening of July 17 that was not observed elsewhere. The synoptic meteorological situation that developed and potentially allowed smoke from wildfires over northwestern Ontario to be transported to receptors in northeastern Ontario (as well as in Michigan and New York) included a high pressure ridge at upper and middle layers of the atmosphere over the western Great Lakes region and northwestern Ontario during July 19, as well as a low pressure trough over northern Quebec. This pattern resulted in a north-westerly flow and transport of pollutant-laden air parcels from northwestern Ontario toward the air monitoring stations in northeastern Ontario. Figure 3b shows the pattern of geopotential heights at the 700 hPa level with wind flags at 850 hPa. Remote sensing evidence for the movement of smoke along this transport direction is shown in the plot of total column CO (Figure 3c) and the GASP aerosol plot (see the SM, Figure S3). Both figures clearly show evidence of smoke products in the troposphere spreading downstream, from the location of the fires in northwestern Ontario, toward northeastern Ontario. A surface high pressure ridge was over the Great Lakes region and subsidence on the east side of the ridge allowed downward transport from middle layers of the atmosphere toward the surface.

The result of the joint sQTBA analyses for July 15 – 25, 2011 at these five stations (minimum number of receptors was set at three) is in agreement with Figure 3b and Figure S3 and it reveals that this area of northwestern Ontario (northwest of Thunder Bay and near the border regions of Manitoba and Ontario) was among the strongest potential source regions of PM_{2.5} (Figure 3d). Other potential PM_{2.5} sources represented in the sQTBA map include the

active fire sources around James Bay and Hudson Bay (also shown in Figure 3d) as well as fires in Idaho that were plotted on the NOAA HMS fire and smoke charts which are publicly available (NOAA, 2013b). The strong US Midwest contributions also observed may be due to PM_{2.5} sources not related to forest fires.

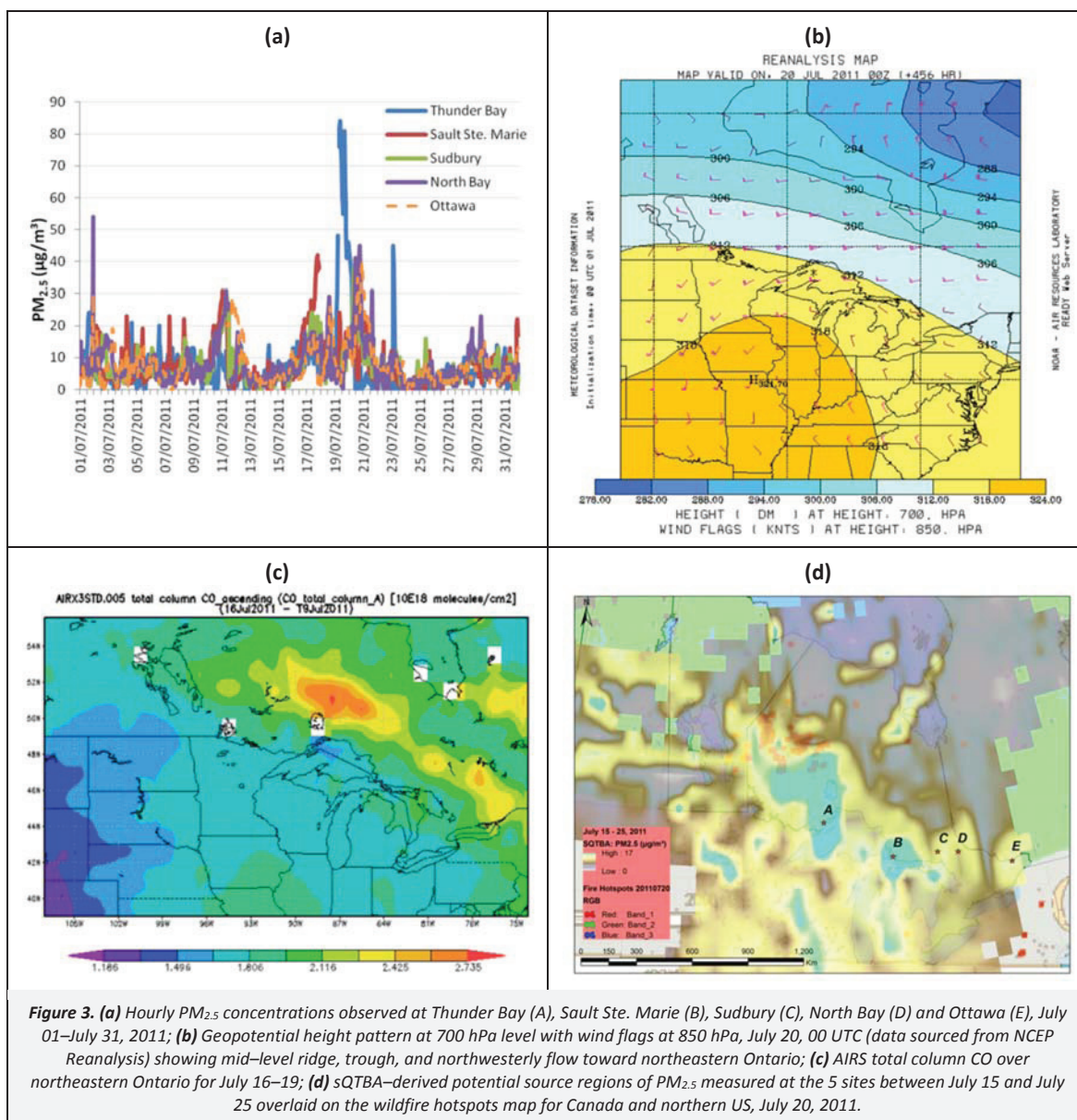
Local sources of PM_{2.5} could also be observed around Sault Ste. Marie in Figure 3d. These local sources, which were possibly not related to any forest fires around Sault Ste. Marie (Figure 3d shows no fire hotspot in this area), may be responsible for the elevated levels of PM_{2.5} observed on the evening of July 17, 2011. As with the 2012 event described earlier, back trajectories appear to originate from the local high pressure cells that develop over the lake surfaces during summer.

5. Summary and Conclusions

The success of the complementary techniques of sQTBA analyses combined with meteorological analyses in the events examined above illustrates the applicability of the technique to events of varying complexity. The events examined included cases where the sources seemed easy to identify by meteorological analysis but the presence of other potential sources (from other active forest fires and from non-wildfire-related sources) made the situation more complex.

The impact of forest fires on particulate matter pollution in a given region may often be difficult to quantify due to the intermittent and unpredictable nature of wildfire occurrences and the meteorological conditions prevailing during periods of emissions release. Other variable factors, such as the geographical extent of the fires and the nature of biomass burned could also make it challenging to summarize the forest fire contributions to particulate matter pollution in an emissions inventory. Thus, in Ontario, potential impacts of forest fires are forecasted by determining whether prevalent meteorological conditions during these fires are suitable for atmospheric transport of released pollutants and further analyzed after allowing a reasonable period of time to elapse for potential incidence by observing the fluctuations of 'marker' pollutant species measured at receptor sites. While tracer species unique to forest fires/biomass burning exist, their use in large monitoring networks is not widespread due to logistical limitations. Thus, combinations of meteorological analyses and ground-based measurements of particulate matter (e.g. PM_{2.5}) may be the only available tools when apportioning the sources and contributions of forest fires.

The use of real-time data helps to isolate potential incidences of impacts from ambient sources of pollution with greater resolution than more traditional one-in-six days, 24 h-averaged data. The benefit of this is immediately observed in this work where concentrations of PM_{2.5} were only elevated for a few hours before steeply declining to near-background levels. Clearly, averaging the data over a longer period will greatly weaken the ability to observe the contributions of these forest fire impacts at the receptor sites. Meteorologically-based receptor modeling of these impacts observed in near-real-time can be a powerful tool for source apportionment of forest fires. Combining high resolution air mass back trajectories (12 trajectories each day) with near-real-time pollutant concentrations across multiple receptor sites allows the use of advanced trajectory ensemble models such as sQTBA which produce concentration fields that reveal the potentially contributory regions for the given period of interest. In the examples discussed in this work, it is clear that potential regions of forest fire impacts in Ontario were mainly located in Canada's boreal forests located to the northeast and northwest of the multiple receptor sites, indicating that in addition to forest fires that may occur within the province, fires as far away as the Yukon and the Northwest Territories may also be potential contributors to high PM_{2.5} levels in Ontario.



The techniques described in this work, together with developments in remote sensing, meteorological analysis and pollutant detection instrumentation, could find annual application each wildfire season in analyses of future forest fire events that occur far from the populated regions. Application of the techniques in future events of varying complexity is likely to result in improvements in our understanding of the pathways and transport mechanisms by which smoke plumes affect ground-level pollutant concentrations.

Acknowledgments

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model used in this application. Analyses and visualizations of CO data from the AIRS instrument on the Aqua satellite used in this paper were produced with the Giovanni online data system, developed and maintained by the NASA Goddard Earth Sciences Discovery and Information Services Center (GES DISC). Images of geopotential height patterns (Figures 1b, 2b and 3b) were provided by the NOAA Air Resources Laboratory from their Web site at <http://ready.arl.noaa.gov>.

Supplementary Material Available

Discussion on the simplified quantitative transport bias analysis (sQTBa), time series plots for the periods of greatest $PM_{2.5}$ impacts across the receptor sites of interest in 2011 and 2012, time series plots comparing $PM_{2.5}$ with O_3 and NO_2 at the receptor sites for the 2012 event, the GASP AOD from the GOES satellite for the 2011 event and an annual $PM_{2.5}$ emissions inventory map for both Canada and the US are included in Discussion S1 and Figures S1 to S4 respectively. This information is available free of charge via the internet at <http://www.atmospolres.com>.

References

- Akagi, S.K., Yokelson, R.J., Burling, I.R., Meinardi, S., Simpson, I., Blake, D.R., McMeeking, G.R., Sullivan, A., Lee, T., Kreidenweis, S., Urbanski, S., Reardon, J., Griffith, D.W.T., Johnson, T.J., Weise, D.R., 2013. Measurements of reactive trace gases and variable O_3 formation rates in some South Carolina biomass burning plumes. *Atmospheric Chemistry and Physics* 13, 1141–1165.

- Begum, B.A., Kim, E., Jeong, C.H., Lee, D.W., Hopke, P.K., 2005. Evaluation of the potential source contribution function using the 2002 Quebec forest fire episode. *Atmospheric Environment* 39, 3719–3724.
- Benner, B.A., Wise, S.A., Currie, L.A., Klouda, G.A., Klinedinst, D.B., Zweidinger, R.B., Stevens, R.K., Lewis, C.W., 1995. Distinguishing the contributions of residential wood combustion acid mobile source emissions using relative concentrations of dimethylphenanthrene isomers. *Environmental Science & Technology* 29, 2382–2389.
- Brereton, C.A., Johnson, M.R., 2012. Identifying sources of fugitive emissions in industrial facilities using trajectory statistical methods. *Atmospheric Environment* 51, 46–55.
- Brook, J.R., Johnson, D., Mamedov, A., 2004. Determination of the source areas contributing to regionally high warm season PM_{2.5} in Eastern North America. *Journal of the Air & Waste Management Association* 54, 1162–1169.
- Dempsey, F., 2013. Forest fire effects on air quality in Ontario: Evaluation of several recent examples. *Bulletin of the American Meteorological Society* 94, 1059–1064.
- Draxler, R.R., Rolph, G.D., 2010. http://ready.arl.noaa.gov/HYSPLIT_traj.php, accessed in 2012.
- Gibson, M.D., Pierce, J.R., Waugh, D., Kuchta, J.S., Chisholm, L., Duck, T.J., Hopper, J.T., Beauchamp, S., King, G.H., Franklin, J.E., Leaitch, W.R., Wheeler, A.J., Li, Z., Gagnon, G.A., Palmer, P.I., 2013. Identifying the sources driving observed PM_{2.5} temporal variability over Halifax, Nova Scotia, during BORTAS-B. *Atmospheric Chemistry and Physics* 13, 7199–7213.
- Goody, R., 1995. *Principles of Atmospheric Physics and Chemistry*, Oxford University Press, New York, NY, 324 pages.
- Harrison, R.M., Yin, J.X., 2000. Particulate matter in the atmosphere: Which particle properties are important for its effects on health? *Science of the Total Environment* 249, 85–101.
- Hecobian, A., Liu, Z., Hennigan, C.J., Huey, L.G., Jimenez, J.L., Cubison, M.J., Vay, S., Diskin, G.S., Sachse, G.W., Wisthaler, A., Mikoviny, T., Weinheimer, A.J., Liao, J., Knapp, D.J., Wennberg, P.O., Kurten, A., Crounse, J.D., St. Clair, J., Wang, Y., Weber, R.J., 2011. Comparison of chemical characteristics of 495 biomass burning plumes intercepted by the NASA DC-8 aircraft during the ARCTAS/CARB-2008 field campaign. *Atmospheric Chemistry and Physics* 11, 13325–13337.
- Hopke, P.K., Zhou, L.M., Poirot, R.L., 2005. Reconciling trajectory ensemble receptor model results with emissions. *Environmental Science & Technology* 39, 7980–7983.
- Jaffe, D.A., Wigder, N.L., 2012. Ozone production from wildfires: A critical review. *Atmospheric Environment* 51, 1–10.
- Jordan, T.B., Seen, A.J., Jacobsen, G.E., 2006. Levoglucosan as an atmospheric tracer for woodsmoke. *Atmospheric Environment* 40, 5316–5321.
- Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Leetmaa, A., Reynolds, R., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K.C., Ropelewski, C., Wang, J., Jenne, R., Joseph, D., 1996. The NCEP/NCAR 40-year reanalysis project. *Bulletin of the American Meteorological Society* 77, 437–471.
- Keeler, G.J., Samson, P.J., 1989. Spatial representativeness of trace-element ratios. *Environmental Science & Technology* 23, 1358–1364.
- Lackmann, G., 2011. *Midlatitude Synoptic Meteorology: Dynamics, Analysis, and Forecasting*, American Meteorological Society, Boston, MA, 345 pages.
- Meyer, M.B., Patashnick, H., Ambs, J.L., Rupprecht, E., 2000. Development of a sample equilibration system for the TEOM continuous PM monitor. *Journal of the Air & Waste Management Association* 50, 1345–1349.
- NASA (NASA Giovanni Data and Information Services Center), 2013. <http://disc.sci.gsfc.nasa.gov/giovanni>, accessed in 2013.
- Natural Resources Canada, 2013. http://cwfis.cfs.nrcan.gc.ca/en_CA/index, accessed in 2013.
- Neuberger, M., Schimek, M.G., Horak, F., Moshhammer, H., Kundi, M., Frischer, T., Gomiscek, B., Puxbaum, H., Hauck, H., Auphep-Team, 2004. Acute effects of particulate matter on respiratory diseases, symptoms and functions: Epidemiological results of the Austrian Project on Health Effects of Particulate Matter (AUPHEP). *Atmospheric Environment* 38, 3971–3981.
- NFD (National Forestry Database), 2013. http://nfdp.ccfm.org/fires/national_e.php, accessed in 2014.
- NOAA (National Oceanic and Atmospheric Administration), 2013a. <http://www.ssd.noaa.gov/PS/FIRE/GASP/gasp.html>, accessed in 2013.
- NOAA (National Oceanic and Atmospheric Administration), 2013b. <http://satepsanone.nesdis.noaa.gov/FIRE/fire.html>, accessed in 2014.
- NOAA ARL (NOAA Air Research Laboratory), 2008. http://www.arl.noaa.gov/faq_hg11.php, accessed in 2013.
- NOAA ARL READY, 2013. <http://www.ready.noaa.gov/READYamet.php>, accessed in 2013.
- OMOE (Ontario Ministry of the Environment), 2013. Air Quality in Ontario, Report for 2011, Report No. PIBS 9196, Queen's Printer for Ontario, 96 pages.
- OMOE (Ontario Ministry of the Environment), 2010. <http://www.airqualityontario.com/>, accessed in 2014.
- Ondov, J.M., Buckley, T.J., Hopke, P.K., Ogulei, D., Parlange, M.B., Rogge, W.F., Squibb, K.S., Johnston, M.V., Wexler, A.S., 2006. Baltimore supersite: Highly time- and size-resolved concentrations of urban PM_{2.5} and its constituents for resolution of sources and immune responses. *Atmospheric Environment* 40, S224–S237.
- Patashnick, H., Rupprecht, E.G., 1991. Continuous PM-10 measurements using the tapered element oscillating microbalance. *Journal of the Air & Waste Management Association* 41, 1079–1083.
- Pope, C.A., Burnett, R.T., Thurston, G.D., Thun, M.J., Calle, E.E., Krewski, D., Godleski, J.J., 2004. Cardiovascular mortality and long-term exposure to particulate air pollution – epidemiological evidence of general pathophysiological pathways of disease. *Circulation* 109, 71–77.
- Ramdahl, T., 1983. Retene – a molecular marker of wood combustion in ambient air. *Nature* 306, 580–583.
- Rastogi, A.K., 2013. <http://code.google.com/p/metcor/>, accessed in 2013.
- Seidel, D.J., Ao, C.O., Li, K., 2010. Estimating climatological planetary boundary layer heights from radiosonde observations: Comparison of methods and uncertainty analysis. *Journal of Geophysical Research–Atmospheres* 115, art. no. D16113.
- Seinfeld, J.H., Pandis, S.N., 1998. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, John Wiley and Sons Inc, New York, 1326 pages.
- Simoneit, B.R.T., 2002. Biomass burning – A review of organic tracers for smoke from incomplete combustion. *Applied Geochemistry* 17, 129–162.
- Sofowote, U.M., Rastogi, A.K., Debosz, J., Hopke, P.K., 2014. Advanced receptor modeling of near-real-time, ambient PM_{2.5} and its associated components collected at an urban-industrial site in Toronto, Ontario. *Atmospheric Pollution Research* 5, 13–23.
- Sofowote, U.M., Hung, H., Rastogi, A.K., Westgate, J.N., Deluca, P.F., Su, Y.S., McCarry, B.E., 2011. Assessing the long-range transport of PAH to a sub-Arctic site using positive matrix factorization and potential source contribution function. *Atmospheric Environment* 45, 967–976.
- Statistics Canada, 2013. <http://www12statcan.gc.ca/census-recensement/2011/as-sa/fogs-spg/select-Geo-Choix.cfm?Lang=Eng&GK=CMA&PR=10#PR35>, 2014.
- Stull, R., 2000. *Meteorology for Scientists and Engineers*, Brooks Cole, Pacific Grove, CA, 502 pages.
- Transportation Safety Board of Canada, 2013. <http://www.tsb.gc.ca/eng/enquetes-investigations/rail/2013/R13D0054/R13D0054.asp>, accessed in 2013.
- U.S. EPA (U.S. Environmental Protection Agency), 2011. Air Monitoring Methods – Criteria Pollutants, <http://www.epa.gov/ttn/amtic/criteria.html>, accessed in 2012.

- Val Martin, M., Logan, J.A., Kahn, R.A., Leung, F.Y., Nelson, D.L., Diner, D.J., 2010. Smoke injection heights from fires in North America: Analysis of 5 years of satellite observations. *Atmospheric Chemistry and Physics* 10, 1491–1510.
- Venier, M., Ma, Y.N., Hites, R.A., 2012. Bromobenzene flame retardants in the Great Lakes atmosphere. *Environmental Science & Technology* 46, 8653–8660.
- Wang, Y.G., Hopke, P.K., Rattigan, O.V., Chalupa, D.C., Utell, M.J., 2012. Multiple-year black carbon measurements and source apportionment using Delta-C in Rochester, New York. *Journal of the Air & Waste Management Association* 62, 880–887.
- Wang, Y.G., Hopke, P.K., Rattigan, O.V., Xia, X.Y., Chalupa, D.C., Utell, M.J., 2011. Characterization of residential wood combustion particles using the two-wavelength aethalometer. *Environmental Science & Technology* 45, 7387–7393.
- Wang, Y.G., Huang, J.Y., Znananski, T.J., Hopke, P.K., Holsen, T.M., 2010. Impacts of the Canadian forest fires on atmospheric mercury and carbonaceous particles in Northern New York. *Environmental Science & Technology* 44, 8435–8440.
- Wotawa, G., Trainer, M., 2000. The influence of Canadian forest fires on pollutant concentrations in the United States. *Science* 288, 324–328.
- Yunker, M.B., Macdonald, R.W., Vingarzan, R., Mitchell, R.H., Goyette, D., Sylvestre, S., 2002. PAHs in the Fraser River basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry* 33, 489–515.
- Zhao, W.X., Hopke, P.K., Zhou, L.M., 2007. Spatial distribution of source locations for particulate nitrate and sulfate in the upper-midwestern United States. *Atmospheric Environment* 41, 1831–1847.
- Zhou, L.M., Hopke, P.K., Liu, W., 2004. Comparison of two trajectory based models for locating particle sources for two rural New York sites. *Atmospheric Environment* 38, 1955–1963.