Impacts of the July 2012 Siberian fire plume on air quality in the Pacific Northwest

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Abstract. Biomass burning emissions emit a significant amount of trace gases and aerosols and can affect atmospheric chemistry and radiative forcing for hundreds or thousands of kilometres downwind. They can also contribute to exceedances of air quality standards and have negative impacts on human health. We present a case study of an intense wildfire plume from Siberia that affected the air quality across the Pacific Northwest on 6–10 July 2012. Using satellite measurements (MODIS True Colour RGB imagery and MODIS AOD), we track the wildfire smoke plume from its origin in Siberia to the Pacific Northwest where subsidence ahead of a subtropical Pacific High made the plume settle over the region. The normalized enhancement ratios of O₃ and PM₁ relative to CO of 0.26 and 0.08 are consistent with a plume aged 6–10 days. The aerosol mass in the plume was mainly submicron in diameter (PM₁/PM₂.₅ = 0.96) and the part of the plume sampled at the Whistler High Elevation Monitoring Site (2182 m a.s.l.) was 88 % organic material. Stable atmospheric conditions along the coast limited the initial entrainment of the plume and caused local anthropogenic emissions to build up. A synthesis of air quality from the regional surface monitoring networks describes changes in ambient O₃ and PM₂.₅ during the event and contrasts them to baseline air quality estimates from the AURAMS chemical transport model without wildfire emissions. Overall, the smoke plume contributed significantly to the exceedances in O₃ and PM₂.₅ air quality standards and objectives that occurred at several communities in the region during the event. Peak enhancements in 8 h O₃ of 34–44 ppbv and 24 h PM₂.₅ of 10–32 µg m⁻³ were attributed to the effects of the smoke plume across the Interior of British Columbia and at the Whistler Peak High Elevation Site. Lesser enhancements of 10–12 ppbv for 8 h O₃ and of 4–9 µg m⁻³ for 24 h PM₂.₅ occurred across coastal British Columbia and Washington State. The findings suggest that the large air quality impacts seen during this event were a combination of the efficient transport of the plume across the Pacific, favourable entrainment conditions across the BC interior, and the large scale of the Siberian wildfire emissions. A warming climate increases the risk of increased wildfire activity and events of this scale reoccurring under appropriate meteorological conditions.

1 Introduction

Wildfires emit significant amounts of primary pollutants including fine particulate matter (PM₂.₅), carbon monoxide (CO), nitrogen oxides (NOₓ), and non-methane organic carbons (NMOCs). These pollutants can react to form ozone (O₃) and secondary organic aerosols (SOA), which

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can affect downwind air quality and climate over a wide range of scales from local (Akagi et al., 2011, 2012, 2013; Smolyakov et al., 2014), to regional (Wigder et al., 2013), to intercontinental (Jaffe et al., 2001, 2004; Bertschi et al., 2004; Bertschi and Jaffe, 2005; Lapina et al., 2006; Pfister et al., 2006; Weiss-Penzias et al., 2007; Bein et al., 2008; Strada et al., 2012; Jaffe and Wigder, 2012). Short-term exposures to air pollution from biomass burning have been found to be associated with a range of health impacts, including respiratory symptoms, increased hospital admissions, and emergency room visits and premature mortality (Henderson and Johnston, 2012).

The rate of O$_3$ and SOA formation in wildfire plumes, and hence the potential for adverse air quality and health impacts, can vary considerably due to several factors, including biomass fuel type, combustion characteristics, atmospheric conditions, and downwind distance (Akagi et al., 2011; Jaffe and Wigder, 2012). The O$_3$ production in biomass burning is driven by photochemical reactions of NO$_x$ and non-methane organic carbons (NMOCs) and typically occurs in a NO$_x$ limited environment. O$_3$ enhancement relative to CO, also known as the normalized enhancement ratio (NER; $\Delta$O$_3$/CO), for temperate and boreal wildfires generally increases as the plume ages and is typically in the range of 0.1 to 0.7 (Jaffe and Wigder, 2012). A similar trend is present for the NER of PM$_1$ ($\Delta$PM$_1$/CO) for plumes aged < 2 days; yet for older plumes, SOA formation can be overshadowed by PM loss due to wet and dry deposition, cloud processing, and/or evaporation (Wigder et al., 2013). Further study is still required to better understand the interactions of the above factors on downwind air quality for long-range transport (LRT) events.

The Russian boreal forests represent the largest forested region on Earth with an aerial coverage of approximately 8 million km$^2$ (Stocks, 2004). About 1% of this region is damaged from 10 000 to 35 000 forest fires annually (Isaev et al., 2002). Jaffe et al. (2004) estimated that the active Siberian wildfire season in 2003 caused summertime O$_3$ enhancements of 9–17 ppbv, which led to episodic O$_3$ exceedances in western North America. Other studies have also demonstrated the LRT of wildfire smoke from Siberia can affect air quality over Asia (Jeong et al., 2008; Jung et al., 2016) and contribute to arctic haze (Brock et al., 2011).

This study examines an episodic LRT event of Siberian wildfire smoke to the Pacific Northwest that had significant and widespread air quality impacts on 6–10 July 2012. Record temperatures and dry weather across Siberia in 2012 led to increased wildfire activity. Over 17 000 wildfires were detected in July and August 2012 alone. The Fire Inventory from NCAR model (FINN; Wiedinmyer et al., 2011) estimated that 48 Tg of CO emissions occurred by August 2012, which was already more than double the biomass burning emission for the entire 2010 season and second only to the 72 Tg from the 2003 season (NASA, 2012). Cottle et al. (2014) examined the impact of this event in the Lower Fraser Valley (LFV) in British Columbia, Canada, using lidar measurements taken at the University of British Columbia. HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) trajectory analysis confirmed that aerosol layers subsiding over the LFV had largely originated from over wildfire source areas in Siberia at least 6 days earlier. Aerosol backscatter measurement and low depolarization volume ratios during the event showed the progressive entrainment of smoke into the LFV through 10 July 2012, which coincided with the high PM$_{2.5}$ observed by the region’s fixed air quality monitoring network. This study expands on the Cottle et al. (2014) work in a number of significant ways: it analyzes potential air quality impacts over a much greater geographical area encompassing large parts of British Columbia and Washington State (WA), uses detailed air quality measurements at a high elevation background site to provide insight into plume chemistry, and makes use of photochemical modeling to establish baseline air quality conditions in the absence of any wildfire emissions to determine the smoke plume’s contribution to degraded air quality and exceedances of regional air quality objectives and national standards of O$_3$ and PM$_{2.5}$.

2 Methods

2.1 Monitoring data

2.1.1 Ambient air quality monitoring data

Ambient O$_3$ and PM$_{2.5}$ data were obtained from several air quality monitoring networks (Fig. 1c and d), including 35 stations from the British Columbia Ministry of Environment (BCMoE) network (BCMoE, 2014a), 22 air quality monitors within the LFV Air Quality Monitoring Network (Metro Vancouver, 2014), and 15 stations from the Washington State Monitoring Network (WSMN) (WA ECY, 2012). The air quality monitoring network across British Columbia is, in part, co-managed by the National Air Pollution Surveillance Network (NAPS) (Dabeck-Zlotorzynska et al., 2011). Study sites also included two marine background sites – Amphitrite Point, Ucluelet (site C, 18 m a.s.l., on Vancouver Island; McKendry et al., 2014), and Cheeka Peak (site D, 480 m a.s.l., in WA; Weiss-Penzias et al., 2004) – and two elevated background sites – Whistler Peak (site 1, 2182 m a.s.l., located 120 km north of Vancouver; Takahama et al., 2011; Macdonald et al., 2011) and Mt. Rainier Jackson Visitor Centre (site F, 1782 m a.s.l., located in Pierce County, WA). A description of the data analyzed and sampling methodologies for the various networks and sampling sites is summarized in Table 1.
2.1.2 Lidar sites

Two lidars were used in this study: one located on the University of British Columbia Point Grey campus in Vancouver (site B, 80 m a.s.l.) and a second located at the north end of Whistler village (site A, 650 m a.s.l.), approximately 8 km north of the Whistler Peak High Elevation Site (WHI) (Gallagher et al., 2012; Strawbridge, 2013). The lidars use a Continuum Inlite III laser with an approximate output power of 1.5 W at 10 Hz and operates simultaneously at wavelengths of 532 and 1064 nm. The system measures the return signals through three detection channels: one at 1064 nm and two at 532 nm (one for each polarization). Backscatter information provided by the upward pointing setup has a vertical resolution of 3 m from near the ground up to 15 km and uses a 10 s averaging period. Additional details on the lidar systems can be found in Strawbridge (2013).

2.1.3 Whistler Peak High Elevation Site

Air quality measurements were collected at the Whistler Peak High Elevation Site (see Table 1 for details). CO was measured with Thermo Environmental Instruments Inc. Model 48C Trace Level analyzer and O$_3$ was measured with a Thermo Environmental Instruments Inc. UV absorption monitor (TECO 49C). Particle chemistry was provided both through integrated filter samples and continuously with an Aerosol Chemical Speciation Monitor (ACSM). Inorganic ions (Cl$^-$, NO$_3^-$, Na$^+$, NH$_4^+$, K$^+$, Mg$^{2+}$, and Ca$^{2+}$) were analyzed on 48 h filter samples by ion chromatography (IC) and ICP-MS. OC, POC, and EC measurements were also obtained on a Sunset Lab OC/EC aerosol analyzer (Sunset Laboratory Inc., 2016) using the EnCan Total-900 thermal method (Huang et al., 2006) with an 8-day sampling period (3–11 July) split between day and night. The ACSM provided particle chemical composition for SO$_2^{−}$, NO$_3^-$, Cl$^-$, NH$_4^+$, and organics on a 30 min time resolution. Optical measurements of black carbon (rBC) were acquired using the Single Particle Soot Photometer (SP2). During the study, the SP2 instrument had a steady 6% loss in its laser power; however, internal diagnostics suggest that the power loss is unlikely to significantly affect the readings because heating aerosol during intake was unaffected. SP2 readings were also affected by a leak in the inlet plumbing noticed after the event. Particle size distributions were measured by an optical particle counter (OPC) over the (0.25 to 32 μm) for the entire period and also from a scanning mobility particle sizer (SMPS) (14–572 nm diameter) from 9 July onwards. Details of the trace gas and particle measurements at Whistler Peak are discussed by Macdonald et al. (2011) and Takahama et al. (2011).

2.2 Trajectory and dispersion modeling

A detailed analysis by Cottle et al. (2014) using HYSPLIT trajectories and high-resolution smoke dispersion models generated with the Bluesky framework (Sullivan et al., 2008) concluded that wildfire sources within North America did not significantly influence air quality within the LFV during the 6–10 July 2012 period.

A similar analysis was done using the CMC trajectory model (D’Amours et al., 2015) with meteorological fields from the Global Environmental Multiscale Limited-Area Model (GEM-LAM) 15 km model at 15 min intervals for nearby North America wildfires (Long Draw, Oregon, and Waldo Canyon, Colorado). Forward trajectories were integrated for a 72 h period at release heights of 10, 100, and 1000 m above ground level (a.g.l.). Figure S1 in the Supplement shows the forward trajectories for the Long Draw and Waldo Canyon wildfires released at 00:00 UTC on 9 July 2012 and confirmed that smoke from these sources were unlikely to have contributed to the degraded ambient air quality observed over the larger geographical extent in this study. Trajectory modelling was not done for the few small wildfires within the Interior of British Columbia. Overall, the effects of these sources are expected to be localized and with only minor air quality enhancements until after 9 July 2012.

2.3 Analysis of O$_3$ and PM$_{2.5}$ impacts

Based on measurements collected at over 70 air quality monitoring stations in the Pacific Northwest, the air quality impacts of the Siberian wildfire plume were assessed by examining the O$_3$ and PM$_{2.5}$ concentrations at various averaging periods of 1, 8, and 24 h, depending on the pollutant. For each monitoring station, severe air quality episodes were identified using the regional objective and national standards, applicable at the time of the event (Table 2), as thresholds and were compared to the average historical July daily maxima values from 2000 to 2010. It should be noted that although some of these standards, for example the Canada Wide Standard (CWS) for O$_3$ (CCME, 2014), are based on multi-year statistics, all standards were compared to hourly observations of their respective averaging periods.

To further characterize the timing and spatial extent of these impacts, baseline concentrations of O$_3$ and PM$_{2.5}$ over the Pacific Northwest, in the absence of any wildfire emissions from either Siberia or from within North America, were simulated using the AURAMS (A Unified Regional Air-quality Modelling System) air quality transport model (Gong et al., 2006, with updates from Kelly et al., 2012) and were compared with observations.

2.3.1 AURAMS baseline simulation

AURAMS was run over a 12-day period (5–15 July 2012) using a nested configuration of 12 and 4 km grid spacing with the inner (4 km) domain covering southern British Columbia and northern WA (Fig. 1a and c). Meteorology for the simulations was provided by Environment and Climate Change Canada’s (ECCC) GEM-LAM weather forecast model (Côté
Table 1. Instrument list for various monitoring networks used in the study.

<table>
<thead>
<tr>
<th>Network/site</th>
<th>Measured parameters</th>
<th>Instrument(s)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>British Columbia Minstry of Environment (BCMoe) network and Lower Fraser Valley Air Quality Monitoring Network</td>
<td>O$_3$</td>
<td>Thermo Environmental Instruments Inc. UV absorption monitor (TECO 49C)</td>
<td>Environment Canada (2013); Metro Vancouver (2014)</td>
</tr>
<tr>
<td>Amphitrite Point, Ucluelet (site C)</td>
<td>O$<em>3$, PM$</em>{2.5}$</td>
<td>Thermo 49i O$_3$ analyzer, Thermo Sharp 5030 particulate monitor</td>
<td>McKendry et al. (2014)</td>
</tr>
<tr>
<td>UBC lidar (site B) Whistler village lidar (site A)</td>
<td>Aerosol backscatter</td>
<td>Lidar</td>
<td>Strawbridge (2013)</td>
</tr>
<tr>
<td>Whistler Peak High Elevation Site (site 1)</td>
<td>CO, O$_3$, Refractory black carbon, Particle chemistry &lt; 1 micron (SO$_4^{2-}$, NO$_3^-$, Cl$^-$, NH$_4^+$, Organics), Particle size distributions from 0.25 to 32µm diameter, Particle size distributions in range ~ 14 to 572 nm diameter, Integrated inorganic particle chemistry (in this study: SO$_4^{2-}$), Integrated organic chemistry (OC and EC)</td>
<td>Thermo Environmental Instruments Inc. Model 48C Trace Level analyzer, Thermo Environmental Instruments Inc. UV absorption monitor (TECO 49C), Droplet Measurement Technologies Inc. Single Particle Soot Photometer (SP2), GRIMM portable aerosol spectrometer 1.109, TSI 3081 scanning mobility particle sizer (SMPS) with a TSI 3775 condensation particle counter (CPC) operated at a low flow setting, Teflon filters; 48 h samples</td>
<td>Macdonald et al. (2011), Macdonald et al. (2011), Takahama et al. (2011), Takahama et al. (2011), McKendry et al. (2008)</td>
</tr>
<tr>
<td>Integrated organic chemistry (OC and EC)</td>
<td>Sunset Lab OC-EC aerosol analyzer (EnCan Total-900 Thermal method); 8-day sample</td>
<td>Sunset Laboratory Inc. (2016); McKendry et al. (2008)</td>
<td></td>
</tr>
<tr>
<td>Washington State Monitoring Network (WSMN)</td>
<td>O$<em>3$, PM$</em>{2.5}$</td>
<td>Teledyne-API T400 Nephelometer</td>
<td>WA ECY (2008, 2012)</td>
</tr>
<tr>
<td>Cheeka Peak (site D)</td>
<td>O$<em>3$, PM$</em>{2.5}$</td>
<td>Teledyne-API T400 Nephelometer, Radiance Research M903</td>
<td>WA ECY (2008, 2012)</td>
</tr>
<tr>
<td>Mt. Rainier Jackson Visitor Center (site F)</td>
<td>O$_3$</td>
<td>Teledyne-API T400 Nephelometer</td>
<td>WA ECY (2008, 2012)</td>
</tr>
</tbody>
</table>
Figure 1. Map of the monitoring network data and AURAMS modelling domain used in the study. The pink and blue regions in (a) highlights the two geographical regions of interest, British Columbia and Washington state, respectively. The black outlined area in (a) highlights the geographical location of the MODIS True Colour image on 29 June 2012 from the NASA earth Observatory site (b). The blue and red outlined areas in either (a) and/or (c) highlight the 12 and 4 km AURAMS grid domains, respectively. The green outline in (c) highlights the Northern Interior sites that fall outside the gridded domain. The Lower Fraser Valley (LFV; black outline in e) inset (d) displays the Metro Vancouver air quality monitoring network and the location of the UBC lidar. Abbreviations used in this figure are defined in Table S1 and throughout the text. Topography shaded in panels (c) and (d).

Table 2. 2012 Ambient air quality standards and objective for $O_3$ and PM$_{2.5}$.

<table>
<thead>
<tr>
<th>Standard/objective</th>
<th>Pollutant</th>
<th>Averaging period</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Canada NAAQS</td>
<td>$O_3$</td>
<td>1 h</td>
<td>82 ppbv</td>
<td>Health Canada (1999)</td>
</tr>
<tr>
<td>Canada CWS</td>
<td>$O_3$</td>
<td>8 h</td>
<td>65 ppbv</td>
<td>CCME (2014)</td>
</tr>
<tr>
<td>BC provincial and Metro Vancouver</td>
<td></td>
<td></td>
<td>65 ppbv</td>
<td>BCMoE (2014b)</td>
</tr>
<tr>
<td>Vancouver</td>
<td></td>
<td></td>
<td></td>
<td>Metro Vancouver (2014)</td>
</tr>
<tr>
<td>Canada CWS</td>
<td>PM$_{2.5}$</td>
<td>24 h</td>
<td>30 µg m$^{-3}$</td>
<td>CCME (2014)</td>
</tr>
<tr>
<td>BC provincial and Metro</td>
<td></td>
<td></td>
<td>25 µg m$^{-3}$</td>
<td>BCMoE (2014b)</td>
</tr>
<tr>
<td>Vancouver</td>
<td></td>
<td></td>
<td></td>
<td>Metro Vancouver (2014)</td>
</tr>
<tr>
<td>US EPA NAAQS</td>
<td></td>
<td></td>
<td>35 µg m$^{-3}$</td>
<td>US EPA (2014)</td>
</tr>
</tbody>
</table>
et al., 1998) run at a 2.5 km resolution and then interpolated to the AURAMS domains. GEM-LAM was run in a series of 30 h simulations starting from 00Z with the first 6 h of each simulation discarded as “spin up” in order to allow model fields to reach steady state. The model was run with anthropogenic emissions based on the 2010 Canadian and 2008 US emission databases. Emission totals for both databases were adjusted to 2012 levels using Metro Vancouver forecasted and backcasted LFV emission estimates (GVRD, 2007). The model used biogenic emissions that calculated using the Biogenic Emissions Inventory System (BEIS) version 3.0.9 emissions algorithms and Biogenic Emissions Landuse Database 3 (BELD3) data. AURAMS employed the ADOM-II gas-phase chemical mechanism (Stockwell et al., 1989) and used a chemically specified 12-bin sectional distribution to characterize particulate matter from 0.01 to 40.96 µm in diameter.

Lateral boundary conditions to the AURAMS model along its outer (12 km) domain were supplied by seasonally representative climatological values for some species and for O3 an additional adjustment was made to the climatological boundary conditions in response to the local tropopause height (Makar et al., 2010). Lateral boundary conditions to the inner 4 km domain were supplied from the 12 km model output. AURAMS runs did not include any wildfire emissions from either Siberia or from within North America.

The reliability of the AURAMS baseline simulation (Table S2) was examined by comparing the 1 h observed and modelled O3 and PM2.5 over non-event days (5 July and from 12 to 15 July). Within the 4 km domain, the model bias in 1 h O3 and PM2.5 is relatively low (2.35 ppbv and 1.14 µg m$^{-3}$, respectively). Over the 12 km domain, the O3 and PM2.5 performance degraded, with O3 bias and RMSE of 14.3 ppbv and 20.86 ppbv, respectively, and PM2.5 bias and RMSE of 5.84 and 10.21 µg m$^{-3}$, respectively. However, it should be noted that the majority of the study sites fall within the 4 km domain. Overall, the 1 h model performance of the baseline simulation is comparable to the assessed reliability of the AURAMS model from other studies (Makar et al., 2014).

### 2.3.2 LRT enhancements analysis

LRT enhancements were identified when ambient air quality conditions in the Pacific Northwest departed from the baseline conditions as modelled by AURAMS, beyond a reasonable margin of error. As the model bias can affect the magnitude of the LRT enhancement estimates, the uncertainty in these enhancements was quantified in a conservative manner in this study. This was done by examining the range of differences between the 8 and 24 h observed and modelled values for O3 and PM2.5, respectively, during non-event days (5 July and from 12 to 15 July) at each monitoring location.

Table S1 in the Supplement provides uncertainty estimates in 8 h O3 and 24 h PM2.5 for selected sites. For reporting purposes, enhancements for multiple monitoring locations were aggregated, when appropriate, to reflect the average contribution of the Siberian plume across the different regions in the Pacific Northwest.

### 3 Results and discussion

#### 3.1 July 2012 smoke event overview

On 29 June 2012, smoke originating from wildfires over eastern Russia, as noted by the NASA Earth Observatory group, drifted eastward across the Sea of Japan, the Sea of Okhotsk, and Kamchatka (Fig. 1b). Moderate Resolution Imaging Spectroradiometer (MODIS) Aerosol Optical Depth (AOD) imagery between 1 and 6 July, shown in Fig. S2a–f, illustrates the progression of the aerosol plume (AOD $>$ 1) across the Pacific to western North America. Long-range transport of aerosols was aided by strong zonal flow established as a developing storm tracked from the Bearing Sea to the Gulf of Alaska. Subsidence ahead of the subtropical Pacific high is thought to have contributed to the descent of the plume over the study area. In western North America, subsidence and mountain wave activity are found to be important factors in bringing mid-tropospheric dust layers in range of boundary layer entrainment process (McKendry et al., 2001; Hacker et al., 2001).

On 7 July, the MODIS True Colour imagery (Fig. 2) shows the smoke plume oriented northeast to southwest across the southern British Columbia and Washington State. At this time, degraded air quality conditions were observed inland over the Interior of British Columbia, where both the 8 h O3 and 24 h PM2.5 exceedances occurred at Kamloops (site 8). From 8 to 9 July, poor air quality conditions continued to spread across the Interior, coinciding with the northward and inland progression of the plume as collaborated by AOD imagery (Fig. S2g), and caused air quality exceedances at several communities (see Table 3). Over in the coastal region, elevated O3 and PM2.5 also occurred on 8 July, with isolated 8 h O3 exceedances at Chilliwack (site 3) and Enmoreclaw (site 5). MODIS AOD on 10 July (Fig. S2h) showed the plume remnant had shifted southeastward and out of the Pacific Northwest domain.

Figure 3 provides an overview of the maximal enhancements to 8 h O3 and 24 h PM2.5 estimated based on the differences between the ambient air quality observation and the baseline AURAMS air quality modelling for the event. Overall, the baseline modelling suggests that the greatest air quality impacts from the Siberian wildfire smoke were in the Interior of British Columbia and at the Whistler Peak High Elevation Monitoring Site with peak increases of 34–44 ppbv for 8 h O3 and 10–32 µg m$^{-3}$ for 24 h PM2.5. The coastal portions of British Columbia and Washington State saw lesser enhancements of 10–14 ppbv and 4–9 µg m$^{-3}$, respectively (Table 4). The regional disparity in the enhancement may be attributable to difference in subsidence across the region.
Coastal regions had stable atmospheric conditions, leading to stagnant conditions under a strong thermal inversion between 500 and 1000 m a.s.l. (Fig. 4a). Stable atmospheric conditions likely limited the entrainment of the plume over the coastal area and caused a buildup of local anthropogenic emissions. Over the Interior of British Columbia, dry and less stable conditions existed with daytime mixing heights over 3 km a.s.l. (Fig. 4b). The variations in 8 h O$_3$ and 24 h PM$_{2.5}$ enhancements (with uncertainty estimates) across the Pacific Northwest are listed in Table 4 and are discussed in more detail in the following sections.

### 3.2 Plume chemistry at Whistler High Elevation Monitoring Site

The Whistler Peak High Elevation Monitoring Site is situated to monitor air chemistry in the free troposphere, such as background air pollutants, and to study long-range transport events across the Pacific (Leaitch et al., 2009; Macdonald et al., 2011). On 6 July, significant increases in 1 h O$_3$, CO, and PM$_{2.5}$ mark the onset of the smoke event at Whistler Peak (Fig. 5) and coincide with the timing of elevated aerosol layers detected by the Whistler lidar. Aerosol backscatter ratios show that the plume persisted over Whistler from 6 July 14:00 ST (WHI1) to 8 July 06:00 PST (WHI2) at an elevation of 2–3.6 km a.s.l. Backscatter ratios also indicate the entrainment of the aerosols deeper into the Whistler Valley as hourly observed O$_3$, PM$_{2.5}$, and CO reached peaks of 86 ppbv, 24 µg m$^{-3}$, and 276 ppbv, respectively. The maximum 1 h O$_3$ and CO values were approximately 50 and 167 ppbv above the average July background levels as reported in Macdonald et al. (2011) for 2002–2006. O$_3$ conditions prior to the event were consistent with July background levels.

The ratio of O$_3$ to CO enhancements ($\Delta$O$_3$/$\Delta$CO) provides some insight into the relative photochemical production of O$_3$ in wildfire plumes (Jaffe and Wigder, 2012) and is strongly influenced by the travel time between the fire and measurement location (Mauzerall et al., 1998). For this study,
the plume age is estimated to be between 7 and 10 days old based on a Siberian origin of 29 June 2012. The $\Delta$O$_3$/ΔCO ratio was estimated using observations from WHI1 to WHI2 and excluding cases where CO was lower than the July background (110 ppbv). The regression slope of O$_3$ and CO for the event ranged from 0.15 to 0.24 with a mean value of 0.26 ($n$ = 20, $r^2$ = 0.80) and is comparable to other similarly aged Siberian plumes with excess ratios of 0.22–0.36 (6–10 days; Bertschi et al., 2004) and 0.15–0.84 (7–10 days, $n$ = 5; Bertschi and Jaffe, 2005).

As shown in Fig. 5d, during the smoke event (WHI1 to WHI2) the PM$_{2.5}$ mass was dominated by submicron aerosol with an average PM$_1$/PM$_{2.5}$ ratio of 0.96. The particle composition as determined from the ACSM (Fig. 6) during the event was on average approximately 87.6 % organic mass, 3.7 % NO$_3^-$, 5.9 % SO$_4^{2-}$, 2.5 % NH$_4^+$, and 0.3 % Cl$^-$. PM$_1$ mass is estimated from the ACSM, since the size distribution data indicate that most of the mass is below 0.7 µm; in Fig. S3, an example is shown for 9 July 2012, when the SMPS became operational again. Further, a comparison of the mass concentrations estimated from the SMPS, based on the assumptions of spherical particles and a particle mass density of 1.2 g cm$^{-3}$ as a result of the dominant organic composition during that period (Fig. 6), with the ACSM for 9–31 July is illustrated in Fig. S4. The comparison is strong over the 518 1 h averaged data: $r^2$ = 0.95; slope = 1.10. As in the example in Fig. S3, the PM$_{2.5}$ mass is estimated with the addition of the OPC data.

The evolution of PM concentrations in wildfire plumes is largely affected by the rate of SOA formation (Akagi et al., 2012). Using the regression methods described in Wigder et al. (2013), APM$_1$/ΔCO is estimated at 0.08 ($n$ = 20, $r^2$ = 0.94) during the WHI1 to WHI2 period. This ratio is comparable to those of long-range transported plume (> 740 km) of 0.03 and 0.06 found in Weiss-Penzias et al. (2007) and Wigder et al. (2013), respectively. Compared to the median ratios of 0.17, 0.29, 0.29, and 0.19, corresponding to distances of < 140, 140–340, 340–540, and 540–740 km as reported by Wigder et al. (2013), the ratio found in this study is relatively low, suggesting that the net PM$_1$ loss...
Figure 3. Overview of the maximal enhancements to 8 h $O_3$ and 24 h PM$_{2.5}$ estimated for the study area (a and b, respectively) and the LFV (c and d, respectively) based on the differences between the ambient air quality observation and the AURAMS baseline modelling for the 6–10 July 2012 smoke event. Abbreviations used in this figure are defined in Table S1 and throughout the text.

Figure 4. Skew-T atmospheric radiosondes for Quillayute (site E) and Kelowna Airport (site G) at 00Z on 8 July 2012. The atmospheric dry bulb temperature ($T$) and dew point temperature ($Td$) are shown from the surface up to 100 mb. Black dashed line is a trace of a surface parcel lift to estimate the mixed layer depth at the height where it intersects the environmental dry bulb temperature.
Figure 5. Whistler lidar (site A, 650 m a.s.l.) measured backscatter ratio of the 1064 nm channel to that of clear air for the July 2012 Siberian smoke plume (a) and observed (solid) 1 h \( \text{O}_3 \) (b), 1 h \( \text{CO} \) (c), and 1 h PM\(_1\) (based on the ACSM) and PM\(_{2.5}\). (d) at Whistler Peak High Elevation station (site 1, 2182 m a.s.l., indicated as a horizontal white line in panel a). AURAMS baseline (dashed) 1 h \( \text{O}_3 \) and 1 h PM\(_{2.5}\) are shown in panel (b) and (d), respectively. Elevated aerosol backscatter in panel (a) are noted from 6 July 14:00 PST (WHI1) to 8 July 06:00 PST (WHI2) by red dashed lines. The shaded regions indicate nighttime hours between 18:00 and 07:00 PST.

 exceeds SOA formation during transport as the plume ages. A recent wildfire study (Jolleys et al., 2012) found little SOA production in aged biomass burning plumes; however, it was noted that other aircraft campaigns had found a large variability in OA enhancements.

The OC / EC ratio is also considered as its increase may indicate a contribution from SOA formation to the OC as a smoke plume ages. From the 8-day integrated PM\(_1\) quartz filter samples, separated by day and night, the value of OC / EC was 3.8 (day) and 3.4 (night) at Whistler Peak for the period 3–11 July. During the WHI1–WHI2 event, an OC / EC ratio
of 3.7 was also estimated by scaling the average ACSM OM and rBC to the 8-day integrated OC and EC sample, respectively. In the week prior to 1 July 2012, a large variation in OC / EC (approximately 1.2 to 7) was observed in the suburb of Novosibirsk, approximately 400–800 km south to southwest of the main sources of the Siberian wildfire (Smolyakov et al., 2014). Due to this large variation in OC / EC close to the source as attributed to variations in burning phase and fuel types, no further inferences of SOA formation in the plume were made. The ratios of m/z 44 and m/z 43 to OM derived from the ACSM, indicate that the organic aerosol during the height of the biomass burning plume (WHI1 to WHI2) was more oxygenated than at any other time during 5–11 July (Fig. S5). If SOA production during the plume transport was insignificant, then this observation implies either a relatively high level of oxygenation near the source, heterogeneous oxidation of the OM during plume transport,
or the loss of less oxygenated components of the OM during transport.

The overall contribution of the 6–10 July Siberian plume event to the air quality standards for $O_3$ and $PM_{2.5}$ at Whistler Peak (site 1) were estimated using AURAMS baseline modelling (also shown in Fig. 5). Generally, the model depicts relatively clean air quality conditions (approximately 30 ppbv and 1.0 $µg\,m^{-3}$ for $O_3$ and $PM_{2.5}$, respectively) on 5 July with an increase trend in baseline $O_3$ (between 5 and 10 ppbv from 5 to 8 July). It is estimated that the Siberian wildfires contributed an additional 44 ppbv to the daily maximum 8 h $O_3$ and 10 $µg\,m^{-3}$ to the 24 h $PM_{2.5}$ concentration (Table 4).

### 3.3 Lower Fraser Valley

The UBC lidar (site B) data indicated a plume arrival time of 12:00 PST on 6 July (LFV1, Fig. 7a) and a plume elevation of between 2 and 3.6 km a.s.l. over Metro Vancouver, which is similar to that found by the Whistler lidar (site A) but with generally weaker aerosol backscatter ratio readings (Fig. 7a). The plume had a shorter overall persistence over Metro Vancouver with the plume’s trailing edge shifting northward by 03:00 PST on 8 July (LFV2). It appears that the conditions favourable for the transport of Siberian smoke to LFV were also conducive to $O_3$ production (e.g. light winds, clear skies, and high temperatures). This makes the attribution of Siberian smoke impacts on local air quality slightly more challenging in the LFV than at Whistler. Figure 7b and c depict the increasing $O_3$ and $PM_{2.5}$ across the LFV during 6–10 July with the network average 1 h $O_3$ and 1 h $PM_{2.5}$ rises to 54 ppbv and 18 $µg\,m^{-3}$, respectively, on 8 July. Peak $O_3$ condition of 79 ppbv observed at Hope Airport (site 4) and $PM_{2.5}$ of 26 $µg\,m^{-3}$ at Port Moody Rocky Point (site 2) arose as inland temperature reached a maximum of 31°C at Chilliwack (site 3) on 8 July. Inland temperatures and $O_3$ conditions moderated subsequently on 9 July following a brief period of elevated convection in the early morning hours that affected parts of WA and LFV. High $O_3$ concentrations were reported over the high elevation (>1000 m a.s.l.) site in North Vancouver, located close to the coast over the northern portion of the LFV, by a separate field campaign (Bart et al., 2014).

The AURAMS model was used to assess the underlying baseline air quality (Fig. 7b and c) and to assess the local impacts through a site-by-site analysis of differences with LFV monitoring network data (Fig. 8a and b). For 6–8 July, the AURAMS baseline simulation without wildfire emissions captured the rise in daytime $O_3$ on average across the LFV network (Fig. 7b) and on a site-by-site basis (Fig. 8a). In contrast, the baseline simulation was consistently lacking in nighttime $O_3$ titration. Previous air quality studies have shown that these discrepancies do not significantly affect the rest of the baseline AQ simulation (Makar et al, 2014). As such, the estimates of $O_3$ enhancement within the LFV are based on daytime hours only. The 1 h $O_3$ enhancements occurring on 9–10 July appear, based on the lidar data, to be about 18–27 h after the plume advected north of the LFV (Fig. 8a). The average enhancement of peak 8 h maximum $O_3$ across the LFV was estimated to be 10 ppbv (Table 4; hourly variability shown as shaded regions in Fig. 8a). Spatial maps of 8 h $O_3$ (Fig. S6) show widespread enhancements across
the LFV on 9 July lingered in the western portion on 10 July. The lack of \( O_3 \) enhancement estimated throughout the LFV on 8 July (Figs. 8a and S6) suggests the exceedance-level \( O_3 \) episode in Chilliwack (Fig. 8c) may have been due to local \( O_3 \) production and not the Siberian wildfire.

The \( PM_{2.5} \) enhancements associated with the LRT event developed in the morning of 8 July, much earlier than with any \( O_3 \) enhancements, and then persisted through 10 July. The Siberian plume is estimated to have enhanced the 24 h \( PM_{2.5} \) by a maximum of 9 \( \mu g \cdot m^{-3} \) for sites across the LFV. Spatially, the largest enhancements to \( PM_{2.5} \) appear to be concentrated over the northern portion of the LFV (Fig. 3 and by day in Fig. S6). This suggests that entrainment of the plume may have been facilitated by slope flows along the northern mountainous terrain, yet appears not to have reached the floor of the LFV, as evidenced by \( O_3 \) data from the fixed monitoring network.

### 3.4 Washington State

In general, the air quality concentrations in WA during the episode were similar to those of the LFV. The average hourly \( PM_{2.5} \) across WA rose to a maximum of \( \sim 15 \mu g \cdot m^{-3} \) on 9...
Figure 8. Contributions of the Siberian wildfire smoke plume based on the differences of observed 1 h O$_3$ (a) and PM$_{2.5}$ (b) and the AURAMS baseline without wildfire emissions on a site-by-site basis for the LFV. For (a) and (b), the average contribution is labelled by the blue solid line. The light grey and dark grey shading represents the range and interquartile range of the contribution across the network at a particular hour. Panel (c) shows the AURAMS baseline and observed O$_3$ time series for Chilliwack, BC (site 3). The light blue shaded regions indicate nighttime hours between 18:00 and 07:00 PST.

Figure 9. The hourly observed (blue) and baseline AURAMS (red, dashed) without wildfire emissions O$_3$ (top) and PM$_{2.5}$ (bottom) for Kamloops (site 8) (a) and Williams Lake (site 9) (b). The shaded regions indicate nighttime hours between 18:00 and 07:00 PST.
July and the peak 8 h O$_3$ of 72 ppbv (Fig. S7) occurred at Enumclaw on 8 July. The LRT smoke enhancements in WA (Table 4) were similar to those in the LFV for O$_3$ with a 10 ppbv increase to the 8 h O$_3$ and a weaker contribution to the 24 h PM$_{2.5}$ of 4 µg m$^{-3}$ (Fig. 3 and by day in Fig. S6). However, the timing and geographical extent of the impacts of PM$_{2.5}$ and O$_3$ differed considerably. In particular, the impacts on PM$_{2.5}$ were minimal over WA until 10 July, while O$_3$ underwent some modest enhancement on 9 July at a limited number of monitoring locations (Fig. S7). It is likely that some of the discrepancies (between the Canadian and US portions of the LFV in the estimated plume impacts) arise from model errors stemming from different methodologies used to create the 2010 Canadian and 2008 US emissions inventories.

3.5 The Interior of British Columbia

The character of the Siberian plume and its impacts over the Interior of British Columbia differed considerably from the enhancements seen over the LFV and WA. The air quality for two communities (Fig. 9), Kamloops (site 8) and Williams Lake (site 9), reflects the dramatic response seen across the Interior. The arrival of the plume at these communities, 19:00 PST on 6 July for Kamloops (site 8) and 12:00 PST on 8 July for Williams Lake (site 9), resulted in a sudden shift in hourly PM$_{2.5}$ and elevated O$_3$ on the subsequent day. Based on the AURAMS modelling, it is estimated that the plume contributed an additional 18 and 38 ppbv at Kamloops (site 8) and Williams Lake (site 9), respectively, to the daily maximum 8 h O$_3$, and an additional 32 and 30 µg m$^{-3}$, respectively, to the 24 h PM$_{2.5}$ (Table 4). The suddenness and magnitude of these impacts suggests that the plume mixed well into the boundary layer after its transition across the Coast Mountains, which also corroborates the entrainment signatures noted on the Whistler lidar that shows the plume mixed down to the base of the Whistler Valley (Fig. 5).

Unfortunately, the 12 km model study domain did not extend north sufficiently to provide baseline estimates of the air quality conditions to Quesnel (site 10), Prince George (site 11), and the Northern Interior (site 12 to 14), which also experienced conditions that exceeded national and provincial air quality standards. Instead, a baseline air quality level was inferred from the observations on 6 July and the enhancements at these sites (listed in Table 4) were found to be consistent with those experienced at Williams Lake (site 9; based on a 6 July baseline, as well as the 12 km domain) except for slightly weaker 24 h PM$_{2.5}$ enhancements of 22–24 µg m$^{-3}$.

On 10 July, as the bulk of the Siberian plume shifted north-eastward out of British Columbia, remnants of the plume shifted over the Southern Interior and Whistler; however, attribution of the plume’s impacts was impeded due to local wildfire activity (Matthew Creek; approx. 100 km west of Kelowna; 155 acres). Overall, the maximal air quality enhancements due to smoke from the Siberian wildfires in the Southern Interior was approximately 15 ppbv and 15 µg m$^{-3}$ for 8 h O$_3$ and 24 h PM$_{2.5}$, respectively, based on the impact analysis conducted at Vernon (site 7) for the 6–9 July period (Fig. S10).

4 Conclusions

The extreme 2012 Russian boreal fire season resulted in the trans-Pacific transport of a large smoke plume across the Pacific in early July which had substantial, yet varied, impacts across large parts of British Columbia and Washington State. Air quality concentrations in excess of the Canada Wide Standard (65 ppbv for 8 h O$_3$) and the British Columbia Air Quality Objective (25 µg m$^{-3}$) of 24 h PM$_{2.5}$ were experienced on 6–10 July at several municipalities across British Columbia.

The aged smoke plume (6–10 days old), as supported from data from Whistler Peak, had a mean normalized enhancement ratio ($\Delta$O$_3$/\Delta CO) of 0.26 and a PM$_1$/CO ratio of 0.08, consistent with other long-range transport smoke studies of similar origin. Fine organic aerosol mass (based on the ACSM measurements) accounted for the majority (~88%) by mass fraction) of the 1 and 24 h PM$_{2.5}$ enhancements of 23 and 10 µg m$^{-3}$, respectively. Analysis of OC/EC did not provide evidence for significant SOA production during transport as the plume ages. However, analysis of the ratios of m/z 44 and m/z 43 to OM derived from the ACSM, the level of oxygenation of organic material in this plume was much higher than during flanking times. If SOA formation during transport was minimal, then the relatively high level of oxygenation may have resulted from three factors: the OM near the source was relatively oxygenated; heterogeneous oxidation of the OM during plume transport; or removal of less oxygenated components of the OM during transport.

Baseline air quality modelling provided by the AURAMS model was used to delineate the scope of the LRT enhancements to air quality under varying atmospheric conditions. Generally, the largest enhancements to 8 h O$_3$ of 34–44 ppbv and 24 h PM$_{2.5}$ of 10–32 µg m$^{-3}$ occurred at Whistler Peak (site 1), Kamloops (site 8), and across the Central and Northern Interior of British Columbia (site 9 to 14). In contrast, lesser LRT enhancements of approximately 10 ppbv to 8 h O$_3$ and 4–9 µg m$^{-3}$ to 24 h PM$_{2.5}$ were estimated for coastal British Columbia and Washington State. Stable atmospheric conditions along coastal area of British Columbia and Washington State likely limited the initial entrainment of the plume. The AURAMS modelling approach accounted for enhancements in anthropogenic air quality due to the smoke plume in the majority of cases with the exception of 8 July at Chilliwack (site 3) and Enumclaw (site 5), where elevated levels may have been due to other factors.

Long-range transport smoke events, such as the 2012 Siberian forest fire plume, highlight the far reaching impacts of wildfires and how air quality impacts can vary with downwind meteorology, plume age, and entrainment path-
ways. This demonstrates the need to include wildfire emissions within chemical transport model and to derive effective parameterizations for the lofting and subsequent reactions in modelling ambient air quality conditions. Lateral boundary for chemistry used in regional air quality modelling application should be responsive to the long-range transport of pollutants. Wildfires sources pose an increasing concern as global climate change is expected to increase wildfire frequency and severity in many regions (IPCC, 2007; Gillett, 2004; Liu et al., 2013). As such, wildfire smoke is expected to contribute more to the ambient levels for O$_3$, PM$_{2.5}$, CO, and other pollutants and can potentially increase the frequency of extended periods of degraded air quality.

5 Data availability

Data sources used in this paper are listed in the supplemental material (Table S3). For ECCC data not readily accessible, please contact the corresponding author (andrew.teakles@canada.ca) for assistance.

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Author contributions. A. Teakles designed the study, developed the methodology, and prepared the manuscript with contributions from co-authors. R. So consolidated and analyzed the data and provided contributions to the methodology. J. Baik provided assistance in developing R code to generate figures. B. Ainslie generated the AURAMS model run. S. Hanna, A. Bertram, and L. Huang provided data and their expertise on data and instrumentation comparability. K. B. Strawbridge provided the lidar data and his expertise in interpreting these data. A. M. Macdonald and R. Leaitch provided and analyzed data collected at Whistler Peak High Elevation Site. D. Toom assisted with the ACSM data and supplementation analysis on OA oxygenation. I. McKendry provided his expertise on the LFV analysis. R. Vingarzan reviewed and provided contributions to the methodology and manuscript composition. C. Schiller reviewed the manuscript and provided her chemistry expertise to the data interpretation. R. Nissen performed the synoptic analysis. D. A. Jaffe contributed to the enhancement ratios analysis and the US analysis.

Competing interests. The authors declare that they have no conflict of interest.

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