The impacts of transported wildfire smoke aerosols on surface air quality in New York State: A case study in summer 2018

Wei-Ting Hung[1, *], Bhupal Shrestha[1], Cheng-Hsuan (Sarah) Lu[1], Hsiao-Chun Lin[1], Chin-An Lin[1], Dustin Grogan[1], Jia Hong[2], Ravan Ahmadov[3,4], Eric James[3,4] and Everette Joseph[5]

[1]Atmospheric Sciences Research Center, University at Albany, State University of New York, Albany, NY, USA

[2]State Key Laboratory of Information Engineering in Surveying, Mapping and Remote Sensing, Wuhan University, Wuhan, China

[3]CIRES, University of Colorado, Boulder, CO, USA

[4]NOAA Earth System Research Laboratory, Boulder, CO, USA

[5]National Center for Atmospheric Research, Boulder, CO, USA

[*]Corresponding author: whung@albany.edu

Abstract

Wildfire smoke aerosols, once emitted, can transport over long distances and affect surface air quality in downwind regions. In New York State (NYS), fine particulate matter ($PM_{2.5}$) concentration continues to decrease due to anthropogenic emission reductions and regulatory initiatives in recent years. Smoke aerosols, however, are projected to increase, making them the dominant source of $PM_{2.5}$. Thus, the influences of smoke aerosols could become more important in the future. In this study, the long-range transport of smoke aerosols, and their impacts on local air quality over NYS in mid-August 2018 were investigated using satellite measurements, ground-

based networks, and model products. Satellite measurements showed extensive fire activities over the northwestern United States (US) during August 8^{th} – 10^{th}. Air quality monitoring sites in NYS reported a threefold increase in average $PM_{2.5}$ concentration (from 8.4 ± 3.4 μg m^{-3} to 24.8 ± 4.0 μg m^{-3}) on August 15^{th} – 16^{th}, while the ground-based profiler network detected aerosol layers at 2 – 5 km across the state. Analysis of backward trajectories revealed that the plumes originated from wildfires, transported through southern Canada and arrived at the east coast during a period of 5 – 7 days. The increased $PM_{2.5}$ in NYS can be attributed to boundary layer entrainment and vertical mixing of the aloft transported smoke aerosols down to the surface. The NYS mesoscale weather network (NYSM), which is originally designed for severe weather monitoring, ~~demostrate~~demonstrate the capability to probe the lower atmosphere and provide the vertical extent information of air pollutants.

1. Introduction

Wildfires have significant impacts on surface air quality by emitting fine particulate matters ($PM_{2.5}$, with radius less than 2.5 μm), so-called smoke aerosols, and trace gases into the atmosphere (Kaulfus et al., 2017; Larsen et al., 2017; Miller et al., 2011; Val Martin et al., 2013; Wotawa and Trainer, 2000). In the United States (US), over 25% of annual $PM_{2.5}$ emissions are attributed to fire activities (Kaulfus et al., 2017). These smoke aerosols are composed of 50 – 60% of organic carbon (OC) and 5 – 10% of black carbon (BC) (Reid et al., 2005). During smoke episodes, increases of trace gases, including carbon monoxide (CO) and nitrogen oxides (NO_x) are observed (DeBell et al., 2004; Dreessen et al., 2016; Mathur et al., 2008; Sapkota et al., 2005; Yurganov et al., 2004, 2011). Increases in ground-level ozone (O_3) concentrations are also reported since these trace gases are precursors for O_3 photochemical production (McKeen et al., 2002; Spichtinger et al., 2001).

Due to the intense energy generated by fires, strong convection enables smoke aerosols and trace gases to be injected above the boundary layer (BL) into the free troposphere. Once the smoke aerosols are entrained into the free troposphere, large-scale circulations can transport them across long distances (Colarco et al., 2004; Duck et al., 2007; Heilman et al., 2014; Miller et al., 2011; Peterson et al., 2014; Val Martin et al., 2010). For instance, smoke aerosols and CO emitted by fire activities in the western US could be transported eastward to the northeastern US by mid-latitude westerlies (Miller et al., 2011). In contrast, the emissions from Alaskan fires could be transported westward toward Russia due to the easterly circulation (Sitnov and Mokhov, 2017).

As the plumes are transported and dispersed in the atmosphere, smoke-laden air masses may be transported downward toward the surface. Smoke aerosols from the free troposphere could be transported to lower altitudes under the influence of synoptic scale subsidence, which could

potentially intersect the BL and lead to an entrainment process (Duck et al., 2007). Once smoke aerosols are entrained into the BL, thermal turbulent mixing further mixes aerosols from the BL top downward to the surface, resulting in increased $PM_{2.5}$ concentrations and poor air quality over regions far from the fire sources (Colarco et al., 2004; Duck et al., 2007; Miller et al., 2011; Veselovskii et al., 2015). The entrainment and mixing process of aerosols have been studied extensively using observations and models (Bravo-Aranda et al., 2015; Clarke and Kapustin, 2002; Clarke et al., 1996, 2001; Nilsson et al., 2001; Pahlow et al., 2005; Wood et al., 2000). For instance, Pahlow et al. (2005) reported an overlap of a smoke layer and growing BL from lidar backscattering profiles during a long-range transport (LRT) smoke event in Baltimore. They also found that the surface $PM_{2.5}$ concentrations peaked about 2 h after the intersection of smoke layer and the BL.

During the transport, the physical properties and chemical composition of the smoke aerosols vary significantly with plume age. During the aging process, the size and mass of smoke aerosols increase. Aged smoke plumes are also enriched by secondary species from anthropogenic emissions, such as sulfate, ammonium and nitrite (Reid et al., 1998, 2005). In addition, previous studies reported significant O_3 and/or $PM_{2.5}$ enhancements within wildfire plumes that mix with urban pollutants during transport (Bein et al., 2008; Forster et al., 2001; Junquera et al., 2005; McKeen et al., 2002; Miller et al., 2011). Therefore, it is challenging to characterize and quantify the impacts of smoke aerosols on air quality over downwind regions that are already burdened by large anthropogenic emissions (Bein et al., 2008; McKeen et al., 2002; Singh et al., 2012; Wotawa and Trainer, 2000).

In New York State (NYS), the air quality has significantly improved, with decreasing $PM_{2.5}$ mass concentrations in both urban and rural regions during the past two decades (Emami et al.,

2018; Rattigan et al., 2015; Squizzato et al., 2018). For instance, Rattigan et al. (2015) analyzed the 2000 – 2014 $PM_{2.5}$ observations at 16 in-situ sites across the state and reported a downward trend, with decreases of 4 – 7 μg m^{-3}. The annual average $PM_{2.5}$ concentrations at Albany, plus the 95% confidence intervals, were 12.4 ± 1.4 μg m^{-3} and 7.6 ± 0.8 μg m^{-3} for 2000 and 2014, respectively. The contribution of anthropogenic species (SO_4, NO_3 and HN_4) also decreased by one third across the state, but the downward trend was less significant for OC, which is the primary component of smoke aerosols.

Episodic transport of smoke aerosols has been reported across NYS (Dutkiewicz et al., 2011; Qureshi et al., 2006; Wang et al., 2010; Wu et al., 2018; Zu et al., 2016). An episode of Quebec fires in May 2010 resulted in an 18-fold increase in $PM_{2.5}$ over the northern NYS (Wang et al., 2010). Similarly, a smoke event originated from Alberta, Canada, in May 2016 brought $PM_{2.5}$ close to, and O_3 in excess of, the air quality standard over New York City (NYC) (Wu et al., 2018). On the other hand, historical records indicate that the frequency, intensity and burned areas of fire activities in Northern America show increasing trends under a changing climate (Kasischke and Turetsky, 2006; Westerling and Bryant, 2007; Westerling et al., 2006), and wildfire emissions are projected to become the dominant source of surface $PM_{2.5}$ in the future (Ford et al., 2018; Gillett et al., 2004; Liu et al., 2016; Val Martin et al., 2015; Wotton et al., 2010). Consequently, the contributions of smoke aerosols in NYS could potentially offset the reduction in total $PM_{2.5}$. Thus, understanding the impacts of wildfire smoke on air quality in NYS becomes increasingly important, given regulatory and compliance implications.

This study investigated the transport of wildfire smoke aerosols and their impacts on local air quality in NYS during mid-August of 2018. Multiple datasets, including satellite measurements, ground-based observations and model products, were used to locate the source regions, understand

105 the horizontal and vertical distribution of the plumes during their transport, and access the impacts
106 on local air quality in NYS. Although the particle aging process during the transport is important,
107 it is beyond the scope of this study.

108

109 **2. Datasets**

110 The datasets used in this study are summarized in Table 1, and are briefly described in this
111 section. Detailed descriptions can be found in the cited references.

112

113 **2.1 Observations from surface networks**

114 *AirNow $PM_{2.5}$*

115 The US Environmental Protection Agency (EPA) AirNow (https://airnow.gov/) collects
116 real-time air quality measurements from over 2,000 surface monitoring sites nationwide
117 maintained by state or local air quality agencies. In NYS, air quality data are collected and quality
118 controlled by the NYS Department of Environmental Conservation (DEC). In this study, $PM_{2.5}$
119 mass concentrations from 17 monitoring sites across NYS, including 10 urban sites near NYC and
120 7 suburban sites (fig. 1; table 2), were used. The $PM_{2.5}$ measurements at three sites (marked with
121 asterisks in table 2) are based on the Federal Equivalence Method (FEM), while others are based
122 on the Tapered Element Oscillating Microbalances (TEOM) technology.

123 *NYS Mesonet*

124 In 2017, University at Albany, State University of New York (UAlbany), in partnership
125 with the Federal Emergency Management Agency (FEMA), the NYS Division of Homeland

Security and Emergency Services, and the National Oceanic and Atmospheric Administration (NOAA) National Weather Service (NWS), has completed the deployment of an advanced, statewide mesoscale network (mesonet). NYS Mesonet (NYSM) consists of 126 standard weather sites, 17 enhanced flux sites, 20 snow sites and 17 profiler sites. Detailed descriptions of NYSM can be found at http://nysmesonet.org/. In this study, hourly precipitation from standard sites, and 10-min averaged atmospheric vertical profiles from profiler sites (fig. 1; table 2) were used. Details about lidar configuration and calibration are described in the Appendix.

2.2 Satellite measurements

VIIRS Aerosol Optical Depth

The Visible Infrared Imaging Radiometer Suite (VIIRS) sensors onboard the Suomi National Polar-orbiting Partnership (S-NPP) satellite and NOAA-20 satellite were launched in October 2011 and November 2017, respectively. The wide swath (3000 km) allows VIIRS to provide full global coverage every day. VIIRS measures 22 spectrum channels in the range of 412 – 12,050 nm, including imagery bands, moderate resolution bands (M-bands) and the day-night band. The M-bands are mainly used for aerosol retrieval with 750 m spatial resolution (Cao et al., 2013). In this study, S-NPP level 3 daily gridded (6 x 6 km) aerosol optical depth (AOD) at 550 nm products were used.

OMPS Aerosol Index

The Ozone Mapping Profiler Suite (OMPS; Flynn et al., 2014), aboard S-NPP and NOAA-20 satellites, consists of Nadir Mapper (OMPS-NM), Nadir Profiler (OMPS-NP) and Limb Profiler

147 (OMPS-LP). OMPS-NM is a total-column ozone sensor, measuring solar backscattering in near-
148 UV bands (300 – 380 nm) with daily global coverage.

149 OMPS aerosol index (AI) product is defined as:

150 $$AI = -100\left[\left(\frac{I_{340}}{I_{378}}\right)_{meas} - \left(\frac{I_{340}}{I_{378}}\right)_{calc}\right]$$

151 where I_{meas} is the measured backscattering, I_{calc} is the backscattering for a Rayleigh atmosphere,
152 and the labeled numbers indicate the spectrum used for calculation. Positive values represent
153 absorbing aerosols, such as mineral dust over deserts and carbonaceous aerosols produced by fire
154 activities. Negative values indicate scattering aerosols, such as sulfate, nitrate and other
155 anthropogenic aerosols. Level 3 daily gridded (0.5° x 0.5°) products were used in this study.

156 *AIRS CO*

157 The Atmospheric Infrared Sounder (AIRS) suite consists of AIRS instrument, the
158 Advanced Microwave Sounding Unit (AMSU) and the Humidity Sounder for Brazil (HSB). It was
159 launched aboard Aqua satellite in 2002. AIRS is an infrared spectroradiometer, measuring 370 –
160 1,540 nm spectrum with 2,378 channels. AMSU and HSB are microwave radiometers with 15 and
161 4 channels, respectively (Aumann et al., 2003). AIRS uses radiances at 450 – 458 nm spectrum for
162 CO retrieval with an estimated accuracy about 15% (McMillan et al., 2005, 2008). Level 3 version
163 6 daily gridded (1° x 1°) daytime CO mixing ratios were used in this study. Since AMSU has been
164 offline since September 2016 due to power issues
165 (https://airs.jpl.nasa.gov/data/amsu_a2_anomaly), data used in this study are from AIRS
166 measurement only.

167

2.3 Model products

HRRR

The High-Resolution Rapid Refresh (HRRR; Alexander et al. 2010) is an atmospheric model developed by the NOAA/Earth System Research Laboratory (ESRL)/Global Systems Division (GSD). It has 3 km horizontal resolution and 51 vertical levels in hybrid coordinate. HRRR provides hourly analysis over the contiguous US (CONUS) and Alaska. The current operational HRRR (HRRRv3) was implemented at the NOAA/National Centers for Environmental Prediction (NCEP) since July 2018. Details about HRRR can be found at https://rapidrefresh.noaa.gov/hrrr/ and http://www.nco.ncep.noaa.gov/pmb/products/hrrr/. In this study, vertical velocity in pressure coordinates (e.g. omega) and horizontal wind at 700 hPa were analyzed.

RAP-Smoke

The experimental Rapid Refresh (RAP; Benjamin et al., 2016) model is running in real-time to provide hourly weather forecasting ~~and smoke forecasting~~ at NOAA/ESRL~~/GSD~~ on experimental mode and NOAA/NCEP operationally. The RAP model domain covers the entire North and Central America, Hawaii, and a part of South America. The RAP model assimilates a wide range of in-situ and remote sensing meteorological data in real-time. Since summer 2018 the smoke forecasting capability has been added to the experimental RAP model at NOAA/ESRL [https://rapidrefresh.noaa.gov/RAPsmoke/]. ~~The model domain covers the entire North and Central America, Hawaii, and a part of South America. The RAP model assimilates a wide range of in-situ and remote sensing meteorological data in real-time. T~~The satellite fire radiative power data are ~~also~~ ingested into the model to estimate the wildfire emissions and smoke transport from

fires. Due to the large spatial coverage, the RAP-Smoke model can be used to study the long-range smoke transport from wildfires in the western US and Canada to the eastern US (Ahmadov et al., 2020). In this study, the simulated vertically integrated smoke concentrations ~~were~~ are used to analyze the smoke transport from the wildfires.

MERRA-2

The Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2; Gelaro et al., 2017) is a global atmospheric reanalysis developed by the National Aeronautics and Space Administration's (NASA's) Global Modeling and Assimilation Office (GMAO). It has 0.5° x 0.625° horizontal resolution and 72 vertical levels in global coverage. MERRA-2 uses Goddard Earth Observing System, Version 5 (GEOS-5) Atmospheric General Circulation Model (AGCM) coupled with Goddard Chemistry Aerosol Radiation and Transport (GOCART; Colarco et al., 2010) model. GOCART model simulates five externally mixed aerosol species, including dust, sea salt, BC, OC, and sulfate. Aerosol and meteorological observations are jointly assimilated in MERRA-2. Details of MERRA-2 aerosol reanalysis can be found in Buchard et al. (2017) and Randles et al. (2017). In this study, smoke aerosol fields (column mass densities and three-dimensional mass concentration) and geopotential heights at 700 hPa were used.

3. Methodology

3.1 Site selection

Fig. 1 shows the locations of AirNow sites, NYSM standard sites and NYSM profiler sites used in this study. To characterize the transport pathway of smoke plumes across the state, these sites were grouped into three regions: western NYS, central NYS and near-NYC regions. NYSM

212 profiler sites at Buffalo (BUFF), Albany (ALBA) and Queens (QUEE) were selected to represent
213 the aerosol distribution over western NYS, central NYS and near-NYC regions, respectively.
214 Adjacent NYSM standard sites and AirNow sites were selected to represent the ambient
215 meteorological conditions and air quality over the three regions. Locations of selected AirNow
216 and NYSM sites are summarized in table 2 and 3, respectively.
217

218 **3.2 Trajectory analysis**

219 Seven-day ensemble backward trajectories were calculated using the HYbrid Single
220 Particle Lagrangian Integrated Trajectory version 4 (HYSPLIT4; Stein et al. 2015) model which
221 is developed by NOAA/Air Resources Laboratory (ARL). The meteorological data was from
222 NCEP Global Data Assimilation System (GDAS) with 0.5° horizontal resolution. The releasing
223 altitude and time were determined by lidar observations at selected sites. Each ensemble run
224 included 27 members. Trajectories were computed online and downloaded from the Real-time
225 Environmental Applications and Display System (READY) website (http://www.ready.noaa.gov)
226 developed by ARL.
227

228 **3.3 Entrainment rate**

229 Entrainment is defined as the mixing of air masses from the free troposphere into the BL.
230 Because the growth of the BL stirs the exchange process, the change of the BL height is generally
231 used to describe the entrainment (Boers et al., 1984; Moeng, 2000; Nilsson et al., 2001; Stevens et
232 al., 2003a). Following Stevens et al. (2003b), the entrainment rate (W_E) was calculated as:

$$W_E = \frac{dh}{dt} + W_h = \frac{dh}{dt} + \int_0^h Div dz$$

$$Div = \frac{du}{dx} + \frac{dv}{dy}$$

where h is the BL height, W_h is the large-scale vertical velocity, and Div is the horizontal divergence at the BL top. In this study, h was given by HRRR analysis and Div was computed from lidar observed horizontal wind in the x and y directions (u and v). Table 3 lists the NYSM profiler sites that were used for Div calculation. For instance, Div at BUFF was calculated from the lidar wind profiles observed at BUFF and WEBS.

4. Results and discussions

4.1 Spatial and temporal variations of aerosols over NYS

Fig. 2 illustrates the timeseries of surface $PM_{2.5}$ concentrations at AirNow sites over western NYS, central NYS and near-NYC regions during August 11th – 20th, 2018. On August 15th – 16th, increases of $PM_{2.5}$ concentrations were observed across the state. The average $PM_{2.5}$ concentrations during the period were 20.4 ± 4.8 μg m^{-3}, 17.2 ± 6.5 μg m^{-3} and 17.8 ± 7.8 μg m^{-3} over western NYS, central NYS and near-NYC regions, respectively. These values were approximately double of the averages during the 2012 – 2013 summertime at multiple sites across NYS (6 – 11 μg m^{-3}) that were reported in Rattigan et al. (2015). The peak hourly $PM_{2.5}$ concentrations of these three regions were 35.6 μg m^{-3}, 35.6 μg m^{-3} and 43.1 μg m^{-3}, which were comparable to or exceeded the current 24 h National Ambient Air Quality Standard (35 μg m^{-3}).

Fig. 3 shows the lidar backscattering profiles at BUFF, ALBA and QUEE during August 14th – 17th. The surface $PM_{2.5}$ and precipitation from AirNow and NYSM standard sites are also

shown. For all sites, backscattering profiles (fig. 3a-c) showed a mostly clear sky during August 15th – 16th, except in early mornings, while clouds were observed at BUFF on the days before and after the event period. Aloft aerosol plumes were observed at 2 – 5 km at BUFF at the nights of 14th and 15th. Aloft plumes were also found at 2 – 3 km at ALBA and 2 – 4 km at QUEE on 15th. The presence of the plumes at BUFF occurred around 12 h earlier than those at ALBA and QUEE. In addition, aerosol layers from surface to around 2 km were observed throughout the event period for all sites.

Fig. 3d shows that $PM_{2.5}$ concentrations at BUFF, ALBA and QUEE increased from 5 – 10 μg m^{-3} to around 26 – 30 μg m^{-3} during August 14th – 16th and the average $PM_{2.5}$ concentration among the three sites increased by threefold (from 8.4 ± 3.4 μg m^{-3} to 24.8 ± 4.0 μg m^{-3}). The precipitation observed on the 17th was associated with the passage of a cold front and the scavenging effect could be the reason for the rapid decreases of $PM_{2.5}$ on that day. Consistent with the aloft plumes observed by the backscattering profiles, $PM_{2.5}$ increases at ALBA and QUEE occurred concurrently while the $PM_{2.5}$ increment at BUFF occurred about 12 h earlier. This is expected as the western NYS first detected the transported polluted plume from the west.

4.2 Source and transport of smoke aerosols

To identify the source regions, HYSPLIT backward trajectories were analyzed. Fig. 4 shows the ensemble 7-day backward trajectories originating at BUFF, ALBA and QUEE. In general, similar transport paths were found for BUFF and ALBA. Under the influence of the synoptic scale circulation, air masses originated from the western states, traveled through northern US and southern Canada, and eventually arrived BUFF and ALBA during a period of 5 – 7 days.

On the other hand, the majority of backward trajectories originating at QUEE were from the Midwest and southern Canada. The heights of the ensemble trajectories were found throughout the troposphere, i.e. from the near surface to over 10 km a.s.l., but the majority of the trajectories traveled below 5 km a.s.l. Therefore, air masses may have picked up anthropogenic aerosols during the transport.

According to VIIRS fire counts (fig. 5), there were extensive fire activities over the Pacific Northwest and British Columbia on August 8^{th} – 10^{th}, which was 5 – 7 days before the $PM_{2.5}$ enhancement observed in NYS. VIIRS true color images also revealed that aerosol plumes (grey areas indicated by yellow arrows) over the northwestern US transported northward to merge with the smoke plumes in British Columbia, and then moved eastward through southern Canada. Moreover, persistent fires in the Midwest were observed during the time period. These fire activities provided fresh emissions that were likely entrained into the smoke-laden air masses as they traveled from the west toward the east coast. Therefore, without considering the aging process, which affects the physical and chemical properties of smoke aerosols, the transport and dispersion of air masses could potentially modify plume characteristics by mixing with the fresh emissions and anthropogenic aerosols. However, it is challenging to determine the extent of fresh/aged smoke aerosols and anthropogenic pollutants without direct measurements of chemical composition.

Fig. 6 illustrates RAP-Smoke, VIIRS AOD, OMPS AI, and AIRS CO on August 15^{th} and 16^{th}. In fig. 6a, RAP-Smoke predicted smoke plumes over the northwestern states, the Midwest and southern Canada. The OMPS AI (fig. 6c) and AIRS CO (fig. 6d) also showed significant smoke plumes that originated from the northwestern US and British Columbia, extended toward southern Canada and eventually reached the east coast. Similarly, VIIRS AOD (fig 6b) indicated

298 the presence of aerosol plumes over the northwestern US, Midwest and northeastern US, despite
299 missing data due to cloud coverage.

300

301 **4.3 Three-dimensional distribution of smoke plumes**

302 In this section, the horizontal distribution and vertical structures of smoke aerosols are
303 investigated. Figs. 7 – 10 illustrate the horizontal and vertical distributions of smoke plumes from
304 MERRA-2 reanalysis on August 10th, 13th, 15th and 16th. Each figure shows the smoke column
305 mass densities in the domain of 25N – 60N and 60W – 135W, and meridional cross-sections of
306 smoke mass concentrations at the source region (122W), the transport over the Midwest (100W)
307 and the transport over the northeastern US (77W).

308 On August 10th (fig. 7), smoke aerosols were transported from the northwestern US (e.g.
309 northern California, Oregon and Washington states) toward central Canada due to a clockwise
310 circulation of a continental high pressure system. The geopotential height also shows a ridge
311 locating between 115W – 105W. The cross-section at 122W shows smoke aerosols across 35N –
312 50N, with an increasing vertical extent to the north. A smoke layer formed and expanded from
313 surface to 600 hPa, which was potentially due to enhanced convection generated by the fire
314 activities. Similarly, the cross-section at 100W shows significant vertical extent of smoke aerosols,
315 revealing that the majority of smoke aerosols were transported through southern Canada. Further
316 downstream at 77W, the cross-section shows low smoke concentrations of ~10 μg m^{-3} near the
317 surface and maximum values of 30 μg m^{-3} at 700 hPa near 42N.

318 On August 13th (fig. 8), the majority of smoke plumes were transported on the periphery
319 of the high pressure system toward southeastern Canada as the ridge broadened and propagated

eastward. This can also be seen from the smoke distribution at 100W. For instance, the suppressed vertical extent and reduced smoke loading at higher latitudes (> 47.5N) represented that the eastward plume had left the Midwest. Meanwhile, part of the plume, which was closer to the center of the high pressure system, dispersed and remained in the Midwest (black circle). In addition, as a trough formed between 115W – 105W, the smoke cross-section at 100W shows that the smoke plume at lower latitudes (40N – 47.5N) moved ~2 degrees southward compared to August 10th. On the other hand, over the Northeast (77W), smoke aerosols transported at the level between 850 – 650 hPa and their loadings were low near the surface.

On August 15th (fig. 9), the majority of the plume continued to move toward the Atlantic Ocean. The geopotential heights at 700 hPa show a trough locating between 80W – 75W, which pushed the dispersed plume (black circle) southward toward NYS. The cross-section at 77W showed that smoke aerosols transported southward, consistent with the spatial distribution. Smoke aerosols also showed a downward movement from the level between 850 – 600 hPa to low levels (altitude < 850 hPa).

On August 16th (fig.10), the dispersed smoke plume continued travelling eastward toward NYS along with the trough. In addition, since the trough moved southward from north of 50N to between 45N – 50N, the cross-section at 77W shows that the smoke plume also moved ~7 degree southward and formed a smoke layer from the surface to 600 hPa. These near-surface smoke aerosols were in position to potentially affect the local air quality over NYS.

4.4 Linkage between the long-range transported smoke plumes and surface air quality

In this section, the synoptic flow and BL evolution over NYS were investigated to understand the linkage between aloft smoke plumes and surface air quality. The synoptic pattern is one possible link for mixing aloft plumes downward through broad descent. In most portions of NYS, the vertical velocities from the HRRR analysis (fig. 11) showed mid-level (700 hPa) ascent (blues) on August 15th (fig. 11a) and descent (reds) on August 16th (fig. 11b). The ascent was associated with a passing trough, while the descent was associated with the broad upstream ridge that followed. As for the smoke aerosols, the majority of the plumes from the Midwest was contained in the ridge (fig. 10a), indicating that the synoptic pattern over NYS may have contributed to the aerosol concentrations shown at low-levels in the previous section (i.e., fig. 10d).

BL entrainment is another possible mechanism to transport smoke aerosols to affect surface air quality. Fig. 12 shows the lidar measurements (i.e. CNR and horizontal winds), the BL height, entrainment rate and $PM_{2.5}$ concentrations at QUEE during August 15th – 16th. Two-day averaged divergence profiles during this period were calculated for the entrainment rate estimation described in section 3.3. On August 15th, the BL started to develop after sunrise at 0700 LT (fig. 12b). Positive entrainment rates indicated the downward flow of aloft air masses, along with smoke aerosols, from the free troposphere into the BL. Meanwhile, an increasing trend in $PM_{2.5}$ concentration was observed concurrently. By midafternoon, near 1400 LT, entrainment rates reduced to near 0 after the BL reached its maximum height and this cut off the entrainment of smoke aerosols from the free troposphere. However, with strong vertical mixing (fig. 12a), smoke aerosols that reached the BL mixed with local air pollutants, which collectively lead to a slight increase of $PM_{2.5}$. After sunset (2000 LT), the BL height dropped rapidly from ~2000 m to around 50 m. Because of the stable conditions and weak vertical motions (fig. 12a), smoke aerosols remained in the residual layer below 2 km at 2100 – 0600 LT (fig. 12b). As a result, the $PM_{2.5}$

concentration showed a slight increase during this time period, which is expected as the aerosol layer was close to the surface.

A similar BL evolution was observed on August 16th, with increased BL height and positive entrainment rate in the morning, and vertical mixing in the afternoon. However, compared to the 15th, the entrainment showed a weaker influence on $PM_{2.5}$ concentrations in the early morning. Since the polluted air mass was located at higher altitudes (2 – 3 km), the overlap of the BL and smoke plume occurred near noon, when the BL was well-developed. In addition, the vertical motions were weaker at 0800 – 1200 LT compared to 15th. These two factors may have suppressed aerosol entrainment, which could be a possible reason for the consistent $PM_{2.5}$ level of 15 – 20 μg m^{-3} during this time period. On the other hand, the weak vertical mixing in late afternoon (1800 LT) and the reduced BL height after sunset (2000 LT) trapped smoke aerosols in the BL, which resulted in the enhancement of $PM_{2.5}$ concentrations.

5. Summary

In this study, ground-based observations, satellite measurements and model products were used to characterize the transport pathway and impacts on the local air quality over NYS during a transported smoke event in mid-August 2018. During the episode, the average surface $PM_{2.5}$ concentration at Buffalo, Albany and Queens increased by 3 times (from 8.4 ± 3.4 μg m^{-3} to 24.8 ± 4.0 μg m^{-3}). To understand the transport pattern of smoke plumes, three sub-regions in NYS were defined: western NYS, central NYS and near-NYC region. The average $PM_{2.5}$ concentrations over western NYS, central NYS and near-NYC regions during the event period were 20.4 ± 4.8

385 μg m^{-3}, 17.2 ± 6.5 μg m^{-3} and 17.8 ± 7.8 μg m^{-3}, respectively. The increases of $PM_{2.5}$ occurred in
386 the western NYS 12 h before the increases occurred in central NYS and near-NYC regions.

387 The lidar observations for the three regions showed aloft aerosol plumes at 2 – 5 km altitude
388 on August 15th – 16th. The plumes in the western NYS arrived about 12 h earlier than those in
389 central NYS and near-NYC regions, which was consistent with the increases in $PM_{2.5}$. The
390 ensemble 7-day backward trajectories for the western NYS and central NYS revealed that the
391 plumes originated from western Northern America, transported through the continent, and arrived
392 the northeastern US. However, trajectories for the near-NYC regions were primarily from the
393 Midwest and southern Canada. In addition, satellite fire counts showed massive fire activities
394 occurred over the northwestern US and British Columbia about 5 – 7 days before the event
395 observed in NYS. RAP-Smoke forecast and satellite measurements, including VIIRS AOD, OMPS
396 AI and AIRS CO, also showed significant smoke plumes over the northwestern US and British
397 Columbia, that later appeared across the continent and the east coast.

398 MERRA-2 reanalysis was analyzed to characterize the transport and three-dimensional
399 distribution of the smoke plumes. On August 10th, the majority of smoke aerosols were observed
400 over western Northern America. These smoke aerosols were then transported from the
401 northwestern US toward central Canada driven by a continental high pressure system. On August
402 13th, the smoke plumes expanded across the continent from the west coast to southeastern Canada,
403 as the ridge continued propagating eastward. Meanwhile, a trough formed over the Midwest and
404 pushed a dispersed smoke plume southward. These smoke aerosols were mainly transported at 850
405 – 650 hPa and the concentrations were relatively low near the surface in the northeastern US. On
406 August 15th, the majority of smoke plumes, along with the ridge, further moved toward the Atlantic
407 Ocean, while the dispersed plumes driven by the trough were transported southward and reached

NYS. Vertical distributions showed a downward movement from the upper troposphere (850 – 600 hPa) to low levels (altitude < 850 hPa). On August 16th, the smoke plumes were transported southeastward by the trough. The meridional cross-section at NYS showed a smoke layer from surface to 600 hPa, indicating the possible influence of aloft smoke on surface air quality. During the complicated transport pattern to NYS, air masses were potentially affected by fresh fire emissions, aging processes and anthropogenic pollutants.

The vertical velocities from HRRR analysis were used to understand the synoptic pattern over NYS during the smoke episode. The descent, which was associated with a upstream ridge, may have contributed to the aerosol concentrations in the low-level atmosphere. Additionally, NYSM lidar measurements were used to investigate the BL evolution and entrainment process, which were the possible linkages between aloft smoke aerosols and surface air quality. After sunrise on August 15th and 16th, positive entrainment rates during the BL development indicated the downward flow of the transported smoke aerosols from the free troposphere into the BL. Strong turbulent mixing also favored the dispersion of smoke aerosols in the BL, along with a $PM_{2.5}$ enhancement around 10 $\mu g\ m^{-3}$. In the afternoon, smoke aerosols and local air pollutants mixed in the BL due to the strong mixing which collectively led to further enhanced $PM_{2.5}$ concentrations. Afterward, the stable condition at nighttime kept most of the smoke aerosols in the residual layer, resulting in an enhanced $PM_{2.5}$ concentration overnight.

In NYS, annual $PM_{2.5}$ levels showed a decreasing trend under emission controls and regulatory initiatives. However, since the frequency, intensity and burning areas of fire activities over Northern America have increased in recent years, the influences of transported smoke aerosols could become more significant. This study demonstrated the capabilities of satellites and new ground-based network in NYS toward better characterizing transported smoke events.

Recently, new generation satellites, which use state-of-the-art techniques with improved resolution and coverage, were launched and became operational. In addition, the deployment of a mesoscale network over NYS was completed in 2017. Although the network is designed to monitor severe weather and improve regional weather forecasts, the network of Doppler lidars is capable of monitoring aerosol vertical distribution. These multi-platform observations, in conjunction with backward trajectory analysis and aerosol reanalysis, provide valuable information on source regions, plume transports, horizontal and vertical distributions, associated synoptic features, and their air quality impacts. In addition, BL entrainment and vertical mixing played important roles in local smoke vertical transport and $PM_{2.5}$ variations. The investigations of the BL structure are critical to understand the impact of transported polluted plumes on surface air quality.

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Appendix

The Leosphere Windcube WLS-100 series Doppler Lidars are currently deployed in NYSM profiler sites. They are pulsed lidars using coherent detection that emits laser pulses at 1.54 μm wavelength with pulse repetition frequency from 10 KHz to 40 KHz depending upon the configuration. The basic Doppler lidar working principle is based on the assumption that aerosols move according to wind speed and those moving aerosols induce an optical frequency change of the backscattered pulse due to the Doppler Effect. The Doppler shift in frequency is proportional to radial wind speed and hence, the signal processing of returned signal provides radial wind speed and Carrier-to-Noise Ratio (CNR). CNR is the integral of signal spectra normalized by noise spectra and hence, the analysis of CNR provides boundary layer height information and aerosol parameters such as backscattering and extinction coefficients by the inversion of the lidar equation:

$$CNR(z) = K * F(z) * \beta(z) * exp(-2 \int_0^z \sigma(r)dr)$$

where z is altitude in meter, K is instrumental constant, $F(z)$ is instrumental function, $\beta(z)$ is backscattering coefficient and $\sigma(r)$ is extinction coefficient at range r. CNR values obtained from the Doppler Lidar is the result of effects of aerosols and $F(z)$ along the line of sight. $F(z)$ is related to the propagation of laser beam and takes maximum value at the focal length of the beam which results in relatively higher CNR values at that range. To remove such instrumental effect, the Doppler Lidar is calibrated with the scan at low elevation angle (e.g. 1° or 2°) in full circle. With the assumption of the homogenous atmospheric condition, i.e. constant backscattering and extinction coefficient at all ranges, CNR profile takes a Lorentzian shape and is fitted with Lorentzian function to obtain $F(z)$ and then retrieve relative attenuated backscatter coefficient, which is defined as the calibrated CNR:

$$\beta_{rel.att.}(z) = \frac{CNR(z)}{F(z)} = K * \beta(z) * exp(-2\int_0^z \sigma(r)dr)$$

Currently, all the Doppler lidars at NYSM profiler sites are running in Doppler Beam Swinging (DBS) mode with the physical resolution of 50m, measurement range from 100m to 7000m, and PRF of 20KHz. In the DBS mode, four line of sight (LOS) measurements are made at four cardinal direction and one LOS measurement at vertical direction. Combination of five LOS measurements allow us to retrieve 3D wind vector using an assumption of homogeneous wind at given altitude at five LOS measurements.

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