

1 **Intra-continental wildfire smoke transport and impact on local air quality** 2 **observed by ground-based and satellite remote sensing in New York City**

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7 **Abstract** The wildfires in Fort McMurray of Alberta, Canada, injected large amounts of smoke aerosols
8 in May 2016 and were identified as being one of Canada's major weather events of the year. This paper
9 presents a synergistic remote sensing and in-situ measurement of the resultant smoke plume transport,
10 optical properties, and its impacts on local air quality in New York City (NYC). Comparisons with the
11 operational air quality model forecast (the NOAA National Air Quality Forecasting Capability, NAQFC)
12 performance are presented. The aloft plume intrusions on May 9-13 and 25-29, 2016, and their mixing
13 down into the planetary-boundary layer (PBL) were observed by a combined lidar, ceilometer and other
14 measurement. A decrease in single-scattering albedo and absorption Angstrom exponent near one indicates
15 that the plumes were absorbing aerosol dominated. Dramatic impacts of smoke transport on the ground air
16 quality are demonstrated with a coincident increase of ground PM_{2.5} (from 5- to 25~30 µg/m³) in NYC
17 urban and upwind rural area, enhancement of the PM_{2.5} speciation (organic carbon, elemental carbon,
18 potassium ion (K⁺)) and the ozone exceedance of NAAQS. Using the satellite and model product, we show
19 regional spatial distribution of smoke, multiple transport paths and wildfire sources. Finally, with the lidar
20 vertical profiling observations, we evaluate the model PBL-height (PBLH) and PM_{2.5} during May 24th to
21 30th, 2016. The model PBLH shows consistent diurnal variation with the observed mixing layer height
22 (MLH), but is clearly overestimated during the convective daytime hours. On the other hand, when
23 estimating the MLH directly from the model PM_{2.5} profile, better agreement with observation was indicated.
24 This helps explain the good agreement between the model PM_{2.5} and surface measurements except for the
25 model overestimate during the morning of May 25 and 26, 2016.

26 **Key words:** Wildfire smoke, transport, air quality, lidar, model

27 **1. Introduction**

28 Wild fires emit large amounts of aerosols or particulates including black carbon (BC) and organic carbon
29 (OC), as well as trace gases such as carbon monoxide (CO), carbon dioxide (CO₂), nitrogen oxides (NO_x),
30 volatile organic compounds (VOCs), etc. (Liu et al., 2014). Critical for health concerns, smoke particulates
31 can be extremely small and result in increased health risk when mixed down into the surface layer. National
32 ambient air quality standards (NAAQS) for fine particulates limit exposure to PM_{2.5} (particle diameters
33 <2.5 μm) to 24-hr average 35 μg/m³ set by US Environmental Protection Agency (EPA) (Dawson et al.,
34 2014). Wildfire emissions and associated ozone (O₃) precursors may result in exceedance of O₃ and/or
35 PM_{2.5} that is referred to the exceptional event in regulatory decisions regarding the NAAQS (Jaffe et al.,
36 2013; Dreessen et al., 2016; Sapkota et al., 2005; Sofowote et al., 2015).

37 The frequency and intensity of wildfire events is expected to increase with climate change (Dennison
38 et al., 2014; Schoennagel et al., 2017). Due to the need to forecast such events and issue warnings to
39 vulnerable populations in urban areas, accurate Chemical Transport Models (CTM) that can quantify the
40 effect of these events and follow the transport and chemical transformations to the surface level are needed.
41 Unfortunately, the limitations of these models as well as difficulties in identifying and quantifying emission
42 from fires make predictions very challenging. Adding to the difficulty is the nature of the urban environment
43 where a complex urban surface is hard to parameterize in forecast models. For these reasons, model
44 performance over urban areas have significant issues. For example, Hogrefe et al (2007) show that total
45 PM_{2.5} mass was strongly overestimated in the NYC metropolitan area; the analysis of species observations
46 and model predictions shows that most of this over-prediction stems from organic aerosols and crustal
47 material. A later study by Doraiswamy et al. (2010) demonstrates that the CMAQ model significantly over-
48 predicts PM_{2.5} in NYC during the summer both in the pre-morning and post-sunset hours. In trying to
49 determine the root cause of these overestimations during specific periods, the need for vertical profiling
50 studies becomes clear. In addressing these issues more directly, Zhang et al. (2012) indicate that the

51 parameterizations for urban sublayer process and physiographic data are challenging but critically
52 important for the $PM_{2.5}$ forecast in the urban areas since they effect pollutant turbulent mixing, dispersion
53 and deposition. On the other hand, Gan et al (2011) use vertical profiling data from a ceilometer to explore
54 the mass concentration over different vertical ranges and show that the integrated $PM_{2.5}$ mass diurnal pattern
55 by the model was in agreement with the ceilometer observation. Further, it was shown that the periods of
56 highest $PM_{2.5}$ over-biases in the model were due to under-predictions of the PBL height which itself was a
57 consequence of incomplete modeling of urban heating processes in the PBL.

58 In addition, smoke from wildfires or biomass burning is one of the major sources in uncertainty of air
59 quality model forecast. Smoke is a critical air pollutant subject to the NAAQS (Zhang et al., 2008; Huang
60 et al., 2017; Miller, 2011). Many efforts have been made to improve modeling the impacts of wildfire
61 emissions on air quality. National Air Quality Forecasting Capability (NAQFC) is a joint NOAA-NCEP
62 regional operational weather forecasting model and EPA-CMAQ model (Lee et al., 2016; Huang et al.,
63 2017). A major feature is the incorporation of real-time intermittent sources for particles emitted from
64 wildfires within the NAQFC domain and windblown dust from outside the CMAQ by coupling to the
65 NEMS global aerosol capability (NGAC) model, as well as a further upgrade for emission sources using
66 the U.S. EPA's 2011 National Emission Inventory (NEI). Ingesting wildfire sources in particular are from
67 satellite retrievals of fires sources and the estimation of the biomass emission footprints (Lee et al., 2016).

68 While urban local sources can lead to high $PM_{2.5}$ levels, the impact of continental transported plumes
69 on $PM_{2.5}$ under conditions when smoke plumes advect into the PBL is not well understood. To explore this
70 in more details, we study intense wildfire that began on May 1, 2016 in Fort McMurray, Alberta, Canada,
71 forcing the largest wildfire evacuation in the province's history. The wildfire spread across approximately
72 590,000 hectares in northern Alberta and into Saskatchewan before it was declared to be under control on
73 July 5, 2016. Such wildfires are mainly due to the dry and warm weather in spring. The smoke from these
74 fires travelled across the U.S. to the Gulf coast and even to Europe according to multiple satellite images
75 (https://en.wikipedia.org/wiki/Fort_McMurray).

76 While efforts have been made to use direct aerosol observations from satellites to estimate surface
77 level $PM_{2.5}$ (Hoff et al., 2009), these studies assume the surface $PM_{2.5}$ is due to convective mixing of
78 aerosols within the PBL. However, it is clear that aloft aerosol plumes (above the PBL) that can contribute
79 to column total aerosol optical depth (AOD) have little connection with ground $PM_{2.5}$ (Engel-Cox et al.,
80 2006; Han et al., 2015). Thus, vertical distributions and spatiotemporal variations of aerosols are critical to
81 satellite remote sensing of ground $PM_{2.5}$ (Liu et al., 2011; Li et al., 2016; Zhang et al., 2015).

82 In this study, a synergistic remote sensing of wildfires smoke transport, sources, optical characteristics
83 and impacts on the local air quality in New York City is presented by using a ground-based multi-
84 wavelength elastic-Raman lidar, a ceilometer, a sun/sky radiometer, in-situ, satellite and model product in
85 May 2016. The time-height distributions and optical properties of aerosols are presented to identify the
86 intrusions of aerosol plumes and the mixing down of particulates in the PBL. The temporal variations of
87 ground $PM_{2.5}$ and its main speciation (OC, EC, K^+ , sulfate and nitrate, etc.) in the urban and upwind rural
88 areas are analyzed to assess the smoke effects. Different transport paths and origins of smoke are
89 investigated with the NOAA-HMS (hazard mapping system) product and HYSPLIT (Hybrid Single Particle
90 Lagrangian Integrated Trajectory) back-trajectory analysis. Regional-scale spatial distribution of smoke
91 plumes in the eastern US are shown from the satellite CALIPSO lidar. Using the remote sensing and in-situ
92 observations, we evaluate the NAQFC product of $PM_{2.5}$ and PBLH. This paper is organized as follows. In
93 Section 2, the observation instruments and the retrieval method of aerosol optical properties are discussed.
94 In Section 3, the results and discussions on the transported smoke aerosols, as well as the variations of
95 ground $PM_{2.5}$ and species are presented. Finally, Section 4 outlines the study's conclusions.

96 **2. Methodology and Observations**

97 **2.1 Ground-based observations**

98 A suite of ground-based remote sensing instruments were deployed for atmospheric observations at CCNY
99 (40.821° N, 73.95° W), including a 3-wavelength Elastic-Raman lidar, a ceilometer (Vaisala CL-51), an
100 AERONET Cimel sun/sky radiometer, a multi-filter shadow band radiometer (MFR-7) and a microwave

101 radiometer (Radiometrics MP-3000A). Meanwhile, a standard surface air quality monitoring station is
102 operated on the City College campus in NYC by the New York State Department of Environment
103 Conservation (NYSDEC) making hourly PM_{2.5}, O₃ and CO measurements. Downwind of CCNY site (54
104 km northeast), there is another AERONET-site (LISCO, 40.955° N, 73.3419° W) and a national
105 meteorological station at Upton New York (OKX, 40.87° N, 72.86° W) together with routine radiosonde
106 launch for meteorological profiling observation.

107 The multi-wavelength Elastic-Raman lidar emits three wavelengths (355-, 532- and 1064-nm) at a
108 repetition rate of 30 Hz (Spectra-physics Quanta-Ray PRO-320) (Wu et al., 2009). A receiver telescope
109 (Ø50-cm) collects three elastic scattering and two Raman-scattering returns by nitrogen (N₂) and water
110 vapor molecules excited by 355-nm laser output. The signals are acquired by a LICEL transient recorder
111 (TR40-160) and recorded with 1-min average and 3.75-m range resolution. The signal at 1064-nm is highly
112 sensitive to the aloft and thin aerosol layer since the molecular backscatter at this wavelength is much
113 weaker than the aerosol. N₂-Raman returns can be used to derive aerosol extinction coefficients at 355-nm
114 without an assumption of a lidar ratio constant but suffer from small signal-to-noise ratio (SNR) in the
115 daytime. The elastic scattering signals are usually used for the daytime measurements (Fernald, 1984); and
116 we use sunphotometer-measured aerosol optical depth (SP-AOD) to constrain the lidar-derived aerosol
117 extinction profile or lidar-ratio. The lidar signals below 0.5-km altitude need to be corrected with the overlap
118 function because of incomplete overlap between the transmitting beams and receiver's field-of-view. The
119 lidar aerosol products include - aloft aerosol layer and cloud height, aerosol extinction coefficient and
120 extinction-related Angstrom exponent (Gan et al., 2011; Wu et al., 2012). Generally, fine particulates such
121 as smoke aerosols have relatively larger Angstrom exponent than the large coarse particles such as dust and
122 sea salt (Eck et al., 1999). Due to eye safety limitations and the need for human observers, the system is
123 usually operated for 2 to 3 days per week and mostly in the daytime on weekdays except for specific events
124 where more extensive time observations are made. To help address this observational limitation, we also
125 deploy a ceilometer (Vaisala CL-51) which measures laser backscatter at a wavelength of 910 nm as a
126 function of height with a range resolution of 10 m (Münkel, 2004). It employs a pulsed InGaAs diode laser

127 as the transmitter that emits eye-safe laser pulses (3 $\mu\text{J}/\text{pulse}$) at high repetition rate (6.5 kHz). A single lens
128 telescope is used for laser transmission and as the lidar receiver (Diameter=14.8 cm, F#3.0). The total
129 attenuated backscatter or calibrated range-corrected backscatter coefficient is produced, which provides a
130 possibility for studying dynamic PBL processes and evolution that can benefit long-term air pollution
131 studies. Lower power allows for eye safety so that the CL-51 is fully automatic providing 24-hr/7-day
132 operation in all-weather condition.

133 Planetary boundary layer (PBL) is the lowest level of the atmosphere where is directly and strongly
134 influenced by the underlying surface (Stull, 1988). The PBL-height (PBLH) is a key parameter in the air
135 quality and weather modeling. The mixing-layer height (MLH) is the height up to which atmospheric
136 properties or substances (e.g. aerosols and water vapor) originating from the Earth's surface or formed
137 within this layer are dispersed almost uniformly over the entire depth of this layer by turbulent vertical
138 mixing processes. According to Stull (1988), a convective boundary layer (CBL) that occurs during the
139 daytime is usually referred as a mixing layer. At top of CBL, there is an entrainment zone (EZ) where air
140 aloft is entrained and mixed into the aerosol-laden and moister PBL. The lidar-measured MLH in the
141 daytime represents the convective PBLH (Cohn et al., 2000; Menut et al., 1999), usually taking as the
142 middle of EZ. In this study, a wavelet transform technique is used to estimate the MLH where the lidar or
143 ceilometer backscatter profile often shows the largest gradient associated with the aerosol loadings (Gan et
144 al., 2011).

145 In addition, AERONET sun/sky radiometer provides direct measurements of column AOD and
146 Angstrom exponent (AE), and can retrieve relevant aerosol microphysical parameters such as volume size
147 distribution, refractive index and single scattering albedo (SSA) through multi-angle sky radiance
148 measurements (Holben et al., 1998; Dubovik et al., 2000). Absorption AOD and its spectral dependence
149 (i.e. Absorption Angstrom exponent or AAE) can be also derived. For "pure" BC in the atmosphere, the
150 AAE is assumed to be one (Bond and Bergstrom, 2006) and observations of AAE greater than one are often
151 taken as evidence of brown carbon (BrC) or dust (Russell et al., 2010). Typically, the uncertainty in
152 AERONET-sun photometer AOD under cloud-free conditions is within ± 0.01 for $\lambda > 440$ nm and less than

153 ± 0.02 for shorter wavelengths (Hoblen et al., 1998; Eck et al., 1999). Error for particle size distribution is
154 estimated to be 15~25% of the radius between 0.1 and 7 μm (Dubrovnik et al., 2000). In this study, Level-
155 1.5 cloud-screen product of AOD and AE are used (<http://aeronet.gsfc.nasa.gov/>). The data at the LISCO-
156 site (a downwind area near the lidar site) are used since the instrument at CCNY site was taken down for
157 the calibration during the period of this study.

158 Additionally, New York State Department of Environment Conservation (NYSDEC) samples $\text{PM}_{2.5}$
159 and PM speciation (OC, EC, Sulfate, etc.) in both urban and non-urban sites (Rattigan et al., 2010, 2016).
160 At CCNY campus, there is an air quality-monitoring site where the $\text{PM}_{2.5}$, O_3 and CO are reported hourly.
161 The $\text{PM}_{2.5}$ and its speciation are routinely monitored at Queens College site (QC, $40.736^\circ\text{N}/73.822^\circ\text{W}$, 14
162 km southeast from CCNY as shown in Fig.1a below) in NYC. The Newburgh site ($41.499^\circ\text{N}/74.01^\circ\text{W}$)
163 located in the north of CCNY (~76 km away) is herein referred to a non-urban or rural site, and the data are
164 useful for evaluating the transport effect. On the other hand, the site of Division Street is located in southern
165 Manhattan, NYC. The hourly OC, EC and sulfate of $\text{PM}_{2.5}$ are measured using the Sunset OC/EC field
166 analyzer (Sunset Lab, Inc.) and sulfate particulate analyzer (SPA, Thermo Electron Company, model 5020),
167 respectively. In addition, the daily average $\text{PM}_{2.5}$ species are measured using integrated filter-based method
168 once-every-3-day (1-in-3 day) following the planned schedule in the EPA's Chemical Speciation Network
169 (CSN) (Oliver et al., 2010). Measurement errors of $\text{PM}_{2.5}$ include uncertainty in cut point tolerances, particle
170 bounce and re-entrainment, impaction surface overloading, and losses to sampler internal surfaces. Relative
171 uncertainty is generally $\pm 15\%$ for Federal Reference Method (FRM) $\text{PM}_{2.5}$ mass measurement based on the
172 previous studies (Rees et al., 2004; Hains et al., 2007).

173 **2.2 Satellite products**

174 The NOAA Hazard Mapping System (HMS) was developed in 2001 by the National Environmental
175 Satellite and Data Information Service (NESDIS) as an interactive tool to identify fires and the smoke
176 emissions over North America in an operational environment. The system utilizes two geostationary and
177 five polar orbiting environmental satellites (Ruminski et al., 2016). The result is a quality-controlled display

178 of the locations of fires and significant smoke plumes. HMS has a number of detection limitations such as
179 clouds hindering detections, no vertical structure information and no quantitative amount or density of
180 smokes and only available during daylight. In addition, the NOAA-HYSPLIT model is used to compute air
181 parcel trajectories and model the dispersion and the route of airborne particles (Draxler et al., 1997) and
182 can be used either in a back-trajectory mode to analyze sources or in forecast mode.

183 CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) instrument, on board the CALIPSO
184 satellite platform, is a spaceborne polarization-sensitive two-wavelength (532- and 1064-nm) lidar (Winker
185 et al., 2009). It observes global aerosol/cloud vertical distribution and provides aerosol type classification
186 and optical properties products. CALIPSO has a narrow laser footprint (~70 m) at the earth's surface and a
187 16-day revisit cycle. The laser pulse repetition frequency of 20.16 Hz produces profile every 335 m along
188 the ground. In this study, the latest release product (Version 4.10, Level-1 attenuated backscatters, linear
189 volume depolarization ratio and Level-2 aerosol extinction and aerosol type classification) are used.

190 **2.3 NAQFC (the NOAA National Air Quality Forecasting Capability) model**

191 NAQFC consists of the NOAA-NCEP regional operational weather forecasting model (NAM-North
192 America Model) and EPA-CMAQ model (Lee et al., 2016; Huang et al., 2017). It is designed to provide 2-
193 day model forecasts of O₃ and fine particulates (PM_{2.5}) twice per day at the 06 and 12 UTC cycles. For the
194 study, products with spatial resolution of 12 km at the 06 UTC are used. The NAQFC program performs
195 incremental tests and evaluations verified against the U.S. EPA AIRNow surface monitoring network.

196 A modified version of the U. S. EPA CMAQ model (version 4.6) dubbed CMAQ v4.6.5, is run with
197 12 km horizontal grid spacing with a Lambert Conformal Conic (LCC) map projection for the product used
198 in this study. The offline coupling between NWS/NCEP NAM meteorological model and CMAQ is
199 achieved by two pre-processors. In addition to the coupled NMMB-CMAQ system, there are other
200 components such as the emission module and the chemical lateral boundary condition builder as well as the
201 product generating post-processing components. Emission inventories are processed by sectors, but the fire
202 sectors do not include prescribed burns and wildfires from the National Emission Inventory (NEI). The U.S.

203 EPA-NEI 2011 version-1 is being incorporated into Premaq's emission projection schemes. The 2006
204 Environment Canada National Inventory sources were used for Canada, and the 2012 Mexico NEI non-
205 road sources were used for Mexico. The emissions from wildfires, prescribed agricultural burns, and land
206 clearing fires were computed using the dynamic fire emission modeling U.S. Forest Service BlueSky smoke
207 emission package (O'Neill et al., 2009) and the NOAA-HMS for fire locations and strength. Wildfires that
208 are estimated to last at least 24 hours are used as emission sources into the NAQFC.

209 The NAQFC CMAQv4.6.5 follows largely the U.S. EPA Aero4 module and the related emission and
210 removal processes found in the U.S. EPA-CMAQ version 4.6 (Foley et al., 2010). Gas to particle conversion,
211 heterogeneous reactions, depositional growth, and coagulation are included (Kelly et al., 2009). The Aero4
212 module simulates particle formation, condensational and coagulation growth or evaporative dissipation of
213 existing particles due to ambient chemical, temperature and humidity conditions. The detailed configuration
214 for NAM-CMAQ system can be found in Lee et al. (2016). The Mellor Yamada Janjic (MYJ) PBL scheme
215 (Janjie et al., 2001) is used in this version of NAM.

216 **3. Results and discussions**

217 **3.1 Temporal variation of PM_{2.5}**

218 Figure 1 shows the geolocation of PM_{2.5} sites and temporal variation of ground PM_{2.5} in the urban and
219 non-urban areas of NYC in May 2016. As shown in Fig.1 (a), Newburgh-site represents a non-urban site to
220 the north of the CCNY-site while other sites are located in the NYC urban area. The average PM_{2.5} over the
221 15 urban sites are also given. It can be seen that there are two significant elevated PM_{2.5} events (episode-1
222 on May 9-13 and episode-2 on May 25-29). The comparison of PM_{2.5} among the urban and non-urban sites
223 for these two cases shows the strong regional footprint of the enhanced PM_{2.5}, indicating that this is not due
224 to local sources but is likely due to transported sources into the NYC area. The dramatic nature of these
225 events can be seen by the large increase in PM_{2.5} from the background levels. For example, at the CCNY
226 site, the PM_{2.5} concentrations increased from 5- to 30- $\mu\text{g}/\text{m}^3$ on May 25-27.

227 Figure 2 gives the temporal variation of the PM_{2.5} speciation and O₃ at Queens-College (QC) site. In
228 Fig.2 (a), both the OC and EC show consistent increase with a dominant OC concentration for the two
229 episodes. For instance, the hourly OC increases from 2 µg/m³ to 8 µg/m³ while the hourly EC increases up
230 to 4 µg/m³ during May 25-27. Meanwhile, the sulfate also indicates a clear increasing trend from May 25
231 to 27. These values are much larger than the monthly average of OC (1.0~ 4 µg/m³), EC (0.5~1.4 µg/m³)
232 and sulfate (1~3 µg/m³) in NYC based on the multi-year data (Rattigan et al., 2010, 2016; Masiol et al.,
233 2017). Importantly, the O₃ concentrations were in exceedance of NAAQS (70-ppb) for the episode-2 on
234 May 25-26. The daily average of PM_{2.5} species from Chemical Speciation Network monitors (1-in-3 day)
235 at Queens College are given in Fig.2 (c), which indicate consistently high OC on May 9 and 27, respectively.
236 In addition, Potassium ion (K⁺) acts as a useful tracer of wildfire smoke because there are few
237 anthropogenic sources, and its concentrations above background levels are a signature of wildfire emissions
238 (Pachon et al., 2013; Dreessen et al., 2016). As shown in Fig.2 (d), the K⁺ concentrations are amplified in
239 the period of episode-I and II. Daily average concentration of K⁺ on May 9-12 (20-22 ng/m³) and May 24-
240 27 (26-42 ng/m³) are much bigger than the value (2.7-6.0 ng/m³) on the other days in the same month.
241 These results indicate the smoke-dominated plume transport and possible mixture with the upwind
242 industrial aerosols (i.e. sulfate) along the transport path.

243 The correlation between the PM_{2.5} concentrations at the CCNY (urban) and Newburgh (rural) is very
244 high with a correlation coefficient (R~0.84) and the linear regressions slope of 0.94 (see Fig.3). In addition,
245 the PM_{2.5} at the upwind rural sites of Whiteface and Pinnacle (atmosphere background site operated by
246 NYSDEC) also shows similar trends to the data at CCNY. Thus, such coincident increase and good
247 correlation are good indicators that the PM_{2.5} at those sites are probably from the similar sources that is
248 associated with the long-range transport of smoke.

249 **3.2 Time-height distribution and optical properties of aerosols**

250 **3.2.1 Episode-1 on May 9-13**

251 The time-height distributions of aerosols and clouds are shown in Fig. 4 from the lidar-ceilometer measured
252 attenuated backscatter during May 9-13, 2016. Some clouds are labeled (dark-red in color) by showing
253 much stronger backscatter signals than the aerosols but much weaker signals above the cloud due to strong
254 attenuation of cloud. As shown in Fig. 4(a), the lidar data in the daytime of May 9-12 shows deep mixing-
255 layer height (MLH) at ~2.5 km on May 9, and the stratification structures or multiple layers of aerosols can
256 be clearly seen in the free troposphere on May 10-12. The MLHs are generally lower than 1.5 km on May
257 11-12. As shown in Fig.4 (b) from the ceilometer measurement, in the early morning of May 9, an aloft
258 layer of aerosols showed a gradual subsidence and then mixed down into PBL at 6 am. A regional sinking
259 air in the northeast coast of US was indicated from the NOAA/NCEP vertical wind velocity at the 850-
260 mbar level at 6:00 UTC on May 9, 2016 (see Fig.10 (d)), which in combination with strongly convective
261 PBL provides a favorable condition for the plume mixed and transported to the ground. These dense aloft
262 aerosols are likely from the long-range transport of plumes due to their isolation from the local near-surface
263 emissions or pollutants. Meanwhile, the mixing and dispersion of aerosol plumes in the PBL was expected
264 in the convective PBL during solar heating of the atmosphere after sunrise. Figure 4(c) shows the
265 continuous 24-hr measurements during May 8-13. Some rainfall occurred in the morning of May 8 and then
266 followed with the clean sky. During May 11 and 12, other aloft aerosol layers were persistently observed
267 around 2-km; and strong backscatters in the PBL indicate generally larger aerosol concentrations.

268 The aerosol extinction coefficients and AOD are derived from the CCNY-lidar and shown in Fig. 5.
269 On May 9 in Fig.5 (a), the peak extinction of aerosols attains 0.1 km^{-1} at 532-nm, and the aloft aerosol
270 layers generally show the extinction coefficient less than 0.05 km^{-1} . The extinction-related Angstrom
271 exponents are estimated in range of 1.5 to 2.5 on May 9-12, which imply the fine-mode dominated aerosols.
272 Figure 5(b) gives the aloft-layer fraction in the total AOD. The aloft-layer-fraction in the total AOD is
273 generally lower than 0.2 on May 9 but becomes larger on May 11-12 with the value of 0.4-0.6. Such AOD
274 contribution by the aloft-layer may cause major difficulties in using satellite-measured column AOD to
275 estimate ground $\text{PM}_{2.5}$.

276 3.2.2 Episode-2 on May 25-29

277 Figure 6 shows the time-height distributions of aerosols during May 24-29, 2016. In Fig.6 (a), the lidar
278 measurements indicate the deep mixing-layer-heights (dash line in the figure) over the convective daytime
279 period on May 25, and the aloft aerosols are clearly visible above the PBL on May 26 and 27. As shown in
280 Fig.6 (b) from the ceilometer measurement, starting from 5:00 am on May 25, two aloft aerosols layers
281 appeared in the PBL, one below 1-km and other at 2~3 km at noon. In particular, the PBL heights show a
282 dramatic increase and reaches up to 3 km in the afternoon, which indicate strong convective or turbulent
283 process resulting in plume mixing within the PBL. Figure 6(c) shows the 24-hr measurements during May
284 24-29. Some rainfall occurred in the morning of May 24. During May 25-28, the PBL-tops show strong
285 diurnal variation with the maximum height of 2.5~3 km at midday but below 1-km at night. Some aloft
286 aerosol layers are indicated in the morning of May 27, and then descend to the surface in the afternoon and
287 night of May 27. Finally, on May 29, the PBL heights become lower at 1.5 km with the smaller attenuated
288 backscatters, and later some rainfall occurs by mid-night.

289 The aerosol extinction coefficients are derived from the CCNY-lidar and shown in Fig. 7. In Fig.7 (a)
290 on May 25, the peak extinction of aerosols attains 0.2 km^{-1} at 532-nm with the Angstrom exponents of
291 1.5~1.7 in Fig.7 (b) that indicate fine-mode particles dominated. The dense aerosol layers at 1.0-2.5 km can
292 be clearly seen before 14:00. Meanwhile, Figure 7(b) shows the profiles of aerosol extinction coefficients
293 at three-wavelength and Angstrom exponents over half-hour average. They show similar structure of
294 aerosols; and the good consistency of 355-nm extinctions between the two independent retrievals from the
295 Raman- and elastic-scattering signals verifies the reliability of retrievals. In addition, the lidar-derived
296 aerosol optical depths vary from 0.2 to 0.4 at 532-nm that indicate more aerosol loading than the one in
297 episode-I. On May 26, the aerosol extinction coefficients in the PBL are in the range of $0.05 - 0.2 \text{ km}^{-1}$
298 while the PBL-tops reach 2.5 km at 11:00 am-6:00 pm. On May 27, more aloft plumes can be seen below
299 4-km while the MLHs are around 2-km altitude. In addition, the mean of the Raman-lidar derived lidar-
300 ratio in the PBL was found to be around $61.4 \pm 5.9 \text{ sr}$ at 355-nm that denotes smoke-like particles (Omar
301 et al., 2009).

302 The correlation between the ground $PM_{2.5}$ and ceilometer-measured attenuated backscatter at near
303 surface is analyzed, and the results in Fig. 8 indicate the modestly high correlation coefficient of 0.7. The
304 relative humidity is generally lower than 70% that means that the moisture is not playing a major role in
305 enhancing the aerosol backscatter. Thus, the ceilometer-measured attenuated backscatter can serve as a
306 reasonable proxy for $PM_{2.5}$ comparisons with models.

307 Figure 9 shows the column aerosol optical properties from the AERONET-measurements. A consistent
308 increase of AOD from 0.18 to 0.45 at 532-nm can be seen on May 25 while the Angstrom exponents are in
309 range of 1.6 - 1.4 at the wavelength pair of 440-870-nm. The single scattering albedos in Fig.9 (b) decreases
310 from the morning to the afternoon (15:52 local time or eastern daylight saving time). We calculate the
311 absorption Angstrom exponents (AAE) at 440-870 nm; it shows a decrease from 1.4 to 1.08 along the plume
312 intrusion. The AAE value with the aerosol plume is close to the value of 1.0 for black carbon (Russell et al.,
313 2010). These results suggest the increase in absorbing particles due to intrusion of transported smoke on
314 May 25. On May 26, the AODs are in the range of 0.25 - 0.45 while the extinction-related Angstrom
315 exponent are smaller than 1.6. On May 27-28, the Angstrom exponents become relatively higher (1.6 - 1.8)
316 but then decrease (1.2-1.6) on May 29. Such temporal variation of Angstrom exponents might correspond
317 to different types of aerosols associated with the transport and mixture with local aerosols.

318 **3.3 Source and transport path of wildfire smoke**

319 Figure 10 shows the MODIS RGB image that indicates the wildfires and smoke over southwestern Canada
320 (Alberta, Saskatchewan, and Manitoba) and northern boundary of U.S on May 8 (Episode-1). The wildfire
321 points and smoke dispersion areas are shown in Fig.10 (b) from the NOAA-HMS product, where the red
322 symbol 'x' represents the fire burning points and the yellow areas denote the smoke dispersion/diffusion
323 region. The results indicate the wildfires at Fort McMurray of Alberta spreading into Saskatchewan and
324 U.S. The numerous severe wildfires burned around Fort McMurray, Alberta, Canada since May 4. In
325 addition, Fig.10 (b) shows the air backward trajectories with the end points at CCNY-lidar site (6:00 UTC
326 on May 9, 72-hr long at the altitude of 1-km and 2-km). The results verify that the air parcels in the PBL in

327 NYC are transported from the wildfire regions in Canada and northwest U.S. Figure 10 (c) shows the
328 surface smoke content from the NAAPS (Navy Aerosol Analysis and Prediction System) model and air
329 backward trajectories at 1-2 km on May 9. The results indicate the relatively high concentration of smoke
330 along the transport path from the northwest US and Canada to NYC.

331 Figure 11 gives the wildfires and smoke sources on May 25-27 (Episode-2), as well as the backward
332 trajectories with the end points at the sites of CCNY, QC and Newburgh (12:00 UTC, 72-hr long, at the
333 altitude of 0.5-km, 1-km and 2-km), respectively. Interestingly, similar trajectories of transport are indicated
334 among the three sites and at the different altitudes. However, the transport paths show significant shift
335 during May 25-27, from western Canada to western and southeast US. There are still numerous wildfires
336 at Fort McMurray, Alberta, Canada and some wildfires from Mexican, Central American and Arizona.
337 Please note that the extensive cloud cover hinders the smoke detection by satellites on these days that are
338 clearly shown on the satellite visible images. It can be seen in Fig.11 that the smoke over NYC on May 25
339 was transported from Canada. On May 26-27 in Fig.11 (b-c), the air was transported from the western and
340 southeast US where there are the smoke originated from Canada.

341 **3.4 Regional aerosol distribution in the northeast U.S.**

342 Spatial and range-solved distribution of aerosols are shown in Fig. 12 from the satellite NOAA-GOES and
343 NASA-CALIPSO measurements on May 12, 2016. Figure 12 (a) displays the relatively high AOD in the
344 northeast US that are associated with the smoke transport, but there are many clouds in the North US and
345 Southern Canada, which prevent the smoke detection from the satellite measurement. The CALIPSO data
346 in Fig.12 (b) show that the aloft plumes are located below 5-km in the latitude of 41°N ~ 50°N in the early
347 morning of May 12. The aerosol extinctions are in range of 0.05~0.2 km⁻¹ at 532-nm in Fig. 12(c) and the
348 elevated layers are mostly classified as smoke in Fig.12 (d).

349 Figure 13 shows the aerosol spatial distribution on May 26 in the eastern US. In this case, the aerosol
350 layers are mostly located in the PBL (< 3-km) in the latitude of 30° N ~ 44° N. There are some low- and
351 high-level clouds blocking the detection of aerosol layers as shown in Fig.13 (b). The aerosols in the PBL

352 are partly classified as polluted continental/smoke or polluted dust nearby NYC (arrow position in the x-
353 axis in Fig.13(c). This seems not consistent with the fact that the organic carbon are major components of
354 $PM_{2.5}$ with less soil component in NYC (see Fig.2a and Fig.2c). The classification of polluted dust depends
355 on an arbitrary threshold at 0.075 of depolarization ratio (Omar, et al., 2009); and the over-classification of
356 polluted dust for the smoke-dominant events have been observed in the previous literatures (Burton et al.,
357 2013; Wu et al., 2017). The average depolarization ratio in the PBL (<2.5 km) is mostly in the range of
358 0.05 to 0.09 at $30^\circ \sim 45^\circ$ N latitude, and 0.01 to 0.04 at $46^\circ \sim 52^\circ$ N latitude. The causes of depolarization
359 by smoke are not well understood, which may be related to coated soot aggregates, lifting of surface soil
360 into the smoke plume and asymmetry of smoke particles (Burton et al., 2015). As the same lidar-ratio (70-
361 sr at 532-nm) is used in retrieving aerosol extinctions for the type of polluted continental and smoke aerosols,
362 such misclassification of smoke as polluted continental aerosols will not affect the retrievals. Figure 3(d)
363 show good consistency of the aerosol extinction coefficients between the ground lidar and CALIPSO
364 product nearby CCNY-site.

365 **3.5 Model $PM_{2.5}$ and PBL-height verification**

366 Besides getting a better understanding of the properties of transported aerosol plumes with our vertical
367 profiling instruments, a more direct assessment of the NAQFC product of PBL-height (PBLH) and ground
368 $PM_{2.5}$ can be made with our measurements. Only the episode-II (May 25-29) is selected because the current
369 NAQFC considers the wildfire emissions only over the continental US. To verify the reliability of
370 ceilometer-observed PBLH, we firstly analyze the correlation between the ceilometer and lidar-measured
371 convective PBLH; they show excellent linear correlation (Slope= 0.96 and correlation $R= 0.81$) as shown
372 in Fig.14 (a) even to heights of 3 km. The comparisons of the PBLH between the model and ceilometer
373 observations are shown in Fig.14 (b). Both show similar diurnal variation of PBLH, e.g. high PBLH at noon
374 and afternoon but low PBLH at night. The model convective PBLH at noon are much higher than the
375 observation. For instance, the model PBLH on May 25 attains 4.5 km while the measured PBLH is around
376 3.0 km. We further calculate the PBLH using the gradient variation of the model $PM_{2.5}$ mass vertical

377 distributions with a wavelet transform technique (Gan et al., 2011). The results in Fig.14(c) indicate lower
378 than the original or meteorological-based PBLH from the NAM model product in the daytime and are closer
379 to the observation. Figure 14(d) shows the hourly $PM_{2.5}$ from the model and observation at CCNY-site. At
380 first, the model product shows generally good agreement with the observations except some overestimation
381 in the model during the morning of May 25 and 26. In fact, taken over the whole event, the model $PM_{2.5}$
382 shows similar temporal variation to measurements before (May 24) and after (May 29-30) the plume
383 intrusion. Additionally, the comparison of model $PM_{2.5}$ at the other two sites (QC and IS-52) also indicate
384 a similar consistency (not shown here).

385 As seen in Fig. 14(c) for the time-height distribution of $PM_{2.5}$ from the model, the aerosols show some
386 elevated layers up to 4-km in the daytime of May 25 and 26, then become lower at 2-km on May 27-29,
387 which is consistent with the lidar- ceilometer profiles in Fig.6. In particular, this shows the danger of using
388 the meteorological-based PBL height output as the only indicator of vertical aerosol extent. Even the small
389 overestimations of the $PM_{2.5}$ seem to be at least partially explained by the model placement of the $PM_{2.5}$
390 mass that is actually somewhat compressed by the low PBL.

391 **3.6 Discussions**

392 The wildfire smoke transport to NYC was proposed by a synergistic ground-based, satellite remote sensing
393 and in-situ measurement in May 2016. In particular, continuous time-height distribution of aerosol
394 measured by a lidar and ceilometer indicate the aloft smoke intrusion and mixing down into PBL. The
395 mechanisms of plume mixing into PBL and ground depend on the synoptic weather process (e.g. air
396 subsidence), plume height, PBL height and entrainment process. Our study indicates that regional air
397 subsidence in the NE US, high PBLH (2.5~3.0 km) and PBL entrainment in the daytime provide favorable
398 condition for plume vertical mixing down and transport to the ground. Colarco et al. (2004) simulated the
399 mechanism of a Canadian Quebec wildfire plume aloft mixing down to the surface using a model on July
400 7, 2002 in Washington DC area of US. The results indicate that the PBL entrainment plays a major role
401 while the subsidence alone was not enough to explain how the fire smoke was transported from an elevated

402 layer to surface. However, large uncertainties in the model simulations are from the magnitude and injection
403 altitude of fire emissions. On the other hand, Pahlow et al (2005) observed the smoke layer from Canadian
404 Quebec wildfire descending to lower altitude in the PBL in the morning of July 6-8 2002 at Baltimore, MA
405 (northeast US); and attributed this to the thermals overshooting at the PBL-top that coincide with upward
406 and downward movements of air. In addition, Duck et al. (2007) showed that the arrival of biomass burning
407 emissions at the surface from Alaska to the northeast coast of North America was associated with the
408 synoptic-scale conditions (e.g. high-pressure ridge).

409 During the period of the two episodes, the coincident increase of ground $PM_{2.5}$ and its species (OC,
410 EC, K^+) in the urban and rural area indicate the regional transport of the smoke. The OC/EC ratio has been
411 used to confirm the profiles of biomass burning and mobile sources since biomass burning usually
412 has higher OC/EC ratios (7-15) than gasoline (3.0-4.0) or diesel vehicles (<1.0) (Pachon et al., 2013).
413 The OC/EC ratio in this study is in range of 3.0-8.0 for the episode I and II (not shown), which indicates
414 the wildfires smoke. Importantly, the ozone concentrations are in the exceedance of NAAQS ($O_3 > 70$ ppb)
415 on May 25-26 that demonstrates the big impact of wildfire smoke on the local air quality. Thus, the episode-
416 II may be referred to an exceptional event due to the wildfires smoke transport (Geigert et al., 2017). It is
417 difficult to quantify wildfire smoke contribution to the ground $PM_{2.5}$ and O_3 concentrations, particularly
418 when mixed with urban pollution and the secondary aerosol and ozone formed. It is challenging to separate
419 the transported smoke from the local emission though the temporal variations of ground $PM_{2.5}$ before and
420 after the plume intrusions can verify the smoke-associated enhancement of $PM_{2.5}$.

421 In addition, it is challenging to model $PM_{2.5}$ and PBLH in the complex urban environment. The
422 comparisons of $PM_{2.5}$ and PBLH between the NAQFC product and our observations generally show
423 consistent diurnal variation, but the meteorological-based PBLH from the model output is overestimated in
424 the mid-day with a strong convection. When we calculate the PBLH using the model $PM_{2.5}$ vertical profile,
425 the estimated PBLH agree well with the lidar observation. This implies that the meteorological-driven
426 PBLH may not always represent the aerosol volumes in the atmosphere, which partly explain why the
427 model $PM_{2.5}$ are over-predicted in the night. Dreessen et al (2017) show a case study on the Canadian

428 wildfire smoke affected air quality in Maryland, US during June 9–12, 2015, which in particular resulted
429 in a multiday O₃ exceedance of the NAAQS. The results indicate that CMAQ model under-predicted ozone
430 formation by around 14 ppb in Maryland, and need adequate means to quantify and justify ozone impacts
431 from wildfires.

432 **4. Conclusion**

433 This paper has analyzed the Canadian wildfire smoke transports that affect the local air quality in NYC in
434 May 2016. The results demonstrate the intrusions of smoke plumes and mixing down into the PBL. The
435 high MLH at 2.5~3 km and regional air subsidence imply favorable vertical transport and mixing condition.
436 The elevated smoke contributes 40~60% fraction to the total column AOD based on the lidar profiling
437 observation on May 12, which indicates the cautions to estimate ground PM_{2.5} using the satellite-measured
438 column AOD. The coincident increase and strong correlation of ground PM_{2.5} in the urban and upwind rural
439 areas indicate that the dominant mechanism is regional transport. The concurrent enhancement of OC, EC,
440 K⁺ and sulfate indicate the smoke transport and mixture with industrial emissions in the upwind area. The
441 ground O₃ concentrations are in exceedance of NAAQS for the episode-II that may be referred as an
442 exceptional pollution event. The severe wildfires and smoke sources in Fort McMurray, Alberta, Canada
443 are verified to be the cause of episode-1 on May 9. The episode-2 on May 25-29 shows the daily variations
444 of aerosol transport path and optical properties.

445 Our assessment of the NAQFC (NAM-CMAQ) indicate a similar diurnal variation of PBLH except an
446 overestimate by the model in the convective daytime. We find that the PBLH estimated from the model
447 PM_{2.5} profiles agrees well with the observation in contrast to the model meteorologically derived PBLH.
448 This demonstrates that the model derived PBLH is not always the best indicator of the vertical depth of the
449 boundary layer aerosols. The model PM_{2.5} agrees well with the observation during May 24-30 except for
450 an overestimate by the model in the morning of May 25-26, which may be associated with the lower PBLH
451 and vertical mixing of aerosol in the model.

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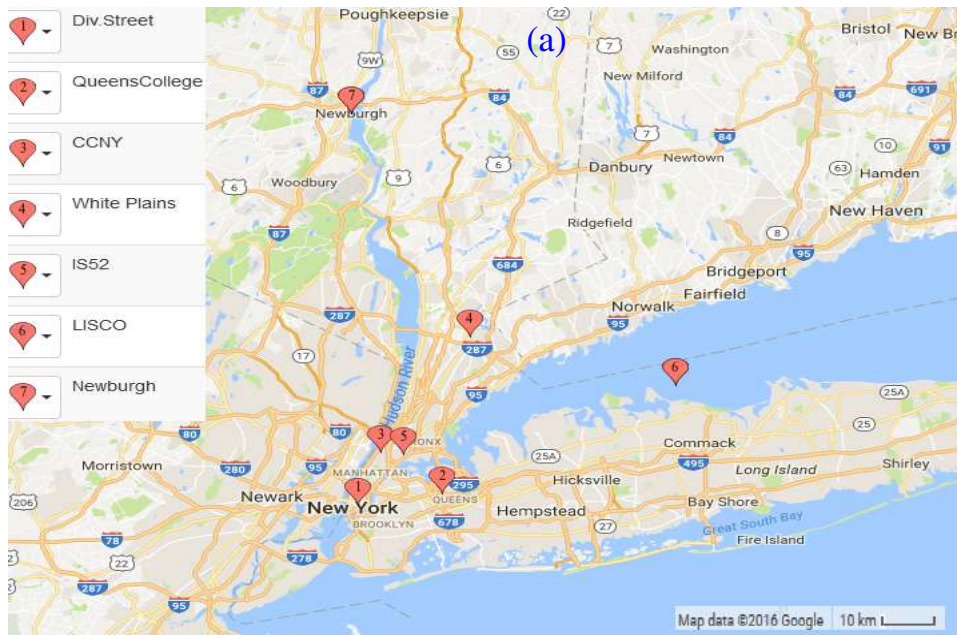
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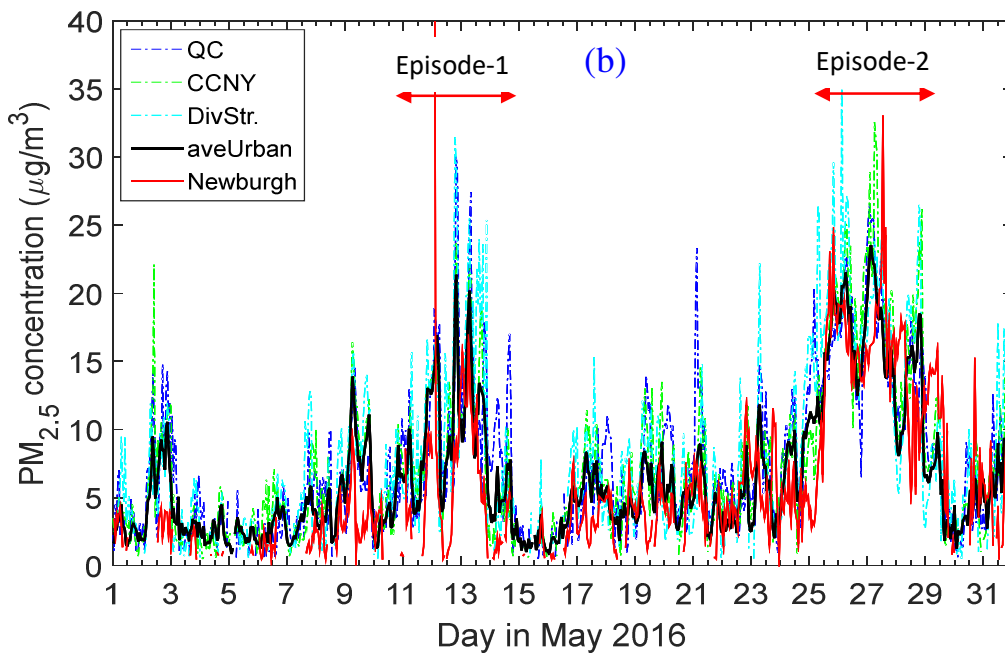
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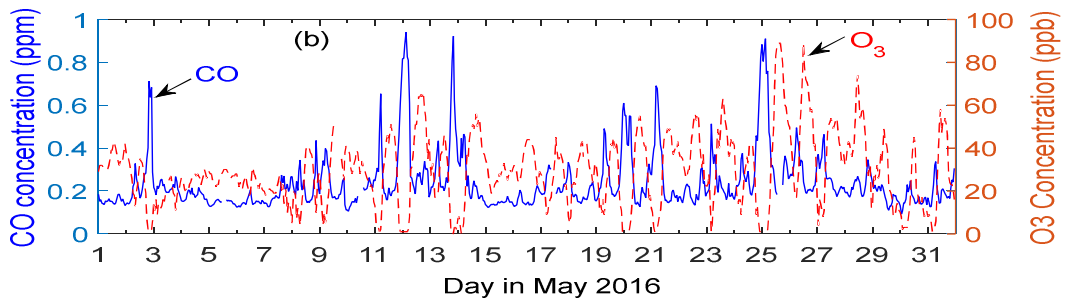
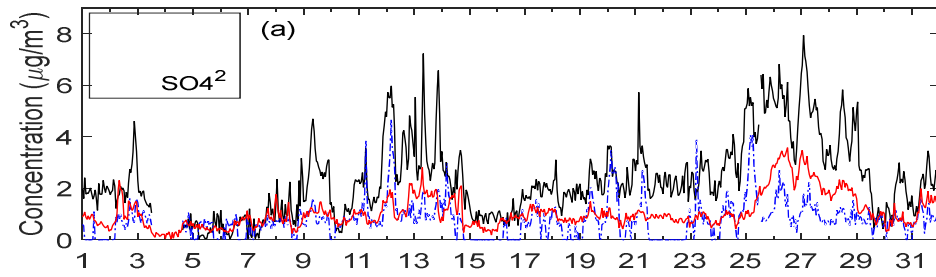


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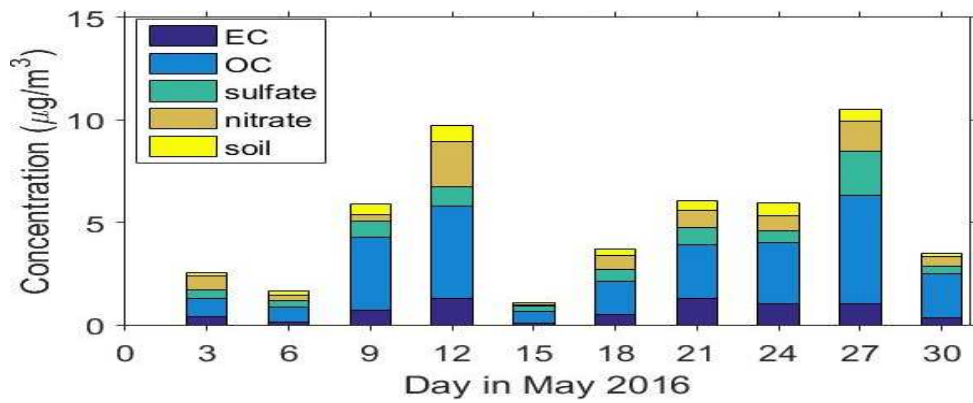
Fig. 1. (a) PM_{2.5}-site geolocations and (b) PM_{2.5} concentration in NYC area, May 2016.

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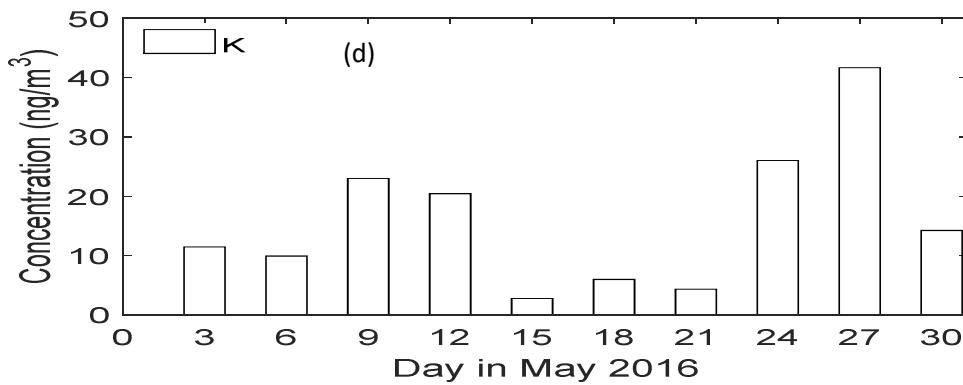
Two episodes of PM_{2.5} increase occurring on May 9-13 and May 25-29, respectively.



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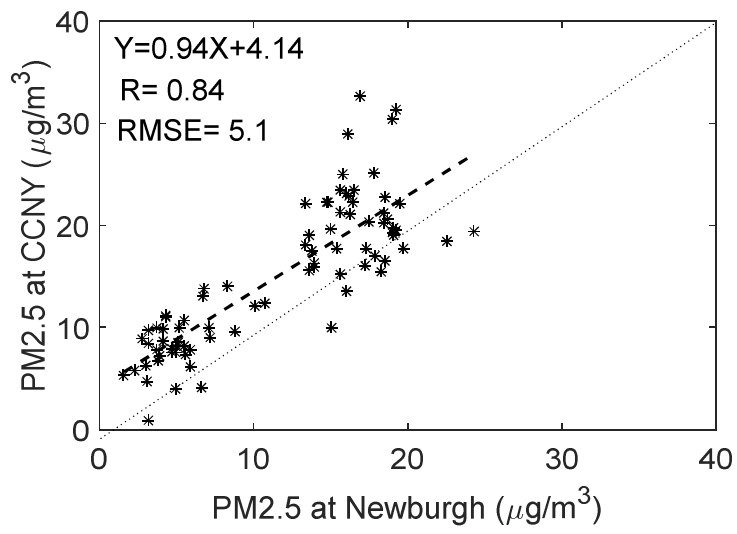


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619 Fig. 2 Temporal variation of PM_{2.5} speciation and ozone. (a) hourly average of OC, EC and sulfate,
 620 (b) ozone and CO, (c) daily average species (1-in-3), (d) Potassium ion (K⁺) at QC site in May 2016.

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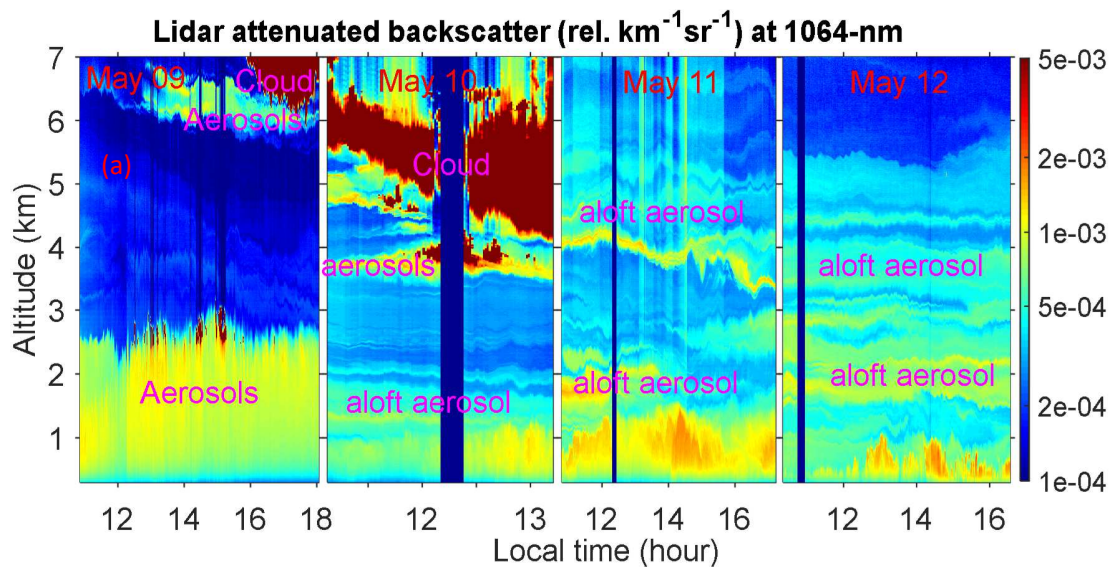
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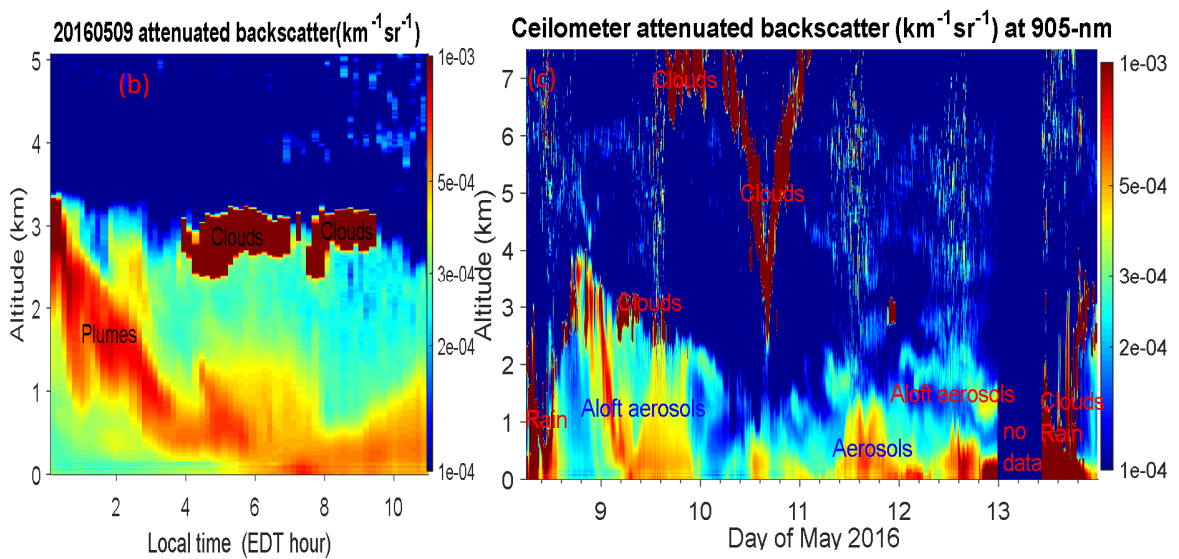
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624 Fig.3 Correlation of $\text{PM}_{2.5}$ at CCNY (urban) and Newburgh (upwind rural) site.

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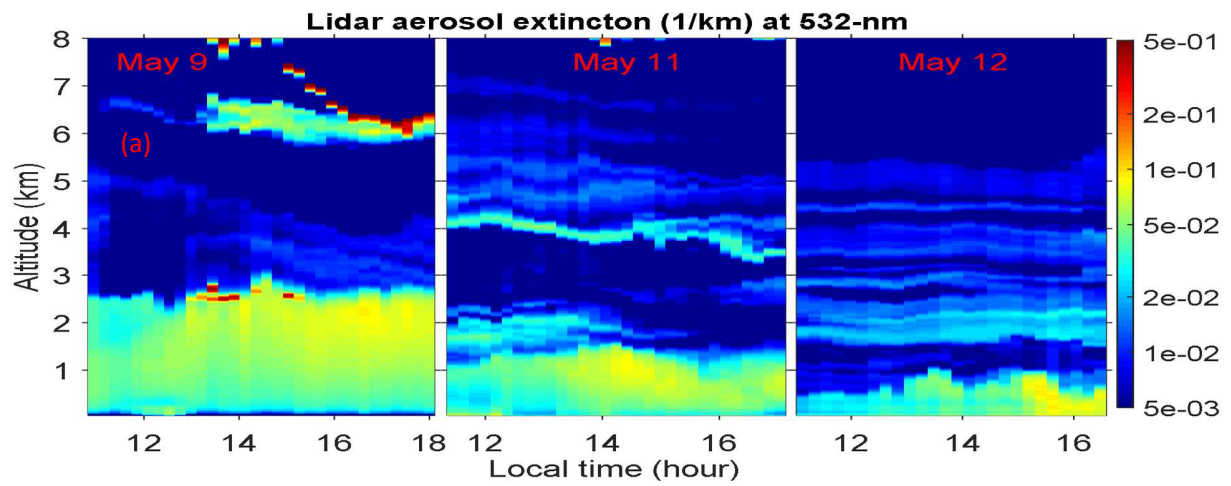


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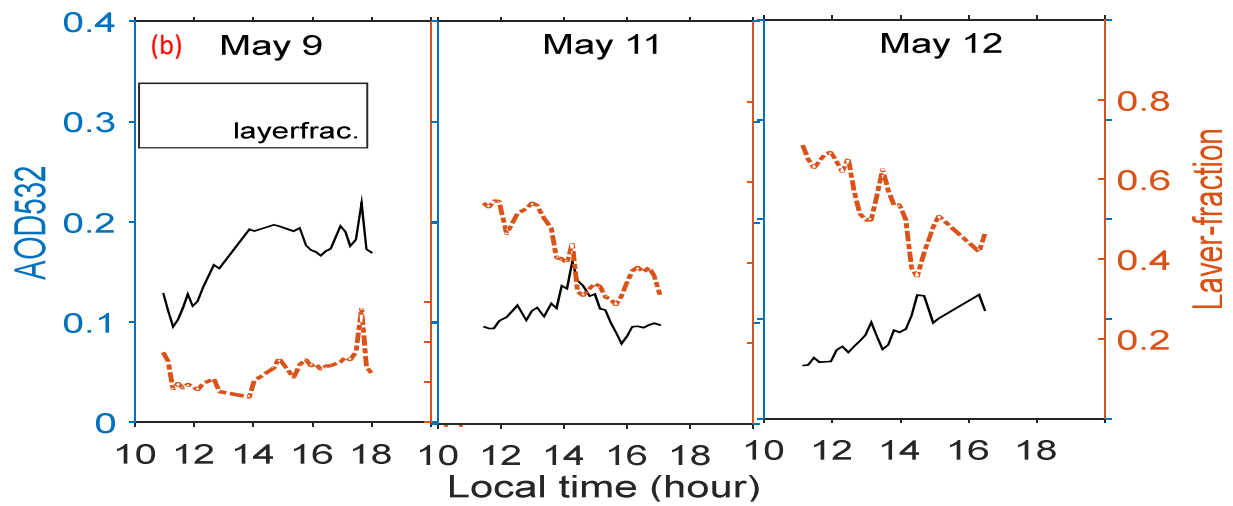


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628 Fig. 4 (a) Range-resolved attenuated backscatter from CCNY-lidar during May 9-12, and (b)-(c)
 629 ceilometer images during May 8-13, 2016 (Episode I). Aloft plumes (yellow-red) and clouds (dark-red)
 630 can be seen.



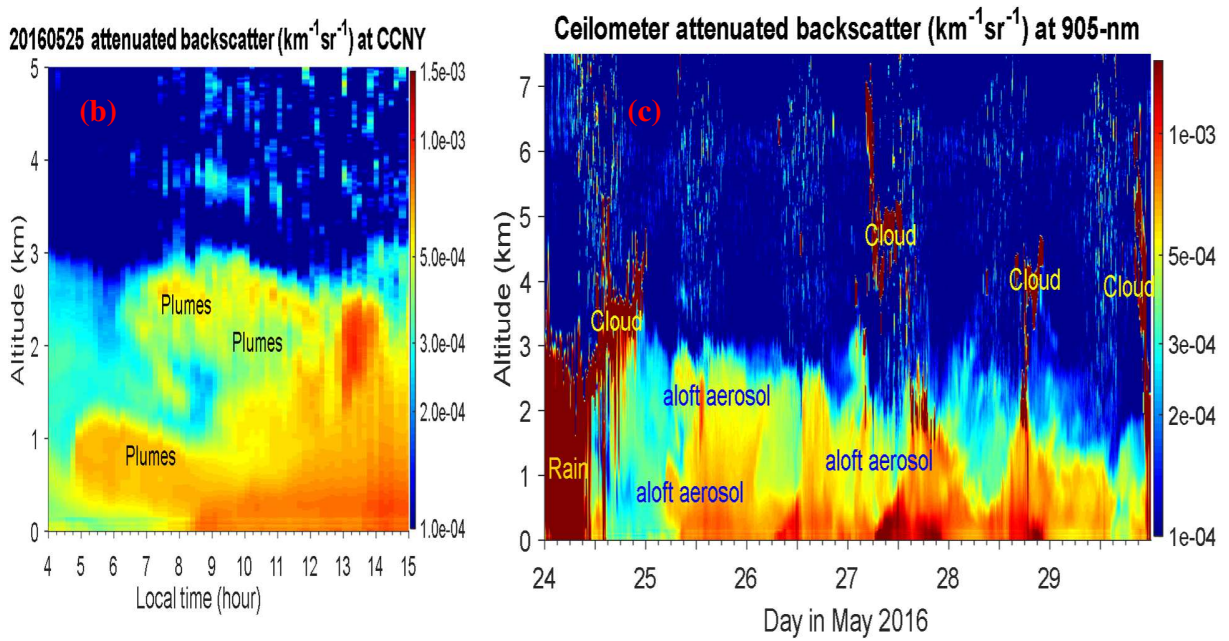
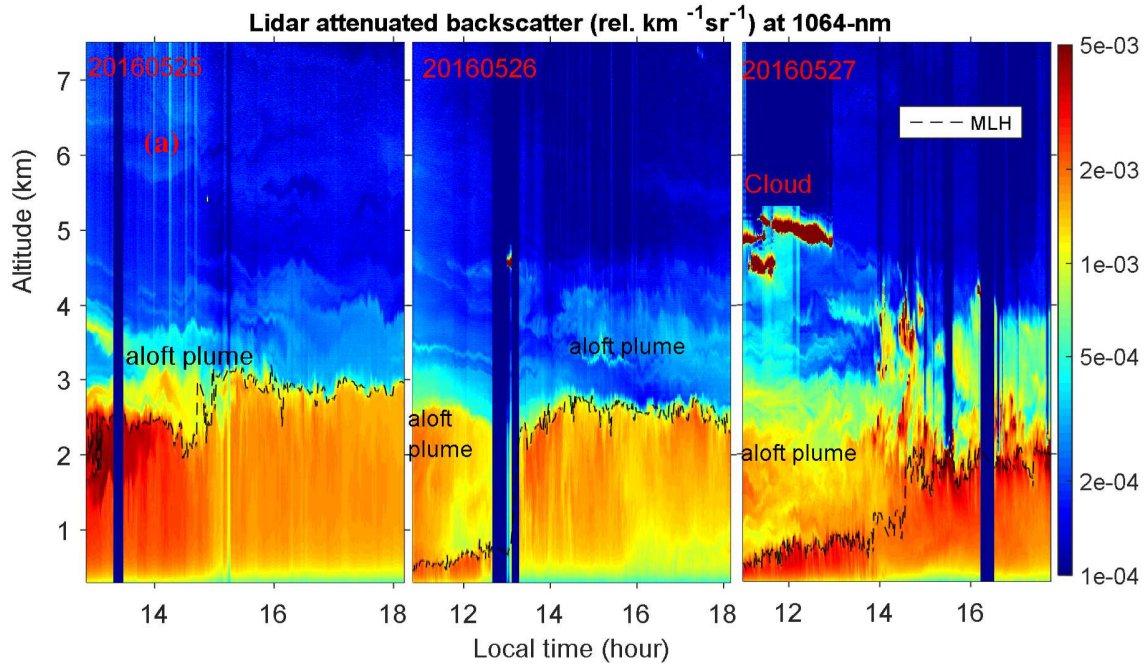
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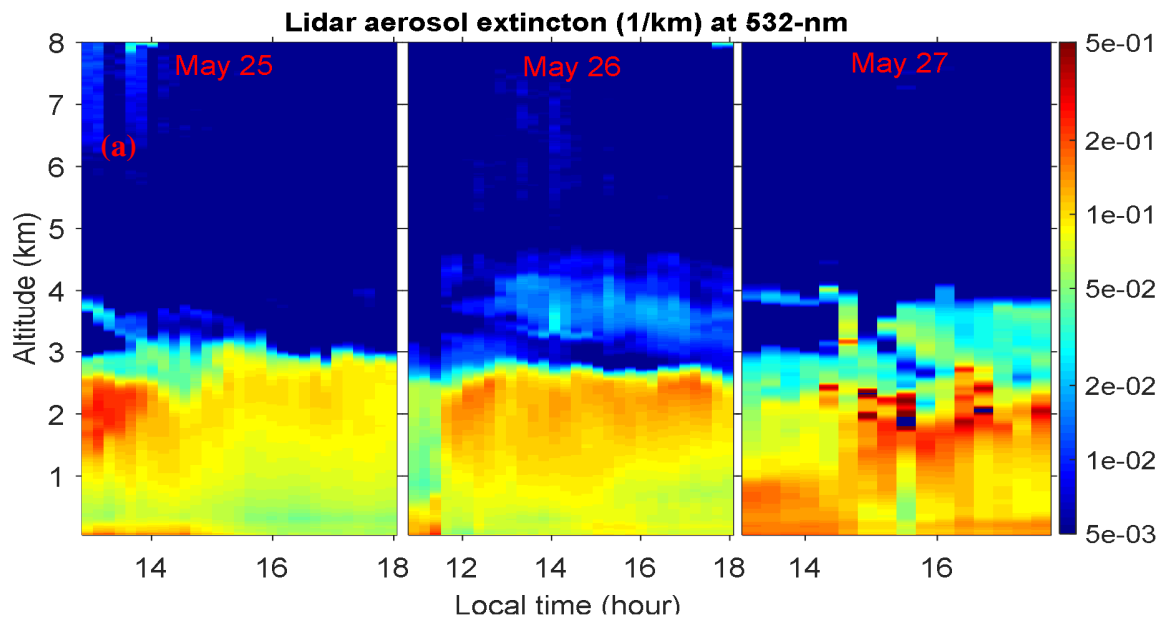
633 Fig. 5 (a) Aerosol extinction coefficients (10-min ave) at 532- nm, (b) total AOD and layer-AOD

634 fraction from the CCNY-lidar during May 9-12, 2016.

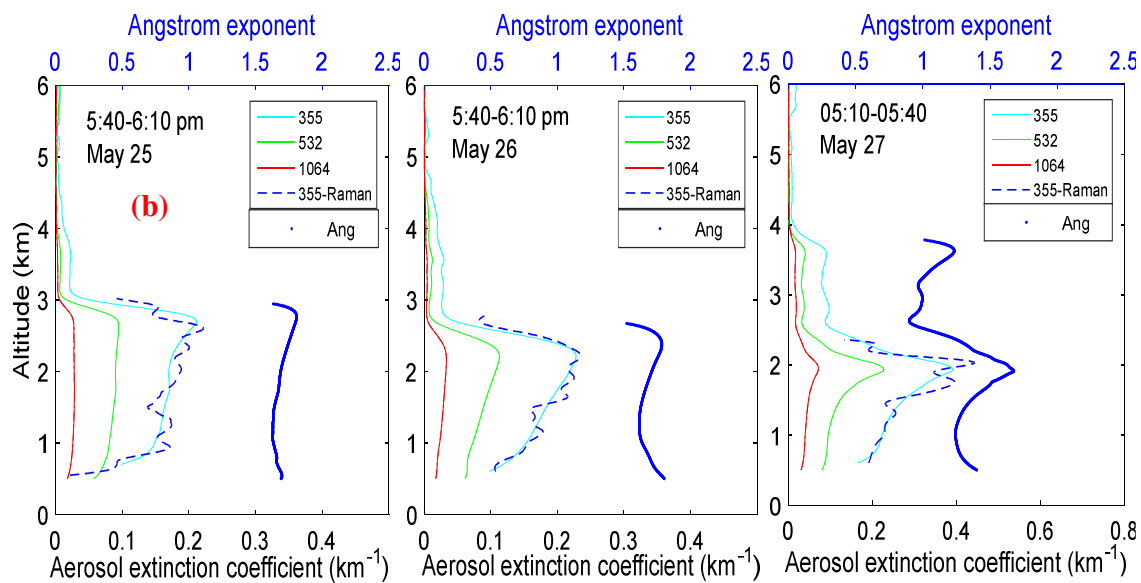


637 Fig. 6. (a) Range-resolved attenuated backscatter (dash line: MLH mixing-layer-height) from the
 638 CCNY-lidar on May 25-27 and (b)-(c) ceilometer data during May 24-29, 2016 (Episode II).

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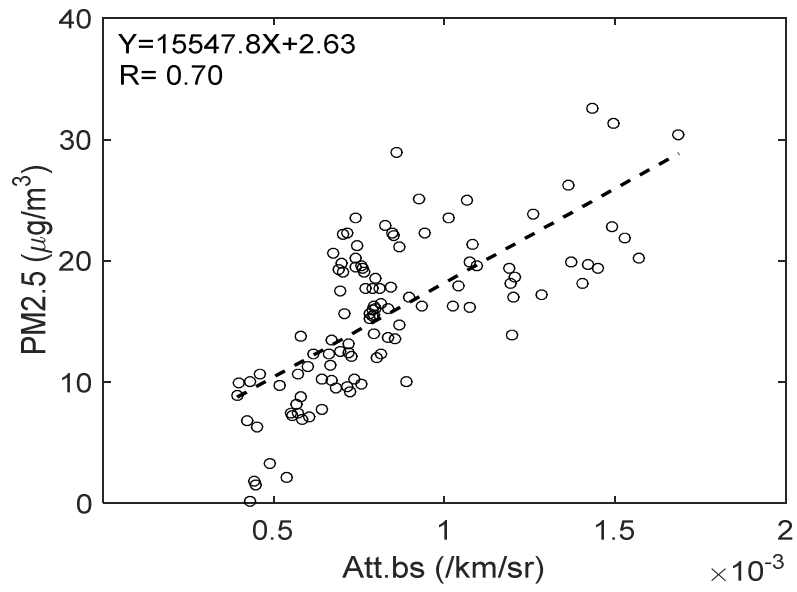


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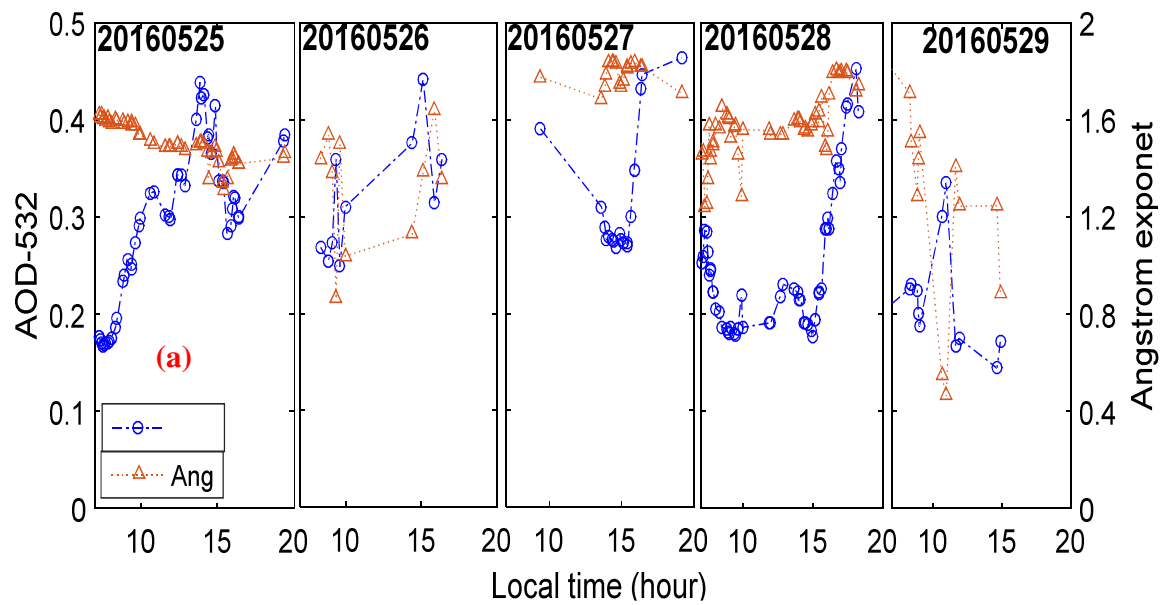
642 Fig. 7. (a) Aerosol extinction coefficient at 532-nm (10-min ave), (b) multi-wavelength
 643 extinction and Angstrom exponent profiles from CCNY-lidar on May 25-27, 2016



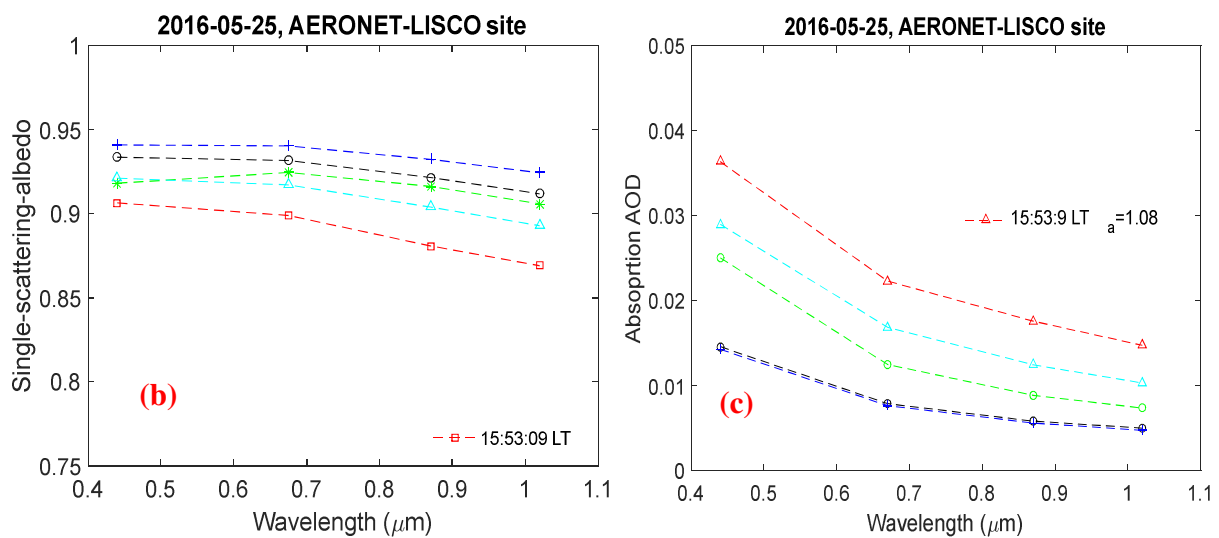
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645 Fig. 8 Correlation of total attenuated backscatter and PM_{2.5} at CCNY

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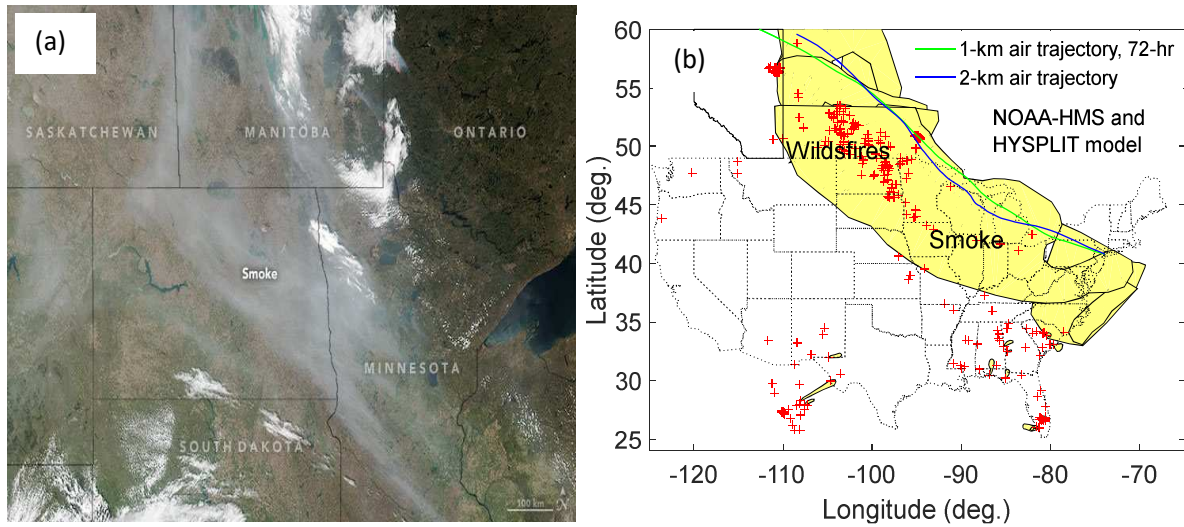


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649 Fig. 9. Aerosol optical properties from the AERONET sunphotometer. (a) AOD and Angstrom
 650 exponent, (b) single-scattering-albedo, (c) absorbing AOD (a_α : Absorption Angstrom exponent).

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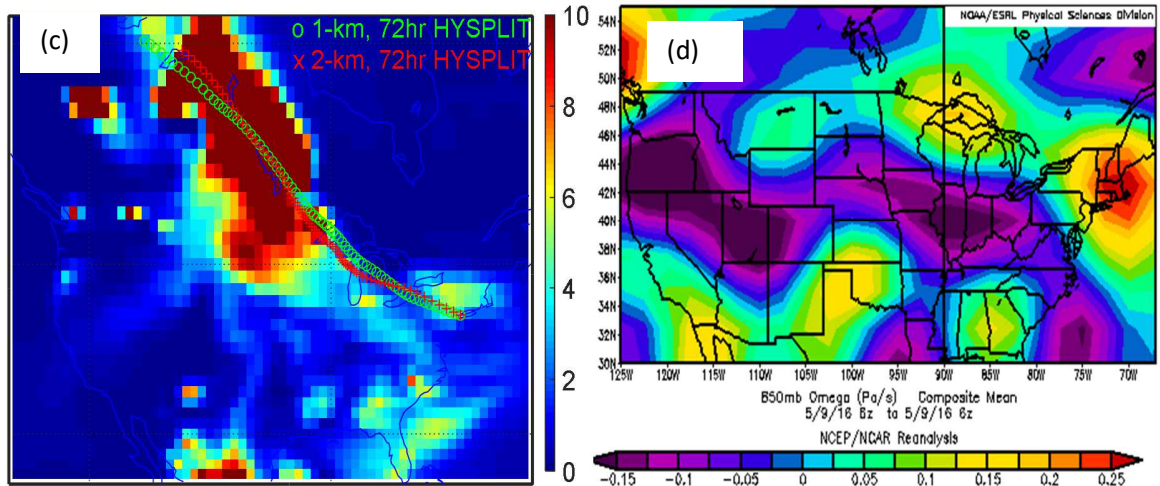
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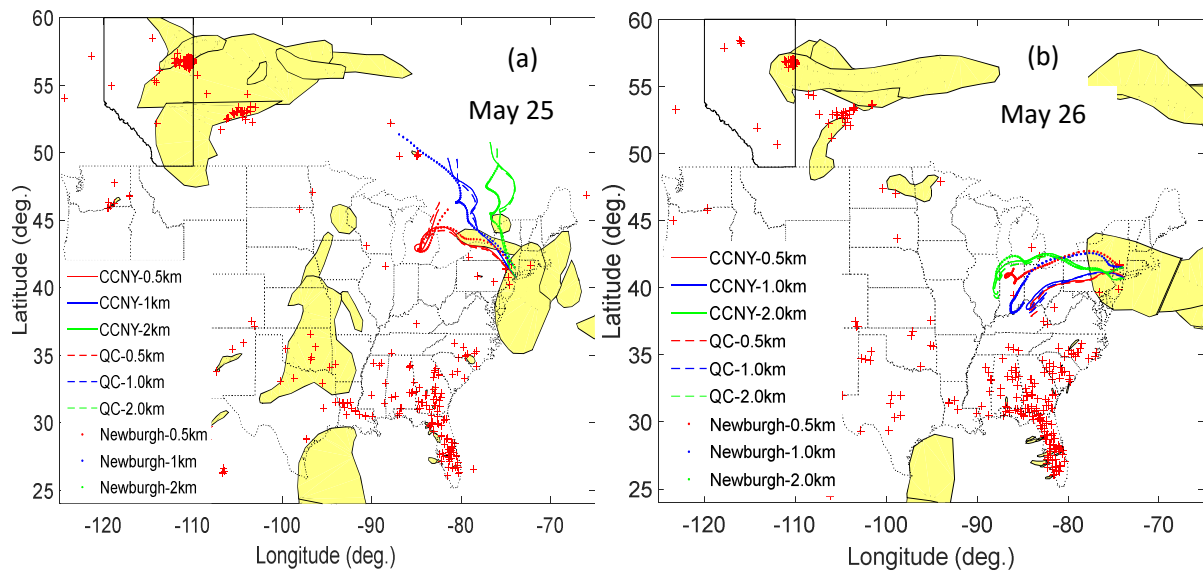
20160509 6UTC NAAPS-model smoke ($\mu\text{g}/\text{m}^3$)



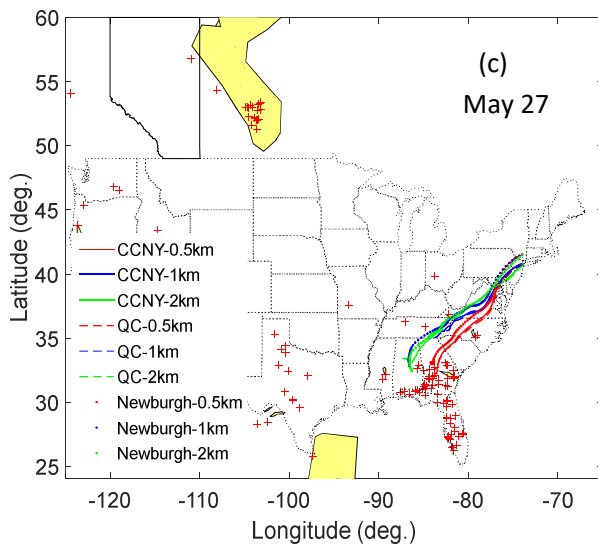
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656 Fig.10 (a) MODIS image showing the smoke over the west Canada and northwest U.S, (b) Fire
 657 sources (red +), smoke areas (yellow) and backward trajectories (green and black lines) by
 658 NOAA-HMS and HYSPLIT model on May 9, 2016; (c) NAAPS model surface smoke at
 659 6:00UTC on May 9, 2016; (d) NOAA/NCEP vertical wind speed at 850-mb level at 6:00 UTC on May
 660 9, 2016 (a positive value corresponds to descending motion and a negative value corresponding to
 661 ascending motion)

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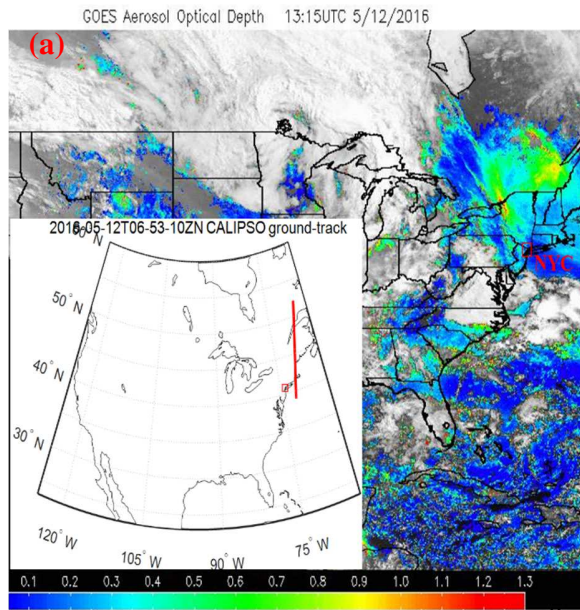


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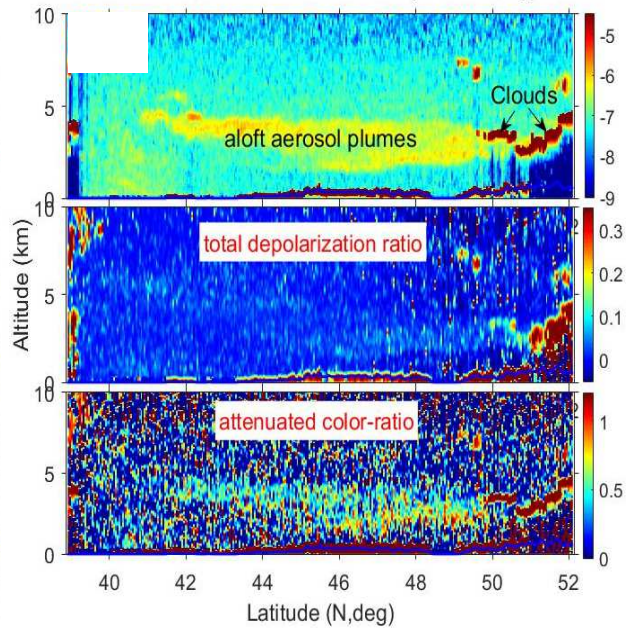
665 Fig.11 (a) -(c) Fire sources (red symbol +) and smoke areas (yellow) from the NOAA-HMS, and
666 air backward trajectories (green-black lines) ending at three sites below 2-km altitude from the
667 NOAA-HYSPLIT model ending at 12: 00 UTC on May 25 (transport from Canada), 26 (transport
668 from the west US) and 27 (transport from the southeast US), respectively. Cloud cover hinders the
669 fire/smoke detection from the satellites.

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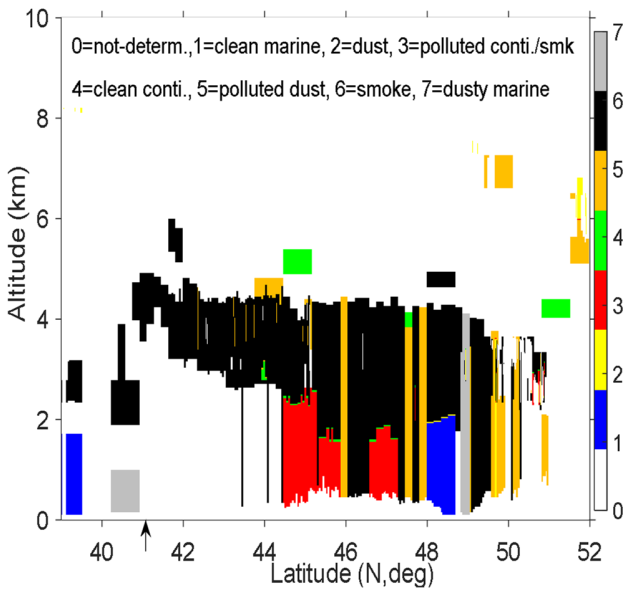
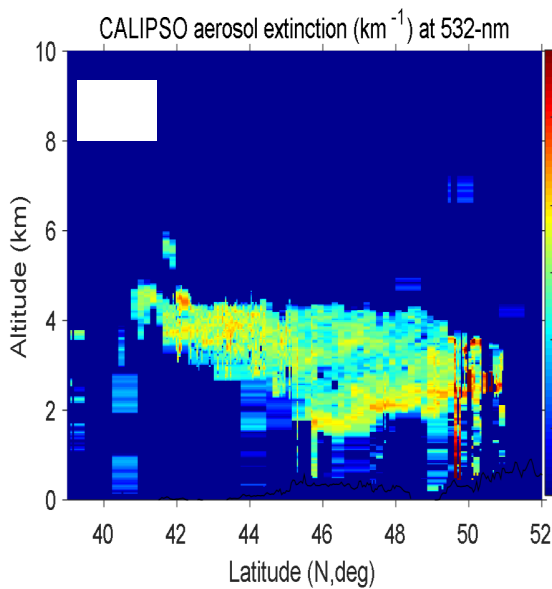
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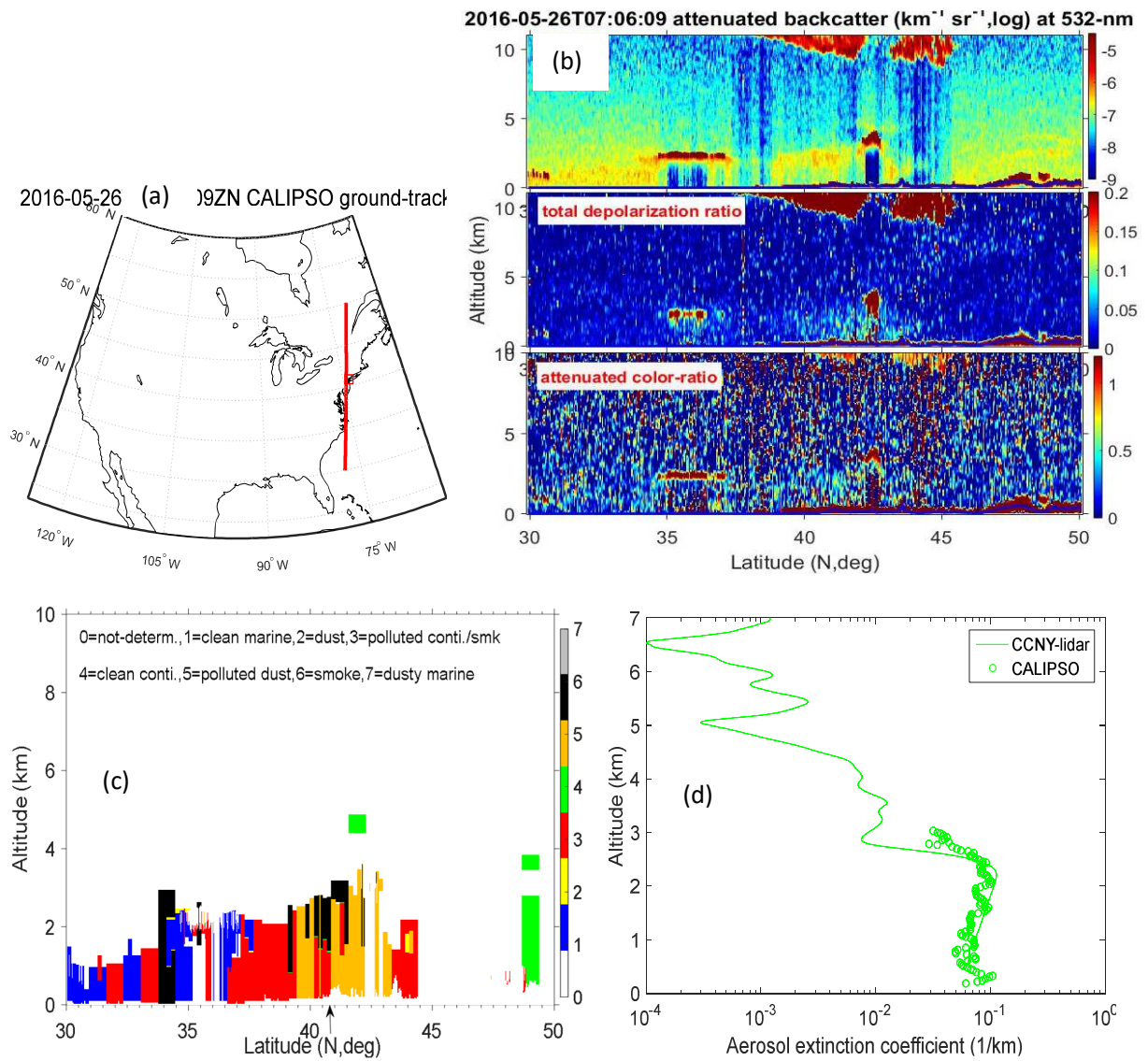
20160512 06:53UTC, attenuated backscatter ($\text{km}^{-1} \text{sr}^{-1}$, log) at 532-nm



672



673 Fig. 12. (a) NOAA-GASP aerosol optical depth (AOD)(Color bar-AOD, high over in the north NYC;
674 gray-cloud cover, a lot of clouds in north US and Canada) and CALIPSO ground track (red line) nearby
675 the CCNY-site (red square), (b) attenuated backscatter, depolarization ratio and color ratio, (c) aerosol
676 extinction and (d) aerosol subtypes (arrow in x-axis: nearest CCNY) on May 12, 2016.

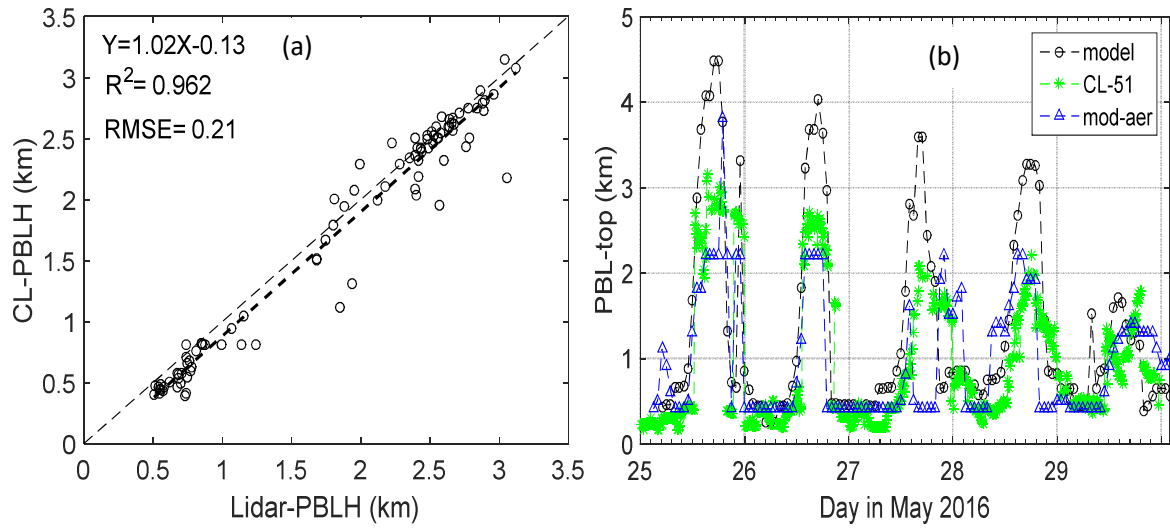


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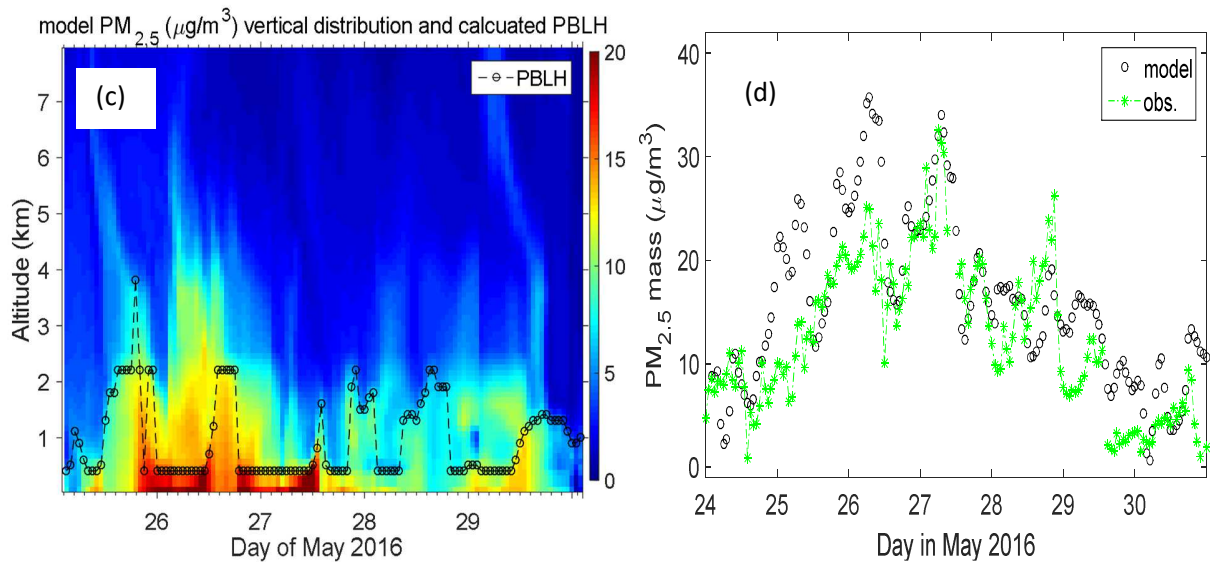
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679 Fig. 13. (a) CALIPSO track (red line) nearby the CCNY site (red square), (b) attenuated backscatter,
 680 depolarization ratio, (c) aerosol subtypes (arrow in x-axis: nearest CCNY), and (d) aerosol extinction
 681 coefficient profiles nearby CCNY-lidar on May 26, 2016.

682



683



684

685 Fig. 14. Comparisons between the model and observations at CCNY site. (a) PBLH correlation
 686 between lidar and ceilometer, (b) PBL-top or PBLH from the model and observation, “model”:
 687 PBLH from the NAM meteorological-based value; “CL-51”: ceilometer-measurement; “mod-aer”:
 688 PBLH estimate from the model $PM_{2.5}$ profile; (c) $PM_{2.5}$ profiles from the model, and (d) $PM_{2.5}$
 689 comparison between the model and ground observation.

690

Dense aloft smoke plume intrusion and mixing down into PBL are observed. A coincident increase of ground $PM_{2.5}$ in the NYC urban/rural area and the enhancement of OC, EC and K^+ indicate the big impacts of transported smoke on the local air quality.

