

1 Aerosol optical characteristics and their vertical distributions under 2 enhanced haze pollution events: effect of the regional transport of 3 different aerosol types over eastern China

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18 **Abstract.** The climatological variation of aerosol properties and the planetary boundary layer (PBL) during 2013–2015 over
19 the Yangtze River Delta (YRD) region were investigated by employing ground-based Microwave Pulse Lidar (MPL) and
20 CE-318 sun-photometer observations. Combining MODIS and CALIPSO satellite products, enhanced haze pollution events
21 affected by different types of aerosol over the YRD region were analyzed through vertical structures, spatial distributions,
22 backward trajectories, and the Potential Source Contribution Function (PSCF) model. The results show that aerosols in the
23 YRD are dominated by fine-mode particles, except in March. The aerosol optical depth (AOD) in June and September is
24 higher due to high single scattering albedo (SSA) from hygroscopic growth, but is lower in July and August due to wet
25 deposition from precipitation. The PBL height (PBLH) is greater (means ranging from 1.23 to 1.84 km) and more variable in
26 the warmer months of March to August, due to the stronger diurnal cycle and exchange of heat. Northern fine-mode
27 pollutants are brought to the YRD at a height of 1.5 km. The SSA increases blocking the radiation to the surface, and cooling
28 the surface, thereby weakening turbulence, lowering the PBL, and in turn accelerating the accumulation of pollutants,

29 creating a feedback to the cooling effect. Originated from the deserts in Xinjiang and Inner Mongolia, long-range transported
30 dust masses are seen at heights of about 2 km over the YRD region with an SSA_{440nm} below 0.84, which heat air and upraise
31 PBL, accelerating the diffusion of dust particles. Regional transport from biomass burning spots to the south of the YRD
32 region bring mixed aerosol particles at a height below 1.5 km, resulting in an SSA_{440nm} below 0.89. During the winter, the
33 accumulation of local emission layer is facilitated by stable weather condition, staying within the PBL even below 0.5 km.

34 1. Introduction

35 Aerosol particles influence Earth's radiation budget and play a significant role in global and regional climate change
36 (Hansen et al., 1997;Che et al., 2015a;Zhang et al., 1998;Li et al., 2016). Despite numerous studies on aerosols, there is still
37 much uncertainty surrounding aerosol optical properties in relation to current assessments and predictions of global climatic
38 change (Ipcc, 2007;Stocker et al., 2013). The frequent occurrence of regional pollution demands worldwide attention
39 because of the serious consequences it can have on visibility and human health (Wu et al., 2012;Sun et al., 2016;Chan and
40 Yao, 2008;Bi et al., 2015).

41 The planetary boundary layer (PBL) is a critical component of Earth's climate system (Medeiros et al., 2005). The PBL
42 is directly coupled with the land surface, and its height (PBLH) plays a significant role in determining the vertical
43 distributions of aerosol particles (Deardorff, 1972). Based on Tang et al. (2016)'s research, the atmospheric mixing layer
44 provide useful empirical information for improving meteorological and atmospheric chemistry models and the forecasting
45 and warning of air pollution. The higher the accumulation of ambient aerosols, the less solar radiation reaches the surface,
46 which will further restrict the development of the PBL, thus compounding the air pollution near the surface during prolonged
47 stagnant weather conditions (Gao et al., 2015;Petäjä et al., 2016;Leng et al., 2015). Lidar measures the intensity of
48 backscattered light as a function of distance from the instrument. The change in backscatter across the top of the boundary
49 layer provides a convenient means of determining the PBLH. Long-term, continuous PBLHs are provided by Micro Pulse
50 Lidar (MPL) observations to ascertain seasonal variations (Ku et al.;Lewis et al., 2013).

51 To analyze the spatiotemporal distribution of aerosol optical properties, multiple measurements from diverse angles

52 have been conducted by researchers worldwide. Satellite remote sensing and ground-based observations are two approved
53 ways of monitoring the long-term variation of Earth's aerosol properties (Holben et al., 2001;Che et al., 2015a;Tao et al.,
54 2014) used ground-based Cimel sun-photometers and found a decreasing trend in aerosol optical depth (AOD) from 2006 to
55 2009, but an increase of ~ 0.03 from 2009 to 2013 in China. Chauvigné et al. (2016) compared aerosol extinction with
56 continuous high-altitude near-surface *in situ* measurements and low-altitude ground-based remote sensing atmospheric
57 column measurements over a one-year period at the Puy de Dôme station, utilizing several *in situ* instruments such as the
58 Cimel sun-photometer for the whole column, and lidar for vertical multi-altitude. Wu et al. (2017) used a variety of
59 ground-based instruments and satellite sensors, including Moderate Resolution Imaging Spectroradiometer (MODIS),
60 Atmospheric Infrared Sounder (AIRS), Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO), and
61 Ozone Monitoring Instrument (OMI) products, to detail an integrated observation of an agricultural biomass burning episode
62 in Nanjing, China. It is significant to estimate the leading pollution contributor using a combination of remote sensing,
63 satellite data (Chen et al., 2017b). Along with the observations, trajectory models (e.g. HYSPLIT) for air mass tracking and
64 potential source apportionment have been applied to analyze the origins of aerosols (Wang et al., 2006b).

65 The Yangtze River Delta (YRD) region is a key economic and cultural hub in China, with progressive development
66 resulting in further particle emissions from industry and other anthropogenic activities. Many studies have been conducted
67 on the aerosol optical properties and their variations in the cities of the YRD, such as Shanghai, Nanjing, Lin'an, and Taihu
68 (Xia et al., 2007;Pan et al., 2010;He et al., 2012;Ding et al., 2013;Liu et al., 2015;Xing et al., 2017). Particularly in
69 Hangzhou, the capital city of Zhejiang province, some researches have focused on determining the seasonal variation of
70 aerosol properties, chemical compositions, and PM concentrations (Qi, 2016;Ming et al., 2017;Xiao et al., 2011), or
71 single-case analyses on features of one pollution process (Chen et al., 2012;Fu et al., 2014;Li et al., 2015). However,
72 research into the sources of pollutants in the YRD region is still needed to understand the mechanisms underlying haze
73 pollution. Among these studies, there is a lack of long-term analysis of aerosols and their sources. In addition, single-case
74 analysis is insufficiently representative for a site that experiences frequent haze occurrence. There is a lack of comprehensive
75 studies in process analyzing from multiple aspects. In particular, studies in the YRD urban area have been based almost
76 exclusively on data from *in situ* measurements, or on whole-column measurements that do not resolve the vertical

77 distribution. This can be obtained from satellite monitoring or ground-based lidar measurements. The retrieved time–height
78 cross section of the extinction coefficient can reflect the vertical distribution and structure of the aerosol layer continuously,
79 which will contribute to optimizing the satellite retrieval algorithm, and to verifying and improving the results from both
80 satellites and models.

81 In this study, multiple data sources are analyzed to further understand the mechanisms underlying haze pollution
82 affected by different aerosol transportation over the YRD region in eastern China. Hangzhou is selected as a representative
83 site. The remainder of the paper is organized as follows: in section 2, the methods and data are presented; in section 3,
84 seasonal aerosol optical properties, PM concentrations, and PBLHs from 2013 to 2015 are analyzed to illustrate the general
85 aerosol characteristics over the urban area of the YRD; in section 4, four typical haze pollution episodes affected by different
86 types of aerosol transported to the urban area of the YRD are analyzed with ground-based sun-photometer and MPL data for
87 the obtaining optical properties and vertical distribution, satellite data from MODIS and CALIPSO for confirming the
88 observed results, PM concentration data to build an overview of the spatial distribution of pollutants in eastern China, and
89 reanalysis data for depicting the wind fields and calculating backward trajectories and potential source contributions; and in
90 section 5, a brief summary is given.

91 **2. Methods and data**

92 **2.1 Ground-based sun-photometer and lidar measurements**

93 In this study, the aerosol optical properties were measured in Hangzhou (30°14'N, 120°10'E, 41.7 m above sea level).
94 This site lies in the West Lake scenic area, which is a commercial and residential area in the southern part of the city (Fig. 1).
95 The AOD at 440, 670, 870, and 1020 nm was measured from January 2013 to December 2015 using a CE-318
96 sun-photometer with a 1.2° full field-of-view (Holben et al., 1998). The validated AOD data presented in this article were
97 obtained using the ASTPwin software developed by Cimel Co. Ltd. (Che et al., 2013) for level 1.5 AOD (cloud-screened).
98 Aerosol optical properties were processed using the method of Dubovik et al. (2000) and Dubovik and King (2000), based
99 on the almucantar measurement data of the Cimel sun-photometer. The Cimel sky radiance measurements taken at 440, 670,

100 870, and 1020 nm, in conjunction with direct sun AOD data collected at same wavelengths, were used to retrieve volume
101 aerosol size distributions ($dV/d\ln(r)$ in the size range 0.05 to 15 μm), single scattering albedo (SSA) values, Ångström
102 exponent (AE), absorption AOD (AAOD), and volume concentrations of fine- and coarse-mode aerosols (Eck et al.,
103 2010;Che et al., 2014). Only SSA and AAOD results with $\text{AOD}_{440\text{nm}} > 0.40$ were used due to the high uncertainties inherent
104 in lower AOD values (Che et al., 2015b).

105 The ground-based lidar instrument used was the MPL installed at the Hangzhou National Reference Climatology
106 Station. The MPL produced by Sigma Space Corporation is an elastic back-scattering lidar, equipped with a 532-nm
107 wavelength laser launcher. In this study, aerosol measurements were obtained with a 30-m range resolution and a 30-s
108 accumulation time, and a blind area of detection existed ranging from 200 to 300 m. The aerosol extinction coefficient
109 profiles, PBLH, and AOD were retrieved during 2013–2015 using the algorithm of (Fernald, 1984), which considers the
110 atmosphere to be comprised of aerosols and air molecules (Liu et al., 2016).

111 2.2 MODIS and CALIPSO data

112 The MODIS instrument operates on both the Terra and Aqua satellites. In this study, to illustrate the spatial distribution
113 of AOD, we used the daily AOD combined dark target and deep blue data at 550 nm for land and ocean from MODIS Level
114 2 Atmosphere Products, during 9–15 October 2015 (Levy et al., 2007a;Levy et al., 2007b). Global MODIS hot spots detected
115 during 6–13 August 2013 in China were obtained from the Fire Information for Resource Management System for fire
116 locations (<https://firms.modaps.eosdis.nasa.gov/>).

117 The products of CALIPSO were used for this study, and these are available at the NASA Langley Research Center
118 (<https://www-calipso.larc.nasa.gov/>). The primary instrument carried on the CALIPSO satellite is the Cloud-Aerosol Lidar
119 with Orthogonal Polarization (CALIOP). In this work, we selected the V4.10 CALIOP Level 2 data products to acquire the
120 images of vertical feature masks and aerosol subtypes (Omar et al., 2009;Winker et al., 2009;Vaughan et al., 2004).

121 **2.3 Reanalysis data for the PBLH and wind field**

122 The reanalysis data for the PBLH and wind fields were downloaded from the European Centre for Medium-Range
123 Weather Forecasts (ECMWF), ERA-Interim (<http://apps.ecmwf.int/datasets/>). The monthly-average PBLH values were from
124 2013 to 2015, and the daily wind fields were analyzed at different pressure levels with a spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$.

125 **2.4 Surface *in situ* PM_{2.5} and PM₁₀ data**

126 In 2012, the China National Environmental Monitoring Center began to make hourly observations of PM_{2.5}, PM₁₀, and
127 other atmospheric pollutants in 367 major cities in China (<http://106.37.208.233:20035/>). In this work, the hourly data were
128 processed to obtain the daily and monthly PM_{2.5} and PM₁₀ concentration results during 2013–2015.

129 **2.5 Backward trajectory, PSCF and CWT analysis**

130 Three-dimensional 72-h air mass backward trajectories at multiple altitudes over Hangzhou every 6 h (0000, 0600, 1200,
131 and 1800 UTC) were calculated using the NCEP FNL (National Centers for Environmental Prediction Final Analyses)
132 archive data. The software TrajStat (<http://www.meteothinker.com/products/trajstat.html>) (Ngan et al., 2015) was used. The
133 cluster methods of air mass backward trajectories from Sirois and Bottenheim (1995) were applied to confirm the major
134 transport pathways.

135 To determine the pollutants' source locations and prevailing transport pathways, backward trajectories combined with
136 PM concentrations [referred to as the Potential Source Contribution Function (PSCF) model and Concentration Weighted
137 Trajectory (CWT) model] were used. A weighting function was applied to better reflect the uncertainty in the values, and this
138 is referred to as WPSCF and WCWT in each model, respectively (Polissar et al., 1999; Hsu et al., 2003; Xin, 2016; Seibert et
139 al., 1994). The specifics of the two models' parameters and algorithms have been described in detail by Wang et al. (2006b).

140 3. Results and discussion

141 3.1. Variation in PM_{2.5} and PM₁₀ mass concentration

142 Figure 2a shows the monthly variation in mass concentrations of PM_{2.5} and PM₁₀ and the proportion of PM_{2.5} to PM₁₀
143 during 2013–2015 in Hangzhou. It can be seen that mass concentrations of PM₁₀ and PM_{2.5} in the YRD region are distinctly
144 higher in January (129.39 µg/m³), November (96.32 µg/m³), and December (119.16 µg/m³) than in other months, while the
145 PM_{2.5}:PM₁₀ ratio during these months plus February also exceeds other months, ranging from 76.1% to 86.6%. Meanwhile,
146 the lowest concentrations occur in July and August, which can result from wet deposition due to the abundant precipitation at
147 this time of year (Jacob and Winner, 2009). These results verify the findings of Ming et al. (2017), Cao et al. (2009), and
148 Wang et al. (2006a), that the highest and lowest PM_{2.5} mass concentrations occur respectively during the winter and summer
149 in the YRD region. For the other months, concentrations of PM₁₀ are high but the PM_{2.5} accounts for less than 65% during
150 March to May.

151 3.2 Variation in aerosol optical properties

152 Figure 2b depicts the monthly means of the AE between 440 nm and 870 nm, and the AOD at four different
153 wavelengths (440, 670, 870, and 1020 nm) in Hangzhou during 2013–2015, retrieved from the CE-318 sun-photometer. The
154 monthly variation trends of AOD means at the four wavelengths are consistent, and the values of AOD_{440nm} are the highest
155 among them. The averaged AOD_{440nm} values during January to May are about 0.8, and the AOD values at the four
156 wavelengths reach their maxima in June at 1.20 ± 0.55 , 0.76 ± 0.37 , 0.54 ± 0.25 , and 0.43 ± 0.20 , respectively. After June,
157 AODs reduce to less than 0.8, and in August the mean values reach their minima at 0.59 ± 0.34 , 0.32 ± 0.21 , 0.23 ± 0.14 , and
158 0.18 ± 0.10 , respectively. In September, the values increase again to 1.07 ± 0.54 , 0.63 ± 0.38 , 0.43 ± 0.26 , and $0.32 \pm$
159 0.20 , respectively, after which the AOD_{440nm} means decrease below 0.8 during October to December.

160 In general, AODs in June and September are relatively high, yet in July and August they are lower than in other months.
161 The results of July and August agree with Qi (2016) that the lowest AOD appears in summer, with a mean value of $0.72 \pm$

162 0.58 at 440 nm in Hangzhou. According to Che et al. (2014), the relatively low PM mass concentrations may result from the
163 substantially increased precipitation in the YRD region in summer (including July and August) due to the summer monsoon,
164 which can decrease the concentration of atmospheric aerosols through wet deposition. This pattern has also been observed in
165 Taihu and Lin'an (Pan et al., 2010). Because the subtropical anticyclone begins to move north and precipitation decreases
166 from the end of August to the beginning of September, the AOD increases in September. Nevertheless, the peak in the
167 monthly AOD means in Pudong occurs in June, which may derive from the stable weather caused by the lack of strong
168 winds (Duan and Mao, 2007).

169 The monthly mean AEs ($AE_{440\text{nm}-870\text{nm}}$) vary almost inversely to the AOD, and are all greater than 0.8, which indicates
170 that the AODs are dominated by fine particles in the YRD region during 2013–2015 (Qi, 2016; Eck et al., 2012). When
171 $AE_{440\text{nm}-870\text{nm}}$ is greater than 0.8 at East Asian sites, fine-mode aerosols emitted from population centers in East Asia
172 dominate the aerosol optical properties even on spring dust days, during which pollution aerosols are mixed with
173 coarse-mode particles. Comparing the bars' heights in the figure, the minimum monthly averaged $AE_{440\text{nm}-870\text{nm}}$ occurs in
174 March (1.16 ± 0.24), and the maximum in September (1.41 ± 0.25). This shows the influence of long-distance dust transport
175 from the desert districts located in northern and northwestern China (Zhang and Li, 2012; Gong et al., 2003), which can have
176 an influence on the YRD region.

177 Figure 2c shows the monthly volume size distributions of aerosols during 2013–2015 in Hangzhou. In general, the size
178 distributions show a bimodal logarithm normal structure: a fine mode with radius $< 0.6 \mu\text{m}$ and coarse mode with radius $>$
179 $0.6 \mu\text{m}$ (Dubovik et al., 2002). Nevertheless, in June and September, the distribution appears tri-modal, indicating a
180 tendency for hygroscopic growth in fine-mode particles in the YRD region (Pilat and Charlson, 1966). In June and
181 September, the fine modes reach maximum peaks at a radius of $0.34 \mu\text{m}$, while other months' peaks occur at a radius of 0.15
182 μm . As for the coarse mode, March, April, November, and December peaks occur at a radius of $2.24 \mu\text{m}$, while the peak
183 radius during the other months is larger than $2.24 \mu\text{m}$. Because of the dust transport, coarse modes dominate in March, while
184 in July, August, and September the coarse mode only contributes a small fraction. Conversely, the fine particles play a
185 leading role in the other months, particularly in June and September. This situation may arise due to the mixed influence of
186 hygroscopic growth, coagulation growth, and different species of aerosols emitted from various sources (Li et al., 2007; Zhu

187 et al., 2014;Che et al., 2014). Hygroscopic properties of ambient particles in urban Hangzhou are mainly a function of their
188 size and chemical composition (Zhang et al., 2011). Since the highest AOD values are seen in June (but not in July and
189 August), it is suggested that the hygroscopic growth in the fine mode leads to enhanced scattering, which significantly raises
190 the AOD values in the YRD region.

191 The SSA is defined as the ratio of the scattering coefficient to the extinction coefficient, and is one of the key
192 parameters used in assessing the radiation effect on climate change (Jacobson, 2000). Highly absorbing particles, including
193 urban-industrial aerosols, have a warming effect, while low absorbing particles such as dust can have a cooling effect
194 (Dubovik et al., 2002). Figure 2d shows the monthly variation of SSA at the 440-, 670-, 870-, and 1020-nm wavelengths.
195 From September to February, the SSAs at the four wavelengths are close and relatively high. However, over the warmer
196 months of March to August, the values decrease and the difference between the wavelengths becomes greater, particularly in
197 July and August with low SSA, which may imply that the low-SSA particles are more sensitive to the wavelength variability.
198 The minimum SSAs occur in August at 0.869 ± 0.095 , 0.839 ± 0.114 , 0.793 ± 0.149 , and 0.762 ± 0.169 , respectively; the
199 maximum is in September, with the highest value of 0.963 ± 0.033 at 440 nm. As a result, the aerosols in the YRD region
200 during August tend to be absorbing, but during September the hygroscopic aerosol particles become more capable of light
201 scattering through condensation and evaporation of water vapor (Adachi et al., 2015), which may result from the variability
202 of the aerosols' composition and sources in the ambient atmosphere. According to the studies by Qi (2016) and Xia et al.
203 (2007), the absence of a heating period in southern China in winter causes higher SSA than in northern cities such as Beijing,
204 where biomass burning can emit black and organic carbon. According to the results of monthly AOD and the volume size
205 distribution presented in this article, the high level of AOD in September may be due to the strong scattering ability provided
206 by the prominent hygroscopic growth.

207 **3.3 Variation in the PBLH measured by lidar**

208 In this study, by utilizing the backscatter signal at 532 nm with an MPL, the PBLHs from 2013 to 2015 at Hangzhou
209 were retrieved at a 10-min temporal and 30-m spatial resolution. To verify the reliability of these PBLHs, we used the

210 monthly averaged reanalysis data of the PBL from ERA-Interim to obtain a linear fit with the PBLHs from MPL (after
211 computing the monthly average). The Pearson's correlation coefficient between these two monthly averaged series is 0.88,
212 which passes the significance test at the $p = 0.1$ level. Despite the strong correlation between the reanalysis- and the
213 MPL-derived PBLH, the values obtained from the reanalysis data are significantly lower than those from the MPL. It is
214 suggested that improvements are needed in the reanalysis data.

215 Since the PBLHs retrieved from the MPL are reliable, we adopt them to examine the monthly variation characteristics
216 in the YRD region during 2013–2015. Figure 3 shows the monthly averaged height of the PBL and its 5th and 95th
217 percentile box plots during 2013–2015 in Hangzhou. Due to the blind detection area of the MPL, the minimum value is
218 about 300 m above the surface. During the warmer months (including March to August), the monthly means of the PBLH are
219 above 1 km, and the range between the 5th and 95th percentiles is greater than 1 km, varying from above 0.5 km to about 2
220 km. Conversely, mean PBLHs in the colder months are almost below 1 km and the range between the 5th and 95th
221 percentiles is half that in the warmer months, ranging from about 0.5 km to below 1.5 km. The wider range in the warmer
222 months may result from a stronger diurnal cycle (Liu and Liang, 2010) and daily exchange of heat and mass in the PBL
223 (Medeiros et al., 2005). These characteristics help to explain the higher *in situ* PM mass concentrations in the winter months
224 than in summer discussed above.

225 **3.4 Case analysis of pollution sources**

226 For a further explanation of the monthly variation in aerosol optical properties, and to examine the variety of pollutant
227 sources affecting the enhanced haze pollution events in the YRD region, four cases with different transportation categories
228 are discussed here, combining ground-based observation, remote sensing, reanalysis, and satellite data, to build a
229 comprehensive analysis (Fig. S6 in the supplementary material).

230 **3.4.1 Pollutants transported from polluted areas in northern China**

231 A few episodes of pollutants transported from northern China were detected by the MPL, as illustrated in Fig. 4a, which
232 shows the time–height cross section of extinction coefficients in Hangzhou. The aerosol vertical distributions were further

233 assessed by extracting profiles of aerosol extinction coefficient from the MPL on a daily basis during this period (Fig. 5). On
234 the first day, 9 October 2015, the thin external aerosol layer is seen around 1 km during nighttime, when the maximum
235 extinction coefficient is more than 0.6 km^{-1} , and this is separate from the lower local pollutants layer. During the following
236 three days, the external aerosol masses are around 1 km at nighttime and above 1.5 km during the daytime, and appear to be
237 multilayer in structure. Meanwhile, the daily averaged PBLH is around 1.2 km and 1.5 km on 11 October and 12 October
238 respectively, sitting above the aerosol layer at nighttime and beneath it during the day (Fig. 6). With the PBL pushing down,
239 the aerosol layer begins to descend from the afternoon of 12 October, continues to descend to below 1 km until 15 October,
240 and then mixes downwards into the local emissions aerosol layer. Concurrently, the peak extinction coefficients vary from
241 0.2 to 0.5 km^{-1} , reaching their maxima when the external and local pollutants mix together in the PBL. To verify the
242 significance of the PBL in this process, the Pearson's correlation coefficient between the daily averaged PBLH from the
243 MPL and the daily mass concentration of $\text{PM}_{2.5}$ during 9–15 October was computed. The result appears negative, at -0.88
244 (passing the significance test at the $p = 0.1$ level), which indicates that the descending PBL accelerates the accumulation of
245 PM mass from the two aerosol layers in it.

246 Aerosol optical properties from the CE-318 retrieval data are exhibited in Fig. 6. According to the pattern of volume
247 size distribution during 11–15 October 2015, the fine mode dominates, which can be referred to the volume of both modes as
248 well, and the peak radius growth from 0.11 to $0.15 \mu\text{m}$ is attributable to the hygroscopic growth, which may help strengthen
249 the scattering ability of the particles (SSA is up to 0.96 at 440 nm). The daily averaged AODs at 440 nm are all above 0.65
250 during this process. On 15 October, the AOD is close to 1.00 at 440 nm when the two aerosol layers mix. The SSAs at all
251 four wavelengths continue to increase from 11 to 14 October, and decrease on 15 October. The values at 440 nm vary from
252 0.95 to around 0.97 . The AAODs show a corresponding variability trend with AOD and the volume of the coarse mode,
253 which dominates the absorbing features. The maximum AAOD at 440 nm is 0.056 on 12 October, and the minimum is 0.026
254 on 14 October.

255 Due to the continuous aerosol-transport signals detected by the MPL during 9–15 October 2015, trajectories over this
256 period were grouped into three clusters, as depicted in Fig. 7a-1. Cluster-1 contributes the maximum proportion of 50.00%
257 and comes from northern China, via Hebei, western Shangdong and Jiangsu province, travelling southwards to the YRD

258 region. This cluster relates to the highest PM_{2.5} concentration ($81.05 \pm 28.18 \mu\text{g}/\text{m}^3$). Cluster-2 travels on the lowest and
259 fastest stemming from north of Anhui province, accounts for 30.95%, and correlates with the PM_{2.5} concentration of $74.45 \pm$
260 $30.17 \mu\text{g}/\text{m}^3$. The highest air mass trajectories associated with cluster-3 account for 19.05% and originate from the eastern
261 edge of Xinjiang at an altitude above 5000 m, also passing through Henan and Anhui province to Hangzhou.

262 The WPSCF reveals the spatial distribution of the probabilities of the potential PM_{2.5} sources obtained by the HYSPLIT
263 model and PM_{2.5} concentrations. As shown in Fig. 7a-2, the most likely source areas with WPSCF values for PM_{2.5} cover
264 most of Henan, Anhui, Jiangsu, and Shangdong provinces, in which the WPSCF values are over 0.8. Figure 7a-3 shows the
265 spatial distribution of the WCWT value, which provides information regarding the relative contribution of source regions
266 potentially affecting PM_{2.5} ($> 35 \mu\text{g}/\text{m}^3$) in Hangzhou, similar to the WPSCF pattern. The northern part of Henan and the
267 western Shangdong region (including the south of the Beijing–Tianjin districts), with rather high WCWT values ($> 75 \mu\text{g}$
268 m^{-3}), is recognized as the area contributing the most PM_{2.5}. Combining the daily spatial distribution of PM_{2.5} concentrations
269 with AOD from MODIS retrieval data over eastern China (Figs. S1–S2 in the supplement), districts where the air mass
270 trajectories pass intensively can be seen to have relatively high pollution levels. These include Henan, Hebei, Anhui, and
271 Jiangsu provinces, situated in the North China Plain, which has been recognized as another center of high PM_{2.5}
272 concentrations (Zhang et al., 2012; Zhang et al., 2013). Besides, the pollution scope from north to the YRD region is wider
273 and more serious day by day.

274 Furthermore, daily wind fields at 850 hPa (height of about 1.5 km, at which the transport is detected by the MPL) from
275 ECMWF (Fig. S1 in the supplementary material) verify the northwest wind prevailing over eastern China during this case,
276 which brought the pollutants from northern China to the YRD region. The wind speed gradually declines, benefiting the
277 formation of stable conditions during the last few days. Note that CALIPSO’s acquisition of the aerosol vertical distribution
278 on 14 October is consistent with the above results that an aerosol layer under 2 km exists over Hangzhou and the areas
279 further north (where intensive trajectories passed over), and is identified as “polluted continental” or “polluted dust” (Fig.
280 8a). Under certain circumstances, however, smoke aerosols can be misidentified as “polluted dust” aerosols (Xia et al.,
281 2013).

282 Although a dry, clean northwestern airstream prevails in eastern China during winter (Tao et al., 2012), northern winds

283 near the surface may not be strong enough to blow the pollutants away from eastern China, but transport the particle
284 pollutants to the central part of the North China Plain. The continuing northwesterly winds blow particle pollutants to the
285 YRD region (Li et al., 2014;Ming et al., 2017). A similar transport mechanism occurs in this case during 9–15 October 2015.
286 Northerly pollutant transport at 850 hPa primarily results in an increase in PM_{2.5} mass concentration, which is facilitated by
287 the occurrence of the descending external aerosol layers, pushed down by the PBL. Pollutants from the North China Plain
288 are dominated by fine particles, which tend towards hygroscopic growth with adequate requisite moisture in the YRD region,
289 increasing the SSA over Hangzhou. As a result, the increasing scattering aerosols transported from haze areas of North China
290 block the solar radiation to the surface, creating a cooling effect. If that is the case, turbulence will correspondingly weaken,
291 and the PBLH will decrease, facilitating the accumulation of pollutants at lower heights. The accumulation of air pollutants
292 can then intensify the scattering ability, forming a positive feedback mechanism.

293 3.4.2 Pollutants transported from desert areas

294 Dust events in spring contribute to high AODs, particularly in northern and eastern China (Che et al., 2015b). They are
295 transported over long distances and their influence can be identified on regional even global scales (Chin et al., 2006;Uno et
296 al., 2009;Chen et al., 2017a;SiYu et al., 2017). For instance, Asian dusts are frequently observed in spring and result in air
297 pollution in the downwind areas of East Asia (Kim et al., 2010;Sakai et al., 2002;Sakai et al., 2003;Liu et al., 2011;Li, 2015),
298 such as the YRD region including Nanjing and Shanghai (Yong et al., 2015;Huang et al., 2013).

299 A series of transported aerosol masses from the northwestern desert area over the YRD region were monitored by the
300 MPL and are seen in the time–height cross section of extinction coefficients at 532 nm during 5–9 March 2013 in Fig. 4b.
301 Throughout this period, the volume size distributions and particle volumes are strongly dominated by the coarse mode (Fig.
302 10). The peak radius of the fine mode is around 0.11 μm , and 1.3 μm for the coarse mode. The AOD on 7 March is around
303 0.88, when the transported aerosol layer is most intense, while the value on 6 March is just 0.47 at 440 nm. Early on 5 March,
304 the intensive layer is detected at 1.5 km and the peak extinction coefficient is around 0.65 km^{-1} , according to the profiles
305 shown in Fig. 9. The SSAs retrieved from the CE-318 are 0.84, 0.91, 0.91 and 0.90 at 440, 670, 870 and 1020 nm,
306 respectively, which are lower than those for the northern transported particles. The corresponding AAODs are 0.13, 0.05,

307 0.04 and 0.04, respectively, which are higher than the values during the northern transportation case. The particles in this
308 case are more strongly absorbing than the type transported from the northern polluted areas discussed in the previous section.
309 The transported mass weakens on the second day until night, when the aerosol layer lies between 1.0 and 2.5 km, with the
310 peak coefficient above 0.4 km^{-1} . Correspondingly, the volumes of both modes decrease, and this is accompanied by a drop in
311 SSA and AAOD to 0.79 and 0.1, respectively, at the 440 nm wavelength. During 7–8 March, a thickening of the external
312 aerosol layer is accompanied by a substantial increase in the volume of coarse-mode particles, coinciding with an increased
313 AAOD to 0.15 on 7 March and 0.13 on 8 March, at 440 nm. The peak extinction is about 0.2 km^{-1} above a height of 2 km.
314 On the last day of this case, no external aerosol layer is detected by the MPL (the noisy signals shown in the profiles in Fig. 9
315 may result from an instrumental malfunction), and the absorption weakens due to the reduced volume. Note that the SSAs
316 show little change during the last three days, which implies that SSA may not be sensitive to the variation of particles in the
317 dominant coarse mode. Meanwhile, the PBLH above the aerosol layer varies from 2.0 km to about 3 km continuously in this
318 case, dissipating the transported dust particles and causing the peak extinction coefficients to decrease each day.

319 From the CALIPSO L2 retrieval results of vertical feature mask and aerosol subtypes (illustrated in Fig. 8b), a 3-km
320 thick aerosol layer can be seen on 5 March 2015 in the northwestern upwind areas of the YRD, with a mixture of “dust” and
321 “polluted dust”. After this, a thin external layer is detected.

322 Assuming that the extraneous aerosol layer was dust, based on the *in situ* and satellite observations, 72-h backward
323 trajectory analysis was applied to determine the source of the dust. Since the dust aerosol layer is most intense at 2 km, the
324 start location of the trajectory calculation was set to be 2000 m. Figure 7b-1 shows three cluster trajectories changing with
325 height. The air masses associated with cluster-1, accounting for 55%, originate from the Gurbantunggut Desert lying in
326 northern Xinjiang, and move southeasterly over the Gobi Desert which is agreed to the results from Bi et al. (2017), and the
327 Badain Duren Desert located in Inner Mongolia. This trajectory is similar to the dust pathway to Xi’an described by (Wang
328 et al., 2006b). The air masses associated with cluster-2, accounting for 35%, follow parallel paths from the Kumtag Desert in
329 western Xinjiang, and pass over the Loess Plateau. The cluster-3 stems from Guangxin province at an altitude below 3000 m,
330 and does not pass over a desert area.

331 The number of trajectories in each cluster and the corresponding PM_{10} mean concentrations for all trajectories are

332 summarized in Table 1. Based on the mean PM_{10} concentrations in each cluster, clusters-1 and 2 take the most primary role
333 in bringing dust to the YRD region. Combining the distribution of WPSCF and WCWT values, the sources most likely to
334 affect the YRD are located in the deserts of northwestern China, termed the “Western High Dust Desert” (Zhang et al. (1998),
335 which include the Gurbantunggut Desert, the Gobi Desert, and the Badain Duran Desert, covered by cluster-1. From the
336 intensive trajectories’ pathways, Henan and Anhui provinces also appear to be highly likely source regions located upwind of
337 the YRD, which just corresponds to the vertical feature mask from the CALIPSO L2 products that show a mixed layer of
338 “dust” and “polluted dust” in Anhui province. The WCWT results make the potential sources explicit. The Gobi Desert, and
339 Henan and Anhui provinces are prominent sources of dust transported to Hangzhou.

340 The daily spatial distributions of PM_{10} concentrations and wind fields at 750 hPa in eastern China during 5–9 March
341 2013 are shown in Fig. S3 in the supplementary material. The *in situ* mean PM_{10} concentration on 5 March is between 200
342 and 250 $\mu\text{g}/\text{m}^3$, while for the remaining days it is between 150 and 200 $\mu\text{g}/\text{m}^3$ in Hangzhou. The wind fields at 750 hPa show
343 northerly and northwesterly winds during 5–8 March 2013, consistent with the primary trajectories to Hangzhou at a height
344 of 2 km. The second day, 6 March, shows the lowest wind speed, coinciding with minimal horizontal transport during that
345 day, as can be seen in the time–height cross section of the MPL extinction coefficient (Fig. 4b). The wind speed over the next
346 two days increases, as does the transport. On the last day, the wind direction turns southwesterly at 750 hPa, which may
347 bring the clean air from the sea to Hangzhou. The wind direction on 9 March is consistent with the existence of cluster-3,
348 which carries little PM_{10} mass and only accounts for 10% among the trajectories.

349 In spring, the long-range transport of dust aerosols from desert regions not only has an impact on northwestern cities
350 such as Xi’an (Wang et al., 2006b), or cities in the north of eastern China such as Beijing, Xianghe, and Shenyang (Jinyuan
351 et al., 2010; Zhang et al., 1998), but also on cities in the YRD region such as Nanjing, Shanghai (Zhang et al., 2016), and
352 Hangzhou. When transported dust particles dominate in the ambient atmosphere over the YRD region, the aerosol optical
353 properties show a relatively high level of absorptive features, much stronger than in aerosols transported from the north.
354 These particles mostly come from moving air masses loaded with dust particles from desert regions located in northwestern
355 China. During the days dust transport enhanced, absorptive dust particles increase AOD and AAOD over Hangzhou, and heat
356 the atmosphere to facilitate the turbulence leading the PBL lifting in the same day, which will accelerate the diffusion in the

357 following days.

358 **3.4.3 Pollutants transported from biomass burning areas**

359 Since the lowest values of SSA occur in August, the aerosol particles over Hangzhou exhibit strong absorption
360 characteristics in that month. Figure 4c illustrates the time–height cross section of extinction coefficient retrieved from the
361 MPL during the transportation of biomass-burning emissions to Hangzhou, and shows little contribution from local
362 emissions. In the first four days, the external aerosols are concentrated at 1.5 to 2 km and the peak extinctions are less than
363 0.2 km^{-1} , even during the daytime (Fig. 11). In the meantime, the PBLH varies from 2.45 km to 2.93 km, above the
364 transported aerosol layer. Over 10–11 August, the external fine particles are noticeably enhanced, with the PBLH decreasing
365 to 2.30 km. Consequently, the layer develops downwards, to a height of around 1 km, which may imply a tendency towards
366 sedimentation. Maximum extinctions exceed 0.2 km^{-1} , and even reach 0.3 km^{-1} during the daytime on 11 August. The
367 transports tend to die down and deposit over 12–13 August, with a higher PBL, thinner aerosol layer, and lower coefficients
368 (below 0.1 km^{-1}). The whole process of this transportation is not as strong as the northern transport process. During 6–9
369 August, AOD values (retrieved from the CE-318) are below 0.5. They increase to 0.75 when the maximum extinction occurs
370 over 11–12 August, and decrease when diffusion and sedimentation contribute. Combined with other aerosol optical
371 properties, fine-mode particles are remarkably dominant during the whole process in terms of the volume and its size
372 distribution (Fig. 12). However, the mass concentration is far less than during the northern transported pollution of 11–15
373 October 2015. In this case, the total volumes are below $0.1 \mu\text{m}^3$ for most of the days, except for the two enhanced transport
374 days in which the total volume exceeds $0.12 \mu\text{m}^3$. The SSAs keep increasing until a peak value of between 0.73 and 0.898 at
375 440 nm before the last two days, which suggests that the increase in external fine particles enhances the scattering ability.
376 The values are similar to the SSAs observed from biomass-burning emissions in the African savanna, Zambia, during 1995–
377 2000 (Dubovik et al., 2002). The AAODs are almost above 0.09 at 440 nm during this period, which is higher than those
378 seen in the northern transported pollution. It is suggested that the particles brought by these air masses are fine mode, with
379 strong absorption characteristics, which is indicative of smoke aerosol.

380 To determine the pollution source, trajectories and primary clusters are depicted in Fig. 7c-1, with the

381 backward-trajectory start locations set to 1000 m, 1500 m, and 2000 m. Considering the significant absorptive ability of
382 aerosol particles transported over Hangzhou, we apply the fire hotspot information (90% confidence) from MODIS
383 Collection 6 and VIIRS 375 m to analyze the possible source of this type of particle. It is assumed that the external particles
384 during these days were created from biomass burning, which is the major source of atmospheric light-absorbing organic
385 carbon and black carbon aerosols, contributing around 42% of the global black carbon emissions (Bond et al., 2004; Bond et
386 al., 2013; Cao et al., 2005). Combining the fire spots detected by MODIS with the trajectory analysis, the majority of
387 trajectories (accounting for 69.79%), which are associated with cluster-1, travel northeasterly to the YRD region, passing
388 over fire spots located in Guangdong, Fujian, Jiangxi, and Zhejiang provinces, with a mean PM_{2.5} concentration of $54.52 \pm$
389 $16.63 \mu\text{g}/\text{m}^3$. According to the study of (Chan, 2017), not only do the biomass burning emissions have local impacts, but they
390 can also be transported intercontinentally and influence the atmospheric system on a global scale. The air masses associated
391 with cluster-2 originate from the sea to the southeast, only passing one spot lying in southern Zhejiang with a mean PM_{2.5}
392 concentration of $39.04 \pm 10.14 \mu\text{g}/\text{m}^3$, bringing clean air from over the sea to Hangzhou during 12–13 August (Fig. S5 in the
393 supplementary material).

394 With reference to the WPSCF and WCWT results, the locations of the potential sources are almost consistent with the
395 regions where fire spots are detected or biomass burning occurs, including the eastern part of Guangdong and Jiangxi
396 provinces, western Fujian, and most of Zhejiang. The occurrence of biomass burning in these areas has been studied
397 previously (Wang and Zhang, 2008). With regard to the retrieval results of the vertical feature mask and aerosol subtype
398 from CALIPSO on 5 August (Fig. 8c), the intensive aerosol layer exists below 2 km along the CALIPSO path southwest of
399 Hangzhou. The aerosol types are mostly identified as “polluted continental” or “polluted dust” over and southwest of
400 Hangzhou, which coincides with the backward trajectories carrying pollutants from the fire spots during 6–13 August 2013.
401 As has been noted, the retrieval results from CALIPSO may misidentify the smoke aerosols as “polluted dust”, but for the
402 majority of days during this case satellite retrieval data verify the results from CALIPSO.

403 The aerosol transported from the southern biomass-burning areas to Hangzhou shows a capacity for strong light
404 absorption, and is dominated by the fine mode. The external aerosol layers observed in the vertical distribution of the
405 extinction coefficient appear around 1–2 km, with the maximum value at 0.3 km^{-1} . The backward trajectory and potential

406 source analysis determine the likely sources. Satellite retrieval results (both MODIS and CALIPSO) verify that the
407 transported aerosol originates from biomass-burning regions, and this is consistent with the aerosol optical properties
408 retrieved from the CE-318 in Hangzhou. In addition, previous studies have shown that aerosol absorption depends on the
409 mixing mechanism of soot with other aerosol components (Ackerman and Toon, 1981;Martins et al., 1998;Jacobson, 2001),
410 so the situation that occurs during 6–13 August 2013 is not unique to the YRD region.

411 **3.4.4 Pollutants emitted from local emissions**

412 While aerosols transported from the northern, southern or desert areas contribute much to the ambient atmospheric
413 pollution in the YRD region, a high concentration of aerosols with diverse properties are also emitted locally (Chen et al.,
414 2012;Zhuang et al., 2015;Liu et al., 2015).

415 Figure 4d shows a series of successive local emissions in the time–height cross section of the extinction coefficient
416 obtained by the MPL, during 27–29 December 2013. The PBLH varies from 1.16 to 0.72 km during this period. It can be
417 seen that the aerosol is composed of two layers, distributed below and above the PBL. An external aerosol layer can be seen
418 at 1–1.5 km, while the layer caused by local emissions lies below 1 km, and even below 0.5 km, due to the stable PBL that
419 exists over cold surfaces, particularly in the winter time (Medeiros et al., 2005). From the vertical profiles of the extinction
420 coefficient in Fig. 13, 27 December 2013 sees the peak extinction of the local emissions layer, at around 0.1 and 0.15 km^{-1} in
421 the nighttime and daytime, respectively. The two separated layers mix downwards on the second day, so that the profiles
422 barely show the separation between them, and the maximum extinction coefficient reaches 0.2 km^{-1} below 0.5 km. On 29
423 December, an intensification is seen both in aerosols transported at higher altitudes and in those emitted locally, and the peak
424 coefficient for the local emissions layer is close to 0.4 km^{-1} during the daytime and 0.2 km^{-1} at night.

425 The AODs retrieved from CE-318 vary from 0.4 to 0.55, according to the results presented in Fig. 14. The volume size
426 distribution during these three days shows a strong bimodal pattern, with the peak volumes at radii of 0.11 μm and 2.94 μm
427 for fine and coarse modes, respectively. Coarse particles noticeably dominate on 27 December, with an SSA of 0.89 at 440
428 nm—lower than the value at other wavelengths. Meanwhile, satellite retrieval results of vertical feature mask and aerosol
429 subtype from CALIPSO (Fig. 8d) verify the dominance of the coarse mode on 27 December. The L2 products confirm the

430 existence of an aerosol layer below 2 km, and identify it as mixed “polluted dust” and “polluted continental” over the YRD
431 region. Furthermore, another layer identified as “dust” exists between the heights of 3-5 km (beyond the detection range of
432 the MPL), composed of high-volume coarse mode particles, which may have an influence on the lower “polluted dust” layer.
433 Over the next two days, the volume of the two modes become more balanced due to enhanced transport and emissions, and
434 the SSAs at 440 nm show an inverse relationship to the values at other wavelengths. On 29 December, the lowest value of
435 SSA (440 nm), at 0.88, and the highest value of AAOD (440 nm), at 0.073, is seen. The maximum SSA value in this case is
436 higher than the peak SSA observed when desert transport occurs, but lower than the peak value seen with the strong
437 biomass-burning transport. The aerosol optical properties of mixed fine- and coarse-mode particles are different from when
438 one mode dominates.

439 With reference to the spatial distribution of PM_{2.5} mass concentration and the wind field at 10 m from the surface (Fig.
440 S5 in the supplementary material), the enhanced emissions seen in the extinction coincide with an increase in the *in situ*
441 observed PM concentration during 28–29 December. The wind is low within the PBL due to the weakening of atmospheric
442 circulation in northern China during winter (Tao et al., 2016). Consequently, the low wind speed at the surface contributes to
443 the accumulation of aerosol particles, resulting in an increase in pollutant concentrations over these days.

444 Pollutants emitted locally stay below 0.5 km during 27–29 December 2013 in the YRD region. Facilitated by the low
445 wind speed at the surface and the downward transport of external dust aerosols, aerosol optical properties show weaker
446 absorption than the strong biomass-burning aerosol, and weaker scattering than transported dust aerosols.

447 4. Conclusion

448 Based on long-term ground-based lidar and sunphotometer observations, variations in monthly averaged aerosol
449 properties and PBLHs during 2013–2015 in the YRD region are presented and discussed. Combining satellite remote sensing
450 data from CALIPSO and MODIS, backward trajectories, and PSCF and CWT analyses, four typical transportation
451 mechanisms are analyzed under enhanced haze pollution events in the YRD region.

452 For the long-term monthly variation of aerosol optical properties in the YRD region, it is found that fine and scattering

453 particles dominate, except in March. The $PM_{2.5}:PM_{10}$ ratio is high, $AE_{440nm-870nm}$ is above 0.8, and SSA exceeds 0.85 during
454 most months. Due to the low-altitude and stable PBL in the colder months (from September to February), PM mass
455 concentrations in January, November, and December are high. Conversely, the lower PM concentrations occur during the
456 warmer months (from March to August), partly due to a higher PBL attributed to a stronger diurnal cycle, but also due to
457 higher precipitation in July and August. The AOD is generally high, implying severe aerosol loading in the YRD. As for the
458 mixed type of aerosol in the YRD, the volume size distributions show a bimodal logarithmic normal structure, except in June
459 and September, when it appears tri-modal due to hygroscopic growth. The highest SSA is also seen during these months.

460 During northern aerosol transportation, particles from polluted areas in North China are seen at a height of about 1–1.5
461 km, within the PBL, and are dominated by the fine mode, with AOD_{440nm} above 0.65 and SSA_{440nm} varying from 0.95 to 0.97.
462 A rising SSA indicates an increase in transported scattering particles, which decrease the radiation to the surface and cool the
463 surface. The weakening turbulence and the declining PBL lowers the aerosol layer and pollutants accumulate at lower
464 altitudes, thereby concentrating the scattering particles and forming a positive feedback mechanism.

465 Although dust transport to the YRD is seasonal and infrequent, it does make a difference to the YRD's aerosol loading.
466 Dust air masses around 2 km are transported from the northwestern desert regions including the Gurbantunggut Desert, the
467 Gobi Desert, and the Badain Duran Desert, and diffuses vertically with the lifting PBL. A significant increase in the coarse
468 mode at high altitudes brings about high concentrations of PM_{10} and higher $AAOD_{440nm}$, illustrating the stronger absorptive
469 feature of transported dust compared to the northern pollutants transported over the YRD region. And the transported
470 absorptive dust particles heat the atmosphere, lifting PBL over YRD region to accelerate the diffusion in the PBL.

471 Biomass-burning pollutants also bring about a seasonal contribution to the aerosol loadings over the YRD region. In the
472 case of biomass-burning transportation, backward trajectories are consistent with the fire spots located in Guangdong, Fujian,
473 Jiangxi, and Zhejiang provinces south of the YRD, bringing absorptive fine particles to heights of about 1.5 km over that
474 region. The SSAs on most days are lower than 0.8 (except one day when the maximum SSA_{440nm} is 0.898), indicating that
475 the biomass-burning pollutants transported from the southwest to the YRD can strengthen the absorptive ability in the
476 ambient atmosphere over the YRD.

477 The accumulation of locally emitted pollutants is facilitated by the low wind speed at the surface (10 m). During the

478 locally emitted aerosol case, the local aerosol layer appears below 1 km, and the low surface wind provides no external
479 transport. The mixture of locally emitted aerosols and dust particles transported downwards shows weaker absorption than
480 the biomass-burning products, and weaker scattering than the transported dust aerosols, with the SSA_{440nm} varying from 0.86
481 to 0.91.

482 The haze pollution events that occur in the YRD region are not only affected by local emissions, but can also be
483 contributed to by regional transport, including pollutants from North China, dust aerosols from the northwestern deserts, and
484 strong absorptive particles from southern biomass-burning areas. Therefore, air quality control should focus not only on local
485 emissions reduction, but also on regional emissions.

486 **Acknowledgments**

487 This work was supported by grant from National Key R & D Program Pilot Projects of China (2016YFA0601901),
488 National Natural Science Foundation of China (41590874&41375153), Natural Science Foundation of Zhejiang Province
489 (LY16010006), the CAMS Basis Research Project (2016Z001 & 2014R17), the Climate Change Special Fund of CMA
490 (CCSF201504), CAMS Basic Research Project (2014R17), the Special Project of Doctoral Research supported by Liaoning
491 Provincial Meteorological Bureau (D201501), Hangzhou Science and Technology Innovative project (20150533B17).

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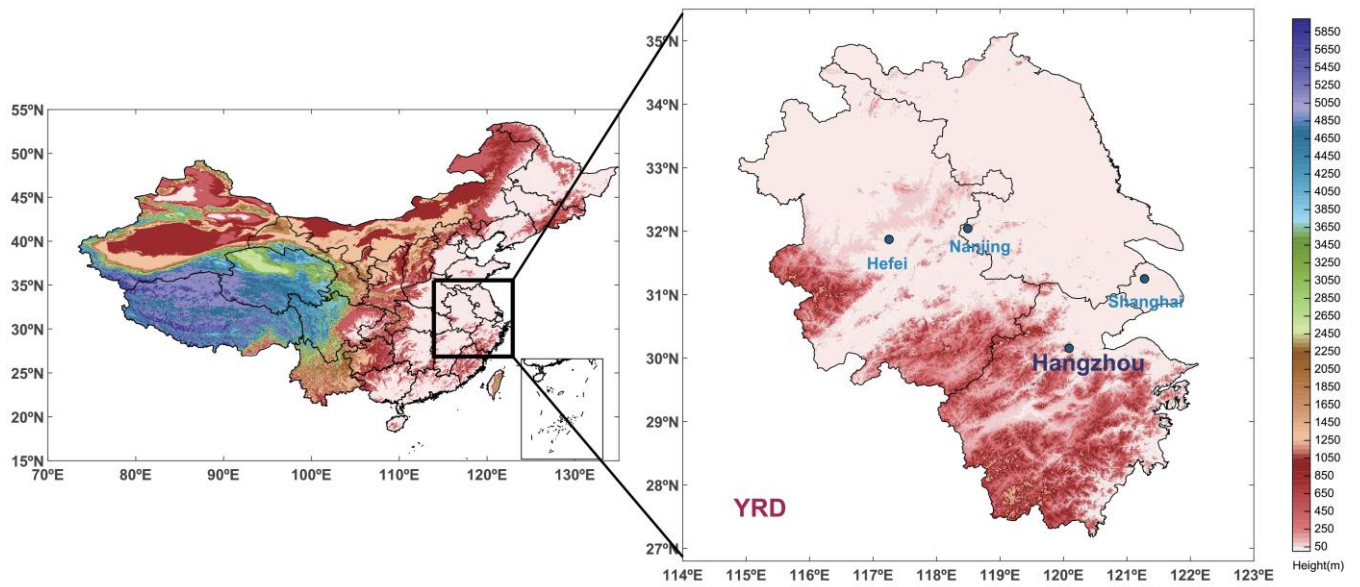
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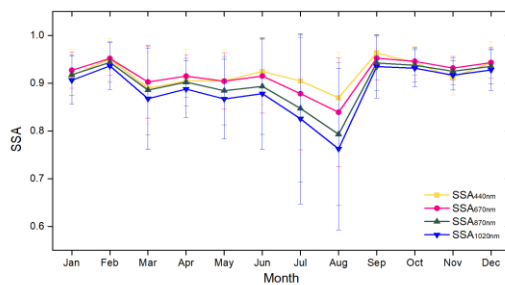
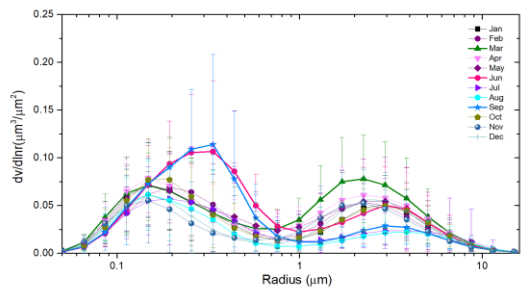
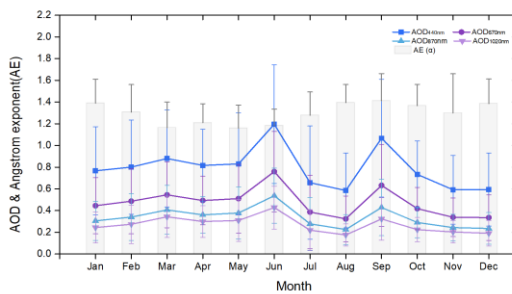
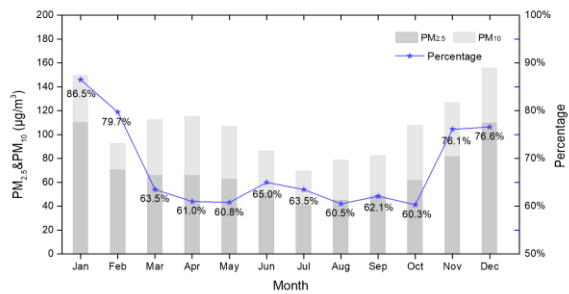
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726 **Figure 1. Terrain elevation and location of the Yangtze River Delta region in China.**

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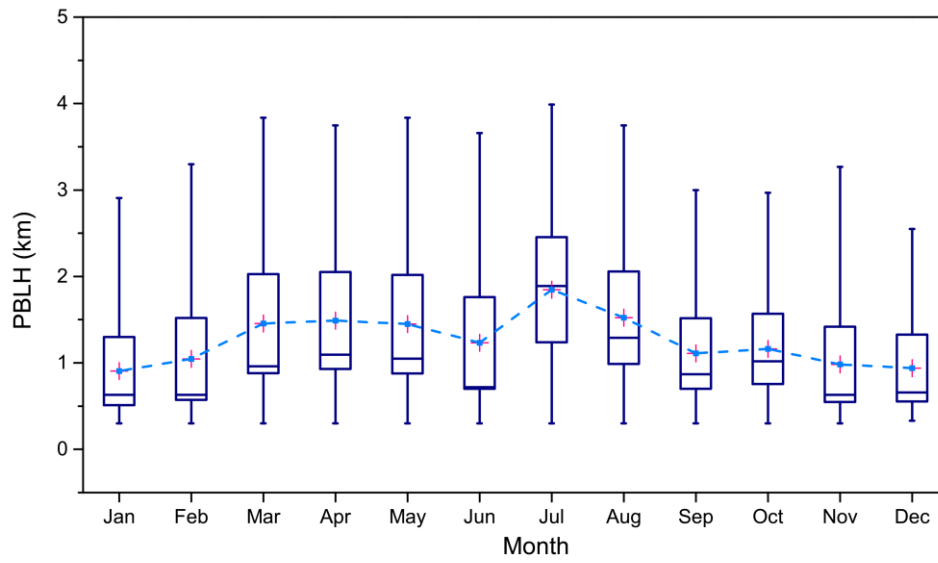
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730 **Figure 2. Monthly variation in averaged (a) mass concentration of PM_{2.5} and PM₁₀, (b) AOD (440, 670, 870, and 1020 nm) and AE**

731 **(between 440 and 870 nm), (c) volume size distribution, (d) SSA (440, 670, 870, and 1020 nm), during 2013–2015 in Hangzhou.**

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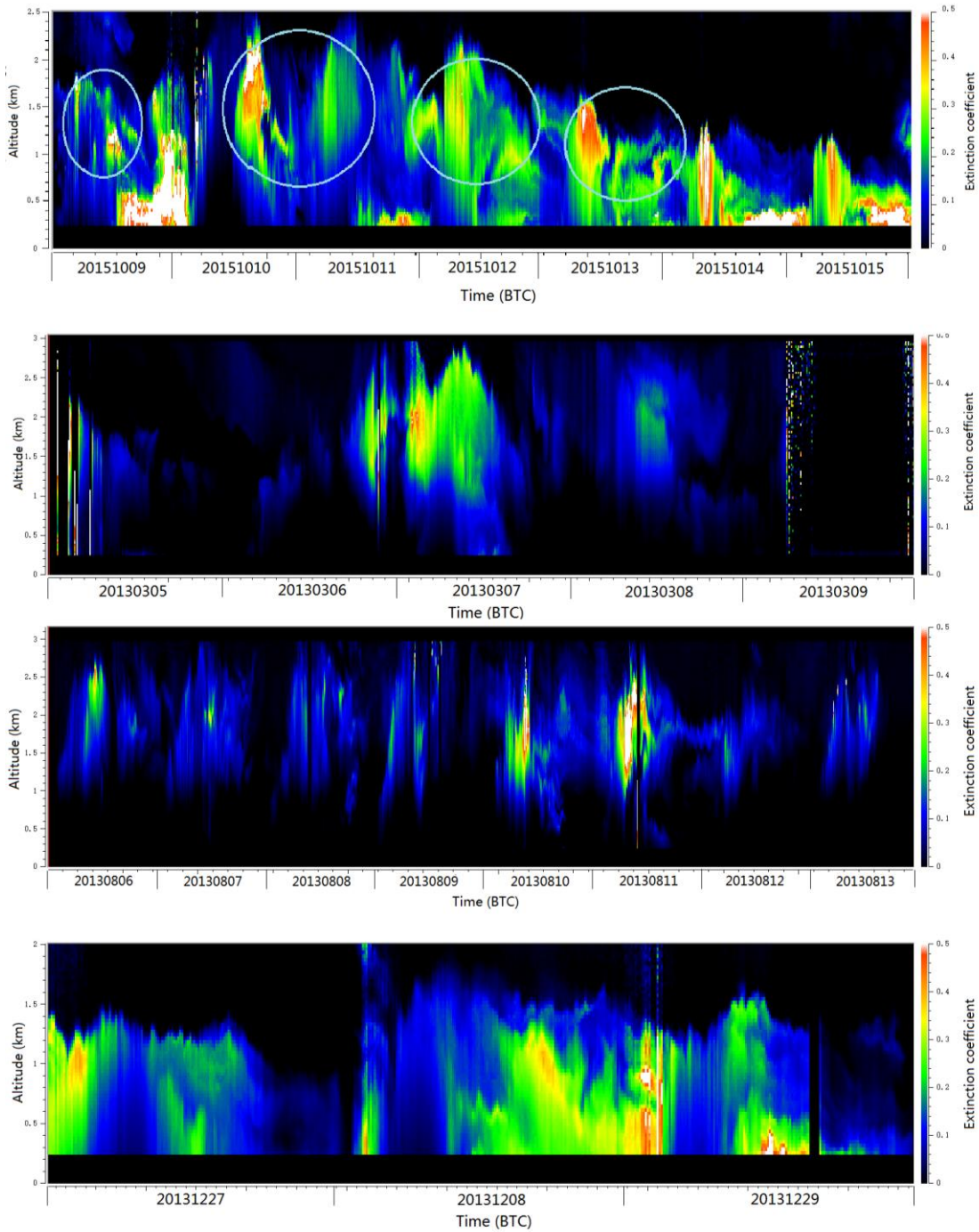
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735 **Figure 3. Monthly variation of averaged PBLH retrieved from the MPL, and their 5th and 95th percentile box plots during 2013–**

736 **2015 in Hangzhou.**

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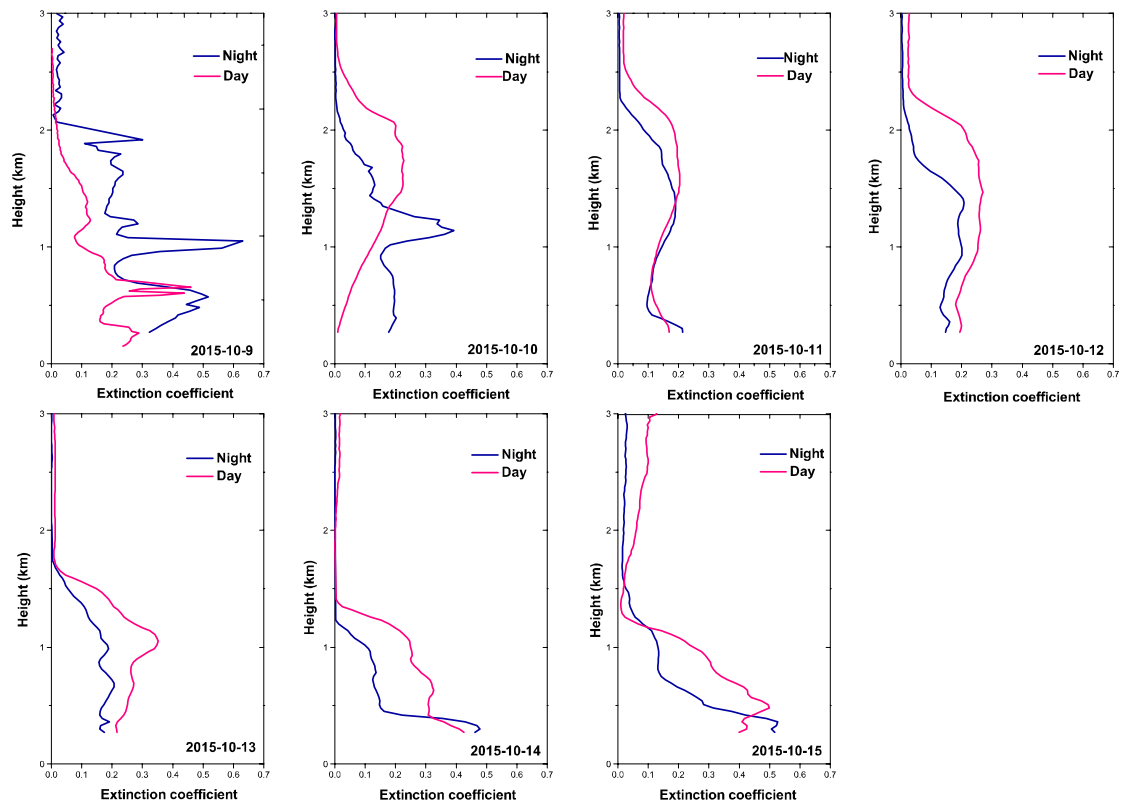


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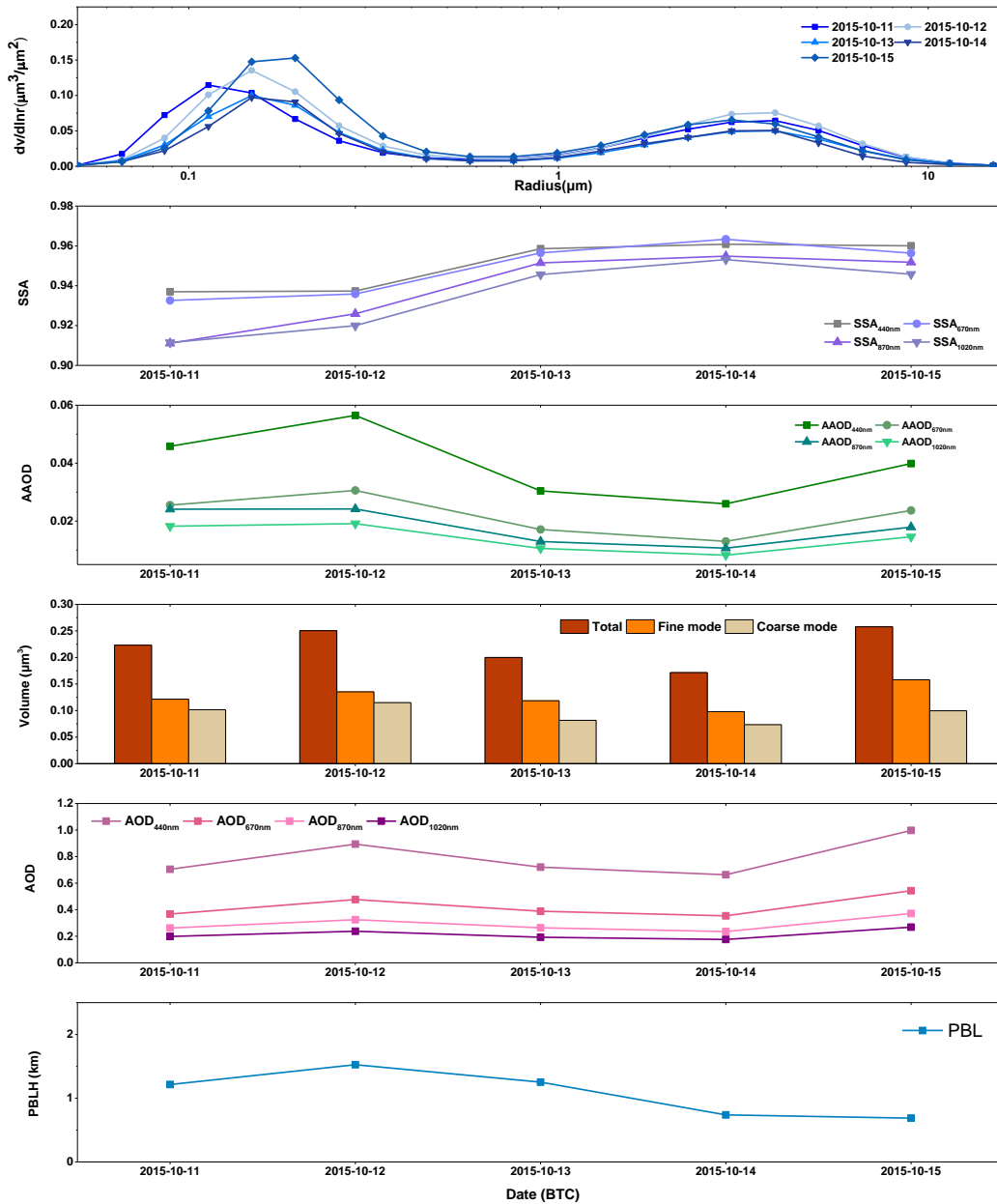
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Figure 4. Time–height cross section of the ground-based lidar-derived extinction coefficient at 532 nm in Hangzhou during (a) 9–15 October 2015, (b) 5–9 March 2013, (c) 6–13 August 2013, and (d) 27–29 December 2013.



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Figure 5. Profiles of the aerosol extinction coefficient from MPL-retrieved data, averaged for daytime and nighttime during 9–15 October 2015 in Hangzhou.

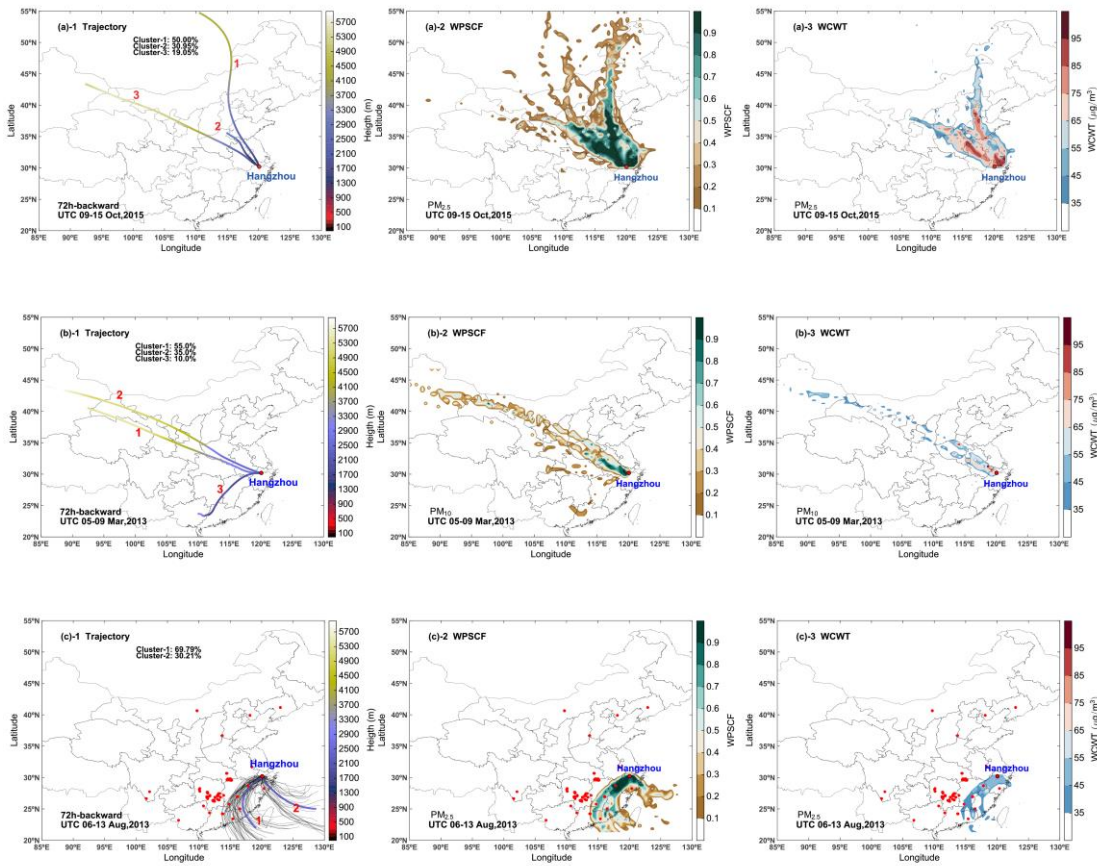


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750 **Figure 6. Daily averaged variation in aerosol optical properties and PBLH in Hangzhou during 11–15 October 2015.**

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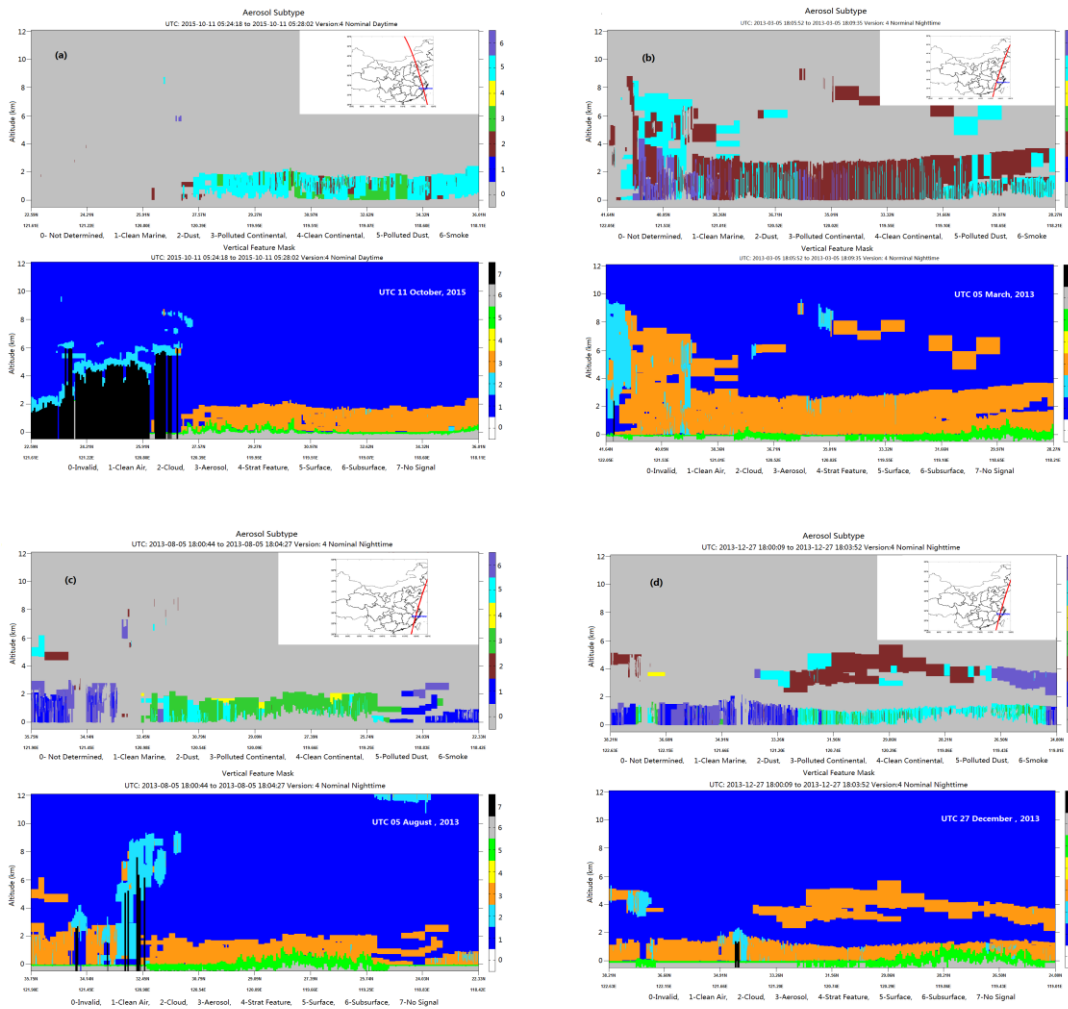
Figure 7. Mean 72-h backward trajectories of each trajectory cluster and spatial distributions of WPSCF and WCWT values for

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PM_{2.5} in Hangzhou during (a) 9–15 October 2015, (b) 5–9 March 2013, and (c) 6–13 August 2013.

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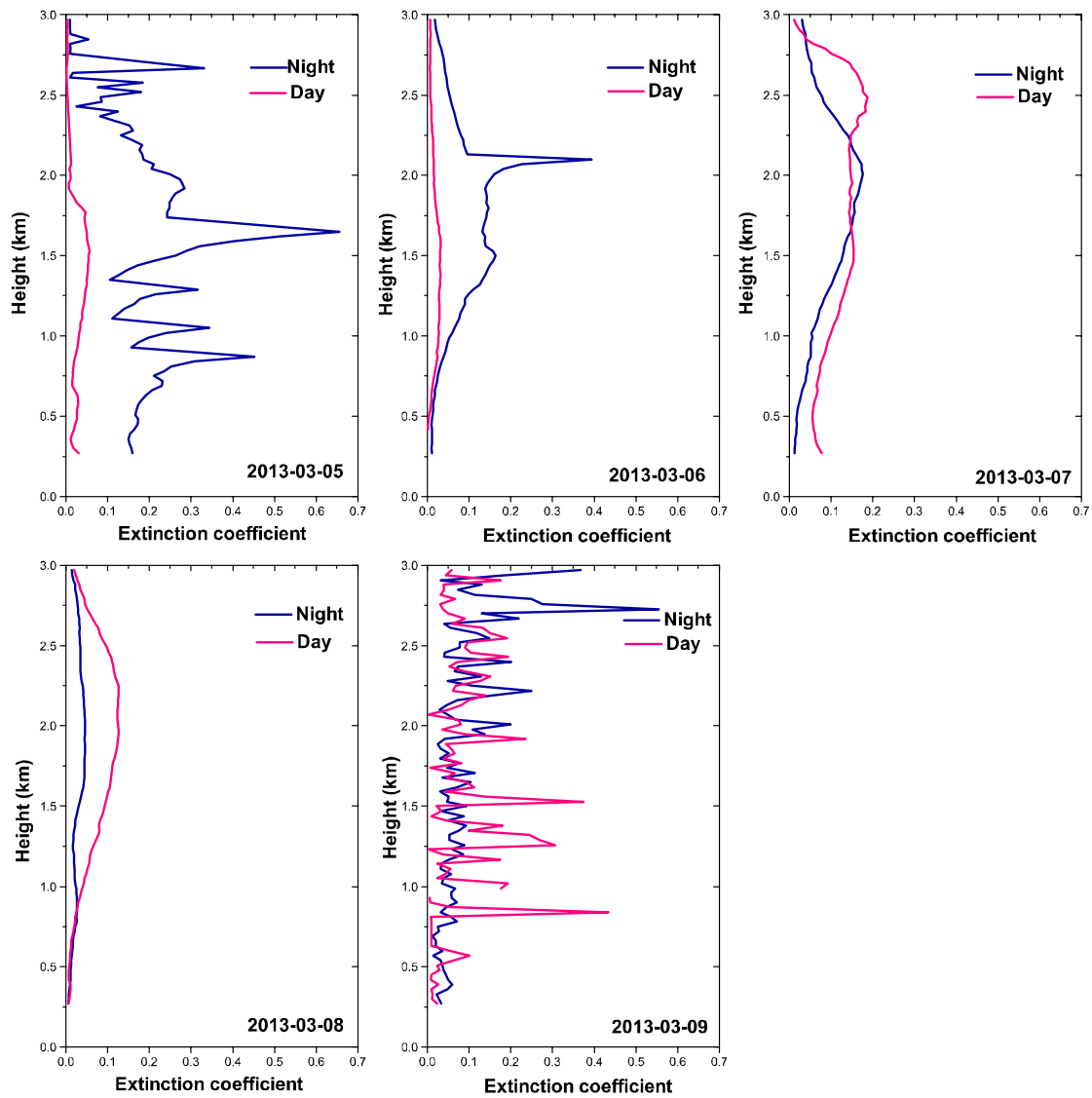
Figure 8. CALIPSO L2 products of vertical feature mask, aerosol subtype, and its ground track on (a) 11 October 2015, (b) 5

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March 2013, (c) 5 August 2013, and (d) 27 December 2013.

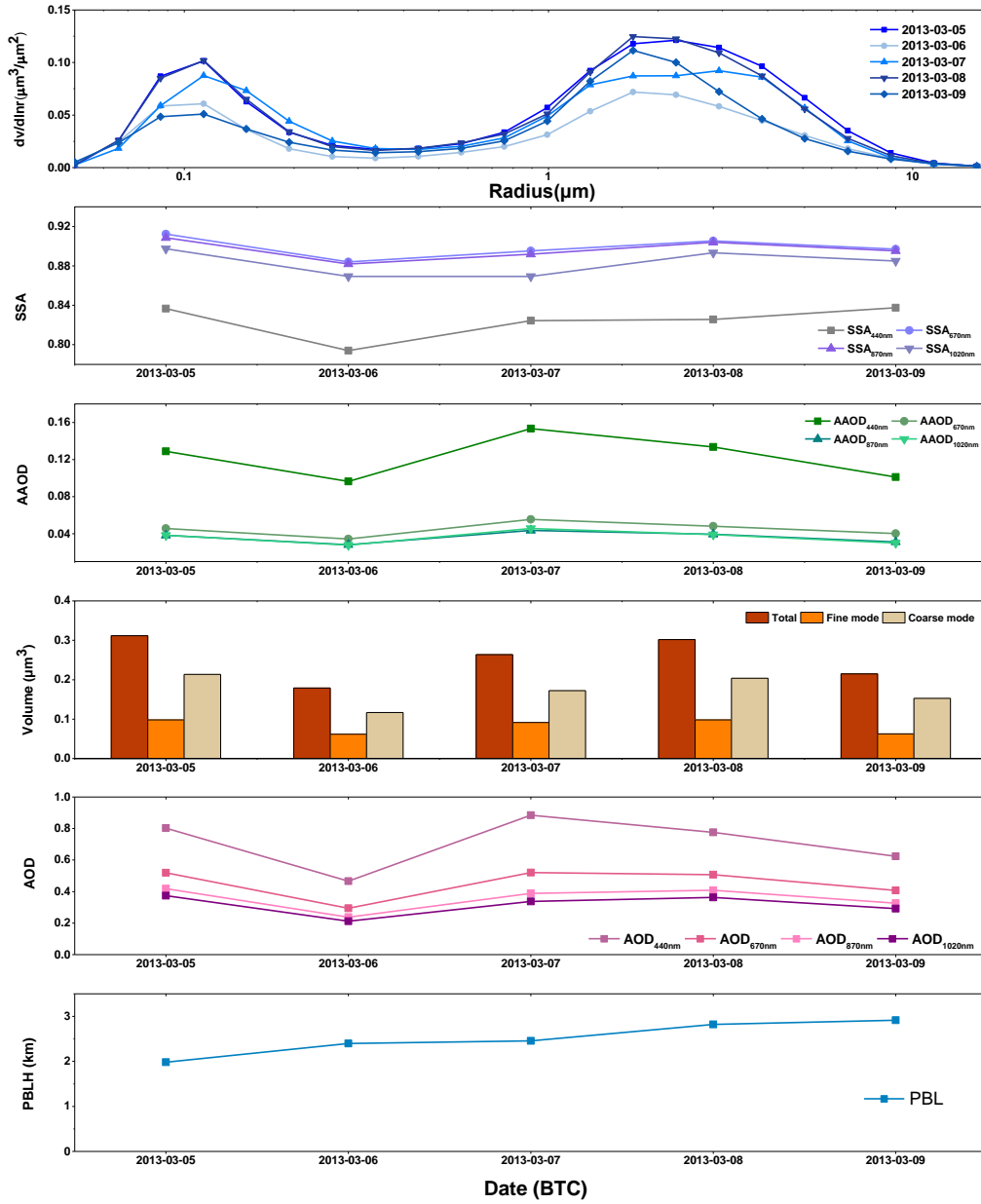
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Figure 9. Profiles of aerosol extinction coefficient from MPL-retrieved data averaged for daytime and nighttime during 5–9 March 2013 in Hangzhou.

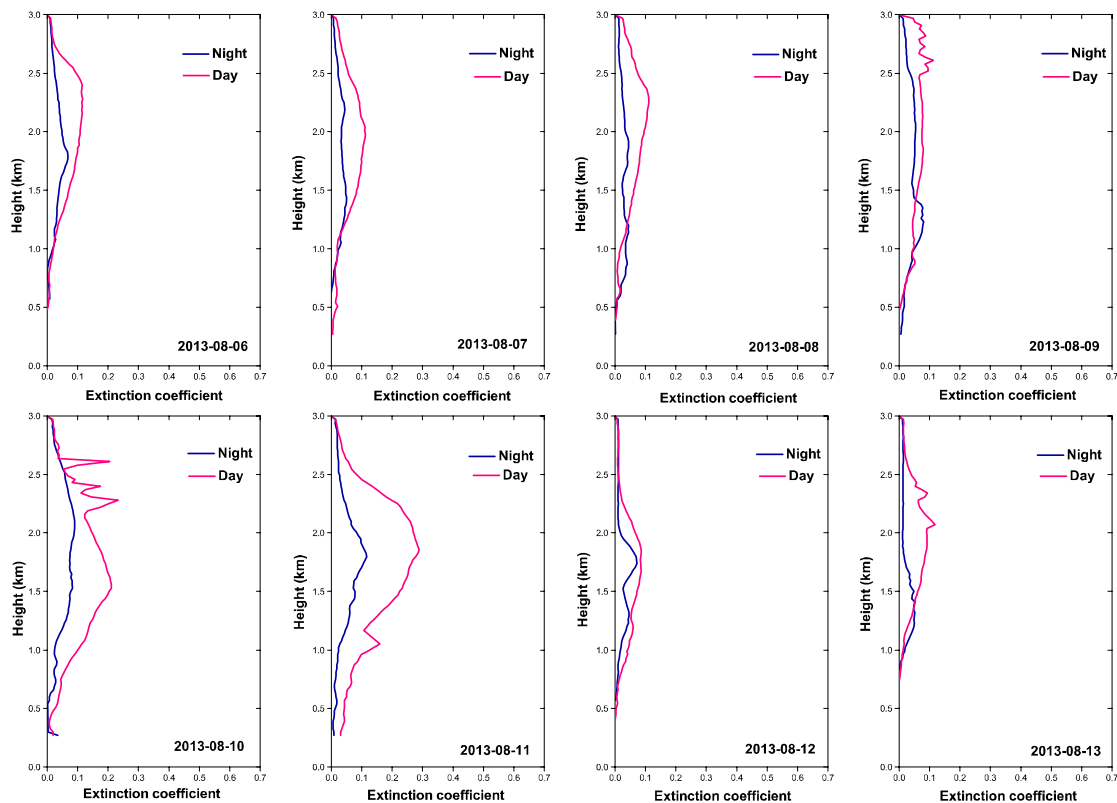


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769 **Figure 10. Daily averaged variation in aerosol optical properties and the PBLH in Hangzhou during 5–9 March 2013.**

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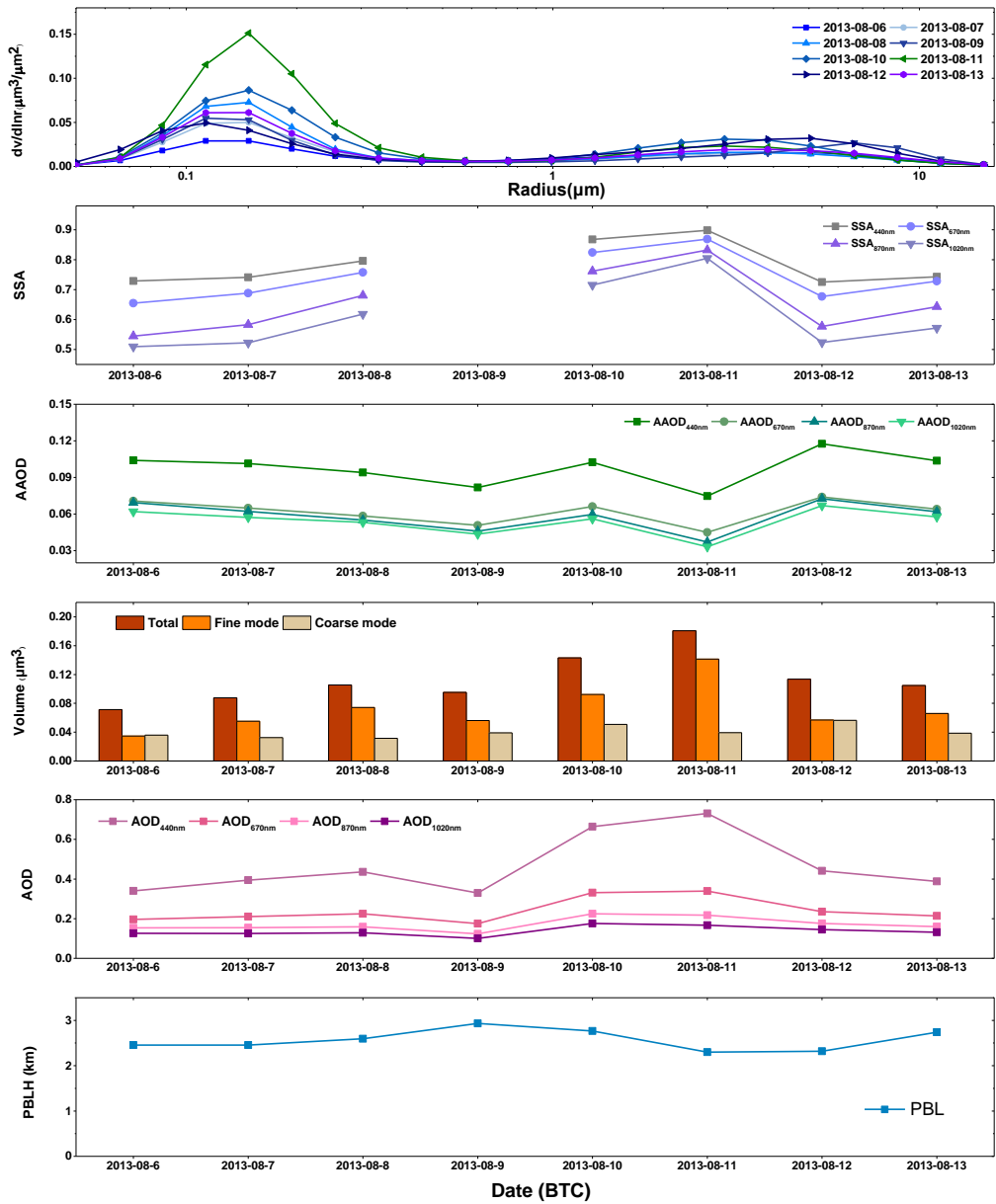
Figure 11. Profiles of the aerosol extinction coefficient from MPL-retrieved data averaged for daytime and nighttime during 6–13

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August 2013 in Hangzhou.

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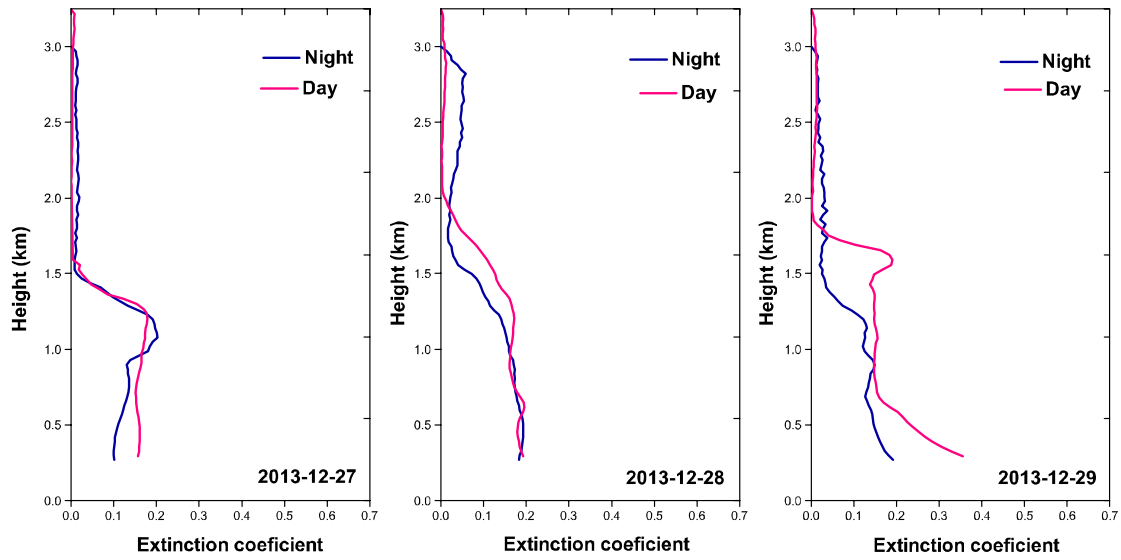


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779 **Figure 12. Daily averaged variation of aerosol optical properties and PBLHs in Hangzhou during 6–13 August 2013.**

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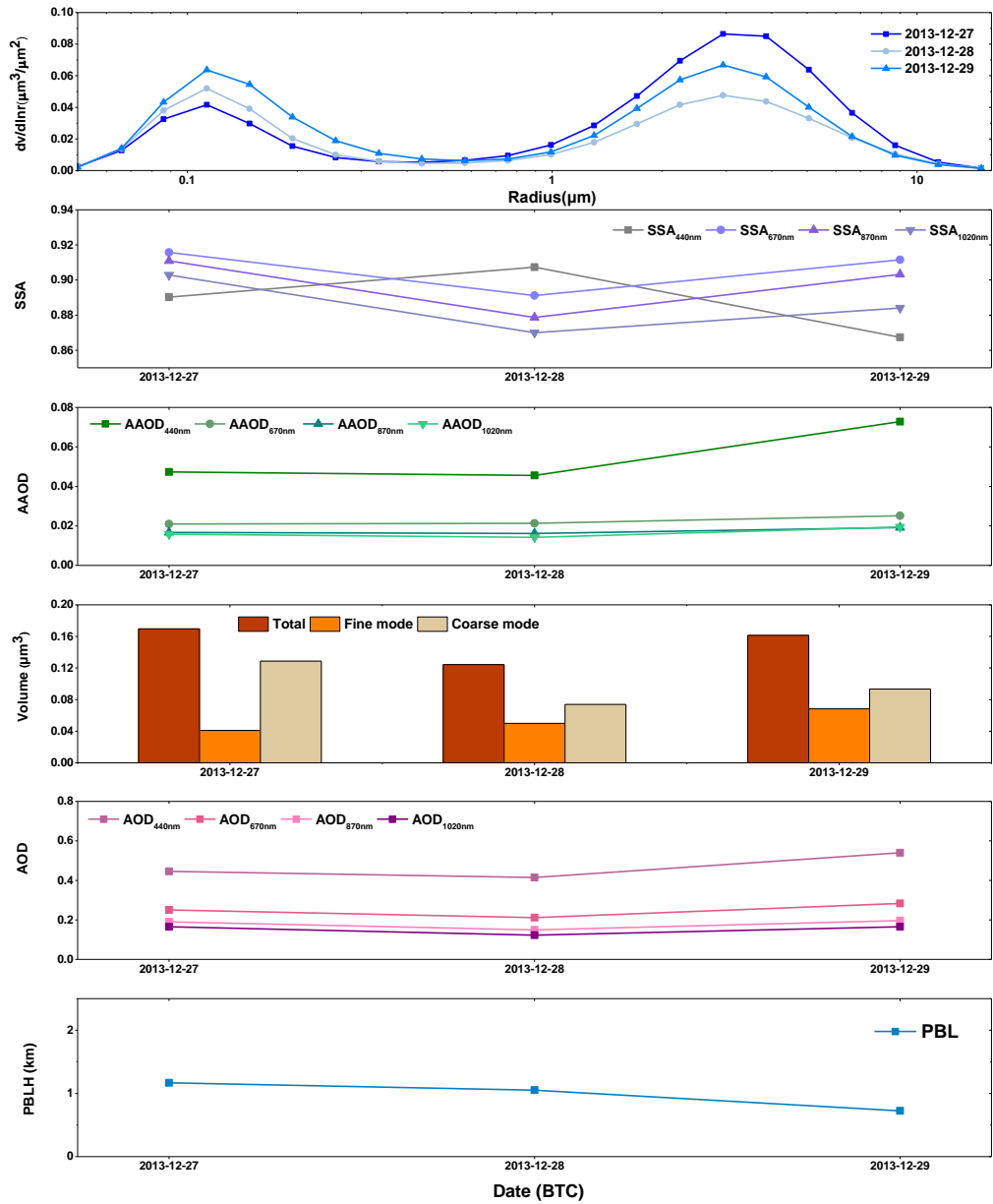
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784 **Figure 13. Profiles of aerosol extinction coefficient from MPL-retrieved data averaged for daytime and nighttime during 27–29**

785 **December 2013 in Hangzhou.**

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789 **Figure 14. Daily averaged variation in aerosol optical properties and the PBLH in Hangzhou during 27–29 December 2013.**

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Case	Cluster	Number	Mean ($\mu\text{g}/\text{m}^3$)
1	1	41	81.05±28.18
	2	22	74.45±30.17
	3	9	76.11±35.51
2	1	12	71.08±60.00
	2	7	84.57±18.26
3	1	61	54.52±16.63
	2	26	39.04±10.14

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793 **Table 1. Trajectory number and PM mean concentration of each cluster in Hangzhou during three cases, including case 1**
794 **(transportation from haze area of North China), case 2 (transportation from northwestern dust area), case 3(transportation from**
795 **southern biomass burning area).**

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