

# 1 **Impact of aerosol hygroscopic growth on retrieving aerosol extinction coefficient** 2 **profiles from elastic-backscatter lidar signals**

3 Gang Zhao<sup>1</sup>, Chunsheng Zhao<sup>1</sup>, Ye Kuang<sup>1</sup>, Jiangchuan Tao<sup>1</sup>, Wangshu Tan<sup>1</sup>, Yuxuan Bian<sup>2</sup>, Jing Li<sup>1</sup>,  
4 Chengcai Li<sup>1</sup>

5 1 Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing,  
6 China

7 2 State Key Laboratory of Severe Weather, Chinese Academy of Meteorological Sciences, Beijing,  
8 100081, China

9 *Correspondence to: Chunsheng Zhao (zcs@pku.edu.cn)*

## 10 **Abstract**

11 Light detection and ranging (lidar) measurements have been widely used to profile ambient  
12 aerosol extinction coefficient ( $\sigma_{\text{ext}}$ ). Particle extinction-to-backscatter ratio (lidar ratio, LR), which  
13 highly depends on aerosol dry particle number size distribution (PNSD) and aerosol hygroscopicity, is  
14 introduced to retrieve the  $\sigma_{\text{ext}}$  profile from elastic-backscatter lidar signals. Conventionally, a constant  
15 column-integrated LR that is estimated from aerosol optical depth is used by the retrieving algorithms.  
16 In this paper, the influences of aerosol PNSD, aerosol hygroscopic growth and relative humidity (RH)  
17 profiles on the variation of LR are investigated based on the datasets from field measurements in the  
18 North China Plain (NCP). Results show that LR has an enhancement factor of 2.2 when RH reaches  
19 92%. Simulation results indicate that both the magnitude and vertical structures of the  $\sigma_{\text{ext}}$  profiles by  
20 using column-related LR method are significantly biased from the original  $\sigma_{\text{ext}}$  profile. The relative  
21 bias, which is mainly influenced by RH and PNSD, can reach up to 40% when RH at the top of the  
22 mixed layer is above 90%. A new algorithm for retrieving  $\sigma_{\text{ext}}$  profiles and a new scheme of LR  
23 enhancement factor by RH in the NCP are proposed in this study. The relative bias between the  $\sigma_{\text{ext}}$   
24 profile retrieved with this new algorithm and the ideal true value is reduced to below 13%.

## 25 **1. Introduction**

26 Atmospheric aerosols can directly scatter and absorb solar radiation, thus exerting significant  
27 impacts on the atmospheric environment and climate change. Vertical distributions of aerosol particles  
28 are crucial for studying the roles of atmospheric aerosols in the radiation balance of the  
29 Earth-Atmosphere system (Kuang et al., 2016), air pollution transportation (Gasteiger et al., 2017) and

30 boundary layer process. However, there remain many problems while determining the spatial and  
31 temporal distributions of aerosols because of their highly variable properties (Anderson and Anderson,  
32 2003; Andreae and Crutzen, 1997) and complex sources. As a result, our knowledge about the vertical  
33 distributions of aerosols is still very limited.

34 Light detection and ranging (lidar) instruments are useful remote sensing tools to monitor profiles  
35 of aerosol optical properties. This kind of instrument involves a pulsed laser beam, which can be used  
36 to detect the back-scatter signals from aerosols and air molecules in the atmosphere (Klett, 1981).  
37 Elastic-backscatter lidar is one of the most frequently used instruments (He et al., 2006; Pietruczuk and  
38 Podgórski, 2009). However, there are some limitations when deriving aerosol extinction coefficient  
39 ( $\sigma_{\text{ext}}$ ) and aerosol **back scattering** coefficient ( $\beta_{\text{sca}}$ ) from elastic-backscatter lidar signals. Many efforts  
40 have been carried out to retrieve the  $\sigma_{\text{ext}}$  profiles from lidar signals (Klett, 1981, 1985). Particle  
41 extinction-to-backscatter ratio, which is usually termed as the lidar ratio (LR), is required when  
42 retrieving  $\sigma_{\text{ext}}$  profiles (Fernald, 1984; Fernald et al., 1972). LR can be derived directly using Raman  
43 lidar (Pappalardo et al., 2004b) and high spectral resolution lidar (She et al., 1992; Shipley et al., 1983;  
44 Sroga et al., 1983) measurements. Raman lidar has low signal to noise ratios (SNR) during the day,  
45 which may lead to significant bias and uncertainties in retrieving lidar signals. High spectral resolution  
46 lidar have high technique requirement and expensive cost. Ansmann et al. (2002) demonstrated that the  
47 profile of LR could be retrieved from Raman lidar and this LR profile can be used to retrieve  $\sigma_{\text{ext}}$   
48 profiles from high SNR elastic-backscattering lidar data. However, there exist many cases when  
49 elastic-backscatter lidar is used without concurrently measured LR profile.

50 Sun-photometer, radiometer and elastic-backscatter lidar data are usually used simultaneously to  
51 retrieve  $\sigma_{\text{ext}}$  profiles (Chaikovsky et al., 2016; He et al., 2006). In these studies,  $\sigma_{\text{ext}}$  profiles could be  
52 retrieved from elastic-backscatter lidar signals by using a constant column-related LR, which is  
53 constrained by measurements of aerosol optical depth (AOD) from sun-photometer. However, many  
54 factors such as aerosol particle number size distribution (PNSD), aerosol refractive index, aerosol  
55 hygroscopicity and ambient relative humidity (RH), have large influences on LR. It is found that the  
56 ratio of  $\sigma_{\text{ext}}$  and  $\beta_{\text{sca}}$  grows linearly but slowly as RH increases when RH is lower than 80%  
57 (Ackermann, 1998; Anderson et al., 2000; Ferrare et al., 2001). Further research found that LR is  
58 likely to change significantly due to the substantial variation of RH in the mixed layer (Ferrare et al.,  
59 1998). Small errors from the initial conditions may lead to large bias of retrieved  $\sigma_{\text{ext}}$  profiles (Sušnik

60 et al., 2014). It is likely that using a constant LR profile instead of variable LR profile to retrieve  
 61 elastic-backscatter lidar data may result in significant bias of retrieved  $\sigma_{\text{ext}}$  profiles. The sounding  
 62 profiles show that RH is highly variable and frequently beyond 80% in the mixed layer in the NCP  
 63 (Kuang et al., 2016) which is one of the most polluted areas around the world (Ma et al., 2011; Xu et  
 64 al., 2011). According to this, it is interesting to know how much  $\sigma_{\text{ext}}$  profiles retrieved from  
 65 elastic-backscatter lidar signals will be deviated if constant column-related LR profile is used in the  
 66 NCP. Few works have been done to assess the bias of using a constant LR profile. This work  
 67 comprehensively studied the possible bias by employing a large datasets of the field measurements.

68 To account for the aerosol hygroscopic growth, the  $\kappa$ -Köhler theory (Petters and Kreidenweis,  
 69 2007) is widely used, in which the chemical composition-dependent variables are merged into a single  
 70 parameter  $\kappa$ . The  $\kappa$ -Köhler equation is expressed as

$$71 \quad \frac{RH}{100} = \frac{GF^3 - 1}{GF^3 - (1 - \kappa)} \cdot \exp\left(\frac{4\sigma_s/aM_{\text{water}}}{R \cdot T \cdot D_d \cdot g f \cdot \rho_w}\right), \quad (1)$$

72 where  $D_d$  is the aerosol dry diameter. GF is the aerosol growth factor, which is defined as the ratio of  
 73 the aerosol diameter under the given RH and dry conditions ( $D_{RH}/D_d$ ). T is the temperature.  $\sigma_{s/a}$  is the  
 74 surface tension of the solution.  $M_{\text{water}}$  is the molecular weight of water. R is the universal gas constant  
 75 and  $\rho_w$  is the density of water.

76 This article is structured in the following way. Section 2 shows all of the data used in this study.  
 77 Section 3 gives the methodology of this research. Mie theory (Bohren and Huffman, 2007) and  
 78  $\kappa$ -Köhler theory (Petters and Kreidenweis, 2007) are used to study the influences of aerosol  
 79 hygroscopic growth on LR. By calculating the LR at different RH, it is found that the RH-related LR  
 80 profiles are significantly different from the constant LR profile as shown in fig. 1(b). We simulate the  
 81 bias of the retrieved  $\sigma_{\text{ext}}$  profiles by using the AOD related constant LR profiles in three steps. Firstly,  
 82 the vertical distributions of the aerosol are parameterized and the corresponding aerosol  $\sigma_{\text{ext}}$  and  $\beta_{\text{sca}}$   
 83 profiles are calculated in section 3.2. Secondly, we calculate the theoretical signals received by the  
 84 elastic-backscatter lidar in section 3.3 by using the  $\sigma_{\text{ext}}$  and  $\beta_{\text{sca}}$  profiles of the first step. Finally, we  
 85 retrieve the  $\sigma_{\text{ext}}$  profiles from the lidar signals of section 3.3 by using the column related lidar ratio  
 86 profiles, in which the method is detailed in section 3.4.1. The retrieved  $\sigma_{\text{ext}}$  profiles are compared with  
 87 the parameterized  $\sigma_{\text{ext}}$  profiles. In section 3.4.2, we proposed a new method of retrieving the  $\sigma_{\text{ext}}$   
 88 profiles, which can account for the variations of LR with RH. Results and discussions are shown in

89 section 4. Section 4.2 shows the bias of retrieved  $\sigma_{\text{ext}}$  profiles by using a column-related LR profile  
90 method. Section 4.2.1 gives the possible bias of the retrieved  $\sigma_{\text{ext}}$  profiles and section 4.2.2 shows the  
91 sensitivity of the bias under different AOD, different aerosol PNSD, different RH profiles and different  
92 aerosol hygroscopicity conditions. In section 4.4, the real-time field measurements results of  
93 micro-pulsed lidar (MPL) are used to validate the feasibility of our new proposed method. The  
94 conclusions of this research come to the section 5.

## 95 **2. Data**

### 96 **2.1 Datasets of aerosol properties**

97 During the periods of Haze in China (HaChi) campaign, the physical and chemical properties of  
98 aerosol particles are measured at the Wuqing meteorological station. Wuqing site is located between  
99 two megacities (Beijing and Tianjin) of NCP, and can represent the pollution conditions of the NCP  
100 (Xu et al., 2011).

101 This study uses the measured datasets of PNSD, black carbon (BC) mass concentrations (Ma et al.,  
102 2012) and aerosol hygroscopicity (Chen et al., 2014; Liu et al., 2014) during the field campaign. The  
103 sampled aerosol particles are selected to have aerodynamic diameter of less than 10 $\mu\text{m}$  by an  
104 impactor at the initial inlet. These particles are carefully dried to below 40% RH and then led to the  
105 corresponding instruments. The aerosol PNSDs with particle diameter in the range from 10nm to 10 $\mu\text{m}$   
106 are measured by jointly using a differential mobility particle sizer (TDMPS, Leibniz Institute for  
107 Tropospheric Research, Germany; Birmili et al., 1999) and an aerodynamic particle sizer (APS, TSI  
108 Inc., model 3321) with a temporal resolution of 5 min. The BC mass concentrations are measured by a  
109 multi-angle absorption photometer (MAAP model 5012, Thermo, Inc., Waltham, MA USA). The  
110 aerosol hygroscopicity is measured by using the humidity tandem differential mobility analyzer  
111 (HTDMA), which measures the aerosol GF as a function of RH at different diameter. The aerosol  
112 hygroscopicity parameter  $\kappa$  can be directly derived from measurements of HTDMA by applying  
113 formula (1).

### 114 **2.2 RH profiles**

115 The intensive GTS1 observation (Bian et al., 2011) at the meteorological bureau of Beijing (39°48'  
116 N, 116°28' E) were carried out from July to September in 2008. With a resolution of 10m in the vertical  
117 direction, the radiosonde data includes profiles of temperature, pressure and RH. During the intensive  
118 observation period, balloon soundings were performed four times a day.

119 Water vapor mixing ratio is almost constant in the mixed layer due to extensive turbulent mixing  
120 ~~existing~~ and decreases rapidly above the mixed layer. RH profiles that exhibit well-mixed vertical  
121 structures ~~are picked out~~ and studied. ~~With this, the maximum RH in the vertical direction can be used~~  
122 ~~as a good representation of RH profiles.~~ RH profiles are classified into four typical groups based on the  
123 maximum RH ranges: 60%-70%, 70%-80%, 80%-90% and 90%-95% (Kuang et al., 2016). These four  
124 kinds of typical well-mixed RH profiles are labeled as P60-70, P70-80, P80-90 and P90-95  
125 respectively. These four kinds of RH profiles, which are shown in fig. 1(a), are used to conduct the  
126 sensitivity studies in this article.

## 127 2.3 MPL signals

128 A single wavelength polarization diversity elastic lidar system is installed on the roof of the  
129 physics building in Peking University. This instrument is a MPL manufactured by Sigma Space, using  
130 a Nd: YVO4 532nm pulsed DC10H-532SS laser source, with a pulse duration of 10.3ns, energy of  
131 6-8uJ and a repetition of 2500Hz. It collects elastically backscattered signals from the atmosphere by  
132 separately detecting its parallel and cross polarization components with respect to the polarization of  
133 laser. We also used the concurrently measured AOD data from the AERONET BEIJING\_PKU station,  
134 which is located at the same place as the Lidar.

## 135 3. Methodology

### 136 3.1 Influences of aerosol hygroscopic growth on LR

#### 137 3.1.1 Calculate the LR values under different RH conditions

138 In this ~~research~~, the Mie model (Bohren and Huffman, 2007) is used to study the influence of RH  
139 on LR. When running the Mie model, aerosol PNSD, aerosol complex refractive index, black carbon  
140 mixing state and black carbon mass concentrations are essential. The results of ~~Mie~~ model contain the  
141 information of the  $\sigma_{\text{ext}}$  and  $\beta_{\text{sca}}$ , which can be used to calculate the LR directly, with  $\text{LR} = \frac{\sigma_{\text{ext}}}{\beta_{\text{sca}}}$ .

142 When exposed to ~~the ambient water content~~, the aerosols ~~get hygroscopic growth~~. To account for  
143 this, ~~the~~ size-resolved hygroscopicity parameter  $\kappa$ , which is derived from the measurements of  
144 ~~HTDMA~~ (Chen et al., 2012; Liu et al., 2011), ~~is used in this study.~~ ~~The used~~ size-resolved  $\kappa$  is shown  
145 in fig. S1. ~~Mean value of size-resolved  $\kappa$  during the Hachi Campaign is used.~~ With this, the aerosol GF  
146 ~~of different size at different~~ RH can be calculated by applying formula (1).

147 Mixing states of BC come from the measurement ~~during~~ the Hachi Campaign. In previous work,

148 BC mixing states during the Hachi campaign were presented as both core-shell mixed and externally  
149 mixed (Ma et al., 2012). Ma et al. (2012) provides the ratio of BC mass concentration under externally  
150 mixed state to total BC mass concentration as follows:

$$151 \quad r_{ext\_BC} = \frac{M_{ext\_BC}}{M_{BC}} \quad (2).$$

152  ~~$M_{ext\_BC}$  is the mass concentration that is externally mixed and  $M_{BC}$  is the total mass concentration of~~  
153 ~~BC.~~ The mean value of  $r_{ext\_BC}=0.51$  (Ma et al., 2012) is used as a representation of the mixing state in  
154 this study. The size-resolved distribution of BC mass concentration is the same as that used by Ma et al  
155 (2012a).

156 The refractive index ( $\tilde{m}$ ), ~~with~~ accounting for the water content in the particle, is derived as a  
157 volume mixture between the dry aerosol and water (Wex et al., 2002):

$$158 \quad \tilde{m} = f_{v,dry} \tilde{m}_{aero,dry} + (1 - f_{v,dry}) \tilde{m}_{water} \quad (3).$$

159  $f_{v,dry}$  is the ratio of the dry aerosol volume to total aerosol volume at given RH condition;  $\tilde{m}_{aero,dry}$  is  
160 the refractive index of dry ambient aerosols and  $\tilde{m}_{water}$  is the refractive index of water ~~content~~  
161 ~~absorbed by aerosols~~. The refractive indices of BC, non-light-absorbing aerosols and water, which are  
162 used in this study, are  $1.8+0.54i$  (Kuang et al., 2015),  $1.53+10^{-7}i$  (Wex et al., 2002) and  $1.33+10^{-7}$   
163 respectively.

164 To sum up, we can calculate the LR of a PNSD under the given RH condition by using the Mie  
165 scattering model. For a dry aerosol PNSD, the corresponding aerosol PNSD at a given RH can be  
166 calculated by applying the size resolved  $k$  and formula (1). Aerosol refractive index can be determined  
167 from formula (3), too. With this information, LR can be calculated. For each aerosol PNSD, we change  
168 the RH from 40% to 95% to calculate the LR values at different RH. Finally, the LR values of  
169 different measured aerosol PNSD at different RH are calculated by using the same method.

### 170 **3.1.2 Parameterizing the variation of LR with RH**

171 When the LR values under different RH are statistically studied, we find that the LR can be  
172 enhanced when the RH increases, which will be discussed in detail in section 4.1.1 and fig 2.

173 The LR enhancement factor is introduced to describe the influence of aerosol hygroscopic growth  
174 on LR at different RH. It is defined as the ratio of LR at a given RH to LR at the condition of RH<40%.  
175 We give the statistically ~~by~~ mean relationships between the LR enhancement factor and RH. The LR  
176 enhancement factor can account for the ~~incensement~~ of LR with RH and the parameterized LR

177 enhancement factor is further used in our proposed method to retrieve the  $\sigma_{\text{ext}}$  profiles.

### 178 3.2 LR profiles and $\sigma_{\text{ext}}$ profiles

179 ~~Assumptions about aerosol properties in the vertical direction are made to calculate LR profiles~~  
180 ~~and  $\sigma_{\text{ext}}$  profiles.~~

181 Liu et al. (2009) studied vertical profiles of aerosol total number concentration (Na) with aircraft  
182 measurements. ~~Vertical distributions of Na are parameterized according to the vertical distribution~~  
183 ~~properties of Na. Results showed that Na is relatively constant in the mixed layer. A transition layer~~  
184 ~~where Na linearly decreases exists in the parameterized scheme. Na also exponentially decreases~~  
185 above the transition layer. The same parameterized scheme proposed by Liu et al. (2009) is adopted by  
186 this study. Both the study of Liu et al. (2009) and Ferrero et al. (2010) manifests that the dry aerosol  
187 PNSD in the mixed layer varies little. The shape of dry aerosol PNSD is assumed constant along with  
188 the height, which means that aerosol PNSD at different heights divided by Na give the same  
189 normalized PNSD.

190 As for the BC vertical distribution, Ferrero et al. (2011) and Ran et al. (2016) demonstrate that BC  
191 mass concentration in the mixed layer remains relatively constant and decreases sharply above the  
192 mixed layer. According to this, parameterization scheme of BC vertical distributions is assumed the  
193 same as that of the aerosol. The shape of the size-resolved BC mass concentration distribution is also  
194 assumed the same as that at the surface.

195 LR profiles and  $\sigma_{\text{ext}}$  profiles can be calculated by Mie theory under these assumptions. Details of  
196 computing  $\sigma_{\text{ext}}$  profiles can be found at Kuang et al. (2015). The calculated LR profiles and  $\sigma_{\text{ext}}$   
197 profiles are used in the following study to provide the theoretical elastic-backscatter signals.

### 198 3.3 Simulated elastic-backscatter lidar signals

199 The intensity of signals received by elastic-backscatter lidar depends on optical properties of  
200 objects and the distance between scattering objects and receiving system. It can be typically described  
201 by the following formula:

$$202 \quad P(R) = C \times P_0 \times \frac{\beta(R)}{R^2} \times e^{\int_0^R -2 \times \sigma(r) \times dr} \quad (4).$$

203 In formula (4),  $P_0$  is the intensity of the laser pulse. R is the spatial distance between scattering  
204 objects and the receiving system. C is a correction factor determined by the status of  
205 elastic-backscatter lidar machine itself.  $\beta(R)$  refers to the sum of aerosol backscattering coefficient

206 ( $\beta_{\text{sca}}$ ) and air molecule backscattering coefficient ( $\beta_{\text{sca,mole}}$ ) at distance R.  $\sigma(R)$  denotes the sum of  $\sigma_{\text{ext}}$   
207 and air molecule's extinction coefficient ( $\sigma_{\text{ext,mole}}$ ).  $\beta_{\text{sca,mole}}$  and  $\sigma_{\text{ext,mole}}$  can be calculated by using  
208 Rayleigh scattering theory when the temperature and pressure are available.

209 In this study, we can theoretically get the intensities of elastic-backscatter lidar signals and the  
210 AOD from each given  $\sigma_{\text{ext}}$  and  $\beta_{\text{sca}}$  profiles with the assumption that C is equal to one. Retrieving  
211 elastic-backscatter lidar signals can result in exactly the same  $\sigma_{\text{ext}}$  profile as the original one when the  
212 profile of LR is available. However, a constant column-related LR profile is used to retrieve  
213 elastic-backscatter lidar signals and the retrieved  $\sigma_{\text{ext}}$  profile would deviate from the given  $\sigma_{\text{ext}}$  profile  
214 when there is insufficient information about the LR profile.

### 215 3.4 Retrieving $\sigma_{\text{ext}}$ profiles from elastic-backscatter lidar signals

#### 216 3.4.1 Retrieving $\sigma_{\text{ext}}$ profiles by using constant column-related LR profile method

217 Traditionally, the AOD from sun-photometer and the elastic-backscatter lidar signals are  
218 combined to retrieve the  $\sigma_{\text{ext}}$  profiles. Additional information is needed to get the mathematical results  
219 of formula (4) because there are two unknown parameters ( $\beta_{\text{sca}}$  and  $\sigma_{\text{ext}}$ ). The commonly used method  
220 of solving this formula is to assume a constant value of column-related LR and then the profiles of  $\sigma_{\text{ext}}$   
221 and  $\beta_{\text{ext}}$  can be retrieved (Fernald, 1984; Klett, 1985). Different values of column-related LR can lead  
222 to different  $\sigma_{\text{ext}}$  profiles and different AOD. A constant column-related LR can be constrained if sun  
223 photometer is concurrently measuring the AOD (He et al., 2006; Pietruczuk and Podgorski, 2009).  
224 Thus,  $\sigma_{\text{ext}}$  profile can be retrieved by using the column-related constant LR profile.

#### 225 3.4.2 Retrieving $\sigma_{\text{ext}}$ profiles accounting for aerosol hygroscopic growth

226 A new method of retrieving  $\sigma_{\text{ext}}$  profiles from elastic-backscatter lidar signals is proposed, in  
227 which the variation of LR with RH can be taken into consideration. This new method requires the  
228 measured elastic-backscatter lidar signals, measured AOD data and RH profiles.

229 A schematic diagram of this method is shown in Fig.2. A parameterized LR profile is used to  
230 retrieve  $\sigma_{\text{ext}}$  profiles instead of an AOD-constrained constant LR profile. Firstly, the LR enhancement  
231 factor are statistically studied and parameterized under different polluted conditions. The results of  
232 mean parameterized LR enhancement factor, which is detailed in section 4.1.1, are used in this study.  
233 LR profile can be calculated by using RH profile, a LR value at dry state and the equations of LR  
234 enhancement factor.  $\sigma_{\text{ext}}$  profile can be retrieved with combination of LR profile and formula (4). Dry  
235 state LR value can be constrained by comparing the integrated AOD value of retrieved  $\sigma_{\text{ext}}$  profile and



236 concurrently measured AOD value. LR profile is determined and  $\sigma_{\text{ext}}$  profile can be retrieved with the  
237 constrained dry state LR.

## 238 4. Results and Discussion

### 239 4.1 LR properties

#### 240 4.1.1 Variation of LR with RH

241 During the field campaign of Hachi, ~~there is a total~~ 3540 different aerosol PNSDs. These aerosol  
242 PNSDs can be used as a good representation datasets of the continental aerosol. LR is calculated by  
243 using different aerosol PNSD and RH values between 30% and 95%.

244 Relationships between dry state LR and concurrently measured  $\sigma_{\text{ext}}$  (sum of the aerosol scattering  
245 and absorption) are shown in Fig. 2(a). It shows that LR can vary across a wide range from 30 sr to 90  
246 sr, which is consistent with the literature values of continental aerosols (Ansmann et al., 2001;  
247 Pappalardo et al., 2004a). This also indicates that calculating the LR by using Mie theory is feasible.  
248 Fig. 2(b) gives the probability distribution function of the LR. Most of the LR lies in the range between  
249 45~65 sr.

250 By calculating the LR values under different RH, we find that the LR tends to increase with RH.  
251 Relationships between the LR enhancement factor and RH are given in Fig. 2(c). The LR enhancement  
252 factor has a mean value lower than 1.2 when the RH is lower than 70%. LR increases linearly with RH  
253 when RH is lower than 80%, which is consistent with ~~the literal~~ results (Salemink et al., 1984).  
254 However, LR can be enhanced by a factor of 2.2 when the RH reaches 92% with mean hygroscopicity  
255 of aerosol.

256 Mean values of LR enhancement factor are parameterized as below:

$$257 \quad RH_0 = RH - 40 \quad (5)$$

$$258 \quad LR = LR_{\text{dry}} \times (0.92 + 2.5 \times 10^{-2}RH_0 - 1.3 \times 10^{-4}RH_0^2 + 2.2 \times 10^{-5}RH_0^3) \quad (6).$$

259 This parameterization equation can be used as a representation of the mean effect of continental  
260 aerosol hygroscopicity on LR.

261 The ~~incensement~~ of LR with RH has been studied before. Ackermann (1998) calculates the  
262 relationships of LR with RH by using the lognormal distribution of aerosols as the input of Mie  
263 scattering theory and finds that the LR increases with RH for ~~the~~ continental aerosols. However,  
264 Ackermann (1998) shows that the LR doesn't show the same properties for maritime aerosols and

265 desert aerosols.

266 We theoretically analyze the reasons of the LR by using the Mie scattering model and the mean  
267 aerosol PNSD of the Hachi campaign. By definition, LR is the ratio of  $\sigma_{\text{ext}}$  to  $\beta_{\text{sca}}$ .  $\beta_{\text{sca}}$  can be written as  
268  $\beta_{\text{sca}} = \frac{\sigma_{\text{ext}} \times \text{SSA} \times \text{PF}(180)}{4 \times \pi}$ , where the SSA is single scattering albedo, which is defined as the ratio of  
269 extinction coefficient and scattering coefficient. PF(180) is the aerosol scattering phase function at the  
270 scattering angle of 180°. Thus,  $\text{LR} = \frac{\sigma_{\text{ext}} \times 4 \times \pi}{\sigma_{\text{ext}} \times \text{SSA} \times \text{PF}(180)} = \frac{4 \times \pi}{\text{SSA} \times \text{PF}(180)}$ . We use the mean aerosol PNSD  
271 as the input of Mie scattering model and calculate the aerosol phase function and SSA values at  
272 different RH. When particle grows, there tends to be larger partition of forward scattering and PF(180)  
273 is smaller, which is shown in fig.S2. The PF(180) decreases 40% from 0.27 to 0.16. At the same time,  
274 the SSA increases 5% from 0.93 to 0.97 and PF(180) as shown in fig.S3. Thus, the LR increases with  
275 the increment of RH.

#### 276 4.1.2 LR ratio profiles

277 Four different types of RH profiles and LR profiles are shown in fig 1. In Fig. 1(a), RH values  
278 increase with height in the mixed layer and decrease with height above the mixed layer. This is a  
279 synthetic result of temperature and water content distributions in the vertical direction. In the summer  
280 afternoon, water vapor is well mixed within the mixed layer and decreases sharply above the mixed  
281 layer. P60-70 can represent the relatively dry environmental conditions. Statistical results show that  
282 P80-90 is most likely to be observed in the environment. P90-95 is a very moist environment condition  
283 and its frequency of being observed is second to that of the P80-90 type.

284 Profiles of LR corresponding to RH profiles of the left column are shown in Fig. 1(b). For each  
285 type of LR profile, LR increases with height in the mixed layer due to the increase of RH. At the  
286 ground, the mean values of LR for each RH profiles are 38.19, 38.28, 39.53 and 40.33 sr, with a  
287 standard deviation of 6.20, 6.22, 6.42 and 6.45 respectively. LR changes little from 38 sr at the ground  
288 to 42 sr at the top of the mixed layer when the ambient RH is low for the RH profile of P60-70.  
289 However, LR grows with a mean value from 40 sr to 60 sr with a relative difference of 50% when the  
290 RH is high for the RH profile of P90-95. With such high variation of LR with RH, the retrieved  $\sigma_{\text{ext}}$   
291 profiles might be greatly deviated when using a constant LR profile instead of a variable one.

292 The black dotted line in Fig. 1(b) is one of the constant column-related LR profiles that are used as  
293 an input of retrieving  $\sigma_{\text{ext}}$  profiles related to the RH profile P70-80. The constant LR has a higher value

294 at the ground and a lower value at the top of the mixed layer when compared with the calculated  
295 variable LR profiles.

296 During the Hachi Campaign, LR values that are calculated by using Mie theory can change from  
297 30 to 55 sr within 12 hours at the ground (about 87% of initial value). With high variation of LR over  
298 time, the LR profile should be updated in time to get an accurately retrieved  $\sigma_{\text{ext}}$  profile. Using only  
299 one measurement of LR profile to retrieve the  $\sigma_{\text{ext}}$  profiles may lead to great bias of retrieved results  
300 (Rosati et al., 2016).

## 301 **4.2 Bias of retrieved $\sigma_{\text{ext}}$ profiles**

302 With the parameterized  $\sigma_{\text{ext}}$  profiles by using the method of section 3.2, we can theoretically get  
303 the AOD and the elastic-backscatter lidar signals. Then the AOD and the elastic-backscatter lidar  
304 signals can be used to constrain a column-related constant LR profile and to retrieve  $\sigma_{\text{ext}}$  profiles.  
305 Finally, the retrieved  $\sigma_{\text{ext}}$  profiles are compared with the parameterized  $\sigma_{\text{ext}}$  profiles and the differences  
306 are statistically studied.

### 307 **4.2.1 Retrieved $\sigma_{\text{ext}}$ profiles vs. original $\sigma_{\text{ext}}$ profiles**

308 Fig. 4 provides an example of the retrieved  $\sigma_{\text{ext}}$  profile by using the variable LR profile method  
309 and that by using the constant LR profile method from simulated lidar signals. These two kinds of  
310 profiles can also be described as a given parameterized  $\sigma_{\text{ext}}$  profile and a retrieved  $\sigma_{\text{ext}}$  profile from  
311 constant LR profile. In Fig. 4(a), the retrieved  $\sigma_{\text{ext}}$  profile by using a variable LR profile method is  
312 demonstrated by solid line. Dotted line shows the retrieved  $\sigma_{\text{ext}}$  profile by using a constant column  
313 related LR method. Fig. 4(b) shows the relative bias of the two retrieved  $\sigma_{\text{ext}}$  profiles at each height.  
314 Fig. 4(c) and (d) are almost the same as Fig. 4(a) and (b) respectively, except that the results of Fig. 4(a)  
315 and (b) come from the RH profile of P70-80 while those of Fig. 4(c) and (d) come from the RH profile  
316 of P90-95.

317 It is shown in Fig. 4(a) that the retrieved  $\sigma_{\text{ext}}$  by using a variable LR profile method increases with  
318 height at a rate of  $92.25 \text{ (Mm}^{-1}\text{km}^{-1}\text{)}$  in the mixed layer, which is consistent with the aerosol loading  
319 and RH distribution. However, the retrieved  $\sigma_{\text{ext}}$  profile by using a constant LR profile method behaves  
320 differently and decreases at a rate of  $-152.87 \text{ (Mm}^{-1}\text{km}^{-1}\text{)}$ . The structure of  $\sigma_{\text{ext}}$  profiles is different by  
321 using two different methods. Moreover, the retrieved  $\sigma_{\text{ext}}$  from RH profile of P90-95 at the top of the  
322 mixed layer is significantly deviated with a relative bias of 40%.

323 Both Fig. 4(a) and (c) show that the retrieved  $\sigma_{\text{ext}}$  is overestimated at ground and underestimated at

324 the top of the mixed layer. From Fig 3(b), it can be concluded that the AOD-constrained constant LR is  
325 larger than the calculated true LR at the ground and smaller at the top of the mixed layer. According to  
326 formula (3), signals of the elastic-backscatter lidar received at any height are proportional to the  
327 backscattering capability of the aerosols. When LR is larger, a larger fraction of the signals transfer  
328 forward and less is scattered back. In order to receive the same amount of signal, the backscattering  
329 coefficient should be larger and this can lead to the result of a larger  $\sigma_{\text{ext}}$  at that layer. Thus, the  $\sigma_{\text{ext}}$   
330 tends to be biased higher than the given parameterized  $\sigma_{\text{ext}}$  when the LR is larger, and vice versa.  
331 Overall, the profiles retrieved by using an AOD-constrained LR can lead to a positive bias at the  
332 ground and a negative bias at the top of mixed layer.

#### 333 4.2.2 Sensitivity Study

334 Simulations are conducted to study the characteristics of the retrieved  $\sigma_{\text{ext}}$  profile bias between  
335 using the constant column-related LR profile and variable LR profile. Different kinds of aerosol PNSD,  
336 AOD, aerosol hygroscopicity and RH profiles are used. Aerosol PNSD data comes from the Hachi  
337 Campaign field measurement. The sensitivity of the bias in aerosol hygroscopicity is evaluated by  
338 changing the size-resolved  $\kappa$  value. Aerosols are defined to have high hygroscopicity when the aerosol  
339 size-resolved  $\kappa$  value is one standard deviation above the mean of the size-resolved  $\kappa$  value. They are  
340 defined as low hygroscopicity if the size-resolved  $\kappa$  value is one standard deviation below mean of the  
341 size-resolved  $\kappa$  value. Four different kinds of RH profiles are also used in this sensitivity study. As  
342 discussed in section 3.2.1, a negative bias at the top of the mixed layer is accompanied by a positive  
343 bias at the ground and the largest bias happens at the top of the mixed layer. It is sufficient to focus on  
344 the relative bias at the top of the mixed layer.

345 Statistical characteristics of the relative bias at the top of the mixed layer are shown in Fig. 5.  
346 Different panels represent the results of different aerosol hygroscopicity. The left column shows the  
347 results of low aerosol hygroscopicity. Middle panel shows results from mean aerosol hygroscopicity.  
348 High aerosol hygroscopicity of particles results in the properties shown in the right panel. For each  
349 panel, relationships between relative bias and AOD are shown. Different colors in each panel show the  
350 results of different RH profiles. Filled colors represent the ranges of the relative bias at one standard  
351 deviation of using different PNSD.

352 Every panel show that relative bias clearly increases with the enhancement of RH in the  
353 surroundings. The relative bias has a mean value of less than 10% for RH profile of P60-70. LR has

354 little variation when the surrounding RH is low and the bias has a low value. For RH profiles of  
355 P70-80 and P80-90, the relative bias increases with RH and increases strongly up to 25% when the  
356 surrounding relative humidity is high. These behaviors of relative difference under different RH  
357 conditions are consistent with the change of LR with RH.

358 Filled color ranges of relative bias at given AOD and RH profile result from the variation of  
359 aerosol PNSD. The LR enhancement factor can have different behavior with different aerosol PNSD  
360 according to Mie scattering theory. Changing the aerosol PNSD leads to a wider range of bias when  
361 the RH is higher. Fig. 5 also shows that different PNSD can change the relative bias by a mean value  
362 of 10% for different polluted conditions.

363 Relative bias increases with AOD value when the AOD is low, while it remains constant when the  
364 AOD is high. When AOD is low, the amount of scattered light by air molecules occupies a large  
365 fraction. Air molecules have a constant LR of  $\frac{8}{3}\pi$  sr according to the Rayleigh scattering theory. The  
366 relative bias of retrieved  $\sigma_{\text{ext}}$  profile is relatively small when the AOD is low. When the AOD has a  
367 larger value, backscattered signals mainly depend on aerosol backscattering and the signals  
368 backscattered by air molecules are negligible. Relative bias mainly reflects the impacts of aerosol  
369 hygroscopicity. The mean relative bias increases from 26% to 32% at high RH conditions with the  
370 increase of aerosol hygroscopicity. Aerosol hygroscopicity should be taken into account under high  
371 RH conditions.

372 To sum up, RH is one of the most important factors that influence the accuracy of retrieving the  
373 elastic-backscatter lidar data. Different PNSD can also lead to a large variation of relative difference.  
374 The relative difference increases with the AOD when the AOD is low, but increases little when the  
375 AOD is high. Under the conditions of both high values of RH and AOD, the relative bias of retrieved  
376 data reaches a maximum due to the influence of aerosol hygroscopic growth.

### 377 **4.3 Evaluation of LR enhancement factor parameterization**

378 Simulations are carried out to test the accuracy of the new methods of retrieving the  $\sigma_{\text{ext}}$  profiles,  
379 which is proposed in section 3.4.2. These simulations employ the elastic-backscattering lidar signals  
380 from section 3.3, the RH profiles, the integrated AOD values of the parameterized  $\sigma_{\text{ext}}$  profiles and the  
381 parameterization scheme of LR enhancement factor formulas (5), (6). With this information, the  $\sigma_{\text{ext}}$   
382 profiles are retrieved by the method of section 3.4.2. We then studied the relative biases between the

383 parameterized  $\sigma_{\text{ext}}$  profiles and the retrieved  $\sigma_{\text{ext}}$  profiles by using the new method.

384 Different kinds of aerosol PNSD, AOD, aerosol hygroscopicity and RH profiles are used in the  
385 simulations. The relative bias are statistically studied and summarized. The values listed in Table 1 are  
386 the mean relative biases under different PNSD conditions. From Table 1, we can see that all of the  
387 relative bias is within the range of 13% for different PNSD, AOD, aerosol hygroscopicity and RH  
388 profiles. This indicates that the algorithm of using the mean LR enhancement factor parameterization  
389 scheme is feasible and can decrease the bias of the retrieved elastic-backscatter lidar data significantly.

#### 390 **4.4 Retrieving the real-time measurement elastic-backscatter lidar signals**

391 MPL data and AERONET data are employed to validate the algorithm of retrieving the  
392 elastic-backscatter lidar data on the day of 5 July 2016. After quality control of data processing,  
393 elastic-backscatter lidar data is retrieved by using both a constant LR profile method and a  
394 parameterized variable LR profile method. Details of retrieving the MPL signals and the auxiliary  
395 information are shown in fig.S5. Fig. 6 gives the retrieved  $\sigma_{\text{ext}}$  profiles ~~using two methods of~~ local time  
396 13:00 (a) and 14:30 (b).

397 Fig. 6(a) is a typical case of the retrieved  $\sigma_{\text{ext}}$  profiles under high values of both RH and AOD  
398 conditions. The retrieved  $\sigma_{\text{ext}}$  profiles by using the constant LR profile method and variable LR profile  
399 method show almost the same properties as the simulations. The relative bias reaches a value of 39.3%  
400 at an altitude of 1.57 km. These differences of retrieved  $\sigma_{\text{ext}}$  profiles may lead to a significant bias of  
401 estimating the mixed layer height and have significant impact on radiative energy distribution in the  
402 vertical direction. Fig. 6(b) shows the retrieved  $\sigma_{\text{ext}}$  profiles of different structures from the same  
403 elastic-backscatter lidar data. The retrieved  $\sigma_{\text{ext}}$  by using variable LR profile method increases with  
404 height within the mixed layer. However, the retrieved  $\sigma_{\text{ext}}$  by using constant LR profile decreases  
405 slightly with height within the mixed layer.

#### 406 **5 Conclusions**

407 The influence of aerosol hygroscopic growth on LR is evaluated by using Mie scattering theory.  
408 Datasets used as input to Mie theory model come from the Hachi Campaign field measurements and  
409 these datasets can be used as a good representation of the continental aerosols. Results show that LR in  
410 the NCP mainly ranges from 30 to 90 sr, which is consistent with literature values of continental  
411 aerosols. LR could be enhanced significantly under high RH conditions, with a mean factor of 2.2 at  
412 92% RH.

413 RH in the mixed layer in the NCP is frequently observed to be higher than 90%. Under these  
414 conditions, large variation of LR in the vertical direction exists. This leads to significant bias of  
415 retrieved  $\sigma_{\text{ext}}$  profile due to a constant LR profile currently used to retrieve the elastic-backscatter lidar  
416 signals. The relative bias of the retrieved  $\sigma_{\text{ext}}$  profiles between the constant LR profile method and the  
417 variable LR profile method can reach up to 40% under high RH conditions and the retrieved  $\sigma_{\text{ext}}$   
418 profile structure can be different under low RH conditions.

419 Sensitivity studies are carried out to test the bias of retrieved  $\sigma_{\text{ext}}$  profiles. The bias increases  
420 linearly with RH at low RH but increases strongly at high RH. PNSD can lead to 10% standard  
421 deviation of the bias. Maximum bias happens under the conditions of both high AOD and RH that  
422 frequently happen in the NCP. The influence of aerosol hygroscopic growth on LR should be taken  
423 into consideration when retrieving the elastic-backscatter lidar data in the NCP.

424 A new algorithm accounting for the aerosol hygroscopic growth is proposed to retrieve the  
425 elastic-backscatter lidar data. A scheme of LR enhancement factor parameterization is introduced in  
426 this algorithm. The bias of retrieved  $\sigma_{\text{ext}}$  profiles by using this algorithm can be constrained within  
427 13%. Real-time measurement of MPL data is employed to validate the algorithm and the results show  
428 good consistency with the simulations.

429 This research will advance our understanding of the influence of aerosol hygroscopic growth on  
430 LR and help to improve the retrieval of  $\sigma_{\text{ext}}$  profile from elastic-backscatter lidar signals.

431

## 432 Acknowledgments

433 This work is supported by the National Natural Science Foundation of China (41590872,  
434 41375134).

435

436

## 437 References

- 438 Ackermann, J. (1998) The Extinction-to-Backscatter Ratio of Tropospheric Aerosol: A Numerical Study. *Journal of*  
439 *Atmospheric and Oceanic Technology* 15, 1043-1050.
- 440 Anderson, T.L., Anderson, T.L. (2003) Variability of aerosol optical properties derived from in situ aircraft measurements  
441 during ACE-Asia. *Journal of Geophysical Research* 108, ACE-15-11-ACE 15-19.
- 442 Anderson, T.L., Masonis, S.J., Covert, D.S., Charlson, R.J., Rood, M.J. (2000) In situ measurement of the aerosol  
443 extinction-to-backscatter ratio at a polluted continental site. *Journal of Geophysical Research: Atmospheres* 105,

444 26907-26915.

445 Andreae, M.O., Crutzen, P.J. (1997) Atmospheric Aerosols: Biogeochemical Sources and Role in Atmospheric Chemistry.  
446 Science 276, 1052-1058.

447 Ansmann, A., Wagner, F., Althausen, D., Müller, D., Herber, A., Wandinger, U. (2001) European pollution outbreaks during  
448 ACE 2: Lofted aerosol plumes observed with Raman lidar at the Portuguese coast. Journal of Geophysical Research  
449 Atmospheres 106, 20725–20733.

450 Ansmann, A., Wagner, F., Müller, D., Althausen, D., Herber, A., von Hoyningen-Huene, W., Wandinger, U. (2002) European  
451 pollution outbreaks during ACE 2: Optical particle properties inferred from multiwavelength lidar and star-Sun photometry.  
452 Journal of Geophysical Research: Atmospheres 107, AAC 8-1-AAC 8-14.

453 Bian, J., Chen, H., ouml, mel, H., Duan, Y. (2011) Intercomparison of humidity and temperature sensors: GTS1, Vaisala RS80,  
454 and CFH. Advances in atmospheric sciences 28, 139-146.

455 Bohren, C.F., Huffman, D.R., (2007) Absorption and Scattering by an Arbitrary Particle, Absorption and Scattering of Light  
456 by Small Particles. Wiley-VCH Verlag GmbH, pp. 57-81.

457 Chaikovsky, A., Dubovik, O., Holben, B., Bril, A., Goloub, P., Tanre, D., Pappalardo, G., Wandinger, U., Chaikovskaya, L.,  
458 Denisov, S., Grudo, J., Lopatin, A., Karol, Y., Lapyonok, T., Amiridis, V., Ansmann, A., Apituley, A., Allados-Arboledas, L.,  
459 Biniatoglou, I., Boselli, A., D'Amico, G., Freudenthaler, V., Giles, D., Jose Granados-Munoz, M., Kokkalis, P., Nicolae, D.,  
460 Oshchepkov, S., Papayannis, A., Perrone, M.R., Pietruczuk, A., Rocadenbosch, F., Sicard, M., Slutsker, I., Talianu, C., De  
461 Tomasi, F., Tsekeri, A., Wagner, J., Wang, X. (2016) Lidar-Radiometer Inversion Code (LIRIC) for the retrieval of vertical  
462 aerosol properties from combined lidar/radiometer data: development and distribution in EARLINET. Atmospheric  
463 Measurement Techniques 9, 1181-1205.

464 Chen, J., Zhao, C.S., Ma, N., Liu, P.F., Göbel, T., Hallbauer, E., Deng, Z.Z., Ran, L., Xu, W.Y., Liang, Z., Liu, H.J., Yan, P., Zhou,  
465 X.J., Wiedensohler, A. (2012) A parameterization of low visibilities for hazy days in the North China Plain. Atmos. Chem.  
466 Phys. 12, 4935-4950.

467 Chen, J., Zhao, C.S., Ma, N., Yan, P. (2014) Aerosol hygroscopicity parameter derived from the light scattering enhancement  
468 factor measurements in the North China Plain. Atmos. Chem. Phys. 14, 8105-8118.

469 Fernald, F.G. (1984) Analysis of atmospheric lidar observations: some comments. Applied Optics 23, 652-653.

470 Fernald, F.G., Herman, B.M., Reagan, J.A. (1972) Determination of Aerosol Height Distributions by Lidar. Journal of Applied  
471 Meteorology 11, 482-489.

472 Ferrare, R.A., Melfi, S.H., Whiteman, D.N., Evans, K.D., Poellot, M., Kaufman, Y.J. (1998) Raman lidar measurements of  
473 aerosol extinction and backscattering: 2. Derivation of aerosol real refractive index, single-scattering albedo, and  
474 humidification factor using Raman lidar and aircraft size distribution measurements. Journal of Geophysical Research:  
475 Atmospheres 103, 19673-19689.

476 Ferrare, R.A., Turner, D.D., Brasseur, L.H., Feltz, W.F., Dubovik, O., Tooman, T.P. (2001) Raman lidar measurements of the  
477 aerosol extinction-to-backscatter ratio over the Southern Great Plains. Journal of Geophysical Research: Atmospheres 106,  
478 20333-20347.

479 Ferrero, L., Mocnik, G., Ferrini, B.S., Perrone, M.G., Sangiorgi, G., Bolzacchini, E. (2011) Vertical profiles of aerosol  
480 absorption coefficient from micro-Aethalometer data and Mie calculation over Milan. Science of the Total Environment  
481 409, 2824-2837.

482 Ferrero, L., Perrone, M.G., Petraccone, S., Sangiorgi, G., Ferrini, B.S., Lo Porto, C., Lazzati, Z., Cocchi, D., Bruno, F., Greco, F.,  
483 Riccio, A., Bolzacchini, E. (2010) Vertically-resolved particle size distribution within and above the mixing layer over the  
484 Milan metropolitan area. Atmospheric Chemistry and Physics 10, 3915-3932.

485 Gasteiger, J., Groß, S., Sauer, D., Haarig, M., Ansmann, A., Weinzierl, B. (2017) Particle settling and vertical mixing in the  
486 Saharan Air Layer as seen from an integrated model, lidar, and in situ perspective. Atmospheric Chemistry and Physics 17,  
487 297-311.



488 He, Q.S., Li, C.C., Mao, J.T., Lau, A.K.H., Li, P.R. (2006) A study on the aerosol extinction-to-backscatter ratio with  
489 combination of micro-pulse LIDAR and MODIS over Hong Kong. *Atmospheric Chemistry and Physics* 6, 3243-3256.

490 Klett, J.D. (1981) Stable analytical inversion solution for processing lidar returns. *Applied Optics* 20, 211-220.

491 Klett, J.D. (1985) Lidar inversion with variable backscatter/extinction ratios. *Applied Optics* 24, 1638-1643.

492 Kuang, Y., Zhao, C.S., Tao, J.C., Bian, Y.X., Ma, N. (2016) Impact of aerosol hygroscopic growth on the direct aerosol  
493 radiative effect in summer on North China Plain. *Atmospheric Environment* 147, 224-233.

494 Kuang, Y., Zhao, C.S., Tao, J.C., Ma, N. (2015) Diurnal variations of aerosol optical properties in the North China Plain and  
495 their influences on the estimates of direct aerosol radiative effect. *Atmos. Chem. Phys.* 15, 5761-5772.

496 Liu, H.J., Zhao, C.S., Nekat, B., Ma, N., Wiedensohler, A., van Pinxteren, D., Spindler, G., Müller, K., Herrmann, H. (2014)  
497 Aerosol hygroscopicity derived from size-segregated chemical composition and its parameterization in the North China  
498 Plain. *Atmospheric Chemistry and Physics* 14, 2525-2539.

499 Liu, P., Zhao, C., Zhang, Q., Deng, Z., Huang, M., Xincheng, M.A., Tie, X. (2009) Aircraft study of aerosol vertical  
500 distributions over Beijing and their optical properties. *Tellus Series B - chemical & Physical Meteorology* 61, 756-767.

501 Liu, P.F., Zhao, C.S., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W.Y., Deng, Z.Z., Ma, N., Mildenerger, K., Henning, S.,  
502 Stratmann, F., Wiedensohler, A. (2011) Hygroscopic properties of aerosol particles at high relative humidity and their  
503 diurnal variations in the North China Plain. *Atmos. Chem. Phys.* 11, 3479-3494.

504 Ma, N., Zhao, C.S., Müller, T., Cheng, Y.F., Liu, P.F., Deng, Z.Z., Xu, W.Y., Ran, L., Nekat, B., van Pinxteren, D., Gnauk, T., Müller,  
505 K., Herrmann, H., Yan, P., Zhou, X.J., Wiedensohler, A. (2012) A new method to determine the mixing state of light  
506 absorbing carbonaceous using the measured aerosol optical properties and number size distributions. *Atmos. Chem. Phys.*  
507 12, 2381-2397.

508 Ma, N., Zhao, C.S., Nowak, A., Müller, T., Pfeifer, S., Cheng, Y.F., Deng, Z.Z., Liu, P.F., Xu, W.Y., Ran, L., Yan, P., Göbel, T.,  
509 Hallbauer, E., Mildenerger, K., Henning, S., Yu, J., Chen, L.L., Zhou, X.J., Stratmann, F., Wiedensohler, A. (2011) Aerosol  
510 optical properties in the North China Plain during HaChi campaign: an in-situ optical closure study. *Atmos. Chem. Phys.* 11,  
511 5959-5973.

512 Pappalardo, G., Amodeo, A., Mona, L., Pandolfi, M., Pergola, N., Cuomo, V. (2004a) Raman lidar observations of aerosol  
513 emitted during the 2002 Etna eruption. *Geophysical Research Letters* 31, 179-211.

514 Pappalardo, G., Amodeo, A., Pandolfi, M., Wandinger, U., Ansmann, A., Bösenberg, J., Matthias, V., Amiridis, V., De Tomasi,  
515 F., Frioud, M., Iarlori, M., Komguem, L., Papayannis, A., Rocadenbosch, F., Wang, X. (2004b) Aerosol lidar intercomparison  
516 in the framework of the EARLINET project. 3. Ramanlidar algorithm for aerosol extinction, backscatter, and lidar ratio.  
517 *Applied Optics* 43, 5370-5385.

518 Petters, M.D., Kreidenweis, S.M. (2007) A single parameter representation of hygroscopic growth and cloud condensation  
519 nucleus activity. *Atmos. Chem. Phys.* 7, 1961-1971.

520 Pietruczuk, A., Podgórski, J., (2009) The lidar ratio derived from sun-photometer measurements at Belsk Geophysical  
521 Observatory, *Acta Geophysica*, p. 476.

522 Pietruczuk, A., Podgorski, J. (2009) The lidar ratio derived from sun-photometer measurements at Belsk Geophysical  
523 Observatory. *Acta Geophysica* 57, 476-493.

524 Ran, L., Deng, Z., Xu, X., Yan, P., Lin, W., Wang, Y., Tian, P., Wang, P., Pan, W., Lu, D. (2016) Vertical profiles of black carbon  
525 measured by a micro-aethalometer in summer in the North China Plain. *Atmospheric Chemistry and Physics* 16,  
526 10441-10454.

527 Rosati, B., Herrmann, E., Bucci, S., Fierli, F., Cairo, F., Gysel, M., Tillmann, R., Größ, J., Gobbi, G.P., Di Liberto, L.,  
528 Di Donfrancesco, G., Wiedensohler, A., Weingartner, E., Virtanen, A., Mentel, T.F., Baltensperger, U. (2016) Studying the  
529 vertical aerosol extinction coefficient by comparing in situ airborne data and elastic backscatter lidar. *Atmospheric  
530 Chemistry and Physics* 16, 4539-4554.

531 Salemink, H.W.M., Schotanus, P., Bergwerff, J.B. (1984) Quantitative lidar at 532 nm for vertical extinction profiles and the

532 effect of relative humidity. *Applied Physics B* 34, 187-189.

533 She, C.Y., Alvarez, R.J., Caldwell, L.M., Krueger, D.A. (1992) High-spectral-resolution Rayleigh–Mie lidar measurement  
534 of aerosol and atmospheric profiles. *Optics Letters* 17, 541-543.

535 Shipley, S.T., Tracy, D.H., Eloranta, E.W., Trauger, J.T., Sroga, J.T., Roesler, F.L., Weinman, J.A. (1983) High spectral resolution  
536 lidar to measure optical scattering properties of atmospheric aerosols. 1: Theory and instrumentation. *Applied Optics* 22,  
537 3716-3724.

538 Sroga, J.T., Eloranta, E.W., Shipley, S.T., Roesler, F.L., Tryon, P.J. (1983) High spectral resolution lidar to measure optical  
539 scattering properties of atmospheric aerosols. 2: Calibration and data analysis. *Applied Optics* 22, 3725-3732.

540 Sušnik, A., Holder, H., Eichinger, W. (2014) A Minimum Variance Method for Lidar Signal Inversion. *Journal of Atmospheric*  
541 *and Oceanic Technology* 31, 468-473.

542 Wex, H., Neususs, C., Wendisch, M., Stratmann, F., Koziar, C., Keil, A., Wiedensohler, A., Ebert, M. (2002) Particle scattering,  
543 backscattering, and absorption coefficients: An in situ closure and sensitivity study. *Journal Of Geophysical*  
544 *Research-Atmospheres* 107.

545 Xu, W.Y., Zhao, C.S., Ran, L., Deng, Z.Z., Liu, P.F., Ma, N., Lin, W.L., Xu, X.B., Yan, P., He, X., Yu, J., Liang, W.D., Chen, L.L. (2011)  
546 Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain.  
547 *Atmos. Chem. Phys.* 11, 4353-4369.

548

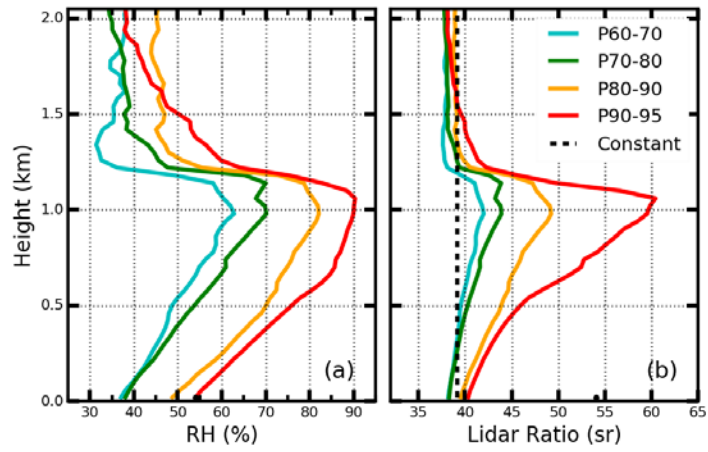
549

550 **Table 1.** Relative difference (%) between the  $\sigma_{\text{ext}}$  profiles by using the proposed new method and the parameterized  $\sigma_{\text{ext}}$   
 551 profiles under different AOD and RH profile conditions

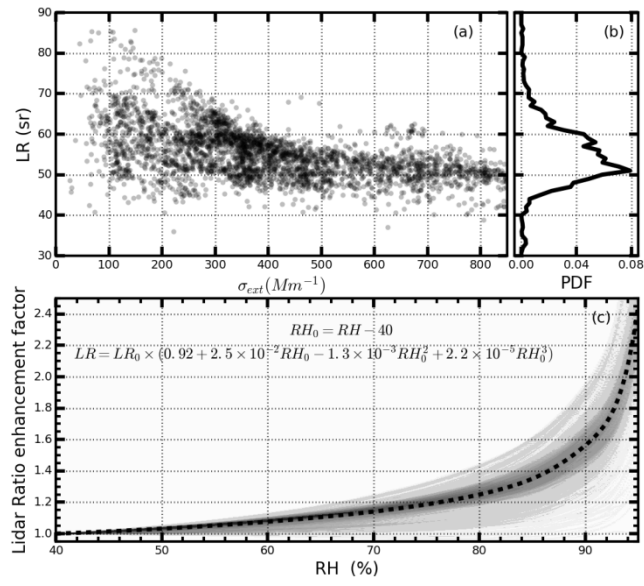
		AOD							
		0.2	0.4	0.6	0.8	1.0	1.2	1.4	1.6
RH profile	P60-70	6	9	11	13	8	8	8	9
	P70-80	7	7	9	12	7	6	7	8
	P80-90	8	5	4	11	6	5	5	6
	P90-95	9	6	6	9	13	7	7	9

552

553



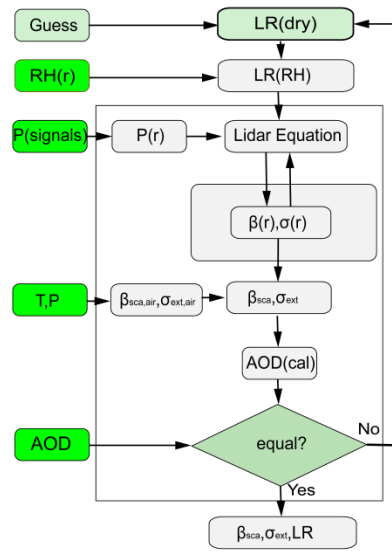
554  
 555 **Figure 1.** (a) Four kinds of RH profiles P60-70, P70-80, P80-90, and P90-95; (b) calculated LR profiles from the  
 556 corresponding RH profiles of (a). Dotted black line is one of the constant LR profiles that are used to retrieve the  
 557 MPL signals.  
 558



559  
 560 **Figure 2.** LR distribution and LR enhancement factor during Hachi campaign. (a) LR distribution under different  
 561 polluted conditions. (b) Probability distribution of the LR. (c) Enhancement factor of the LR. Dotted line is the mean  
 562 fit LR enhancement factor.  
 563  
 564

565

566

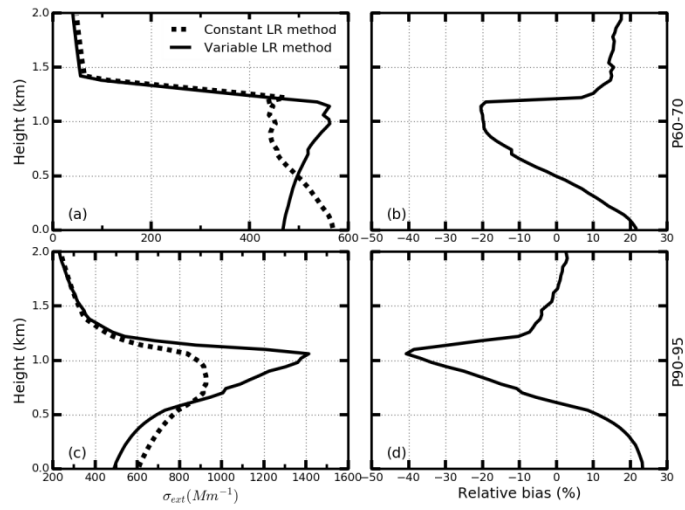


567

568

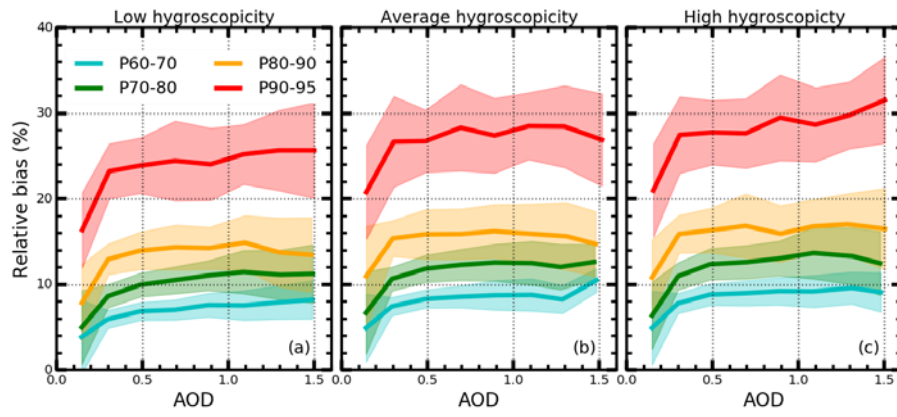
569

**Figure 3.** Schematic diagram of retrieving the  $\sigma_{\text{ext}}$  profile. The input variables are displayed in green background.



571

572 **Figure 4.** (a) Retrieved  $\sigma_{\text{aero}}$  profiles using constant LR profile method (dotted line) and variable LR profile method  
 573 (solid line) from simulated lidar signals. (b) The relative bias of the retrieved  $\sigma_{\text{aero}}$  profile using two different methods.  
 574 (c),(d) are the same as (a), (b) respectively. The LR signals of panel (a) results form P70-80 RH profile, and LR  
 575 signals of panel (b) results from P90-95 RH profile.



576

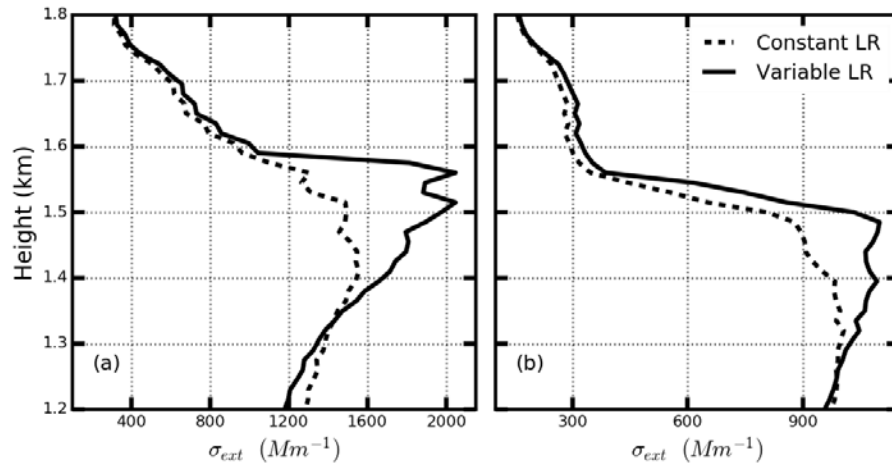
577

578

579

**Figure 5.** Relative bias of the retrieved  $\sigma_{\text{ext}}$  under different AOD, PNSD, and hygroscopicity and RH profiles conditions. Different colors represent different RH profile. Panel (a) is derived from the low hygroscopicity. Panel (b) results from the mean hygroscopicity. Panel (c) is for high hygroscopicity.





580  
 581 **Figure 6.** Retrieved  $\sigma_{\text{ext}}$  profiles from field measurement MPL signals at (a) 13:00 and (b) 14:30 on July 5, 2016. Dotted  
 582 line represents the retrieved  $\sigma_{\text{ext}}$  profiles using constant LR profile method. Solid line represents the retrieved  $\sigma_{\text{ext}}$  profiles  
 583 using variable LR profile method.