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## Multi-peak accumulation and coarse modes observed from AERONET retrieved aerosol volume size distribution in Beijing

Ying Zhang, Zhengqiang Li, Yuhuan Zhang, Yu Chen, Juan Cuesta, Yan Ma

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# Meteorology and Atmospheric Physics

## Multi-peak accumulation and coarse modes observed from AERONET retrieved aerosol volume size distribution in Beijing

--Manuscript Draft--

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	Chinese Major Project of High Resolution Earth Observation System (30-Y20A39-9003-15/17)	Zhengqiang Li
<b>Abstract:</b>	<p>We present characteristic peaks of atmospheric columnar aerosol volume size distribution retrieved from the AErosol RObotic NETwork (AERONET) ground-based Sun-sky radiometer observation, and their correlations with aerosol optical properties and meteorological conditions in Beijing over 2013. The results show that the aerosol volume particle size distribution (VPSD) can be decomposed into up to four characteristic peaks, located in accumulation and coarse modes, respectively. The mean center radii of extra peaks in accumulation and coarse modes locate around <math>0.28(\pm 0.09)</math>-<math>0.38(\pm 0.11)</math> and <math>1.25(\pm 0.56)</math>-<math>1.47(\pm 0.30)</math> <math>\mu\text{m}</math>, respectively. The multi-peak size distributions are found in different aerosol loading conditions, with the mean aerosol optical depth (440 nm) of 0.58, 0.49, 1.18 and 1.04 for 2-, 3- l/II and 4-peak VPSD types, while the correspondingly mean relative humidity values are 58, 54, 72 and 67%, respectively. The results also show the significant increase (from 0.25 to 0.40 <math>\mu\text{m}</math>) of the mean extra peak median radius in the accumulation mode for the 3-peak-II cases, which agrees with aerosol hygroscopic growth related to relative humidity and/or cloud or fog processing.</p>	

**COMMENTS TO THE AUTHOR:**

Reviewer #2: MAP manuscript: Multi-peak accumulation and coarse modes observed from AERONET retrieved aerosol volume size distribution in Beijing

**1. Abstract: line 46 - please add to the end of this sentence: "and/or cloud or fog processing.'**

We have added "and/or cloud or fog processing" to the end of this sentence.

**2. Line 158: Replace 'breaking down' with 'separation'**

We have corrected according to the Reviewer's comments.

**3. Line 239: The Klein and Daba (2014) reference is not pertinent here since this paper mainly focuses on fog droplet size and liquid water content of the fog droplets, not the residual aerosol that remains after fog evaporation. I suggest replacing this with Li et al. (2014)**

Thanks for reviewer's comment. We have corrected according to the Reviewer's comments.

**4. Line 251 : Please add: "and/or cloud or fog processing.'**

We have added "and/or cloud or fog processing" according to the Reviewer's comments.

[Click here to view linked References](#)

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4 1 **Multi-peak accumulation and coarse modes observed from**

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6 2 **AERONET retrieved aerosol volume size distribution in Beijing**

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11 4 Ying Zhang<sup>1</sup>, Zhengqiang Li<sup>1\*</sup>, Yuhuan Zhang<sup>2</sup>, Yu Chen<sup>3</sup>, Juan Cuesta<sup>4</sup>, Yan Ma<sup>1</sup>

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25 **Highlights**

- 26 ● The 3-peak and 4-peak aerosol volume size distributions observed in Beijing.
- 27 ● Characteristics of extra peaks in accumulation and coarse modes respectively.
- 28 ● Statistics on multi-peak cases versus meteorological conditions.

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31 **Abstract**

32 We present characteristic peaks of atmospheric columnar aerosol volume size  
33 distribution retrieved from the AErosol RObotic NETwork (AERONET)  
34 ground-based Sun-sky radiometer observation, and their correlations with aerosol  
35 optical properties and meteorological conditions in Beijing over 2013. The results  
36 show that the aerosol volume particle size distribution (VPSD) can be decomposed  
37 into up to four characteristic peaks, located in accumulation and coarse modes,  
38 respectively. The mean center radii of extra peaks in accumulation and coarse modes  
39 locate around  $0.28(\pm 0.09)$ - $0.38(\pm 0.11)$  and  $1.25(\pm 0.56)$ - $1.47(\pm 0.30)$   $\mu\text{m}$ , respectively.  
40 The multi-peak size distributions are found in different aerosol loading conditions,  
41 with the mean aerosol optical depth (440 nm) of 0.58, 0.49, 1.18 and 1.04 for 2-, 3-  
42 I/II and 4-peak VPSD types, while the correspondingly mean relative humidity  
43 values are 58, 54, 72 and 67%, respectively. The results also show the significant  
44 increase (from 0.25 to 0.40  $\mu\text{m}$ ) of the mean extra peak median radius in the  
45 accumulation mode for the 3-peak-II cases, which agrees with aerosol hygroscopic  
46 growth related to relative humidity [and/or cloud or fog processing](#).

47 **Keywords:** multi-peak aerosol volume particle size distribution, lognormal mode  
48 separation, extra fine and coarse mode peaks, hygroscopic growth

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3 **51 1. Introduction**

4  
5 52 Atmospheric aerosols play important roles in Earth's environment and radiation  
6  
7 53 budget. The aerosols affect the climate system by scattering and absorbing solar  
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9 54 radiation, referred to as direct effects (Stocker et al. 2013). Also, they act as cloud  
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11 55 condensation nuclei and ice nuclei involved in cloud process, referred to as indirect  
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13 56 effects. The increase of aerosol particles can lead to the cloud layer for rapid  
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15 57 adjustments, precipitation reduction and longer cloud lifetime (Lohmann and  
16  
17 58 Feichter 2005; Albrecht 1989; Twomey 1977). Abundant anthropogenic aerosols also  
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19 59 cause frequent air pollution events in developing countries, e.g East of China (Li et  
20  
21 60 al. 2013). Therefore, characterization of key aerosol's parameters in hot spot areas  
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23 61 becomes important due to impacts of aerosols both on climate changes and air  
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25 62 pollution control.

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27 63 Aerosol size distribution is one of the important information of aerosol  
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29 64 properties. The particulates suspended in the air change their sizes by coagulation,  
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31 65 aggregation and condensation, which all vary with the aerosol size distribution.  
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33 66 Although the particle size changes constantly, Willeke and Whitby (1975) found that  
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35 67 atmospheric aerosol particles exist in three modes stably. The aerosol particles with  
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37 68 diameter from 0.1 $\mu$ m to 1 $\mu$ m are referred to as accumulation mode, while those less  
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39 69 than 0.1 $\mu$ m referred to as transient nuclei mode, and those more than 1 $\mu$ m referred to  
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41 70 as coarse mode. Most of researches now follow this definition (e.g. Remer and  
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43 71 Kaufman 2006; Bellouin et al. 2005; Kaufman et al. 2005a, b). The nuclei modal  
44  
45 72 aerosols derived from nucleation process can be observed on the nanometer scale  
46  
47 73 (e.g. Liu et al. 2013; Kulmala et al. 2004). The accumulation modal aerosols are  
48  
49 74 mainly contributed by aging nuclei mode. The collision cross section of  
50  
51 75 accumulation particles is about one order smaller than their geometric cross section,  
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53 76 and thus they are hard to deposit due to viscosity of atmosphere (Park et al. 1999,  
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55 77 Otto et al. 1999). Due to their longer residence time and important impacts on  
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57 78 atmospheric radiation, the accumulation modal aerosols are investigated extensively.

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60 79 Eck et al. (2003a, b) find that accumulation modal volume median radius of  
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3 80 biomass burning aerosol increases with aerosol optical depth (AOD), caused by the  
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5 81 aerosol aging (Reid et al. 1998), and the similar phenomenon is also observed in  
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7 82 urban-industrial and mixed site (Dubovik et al. 2002). Furthermore, two peaks in the  
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9 83 accumulation mode can present simultaneously (i.e. Zhang et al. 2015) in the serious  
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11 84 air pollution conditions, and the second one is considered as the residual of cloud/fog  
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13 85 droplet evaporation (Li et al. 2014; Eck et al. 2012). The coarse modal aerosols can  
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15 86 also present the 2-peak distribution. The first peak of coarse mode with the median  
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17 87 radius from 1 $\mu$ m to 2 $\mu$ m is considered to result from hygroscopic growth or the  
18  
19 88 activated particles with radius less than 1 $\mu$ m (Yang and Wenig 2009; Singh et al.  
20  
21 89 2004). As cloud condensation nuclei or ice nuclei, the first peak of the coarse mode  
22  
23 90 are thought to be involved in the cloud/fog formation and dissipation (Hammer et al.  
24  
25 91 2014). All of these studies analyze the entire aerosol size distribution, rather than  
26  
27 92 extract individual each characteristic peaks, except Li et al. (2014) fitted a lognormal  
28  
29 93 function for extra peak of accumulation mode during heavy pollution condition.

30  
31 94 In this study, we split the columnar aerosol volume particle size distribution  
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33 95 retrieved by Sun-sky radiometer measurements in Beijing over 2013 into lognormal  
34  
35 96 peaks based on a separation method (Cuesta et al. 2008), and analyze the recognized  
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37 97 extra peaks with aerosol optical properties and meteorological conditions. The data  
38  
39 98 and method are described in section 2. In section 3, the properties of extra peaks and  
40  
41 99 ambient atmospheric conditions are analyzed, and the conclusions are provided in  
42  
43 100 section 4.

## 44 45 46 101 **2. Data and Method**

### 47 48 102 **2.1 Data**

49  
50 103 In this paper, the optical and physical properties of atmospheric aerosol are  
51  
52 104 obtained from the Sun-sky radiometer (CE318). The CE318 has 8 channels including  
53  
54 105 340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 1020 nm and 1640 nm for  
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56 106 aerosol measurements, and 936 nm for perceptible water. The direct and diffused  
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58 107 Sun light are measured from ground by CE318 and then used to derive aerosol

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108 properties, such as AOD from extinction of incident direct sunlight, and aerosol  
109 VPSD retrieved from sky light (Dubovik et al. 2000a, b). The temporal resolutions  
110 of these two measurements are about 15 minutes and 1 hour, respectively. The  
111 Sun-sky radiometers have been applied extensively to measure atmospheric aerosol  
112 properties, as known as the AErosol RObotic NETwork (AERONET) (Holben et al.  
113 1998, 2001).

114 A Sun-sky radiometer (#350) belonging to AERONET site Beijing\_RADI  
115 (116.4° E, 40.0° N, 59m) is employed in this study. The AERONET Lev 1.5 data in  
116 2013 is used including AOD, Ångström exponent (AE), single scattering albedo  
117 (SSA) and aerosol volume size distribution. The errors of AOD and SSA provided by  
118 AERONET are about 0.01-0.02 (Holben et al. 1998; Eck, et al. 1999) and 0.03  
119 (Dubovik et al. 2000b), respectively. The AE is computed by linear regression of the  
120 440, 500, 675, and 870 nm AOD data. The Lev 1.5 data is further filtered with  
121 additional quality controls in this paper, including (i) solar zenith angles greater than  
122 50 degrees, (ii) the minimum number of observation angles specified by AERONET  
123 Lev 2.0 data criteria (Holben et al., 2006), and (iii) the total retrieval residual less  
124 than 5%. Moreover, the extra quality control of AOD (440 nm) > 0.4 is applied to  
125 SSA.

126 The meteorological data at World Meteorological Organization (WMO) Beijing  
127 site (54511, 116.5° E, 39.8° N, 31.3m) are used, including temperature, relative  
128 humidity and mean wind speed during 10 minutes, with temporal resolution of 1  
129 hour fitting for AERONET size distribution data.

130 **2.2 Method**

131 In this study, we need to separate the aerosol volume particle size distribution  
132 (VPSD) into Log-Normal Modes (LNM). We employ the separation method  
133 developed by Cuesta et al. (2008). Each LNM is described by 3 parameters: the  
134 modal concentration, modal radius and geometric standard deviation. In order to  
135 obtain these parameters, a kernel function consist of root mean square differences

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3 136 between VPSDs (the one calculated from combination of separated LNMs vs. the  
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5 137 one obtained from AERONET data) is minimized, iteratively using the NelderMead  
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7 138 simplex algorithm (Cuesta et al. 2008). The NelderMead simplex algorithm searches  
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9 139 the convergence by applying the strictly convex function for multidimensional  
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11 140 unconstrained minimization (Lagarias et al, 1998). We choose multi-modal  
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13 141 lognormal distributions to fit the AERONET retrievals by the following formula:

$$142 \quad \frac{dV(r)}{d\ln r} = \sum_{i=1,n} \frac{C_i}{\sqrt{2\pi}(\ln\sigma_i)^2} \exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_i}{\ln\sigma_i}\right)^2\right] \quad (1)$$

143 where  $dV/d\ln r$  (in unit of  $\mu\text{m}^3/\mu\text{m}^2$ ) is aerosol volume particle size distribution,  $C_i$   
144 ( $\mu\text{m}^3/\mu\text{m}^2$ ),  $r_i$  ( $\mu\text{m}$ ) and  $\sigma_i$  are the volume modal concentration, median radius and  
145 standard deviation of each LNM mode, respectively. Here,  $n$  is the total number of  
146 LNM modes.

147 The LNM separation is achieved when the second derivative of a theoretical  
148 Gaussian curve is less than zero. However, some non-significant convex functions  
149 can lead to a larger unreal total number (e.g. 6) of  $n$  (Cuesta et al. 2008). Based on  
150 data statistics and analysis of Beijing 2013 data in this paper, we find that for the  
151 purpose of identifying LNM peaks, one can use the second derivative of -0.003 as  
152 the threshold. Under this threshold, VPSD is only separated into 2 to 4 peaks, with  
153 low convergence errors. In Figure 1, we show the mode separation of an example  
154 VPSD from Beijing 2013 data.

### 155 3. Results

#### 156 3.1 VPSD peak characteristics

157 In 2013, the number of AERONET retrieved size distribution records is 577 in  
158 Beijing site and the proportion of the available [separation](#) cases are 98.6% while the  
159 remainders are mainly non-smooth VPSDs. The recognized 2-, 3- and 4- peak type  
160 VPSD numbers account for 48%, 49% and 3% of total 569 cases (Figure 2),  
161 respectively. In figure 3, we show the statistics (N=277) on the appearance frequency  
162 of the median radii for 3-peak cases. It can be seen clearly that two distinguishable  
163 groups located in accumulation ( $<0.7 \mu\text{m}$ ) and coarse ( $>0.7 \mu\text{m}$ ) particle radius

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4 164 ranges, respectively, which supports the classification of 3-peak-I (i.e., an extra  
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6 165 coarse mode peak) and 3-peak-II (i.e., an extra accumulation mode peak) types in  
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8 166 this study. Therefore, a total of four types of volume size distribution are  
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10 167 summarized in this study and their mean VPSD are shown in Figure 2.

11 168 It can be seen from Figure 2(a) that mean volume of 3-peak-II type, especially  
12  
13 169 the fine modal volume, is larger than others. The 4-peak VPSD numbers are smaller,  
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15 170 but the mean volume is larger (Figure 2b). As listed in table 1, for accumulation  
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17 171 mode range (i.e.  $r_1$  and  $r_2$ ), the first peak is around 0.13-0.15 $\mu\text{m}$  while the second  
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19 172 peaks present at a mean median radius of 0.38  $\mu\text{m}$  ( $\pm 0.11$ ) for 3-peak-II and 0.28  $\mu\text{m}$   
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21 173 ( $\pm 0.09$ ) for 4-peak type, respectively. Moreover, two peak characteristics can also  
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23 174 present in the coarse mode (i.e. 3-peak-I type), with the mean median radius varying  
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25 175 from 1.25  $\mu\text{m}$  ( $\pm 0.56$ ) to 1.47  $\mu\text{m}$  ( $\pm 0.30$ ).

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27 176 The seasonal changes of multi-peak VPSDs are also noticeable. Figure 4 shows  
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29 177 that the 2-peak cases are prevalent in November, December and January, and the  
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31 178 3-peak-I prevalent in April, May. Especially in April, the 3-peak-I and II cases  
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33 179 account for 74% of available VPSDs. In June, due to the impacts of cloud and rain  
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35 180 on remote sensing observation, there are only 8 available VPSDs, while the  
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37 181 summarized percentage of 3-peak-II and 4-peak cases is about 50%, the maximum in  
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39 182 the whole year. It should also be noticed that, the VPSDs with more than 2 peaks (i.e.  
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41 183 3-peak and 4-peak types) appears in all months of the year. Considering that a large  
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43 184 part of these extra peak VPSDs may related to residual of cloud/fog residuals, it is  
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45 185 suggested that cloud contamination (e.g. thin cirrus) on the aerosols occurs (Eck et  
46  
47 186 al., 2014) throughout the year in Beijing 2013.

### 187 **3.2 Optical properties of multi-peak VPSDs**

188 It is important to reveal the aerosol optical properties associated with  
189 multi-peak VPSDs. Figure 5(a) shows the frequency of multi-peak VPSD case  
190 numbers with AOD. The number frequency of the 3-peak-II and 4-peak cases  
191 increases obviously when AOD is more than 0.5. The mean AOD of the 3-peak-II

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3 192 cases is the highest (1.18) in 2013 while much lower for the 3-peak-I (0.49) and  
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5 193 2-peak cases (0.58) as listed in Table 2. The 65% of the 2-peak cases present in the  
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7 194 low aerosol loading conditions (e.g. AOD < 0.5), where probability of the 4-peak  
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9 195 cases is very low (<6%).

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11 196 In figure 5(b), the maximum frequency of 2- and 3-peak-I cases appear at the  
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13 197 positions where AE equals to about 1.4, while AE is 1.3 for the 4-peak cases. For  
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15 198 3-peak-II cases, it presents two maximum frequencies at about AE = 1.2 and 1.4 with  
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17 199 the frequency of 22 and 20%, respectively. This suggests there might be a slight  
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19 200 particle size decrease effect (AE from 1.2 to 1.4) for 3-peak-II type while the more  
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21 201 prevalent cases are with AE = 1.2. It should also be mentioned that although this  
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23 202 standard AEORNET AE are widely used in many studies (Dubovik et al., 2002;  
24  
25 203 Gobbi et al., 2007; Che et al., 2014), more information might be retrieved from AE  
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27 204 computed from the extended wavelengths (i.e. 380-550nm) following analyses of  
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29 205 Eck et al. (2014).

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31 206 In figure 5(c), the mean SSA of 2-peak and 3-peak-I type are lower than those  
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33 207 of 3-peak-II and 4-peak cases, suggesting that the aerosol scattering property is  
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35 208 enhanced in 3-peak-II and 4-peak types, especially the 3-peak-II. The mean SSA at  
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37 209 675nm changes from 0.874 (3-peak-I) to 0.909 (3-peak-II) as listed in Table 2.  
38  
39 210 Moreover, the mean SSA of 3-peak-II type is close to that of 4-peak type at 440 nm  
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41 211 but larger at other three wavelengths. This may indicate more brown carbon and  
42  
43 212 water content in aerosols for 3-peak-II type according to SSA spectral behaviors of  
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45 213 aerosol components (Wang et al., 2013). It should be noticed that this SSA analysis  
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47 214 may have limitations on the sample representativeness, especially for 2-peak type,  
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49 215 considering data quality control of AOD (440 nm) > 0.4 affecting mainly on 2-peak  
50  
51 216 cases. The retrieval of SSA in low AOD conditions may be improved by involving  
52  
53 217 the coincident polarized measurements (Li et al. 2009) in the near future.

### 54 55 218 **3.3 Relationship between VPSD peaks and meteorological factors**

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57 219 Figure 6 shows the number frequencies of multi-peak VPSDs with the  
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220 temperature, relative humidity, and wind speed. The mean temperature of 3-peak-I  
221 cases is about 10.8 °C, higher than that of the other VPSDs (see also Table 2). It is  
222 found that the maximum frequency of 3-peak-II cases occur at a lower temperature  
223 about -8 °C, while that of 4-peak cases is much higher (~18 °C). The rather random  
224 frequency distribution of 2-peak cases confirms that the 2-peak VPSDs are appeared  
225 uniformly throughout the year with varying temperature; while 3-peak-I VPSDs tend  
226 to occur during higher temperature conditions, i.e. summer and late spring. Moreover,  
227 4-peak cases also occur mainly during summer and early spring but with slightly  
228 lower temperature than 3-peak-I type. Particularly, 3-peak-II cases often occur in the  
229 low temperature conditions, which accounts for up to 64% in winter (December,  
230 January and February).

231 The frequency distributions of multi-peak VPSD case numbers on relative  
232 humidity are shown in figure 6b. The 4-peak cases mainly present in the condition of  
233 relative humidity (RH) more than 40%, and the frequency peaks appear near RH of  
234 40%-50% and 70-90%. Meanwhile, the 3-peak-II cases mainly concentrate in the  
235 RH range of 60% to 90%. The number frequencies of other two types present a  
236 roughly symmetric distribution with RH from 10% to 90%. These higher RH  
237 associated with frequency peaks can be related to aerosol hygroscopic growth  
238 (Pilinis et al. 2014; Tang and Munkelwitz 1994), and also contacted with the  
239 cloud/fog processing ([Li et al. 2014](#)).

240 Figure 6(c) shows that the higher number frequency of the 4-peak cases  
241 presents mainly in the condition of lower wind speed ( $WS < 3$  m/s). Together with  
242 Figure 6(a, b), we found that 3-peak-II and 4-peak cases are mainly appear during air  
243 pollution conditions with higher aerosol loading ( $AOD > 0.5$ ), lower wind speed  
244 ( $WS < 3$ m/s), and higher relative humidity ( $RH > 40\%$ ).

### 245 **3.4 VPSD peak correlation with relative humidity**

246 We have seen some complex relationships between multi-peak VPSDs  
247 frequency with relative humidity in Figure 6(b), whereas several researches also

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3 248 suggested possible big influence of RH on extra VPSD peaks through hygroscopic  
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5 249 effects. Here, we show a statistics on the median radius of VPSD peaks with RH bins  
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7 250 in Table 3. It can be seen firstly a general increase of  $r_1$  with RH, agreeing with Eck  
8  
9 251 et al. (2012) results. This can be thought as the classical hygroscopic growth and/or  
10  
11 252 cloud or fog processing. Secondly,  $r_4$  show generally decreasing trends with RH,  
12  
13 253 suggesting the wet scavenging effects on dust-like particles in coarse mode. Thirdly,  
14  
15 254  $r_2$  and  $r_3$  of 3-peak-II and 4-peak VPSDs illustrate generally a positive correlation  
16  
17 255 with RH which can be thought as similar phenomena as hygroscopic growth but  
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19 256 occurring for particles with different size (Li et al. 2014; Reid et al. 1998). The radii  
20  
21 257 of aerosol particles can increase from accumulation to coarse size involved into the  
22  
23 258 cloud/fog process under highly RH conditions (Klein and Dabas, 2014), while the  
24  
25 259 decrease of RH results in the evaporation of cloud/fog droplets and the residual  
26  
27 260 particles (Li et al., 2014) can form the extra peak around 0.25-0.40  $\mu\text{m}$  (Table 3).  
28  
29 261 Fourthly,  $r_3$  of the 3-peak-I VPSD cases present a general negative correlation with  
30  
31 262 RH, in contrast with those of 3-peak-II and 4-peak types. This negative correlation is  
32  
33 263 thought to be associated with hydrophobic fly ash from coal combustion or other  
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35 264 sources.

#### 36 265 **4. Conclusions**

37  
38 266 In this study, the aerosol volume particle size distributions (VPSD) retrieved  
39  
40 267 from Sun-sky radiometer measurements are decomposed into characteristic  
41  
42 268 log-normal peaks in Beijing over 2013. The number frequency of the 2-, 3-I/II and  
43  
44 269 4-peak cases are presented against the aerosol optical properties and meteorological  
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46 270 conditions. It is found that the most part of VPSDs present two characteristic peaks,  
47  
48 271 while the probability of double-peak pattern in a single mode (accumulation and/or  
49  
50 272 coarse modes) increases under pollution conditions. The extra fine and coarse modal  
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52 273 peaks are identified in this study with the mean center radius of about  
53  
54 274 0.28( $\pm$ 0.09)-0.38( $\pm$ 0.11) and 1.25( $\pm$ 0.56)-1.47( $\pm$ 0.30)  $\mu\text{m}$ , respectively, in contrast  
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56 275 with those of standard peaks (0.15 and 3.71  $\mu\text{m}$ ). The 3-peak-I type appears under  
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3 276 dry conditions (mean RH of 54%) and is thought to contain hydrophobic fly ash  
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5 277 particles. The 3-peak-II and 4-peak types prevalently present in the condition of  
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7 278 large aerosol loading and high humidity (mean RH of 72% and 67%, respectively)  
8  
9 279 conditions, with the mean AOD (440 nm) of 1.18 and 1.04, and mean SSA (675 nm)  
10  
11 280 of 0.909 and 0.902, respectively. The correlations between multi-peak VPSD  
12  
13 281 appearance and RH suggest that the aerosol hygroscopic/hydrophobic changes and  
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15 282 cloud/fog process can play important roles in the aerosol size transformation.

17 283 We also noticed that in Beijing the extra coarse peaks (mean median radii  
18  
19 284 around 1.25 and 1.47  $\mu\text{m}$ ) possibly associate with two different sources/mechanisms,  
20  
21 285 considering their opposite RH dependence. The aerosol components are not analyzed  
22  
23 286 in this study, considering component information from in situ measurements is  
24  
25 287 somehow difficult to match exactly remote sensing measurement (i.e. surface  
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27 288 quantities vs. total column values). In the future, with the development of aerosol  
28  
29 289 component inversion techniques based on remote sensing data (e.g. Schuster et al.  
30  
31 290 2015), coupling with the characteristic peak analyses, it is expected to estimate  
32  
33 291 aerosol component information inside each characteristic peaks, which helps  
34  
35 292 understand particulate formation and transformation in the ambient atmosphere.

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#### 40 294 **Acknowledgement**

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50 299 Observation System (30-Y20A39-9003-15/17).

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442 **List of Table Captions**

443 **Table 1.** Three log-normal mode parameters ( $r$ ,  $\sigma$ ,  $C$ ) of VPSD of Figure 2.

444 **Table 2.** The optical, physical and meteorological parameters corresponding to  
445 VPSD types with multiple peaks.

446 **Table 3.** The mean median radius of multi-peak VPSD types under different relative  
447 humidity (RH) bins.

448

449 **List of Figure Captions**

450 **Figure 1.** Aerosol volume particle size distribution and the corresponding separation  
451 into four log-normal modes.

452 **Figure 2.** The annual mean VPSD of 2-peak, 3-peak-I, 3-peak-II cases (a); and  
453 4-peak cases (b) in Beijing 2013.

454 **Figure 3.** The number frequency of median radius for the extra peaks of 3-peak  
455 VPSDs.

456 **Figure 4.** The monthly percentage of multi-peak VPSDs in Beijing 2013.

457 **Figure 5.** The number frequency of multi-peak VPSDs versus AOD (a), AE (b) and  
458 SSA (c). AOD: aerosol optical depth; AE: Ångström exponent; SSA: single  
459 scattering albedo; the x-axis step is 0.1 in (a) and (b).

460 **Figure 6.** The number frequency of multi-peak VPSDs versus temperature (5°C  
461 interval) (a), relative humidity (5% interval) (b) and wind speed at altitude of 10 m  
462 (0.5 m/s interval) (c).

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465 **Table 1.** Three log-normal mode parameters ( $r$ ,  $\sigma$ ,  $C$ ) of VPSD of Figure 2.

VPSD type	N	median radius ( $\mu\text{m}$ )				standard deviation				modal concentration ( $\mu\text{m}^3/\mu\text{m}^2$ )			
		(st.d.)				(st.d.)				(st.d.)			
		$r_1$	$r_2$	$r_3$	$r_4$	$\sigma_1$	$\sigma_2$	$\sigma_3$	$\sigma_4$	$C_1$	$C_2$	$C_3$	$C_4$
2-peak	274	0.15 (0.03)	/	/	3.71 (0.93)	1.49 (0.10)	/	/	1.64 (0.16)	0.053 (0.059)	/	/	0.073 (0.073)
3-peak-I	203	0.15 (0.03)	/	1.47 (0.30)	4.20 (0.62)	1.49 (0.09)	/	2.13 (0.39)	1.54 (0.18)	0.045 (0.040)	/	0.047 (0.035)	0.069 (0.083)
3-peak-II	74	0.15 (0.05)	0.38 (0.11)	/	2.70 (0.51)	1.51 (0.26)	1.72 (0.32)	/	1.90 (0.22)	0.080 (0.049)	0.065 (0.061)	/	0.122 (0.090)
4-peak	18	0.13 (0.03)	0.28 (0.09)	1.25 (0.56)	3.44 (0.97)	1.44 (0.19)	1.57 (0.16)	1.70 (0.64)	1.72 (0.22)	0.054 (0.042)	0.064 (0.044)	0.040 (0.037)	0.088 (0.040)

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468 **Table 2.** The optical, physical and meteorological parameters corresponding to  
469 VPSD types with multiple peaks.

VPSD Type	AOD (440nm)	AE	SSA (675nm)	T (°C)	RH (%)	WS (m/s)
2-peak	0.58	1.28	0.885	5.0	58	1.62
3-peak-I	0.49	1.12	0.874	10.8	54	1.88
3-peak-II	1.18	1.18	0.909	5.2	72	1.31
4-peak	1.04	1.20	0.902	6.6	66	1.58

470 Note: AOD: Aerosol Optical Depth; AE: Ångström Exponent; SSA: Single  
471 Scattering Albedo; T: Temperature; RH: Relative Humidity; WS: Wind Speed.

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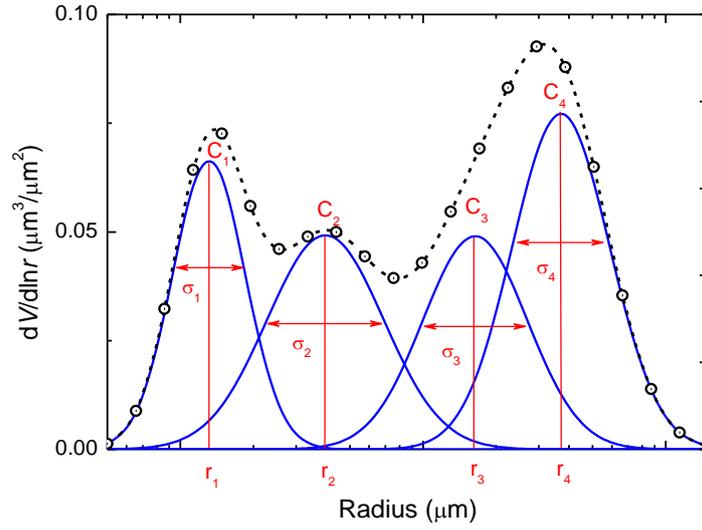
474 **Table 3.** The number frequency of median radius for the extra peaks of 3-peak  
475 VPSDs.

RH (%)		mean median radius (st.d.) [unit: $\mu\text{m}$ ]			
		$r_1$ ( $\mu\text{m}$ )	$r_2$ ( $\mu\text{m}$ )	$r_3$ ( $\mu\text{m}$ )	$r_4$ ( $\mu\text{m}$ )
2-peak	[0,35]	0.14 (0.018)	/	/	4.16 (0.353)
	(35,50]	0.13 (0.019)	/	/	3.76 (0.652)
	(50,70]	0.14 (0.033)	/	/	3.68 (1.381)
	(70,90]	0.16 (0.034)	/	/	3.42 (0.744)
3-peak-I	[0,35]	0.13 (0.014)	/	1.46 (0.200)	4.52 (0.557)
	(35,50]	0.14 (0.021)	/	1.57 (0.248)	4.30 (0.566)
	(50,70]	0.15 (0.024)	/	1.43 (0.347)	4.06 (0.512)
	(70,90]	0.15 (0.035)	/	1.41 (0.339)	3.89 (0.661)
3-peak-II	[0,35]	0.11 (0.012)	0.25 (0.047)	/	2.28 (0.118)
	(35,50]	0.11 (0.021)	0.32 (0.123)	/	2.57 (0.368)
	(50,70]	0.13 (0.048)	0.40 (0.125)	/	2.85 (0.521)
	(70,90]	0.18 (0.049)	0.40 (0.106)	/	2.65 (0.521)
4-peak	[0,35]	/	/	/	/
	(35,50]	0.13 (0.034)	0.29 (0.084)	1.27 (0.497)	3.83 (1.219)
	(50,70]	0.11 (0.025)	0.21 (0.044)	1.00 (0.277)	3.45 (0.871)
	(70,90]	0.13 (0.030)	0.30 (0.096)	1.18 (0.553)	3.06 (0.806)

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478 Figure 1



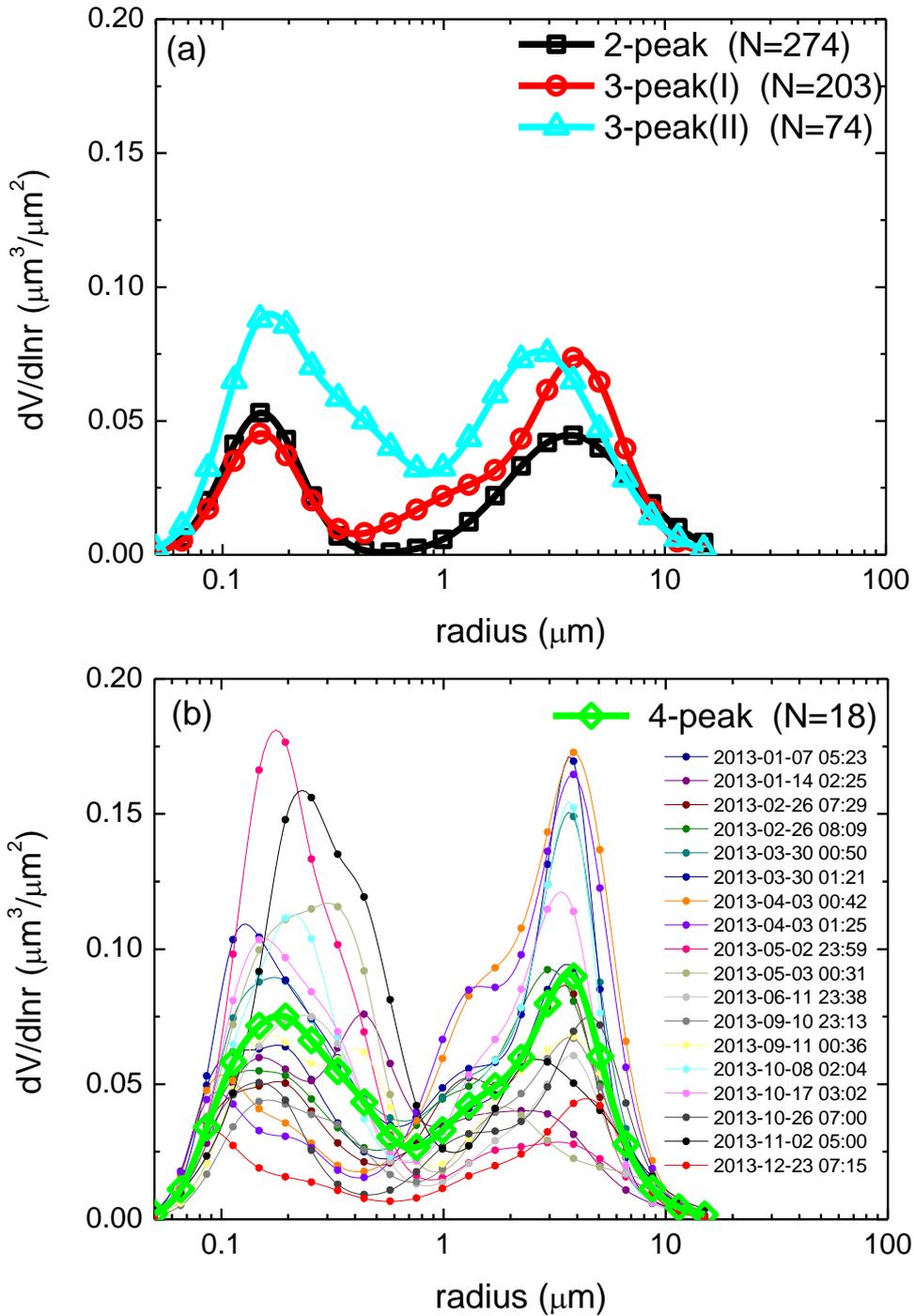
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Figure 1. Aerosol volume particle size distribution and the corresponding separation into four log-normal modes.

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Figure 2



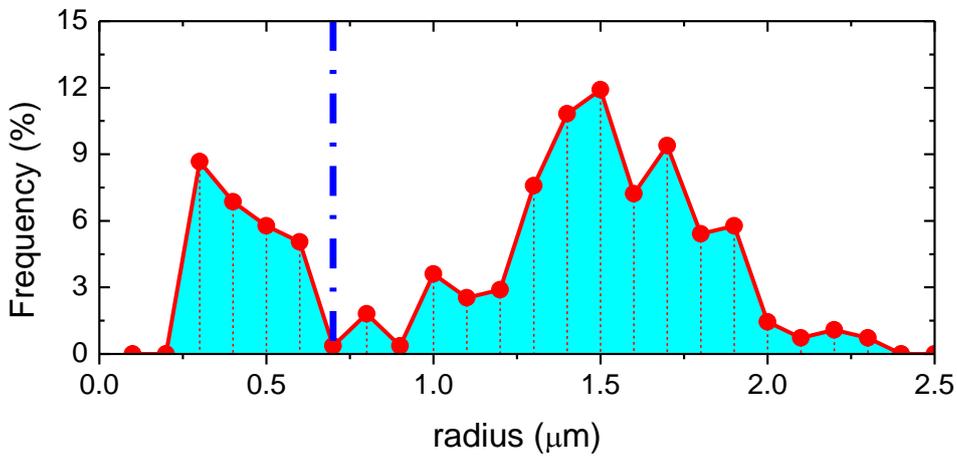
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Figure 2. The annual mean VPSD of 2-peak, 3-peak-I, 3-peak-II cases (a); and 4-peak cases (b) in Beijing 2013.

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491 Figure 3



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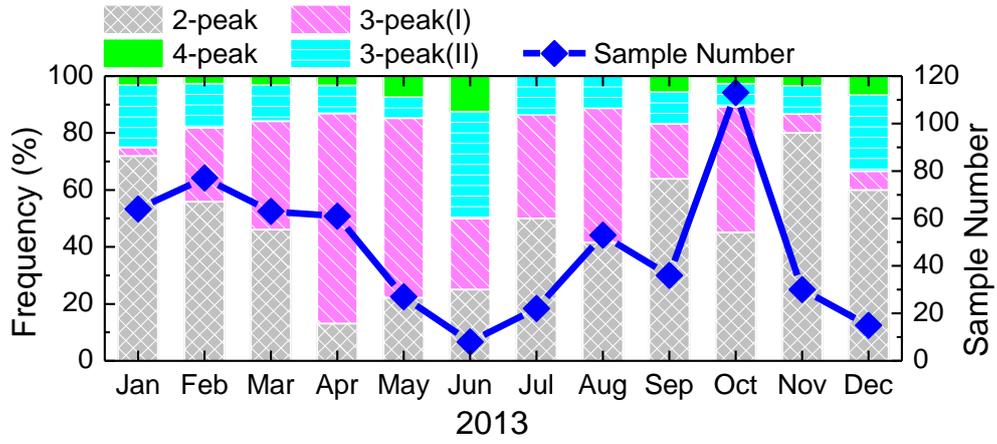
493 Figure 3. The number frequency of median radius for the extra peaks of 3-peak  
494 VPSDs.

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497 Figure 4



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499 Figure 4. The monthly percentage of multi-peak VPSDs in Beijing 2013.

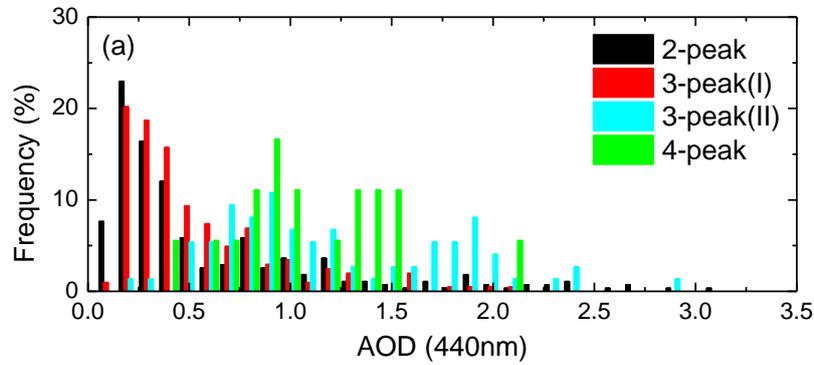
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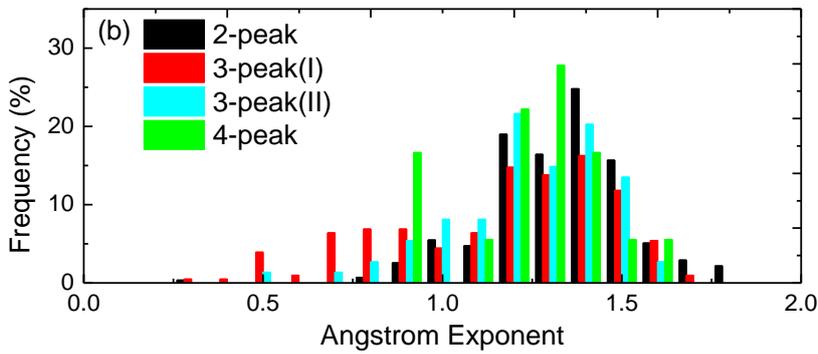
501 Figure 5

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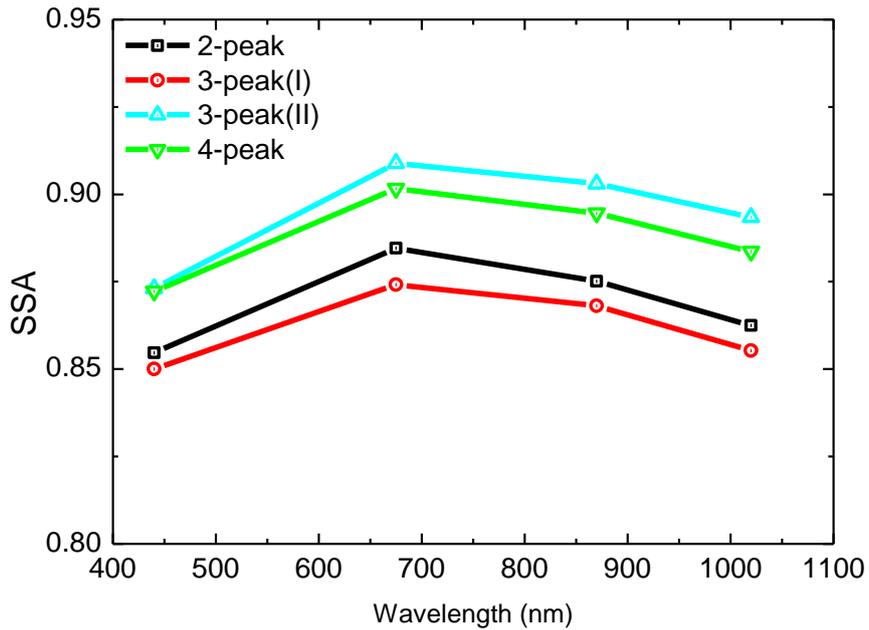
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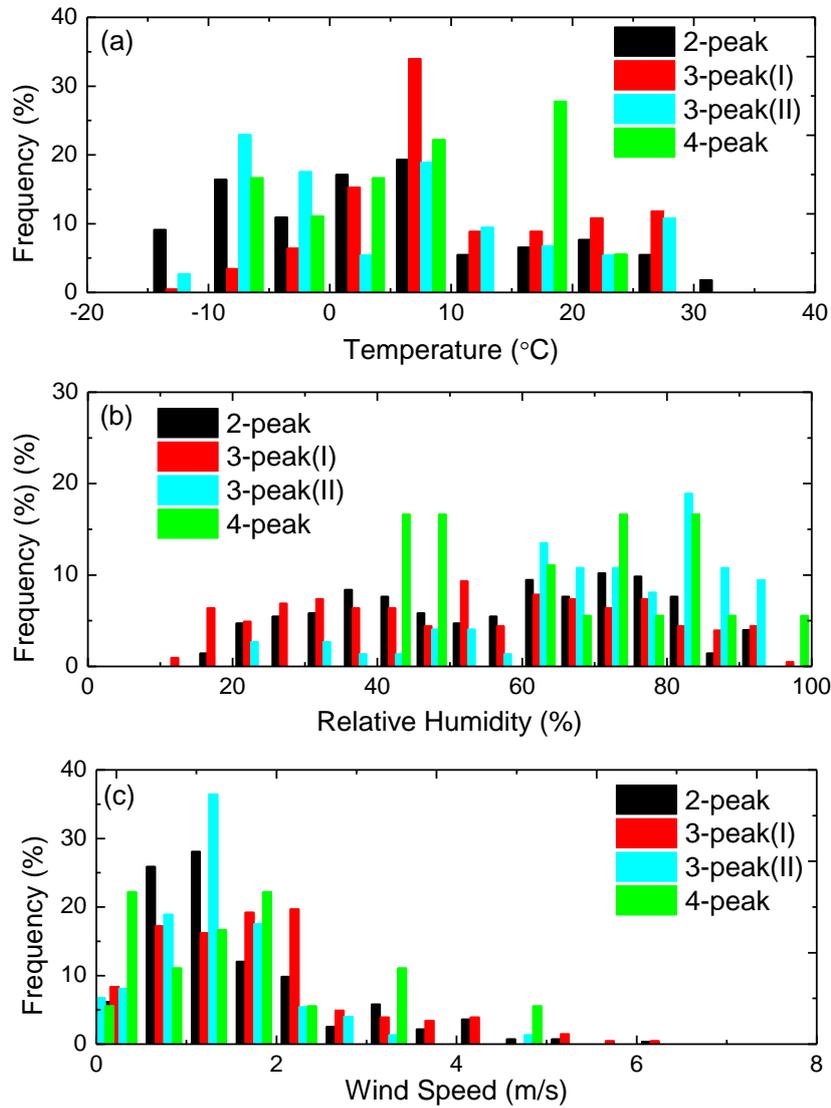
507 Figure 5. The number frequency of multi-peak VPSDs versus AOD (a), AE (b) and  
508 SSA (c). AOD: aerosol optical depth; AE: Ångström exponent; SSA: single  
509 scattering albedo; the x-axis step is 0.1 in (a) and (b).

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511 Figure 6

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516 Figure 6. The number frequency of multi-peak VPSDs versus temperature (5  $^{\circ}\text{C}$   
517 interval) (a), relative humidity (5% interval) (b) and wind speed at altitude of 10 m  
518 (0.5 m/s interval) (c).

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