



42 **1 Introduction**

43 Atmospheric aerosols are solid or liquid particles suspended in the atmosphere that can affect
44 public health, reduce near-surface visibility, decrease direct radiation from the air, and act as
45 condensation nuclei affecting cloud structure and distribution (McNeill, 2017; Rosenfeld et al., 2014).
46 Human activities have dramatically altered air quality, climate and the Earth system. The expansion
47 of urban, agricultural and industrial areas and changes in the nature of land use have increased
48 aerosol concentrations. Due to the complexity of aerosols, many observations have been carried out
49 from different perspectives. However, most of the current observations only measure the state
50 characteristics of aerosols, such as concentration, particle size distribution, and composition, and
51 what is obtained is an average characterization of aerosol properties (Krieger et al., 2012).

52 Aerosol particles in the atmosphere will follow atmospheric motion, which is manifested in the
53 uneven distribution of aerosol particle concentrations in space and time. On the one hand, the
54 unevenly distributed aerosol particles will have a corresponding effect on the light wave propagating
55 in the atmosphere, and on the other hand, we can understand the distribution characteristics of
56 aerosol particles based on the optical effect of aerosol particles and then obtain more information
57 about the transportation of aerosols.

58 Previously, Yuan et al. (2015) introduced the concepts of the atmospheric equivalent refractive
59 index (AERI) and the atmospheric equivalent refractive index structure parameter (AERISP). The
60 AERI includes real and imaginary parts, and accordingly, the AERISP also includes real and
61 imaginary parts of the structure parameters. When the working wavelength is in the atmospheric
62 transparent band, the light wave is almost not absorbed by the gas components in the atmosphere,
63 and the attenuation of the light wave is mainly caused by the extinction of aerosol particles.
64 Theoretical analysis shows that the imaginary part of the AERISP is determined by the fluctuation
65 of aerosol concentration. The real part of the AERISP corresponds to the atmospheric temperature
66 variation. Furthermore, it is assumed that the aerosol concentration variations follow the same
67 pattern as the scalar motion, which is in line with the similar theory of the surface layer, and thus,
68 the vertical transport flux of aerosol particles in the near-surface layer can be obtained by using the
69 imaginary part of the AERISP (Yuan et al., 2016; Yuan et al., 2019). AERISP observations are carried
70 out in many places, and then the aerosol flux is obtained by combining the similarity theory. However,
71 there is a lack of experimental verification of the imaginary structure parameter and aerosol flux
72 observations. Currently, the imaginary part of the AERISPs is only obtained using large-aperture
73 scintillometer (LAS) measurements, so it is necessary to carry out measurements of the imaginary
74 part of the AERISPs based on other different methods, as well as measurements of aerosol fluxes
75 based on different methods.



76 The reference to the measurement of the imaginary part of the AERISP and the aerosol vertical
77 transport flux using the light propagation principle is naturally associated with the current
78 widespread use of LASs based on the light propagation principle for the measurement of the sensible
79 heat flux in the surface layer(Moene et al., 2009). Comparative validation of this measurement is
80 obtained using eddy covariance (EC) techniques(Zhang and Zhang, 2015). EC measurements require
81 a fast response of the sensing elements for wind speed and air temperature. Similar to the validation
82 of LAS measurements of sensible heat fluxes, EC measurements were also considered for aerosol
83 vertical transport flux validation. Measurements of aerosol particle number concentration fluxes
84 using eddy covariance methods have been implemented at many locations(Gordon et al., 2011;Vogt
85 et al., 2011;Ripamonti et al., 2013). We attempted to measure the aerosol particle number
86 concentration flux using an eddy covariance system consisting of a fast-response particle counter
87 with an ultrasonic anemometer, which has a response frequency of up to 10 Hz, and simultaneously
88 measured parameters such as aerosol particle size distribution, mass concentration, forward
89 scattering coefficient, and extinction coefficient to calculate the aerosol mass concentration flux. For
90 half of the experimental period, the trend of the measurements with the two methods was the same,
91 while the other periods differed greatly (experimental results unpublished). The reason may be the
92 very weak extinction of aerosol particles with scales much smaller than the working wavelength.

93 According to the definition of the imaginary part of the AERISP, the validation work can be
94 realized by choosing an instrument for rapid measurement of the atmospheric extinction coefficient
95 and an ultrasonic anemometer to form an EC system. However, the direct and rapid measurement of
96 the atmospheric extinction coefficient is difficult to realize. The imaginary part of the AERI is
97 proportional to the atmospheric extinction coefficient and inversely proportional to the atmospheric
98 visibility, so it is possible to analyze the results of the visibility meter to obtain the imaginary part of
99 the AERISP at a certain point. However, the conventional visibility meter has a slow response and
100 low sampling frequency, which is not suitable for correlating with the vertical velocity to obtain the
101 aerosol flux, and Ren et al. (2020) improved the conventional visibility meter by obtaining visibility
102 data at 1 Hz and then using the eddy covariance method to obtain the vertical aerosol transport flux
103 based on the relationship between visibility and aerosol mass concentration(Ren et al., 2020).
104 Inspired by their work, we use the improved visibility meter in this study to obtain visibility data at
105 a higher frequency of 1 Hz and correlate the data with ultrasonic anemometer measurements to
106 potentially utilize the obtained aerosol vertical transport fluxes to achieve experimental validation
107 of the imaginary part of the AERISP and aerosol flux observations.

108 The theoretical and experimental introduction is given in the second part of the paper, the
109 experimental results are given in the third part, and the conclusions and discussion are given in the
110 fourth part.



111 2 Theoretical methods and experiments

112 The AERISP and the aerosol vertical transport flux are the topics of interest in this paper. In
113 this section, definitions and theoretical expressions for these parameters are given, as well as how
114 the measurements were carried out.

115 2.1 The imaginary part of the AERISP

116 When the beam is transmitted in the atmosphere, in addition to the unevenly spatial distribution
117 of the beam energy due to the refraction diffraction phenomenon caused by the fluctuation of the
118 refractive index caused by the fluctuation of temperature, the beam energy is also attenuated by the
119 absorption and scattering of aerosol particles. Therefore, the atmosphere and aerosol particles can
120 be taken as a whole, called the equivalent medium; thus, the equivalent refractive index n_{equ} concept
121 is introduced (van de Hulst, 1957; Yuan et al., 2021),

$$122 \quad n_{equ} = n_m + i \frac{2\pi}{\eta^3} \int_0^\infty S(0) \frac{dN}{dD} dD \quad (1)$$

123 where n_m is the refractive index of atmospheric molecules, η is the wavenumber of light waves, and
124 i denotes an imaginary number. $S(0)$ is the forward scattering function (0 in parentheses is the
125 scattering angle). N is the number of aerosol particles per unit volume, and dN/dD is the size
126 distribution of aerosol particle sizes.

127 The equivalent refractive index consists of real and imaginary parts denoted by n_{re} and n_{im} ,
128 respectively, i.e., $n_{eff} = n_{re} + in_{im}$. The real part is the refractive index of the molecule, and the
129 imaginary part is.

$$130 \quad n_{im} = \frac{2\pi}{\eta^3} \int_0^\infty \text{Re}[S(0)] \frac{dN}{dD} dD \quad (2)$$

131 It is known that the atmospheric extinction coefficient has a similar form(Liou, 2002):

$$132 \quad \beta_{ext} = \frac{4\pi}{\eta^2} \int_0^\infty \text{Re}[S(0)] \frac{dN}{dD} dD \quad (3)$$

133 From Eqs (2) and (3), we can see that

$$134 \quad n_{im} = \lambda \beta_{ext} / 4\pi \quad (4)$$

135 where λ is the working wavelength ($\lambda = 2\pi/\eta$). Based on the relationship between the aerosol extinction
136 coefficient and visibility (V) ($V = 3.912/\beta_{ext}$), the aerosol extinction coefficient can be deduced from
137 visibility measurements, and then the imaginary part of the AERI can be obtained based on Eq. (4).

138 Experiments show that the temperature fluctuation satisfies the turbulence "2/3" law(Liu et al.,
139 2017), and according to the relationship between temperature and the real part of the AERI, the
140 fluctuation of the real part of the AERI also satisfies the turbulence "2/3" law; thus, we can define
141 the structure parameter of temperature, C_T^2 , and the real part of the AERISP $C_{n,re}^2$. Therefore, general
142 scalars can be extended, such as the fluctuation of the imaginary part of the AERISP and the



143 fluctuation of the atmospheric extinction coefficient. Thus, we can assume that the imaginary part
 144 of the AERI satisfies the turbulence "2/3" law.

$$145 \quad D_{n,Im}(r) = \overline{[n_{Im}(\bar{x}) - n_{Im}(\bar{x} + \vec{r})]^2} = C_{n,Im}^2 r^{2/3} \quad (5)$$

146 Thus, we can introduce the imaginary part of AERISP $C_{n,Im}^2$, a parameter used to describe the
 147 fluctuation intensity of the imaginary part of the AERI. Correspondingly, we can introduce the
 148 structure parameter of the atmospheric extinction coefficient $C_{\beta_{ext}}^2$ and the structure parameter of the
 149 fluctuation of the aerosol mass concentration C_M^2 .

150 **2.2 Two methods of equivalent refractive index structure** 151 **parameter measurement**

152 From the definition of the AERISP in the last part and the relationship between the AERI and
 153 the extinction coefficient, it can be seen that the AERISP has an important influence on light
 154 propagation in the atmosphere, and thus the AERISP can be estimated from the light intensity
 155 fluctuation in light propagation and the monitoring of the extinction coefficient. The section
 156 describes how to measure the AERISP with the help of two methods.

157 **2.2.1 Long-Path Light Propagation Methods**

158 When an approximately collimated light beam in the transparent band of the atmosphere is
 159 selected and propagated over a distance, the light intensity at the receiving end fluctuates. The
 160 fluctuation in light intensity has two causes: one is the uneven distribution of the real part of the
 161 AERI caused by the temperature fluctuation, and the other is the uneven distribution of the imaginary
 162 part of the AERI caused by the uneven distribution of aerosol particles. Assuming that the above two
 163 causes are not related, they can be decomposed. Theoretical analysis shows that the power spectral
 164 density expression for the part of the contribution of the inhomogeneous distribution of the
 165 imaginary part of the AERI to the fluctuation of the light intensity is (Yuan et al., 2015).

$$166 \quad W_{InI,Im}(f) = 64\pi^2\eta^2 \int_0^L dx \int_{2\pi f/v}^{\infty} \Phi_{n,Im}(\kappa) \cos^2\left[\frac{\kappa^2 x(L-x)}{2\eta L}\right] [(\kappa v)^2 - (2\pi f)^2]^{-1/2} \cdot$$

$$167 \quad \left[\frac{2J_1\left(\frac{D_r \kappa x}{2L}\right)}{D_r \kappa x / 2L}\right]^2 \left[\frac{2J_1\left(\frac{D_r \kappa(L-x)}{2L}\right)}{D_r \kappa(L-x) / 2L}\right]^2 \kappa d\kappa \quad (6)$$

168 where f is the frequency of the log-intensity spectrum, η is the wavenumber of the spherical wave
 169 ($\eta=2\pi/\lambda$, λ is light wavelength), x is the position of the propagating wave, L is the length of the
 170 propagation path, κ is the wavenumber of the two-dimensional log-intensity spectrum, and $\Phi_{n,Im}$ is
 171 the spectrum of the imaginary parts of the refractive index, where the subscript n denotes the
 172 refractive index and the subscript Im denotes the imaginary parts of the refractive index. The widely
 173 used von Karman spectral form for $\Phi_{n,Im}$ is adopted in this study (Andrews and Phillips, 2005),
 174 which can be expressed as follows:



$$175 \quad \Phi_{n,\text{Im}}(\kappa) = 0.033 C_{n,\text{Im}}^2 \left(\kappa^2 + \frac{1}{L_0^2} \right)^{-\frac{11}{6}} e^{-\frac{\kappa^2 l_0^2}{5.92^2}} \quad (7)$$

176 Here, L_0 is the outer scale of turbulence, and l_0 is the inner scale of turbulence.

177 Substituting Eq. (7) into Eq. (6) and integrating the right-hand side of Eq. (6) yields,

$$178 \quad W_{\text{InI,Im}}(f) = 0.129 C_{n,\text{Im}}^2 \eta^2 L \nu^{5/3} \left[f^2 + \left(\frac{\nu}{2\pi L_0} \right)^2 \right]^{-4/3} \quad (8)$$

179 Using Eq. (8), the imaginary part of the AERISP can be determined based on the shape of the
 180 spectrum while being constrained by the low-frequency variance of light intensity fluctuation from
 181 the imaginary part of AERISP.

182 To carry out a comparative analysis with the results of the real part of the AERISP, the
 183 expression for the power spectral density of the logarithmic light intensity fluctuation due to the real
 184 part of the AERI is also given here as (Clifford, 1971; Nieveen et al., 1998),

$$185 \quad W_{\text{InI,Re}}(f) = 64 \pi^2 \eta^2 \int_0^L dx \int_{2\pi f/\nu}^{\infty} \Phi_{n,\text{Re}}(\kappa) \sin^2 \left[\frac{\kappa^2 x(L-x)}{2\eta L} \right] [(\kappa \nu)^2 - (2\pi f)^2]^{-1/2} \cdot$$

$$186 \quad \frac{2J_1 \left(\frac{D_r \kappa x}{2L} \right)}{D_r \kappa x / 2L} \left[\frac{2J_1 \left(\frac{D_t \kappa(L-x)}{2L} \right)}{D_t \kappa(L-x) / 2L} \right]^2 \kappa d\kappa \quad (9)$$

187 Integrating Eq. (9) yields the fluctuation variance of the log light intensity as

$$188 \quad \sigma_{\text{InI,Re}}^2 = \int_0^{\infty} W_{\text{InI,Re}}(f) df = 0.89 C_{n,\text{Re}}^2 L^3 D_t^{-7/6} D_r^{-7/6} \quad (10)$$

189 The real part of AERISP is usually calculated using Equ. (10) (Wang et al., 1978).

190 **2.2.2 Spectral analysis methods for single-point** 191 **measurements**

192 Aerosol particles follow atmospheric motion, which is consistent with general atmospheric
 193 motion characteristics, and the "-5/3" law can be used to characterize the fluctuations in aerosol-
 194 related properties. Therefore, in the inertial subregion, the extinction coefficient power spectral
 195 density is

$$196 \quad S_{\beta_{\text{ext}}}(f) = (2\pi/U) S_{\beta_{\text{ext}}}(\kappa) = 0.25 C_{\beta_{\text{ext}}}^2 (2\pi/U)^{-2/3} f^{-5/3} \quad (11)$$

197 The extinction coefficient structure parameter $C_{\beta_{\text{ext}}}^2$ can be converted to the imaginary part of
 198 the AERISP according to equation (4). The coefficient in Eq. (11) is 0.25 (Wyngaard et al., 1971). It
 199 has been suggested in the literature that the coefficient for the spectral density should be 0.125 (Gibbs
 200 and Fedorovich, 2020). The difference between the two coefficients 0.25 and 0.125 is whether the
 201 integral of the spectral density is equal to the variance or half of the variance. If the integral of the
 202 spectral density is equal to the variance, the coefficient of 0.25 is taken; if the integral of the spectral
 203 density is equal to half of the variance, the coefficient is taken as 0.125. According to the spectral
 204 density curve, the coefficients are determined within the inertial subregion, and then the structure



205 parameters $C_{\beta_{ext}}^2$ can be obtained. According to the relationship between the extinction coefficient
206 and the imaginary part of the AERI in Eq. (4), the imaginary part of the AERISP can be obtained as
207 $C_{n,Im}^2$.

208 Similarly, power spectral density profiles with temperature fluctuations that

$$209 \quad S_T(f) = (2\pi/U)S_T(\kappa) = 0.25C_T^2(2\pi/U)^{-2/3}f^{-5/3} \quad (12)$$

210 The actual temperature turbulence spectral density profile often takes the form of a von Karman
211 spectrum as

$$212 \quad S_T(f) = 0.25C_T^2(2\pi/U)^{-2/3} \left(f^2 + \left(\frac{U}{2\pi L_0} \right)^2 \right)^{-5/6} \quad (12')$$

213 Based on the relationship between temperature and the real part of the AERI, we have

$$214 \quad C_n^2 = C_T^2 / R_{TN}^2 \quad (13)$$

215 where R_{TN} denotes the coefficient of proportionality between the change in the real part of the AERI
216 and the change in atmospheric temperature.

$$217 \quad R_{TN} = \frac{dT}{dn_{Re}} = -1.29 \times 10^4 \times \left(1 + \frac{7.52 \times 10^{-3}}{\lambda^2} \right)^{-1} \frac{\bar{T}^2}{\bar{P}} \quad (14)$$

218 where the wavelength λ is in microns, the atmospheric pressure P is in hectopascals, and the
219 temperature T is in kelvin.

220 The real part of the AERISP can be obtained by fitting the experimental data using Eqs. (12) or
221 (12').

222 **2.3 Flux estimation**

223 The method for estimating the AERISP was given in the former sections. The purpose of
224 estimating the AERISP in this paper is to estimate the aerosol flux in the near-surface layer. Here,
225 the method of estimating the aerosol flux based on the AERISP will be given first, and then the
226 method of estimating the aerosol vertical transport flux based on the EC technique will be introduced.

227 **2.3.1 Optical transmission method**

228 Experiments have shown that the AERISPs satisfy the theory of surface layer similarity; thus,
229 (Yuan et al., 2019)

$$230 \quad F_{a_LAS} = \left(\frac{C_{n,Im}^2}{C_{n,Re}^2} \right)^{1/2} \frac{R_{MN}}{R_{TN}} u_* |T_*| \quad (15)$$

231 where u_* is the friction velocity and T_* is the characteristic potential temperature. These two
232 parameters can be determined from wind speed and temperature profiles. The real and imaginary
233 parts of the AERISP are determined from LAS measurements. R_{MN} can be obtained from aerosol
234 mass concentration and visibility measurements ($R_{MN} = M/n_{Im}$, where M is the aerosol mass
235 concentration approximated as PM_{10} and n_{Im} can be determined from visibility measurements)(Yuan



236 et al., 2021), and R_{TN} can be calculated from mean air temperature and other measurements using
237 Eq. (14) again.

238 When turbulence of the surface layer is developed, Eq. (15) can be approximated as (Yuan et al.,
239 2019),

$$240 \quad F_{a_LAS} = a \left(\frac{g}{T} \right)^{1/2} R_{TN}^{1/2} (C_{n,Re}^2)^{1/4} R_{MN} (C_{n,lm}^2)^{1/2} (z-d) \quad (16)$$

241 Here, a is the scale factor with a theoretical value of 0.567 (which needs to be determined by
242 comparative experiments), g is the gravitational acceleration, z is the scintillator height, and d is the
243 zero-plane displacement. Equation (16) does not require measurement of u^* and T^* data. Generally,
244 the measurement heights are high, and the assumption of developed turbulence in the surface layer
245 is easily met during the day under unstable conditions.

246 **2.3.2 Based on single-point eddy covariance**

247 Eddy covariance is a commonly used method for the measurement of Earth air exchange fluxes
248 in the near-surface layer. Using rapid measurements of vertical wind speed and extinction coefficient
249 to obtain the ups and downs of the vertical wind speed and extinction coefficient, the expression for
250 the vertically transported aerosol flux calculated by the eddy-covariance method with the mean
251 vertical velocity close to zero is given by (Wilczak et al., 2001)

$$252 \quad F_{a_EC} = R_{MN} \frac{\lambda}{4\pi} \overline{w' \beta'_{ext}} \quad (17)$$

253 The prime' in Eq. (17) denotes fluctuation.

254 **2.4 Introduction to the experiment**

255 The experiments were performed on the campus of the University of Science and Technology
256 of China (USTC) in Hefei, Anhui Province, China. The campus of the University of Science and
257 Technology of China (USTC) is located in downtown Hefei. Figure 1a gives part of the Hefei city
258 area, where the red rectangle corresponds to Fig. 1b, the campus of USTC. The campus is surrounded
259 by four highways, and the two highways in the west and north have more vehicles, especially the
260 viaduct in the west. The campus is composed of vegetation, roads and teaching buildings. As can be
261 seen in Fig. 1b, green vegetation covers most of the campus. The roofs of the school buildings are
262 almost on a plane with the tree canopy and are approximately 17 meters above the ground. The zero
263 plane displacement in Eq.(16) is taken 17 meters. There are two tall buildings (T and R in Fig. 1b)
264 at the southernmost and northernmost parts of the campus, and the distance between the two
265 buildings is approximately 960 meters. The experiment consists of two parts: one part is to carry out
266 the light propagation experiment using a self-developed large aperture scintillator (LAS), and the
267 other part is to carry out the measurement using the instruments on the meteorological tower in the
268 middle of the beam. The transmitting end of the LAS was installed on the 12th floor of the
269 southernmost building (T in Fig. 1b), the receiving end was installed on the 12th floor of the
270 northernmost building (R in Fig. 1b), and the distance of the beam from the ground was
271 approximately 35 meters. The apertures of the transmitting and receiving ends were 250 mm. The
272 sampling frequency of the receiving end was 500 Hz, and a data file was saved every 30 minutes.
273 The height of the meteorological tower is 18 meters above the roof of a teaching building (P in Fig.



274 1b). The top of the meteorological tower is equal to the height of the beam. The meteorological tower
275 is equipped with 5 layers of wind speed and direction temperature and humidity measurement
276 sensors. At the top of the tower, there is a radiation quadrature sensor, and at the bottom of the tower,
277 there is a rainfall measurement sensor. In this paper, we use data from the top 18-meter height
278 position of the meteorological tower with sensors installed for conventional meteorological
279 parameters, including temperature, humidity, wind speed, and wind direction radiation.
280 Conventional meteorological data are collected at 1-second intervals, average data are obtained
281 every half hour after data collection, and precipitation data are recorded every half hour. A three-
282 dimensional sonic anemometer thermometer was installed at the top of the tower, and a high-
283 frequency sampling visibility sensor CS120A (Campbell, 2012) was upgraded to have the ability to
284 obtain a 1-Hz visibility (Ren et al. 2020). The three-dimensional sonic anemometer thermometer can
285 obtain a sampling frequency of over 10 Hz and is a common instrument used in atmospheric
286 turbulence research; as such, we will not introduce it in depth. To correlate the vertical wind speed
287 with the extinction coefficient to obtain the aerosol flux, the data collected by the sonic anemometer-
288 thermometer at 10 Hz were averaged to obtain 1-Hz data, which were saved in a data file.

289 The time period of the experiment is January 9-23, 2022, a total of 15 days.

290 **2.5 Data quality control**

291 Data obtained from field observations need to be controlled for data quality before further processing
292 (Foken and Wichura, 1996). This study involves several types of data, mean variables, cumulative
293 variables, and fluctuating variables. Mean variables include 30-minute averages of temperature,
294 humidity, wind speed, wind direction, and global radiation. Data quality control for mean variables
295 was performed by comparing measurements at different heights or different sites. The same variables
296 with the same trend at different heights and different locations were considered to be high quality
297 data. All measured mean data were determined to be satisfactory. Cumulative variables refer to 30-
298 minute rainfall data. Rainfall data were qualified with reference to relative humidity, total radiation
299 and air temperature. Fluctuating data included 10-Hz ultrasonic anemometer data and 1-Hz visibility
300 data, as well as high-frequency intensity fluctuation data measured by the LAS, the real and
301 imaginary parts of the AERISP, and calculated aerosol fluxes. Quality control consisted mainly of
302 eliminating spikes and replacing missing data.

303 The reason for the spike points in the light intensity fluctuation data is that the received signal
304 jumps when there are flying birds and other obstructions to the optical signal on the propagation path.
305 This situation is automatically determined by the program. When this occurs, the data for that time
306 period will not be processed. The AERISP and aerosol flux data are judged according to (a) three
307 times the standard deviation (SD) from the mean value and (b) three times the standard deviation
308 from the mean of differences between adjacent moment data. To determine the three times standard
309 deviation from the mean value, the trend is obtained by averaging over a two-hour period, then
310 calculating the difference between the measured value and the trend at each moment, calculating the
311 mean and variance of the difference, and considering a spike point if the difference is outside the 3
312 times standard deviation. The 3 times standard deviation of adjacent differences is determined by
313 first calculating the difference between adjacent moments and then calculating the mean and standard
314 deviation of the difference. Any data that deviates from the mean by more than 3 times the standard
315 deviation is considered a spike point.



316 Data judged to be a spike will be supplemented by the average of adjacent moments. Of course,
317 data processed according to this method appear to be completely missing for longer time periods.
318 For such cases, no further methods to realize supplementation are considered in this paper. There are
319 other errors in measurements made with the LAS that are due to specific reasons (Moene et al., 2009);
320 for example, the effect of spectral shape deviations using the von Karman model and intermittent
321 variations in the properties of this spectrum on the LAS signal is not considered in this study.

322 Similar to CO₂ flux calculation, EC calculations for aerosol flux were performed to obtain
323 aerosol fluxes, and several data quality control work was conducted, such as coordinate system
324 rotations (Wilczak et al., 2001; Yuan et al., 2011), WPL corrections (Webb et al., 1980).

325 **3 Experimental results**

326 In the following, the variation curves of conventional meteorological parameters during the
327 experimental period, individual examples of AERISPs, a comparison of the two methods for the
328 results of multiday continuous observations and a comparison of the two methods for the results of
329 flux measurements will be presented to verify the reliability of the means of light propagation
330 measurement.

331 **3.1 General meteorological parameters and extinction** 332 **coefficients**

333 The variation curves of conventional meteorological parameters during the experiment,
334 including temperature, humidity, wind speed, wind direction, radiation and precipitation, and
335 extinction coefficient are shown in Fig. 2, where the extinction coefficient is calculated from the
336 visibility ($\beta_{ext} = 3.912/V$, V denotes visibility). Seven days during the experiment were sunny, and
337 four of the remaining eight days had rainfall. The temperatures on sunny days were characterized by
338 significant daily variations, with a minimum temperature of 0.4°C, and the maximum diurnal
339 temperature difference could reach more than 9°C. Relative humidity exceeded 80% for only a few
340 periods during the sunny days. The wind speed was generally less than 3 m/s, and there were only
341 very few periods of north wind with a speed of more than 3 m/s. There was no obvious prevailing
342 wind direction during the experimental period, and only the north wind was equivalent to the other
343 directions with a slight predominance. The meteorological conditions during the experiment were
344 similar to those of the local winter season. The extinction coefficient curve with time during the
345 experiment is given in Fig. 2(g). The pollution gradually increased from the 9th to Jan. 13th and
346 decreased on the 13th; from the 14th to the 20th, the pollution gradually increased and decreased on
347 the 20th. The meteorological conditions during the experimental period can be considered typical.

348 **3.2 Example results from measurements of the imaginary part** 349 **of the AERISP**

350 Before carrying out the comparison of the measurement results of the two methods for obtaining
351 the AERISP, the comparison of the measurement results of an individual example is given. The
352 experimental data measured from 2022-01-16 13:00-13:30 will be used here as an example to
353 illustrate the calculation of the AERISP, and the results will be given. This time period is midday on



354 a clear day (shown in Fig. 2e), and both the total radiation and sensible heat fluxes are large, so this
355 time period can be taken as a good typical example.

356 **3.2.1 Structure parameters obtained by light propagation**

357 The AERISP is first given using the light propagation method. The sequence of light intensity
358 signals obtained at the receiving end is shown in Fig. 3a. The time duration is 2022-01-16 13:00-
359 13:30, and the sampling frequency is 500 Hz, so there are 900000 data points in the time series of
360 light intensity fluctuation in Fig. 3a. The curve has both low- and high-frequency fluctuations. Using
361 spectral analysis and correlation analysis, the variance of the low-frequency part of the logarithmic
362 light intensity is $1.08e-4$, and the variance of the high-frequency part is $5.06e-4$. The solid dots in
363 Fig. 3b are the measured spectral densities of the logarithmic light intensity fluctuations, and the
364 black dashed lines and solid lines represent the results calculated by Eqs. (6) and (9), respectively,
365 and represent the contributions of the imaginary part and the real part. As seen from the power
366 spectral density curves of the logarithmic light intensity fluctuations in Fig. 3b, the high-frequency
367 part and the low-frequency part have different characteristics.

368 In the logarithmic plot, the low-frequency part is prominent with a much higher spectral density
369 than the high-frequency part. Theoretical analysis shows that the low-frequency part corresponds to
370 the contribution of the imaginary part of the AERISP. The high-frequency part is flat plus high-
371 frequency attenuation. The high-frequency part corresponds to the contribution of the real part. The
372 part greater than 100 Hz is noise.

373 Based on the previous theoretical approach, the spectral density fitting for the low-frequency
374 part, while constrained by the low-frequency variance, yields an equivalent refractive index structure
375 parameter of $1.14 \times 10^{-25} m^{-2/3}$. Correspondingly, the structure parameter of the real part of the
376 refractive index, based on the high-frequency variance, is obtained as $2.54 \times 10^{-14} m^{-2/3}$.

377 **3.2.2 Obtaining the imaginary part of the AERISP based on** 378 **the spectrum**

379 The coefficients of the power spectral density curves are proportional to the refractive index
380 structure parameters, from which they can be determined. The extinction coefficient structure
381 parameter can be deduced from the power spectral density of the extinction coefficient fluctuation,
382 and the temperature structure parameter can be deduced from the power spectral density of the
383 temperature fluctuation. The extinction coefficient (Fig. 4a) and temperature (Fig. 4b) fluctuation
384 with time for the period 2022-01-16 13:00-13:30 are given in Fig. 4. As shown in Fig. 4, the
385 extinction coefficient curve has more noise, while the temperature curve has less noise. On the
386 temperature fluctuation curve, there are five distinct ramp structures.

387 Power spectral analysis of the data in Fig. 4 was carried out to obtain the power spectral density
388 in Fig. 5. From the extinction coefficient power spectral density curve in Fig. 5a, it can be seen that
389 spectral densities greater than 0.05 Hz exhibit noise, and spectral densities less than 0.05 Hz have
390 inertial subregions. When practically analyzed, the inertial subregion ranges from 0.002 Hz to the
391 noise onset frequency. The motion of aerosol particles in the atmosphere conforms to the "-5/3" law
392 of turbulence. The extinction coefficient structure parameter was obtained by fitting the data in the



393 inertial subregion using Eq. (11) with a value of $3.9 \times 10^{-11} \text{m}^{-2} \text{m}^{-2/3}$, which was then converted
394 to the structure parameter of the imaginary part of the refractive index of $1.04 \times 10^{-25} \text{m}^{-2/3}$.

395 Correspondingly, as seen from the temperature fluctuation power spectrum curve in Fig. 5b,
396 almost no noise appears, which is mainly due to the small noise of the temperature signal itself,
397 while the 1 Hz temperature data here are obtained by averaging the data collected at 10 Hz. The
398 temperature structure parameter of $0.0218^\circ\text{C}^2 \text{m}^{-2/3}$ is obtained by fitting using Eq. (12), which is
399 converted to a refractive index real part structure parameter of $2.1 \times 10^{-14} \text{m}^{-2/3}$.

400 The imaginary part of the AERISP obtained by using a visibility meter and the real part of the
401 AERISP obtained by an ultrasonic anemometer are in good agreement with the previous results given
402 by using optical propagation means.

403 **3.3 Comparison of all results for the AERISP**

404 The previous section gives an individual example. A comparison of all the data during the
405 experiment is given below, as shown in Figs 6 and 7.

406 A comparison of the time series of AERISPs measured by the two methods is given in Fig. 6,
407 where Fig. 6a shows the time series of the imaginary part of the AERISP and Fig. 6b shows the time
408 series of the real part of the AERISP. It can be seen that there are large fluctuations in the imaginary
409 part of the AERISP during the experimental period. This trend is close to that of the aerosol
410 extinction coefficient. From Fig. 6a, it can be seen that there is no obvious daily variation
411 characteristic. The trend agreement of the results obtained by the two methods is very good. From
412 Fig. 6b, it can be seen that the real part of the AERISP on sunny days has obvious daily variation
413 characteristics, which are large during the day and small at night. The agreement of the results
414 obtained by the two methods is good during the daytime, and at night, the results obtained by the
415 light propagation method are larger than those of the large point measurements.

416 Scatter plots of the results of the measurements of the two methods are given in Fig. 7. Figure
417 7a shows the scatter plot of the results of the two methods for the imaginary part of the AERISP with
418 a correlation coefficient R^2 of 0.73, while Fig. 7a shows the scatter plot of the results of the two
419 methods for the real part of the AERISP with a correlation coefficient of real R^2 of 0.62. This shows
420 that the correlation coefficients of the imaginary structure parameters obtained by the two methods
421 are larger than those of the real structure parameters. The reason for the smaller correlation
422 coefficient of the real part of the AERISP is that the difference in the real part of the AERISP obtained
423 by the two methods at night is larger. This shows that the aerosol spatial distribution at night may be
424 more homogeneous than the temperature distribution. The reason for this may be that the temperature
425 distribution in the overlying surface of the campus at night is not uniform, and weak turbulence does
426 not produce strong mixing, resulting in a non-uniform distribution of the real part of the AERISP.
427 There are no strong aerosol emission sources on the nighttime campus, so the distribution of the
428 imaginary part of the AERISP behaves more uniformly.

429 **3.4 Velocity-extinction coefficient correlation for a single point**

430 To calculate aerosol fluxes using EC techniques, delayed correlation of vertical velocity and
431 extinction coefficient is needed. The delayed correlation curves of the vertical velocity and extinction
432 coefficient is given in Fig. 8.



433 The horizontal coordinate of the delay correlation curve in Fig. 8 is the delay time τ and the
434 vertical coordinate is the delay correlation. From Fig. 8, it can be seen that at $\tau = -2$ s, the correlation
435 curve has an obvious extreme value, which is also the minimum value of the delay time in duration
436 of 300 s. The minimum value is $-5.22e-6m^{-1}$. The reason that the extreme value of the correlation
437 curve does not appear at 0 s is that there is a distance of about 0.20 m between the sensing element
438 of the visibility meter and that of the ultrasonic anemometer. Here are given the cases with obvious
439 extremes, and there are some cases where no obvious extremes appear. In such cases where there are
440 no significant extremes, the value associated with a delay time of 0 seconds is taken.

441 3.5 Flux

442 The AERISP was given in the former part, and the aerosol vertical transport flux can be
443 estimated for the duration of 2022-01-16 13:00-13:30 according to Eq. (16),

$$444 F_{a,LAS} = 0.567 * \left(\frac{9.8}{283}\right)^{\frac{1}{2}} * (1.01 \times 10^6)^{\frac{1}{2}} * (2.54 \times 10^{-14})^{\frac{1}{4}} * 6216 * (1.14 \times 10^{-25})^{1/2} * 18 * 10^9 =$$
$$445 1.60 \mu g m^{-2} s^{-1} \quad (18)$$

446 Where $a=0.567$, $T=283K$, $g=9.8m/s^2$, $R_{TN} = 1.01 \times 10^6 K$, $C_{n,Re}^2 = 2.54 \times 10^{-14} m^{-2/3}$, $R_{MN} =$
447 $6216 Kg \cdot m^{-3}$ (Yuan et al., 2015), $C_{n,Im}^2 = 1.14 \times 10^{-25} m^{-2/3}$, $z=35m$, $d=17m$.

448 Similarly, the aerosol flux is obtained from the eddy covariance method according to Eq.(17)

$$449 F_{a,EC} = -0.522 \times 10^{-6} * 6216 * 10^9 * \frac{0.65 \times 10^{-6}}{4\pi} = -1.67 \mu g m^{-2} s^{-1} \quad (19)$$

450 Where $\overline{w'\beta_{ext}'} = -0.522 \times 10^{-6} s^{-1}$, $R_{MN} = 6216 Kg \cdot m^{-3}$, $\lambda = 0.65 \times 10^{-6} m$.

451 From the previous calculations, we can see that during the half hour 2022-01-16 13:00-13:30,
452 the absolute values of the aerosol fluxes obtained by the two methods are very close, but of opposite
453 sign. Since the LAS method based on light propagation cannot determine the direction of flux
454 transport, only the magnitude of the flux can be given. This is similar to the fact that the estimation
455 of surface sensible heat fluxes using a LAS also gives only the magnitude but not the direction. There
456 are some judgments for estimating the direction of sensible heat flux using a LAS, such as those
457 based on sunrise and sunset times and atmospheric stability (Zhao et al., 2018). Here, the flux is
458 negative to indicate the deposition of aerosol particles. Because the experimental site is a campus,
459 there is almost no source of aerosol particle emission in the overlying surface, which is manifested
460 as a sink of aerosol particles inside the city. Therefore, the direction of aerosol flux measurement
461 based on the LAS needs to be judged based on the nature of the subsurface.

462 The results of aerosol flux calculations throughout the experiment, except two days of rain, the
463 22nd and 23rd. are given in Fig. 9. Figure 9a shows the absolute values of the aerosol vertical
464 transport fluxes measured by the two methods based on the imaginary part of the AERISP and EC
465 methods, and Fig. 9b shows the aerosol vertical transport fluxes with sign for transport direction
466 measured, which correspond to the rectangle-point line in Fig. 9a. The trend of aerosol fluxes
467 obtained by the two methods given in Fig. 9a is consistent with a diurnal variation in aerosol fluxes
468 on sunny days, with larger values of aerosol fluxes at noon. At night, the aerosol flux values are
469 smaller. Based on a comparison between Fig. 9a and Fig. 9b, it is shown that the aerosol flux is
470 negative at noon on clear days, indicating that the turbulence is strong at noon, which enhances the
471 downward transport of aerosol particles.



472 This study was conducted in a campus with no emission sources, and the downward flux is
473 reasonable; in fact, there was an upward flux measured by the EC method if there were emission
474 sources in the observation area (Ren et al. 2020).

475 **4 Conclusion and discussion**

476 To validate the previously developed method of measuring the AERISP and the aerosol mass
477 flux, this paper theoretically organizes the concept of the AERISP, introduces two methods of
478 measuring the AERISP and estimating the aerosol vertical transport flux by using the AERISP and
479 EC methods, and carries out field observation experiments in an urban area. The experimental results
480 show that the AERISPs estimated by the two methods are in good agreement, and the aerosol vertical
481 transport fluxes obtained by the two methods based on the AERISP and EC are in good agreement.

482 According to the experimental results, the imaginary part of the AERISP expresses the intensity
483 of the fluctuation of the attenuation of light during transmission. When the air-transparent band is
484 used, the imaginary part of the AERISP characterizes the intensity of the fluctuation of the extinction
485 coefficient of the aerosol.

486 The aerosol flux is related to both the fluctuations in aerosol concentration and the intensity of
487 atmospheric turbulence. When there is an aerosol emission source on the overlying surface, the
488 aerosol flux is positive, transporting aerosol particles upwards. When there is no aerosol emission
489 source in the overlying surface, the overall performance is aerosol particle deposition and
490 downwards transport of flux. In general, urban green lands are areas of aerosol particle deposition,
491 while ocean and desert surfaces can often be viewed as source areas for aerosols.

492 From the experimental results, we can also see that, as a comparison, this paper also gives
493 results for the temperature refractive index structure parameters, and as seen in Fig. 6, the trends for
494 the structure parameters in the real and imaginary parts of the AERISP are different, indicating that
495 temperature fluctuations and aerosol concentration fluctuations are uncorrelated. The purpose of this
496 paper is to illustrate the physical significance of the structure constants of the imaginary part of the
497 AERISP obtained using the LAS technique and to obtain the aerosol vertical transport flux based on
498 the AERISPs. In inverting the imaginary part of the AERISP using the light propagation principle,
499 there is an assumption that the aerosol concentration fluctuations are not correlated with the
500 temperature fluctuations. This assumption cannot be proved theoretically. From the experimental
501 results, as can be seen in Fig. 6, the trends of the real and imaginary parts of the AERISP are different,
502 indicating that the temperature fluctuations and the aerosol concentration fluctuations are
503 uncorrelated. This phenomenon shows that the two sources are different, which is basically
504 consistent with the actual situation. This also shows that the assumptions of the theory for obtaining
505 the imaginary part of the AERISP are reasonable.

506 In order to compare with aerosol transport fluxes obtained based on the AERISPs, this paper
507 uses delayed correlation between the visibility meter and vertical wind speed to obtain aerosol
508 vertical transport fluxes. Currently, a modified visibility meter is utilized to obtain 1 Hz visibility
509 data, and then the extinction coefficient is obtained. From the extinction coefficient power spectrum
510 in Fig. 5a, it can be seen that there is a large amount of noise in the high-frequency part. The signal-
511 to-noise ratio of the extinction coefficient data is too low compared to the temperature fluctuation or
512 velocity fluctuation, which introduces a large error in the calculation of the aerosol flux. Although
513 the overall trend magnitude agreement of the fluxes obtained by the two methods is good enough to
514 show that the two methods can be corroborated with each other, there are still differences in the



515 details, which require technical methods to improve the performance of the instrument and to obtain
516 high-quality aerosol extinction coefficient data in order to carry out the measurements of vertical
517 aerosol transport fluxes based on EC method at a single point.

518 **Data availability.** Requests for data that support the findings of this study can be sent to
519 rmyuan@ustc.edu.cn.

520 **Competing interests.** The authors declare that they have no conflict of interest.

521 **Author contributions.** Renmin Yuan and Hongsheng Zhang designed experiments and wrote the
522 manuscript; Renmin Yuan, Jiajia Hua, Hao Liu, Xingyu Zhu and Peizhe Wu carried out
523 experiments; Renmin Yuan analyzed experimental results. Jianning Sun revised the manuscript and
524 participated in the discussion.

525 **Acknowledgements.** This study was supported by the National Natural Science Foundation of China
526 (42075131) and the National Key Research and Development Program under grant no.
527 2022YFC3700701.

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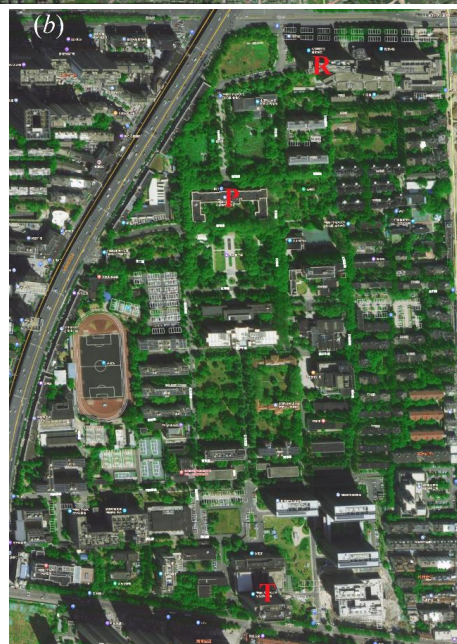
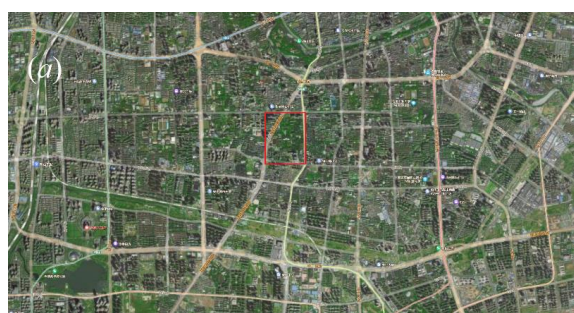


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609 Figures

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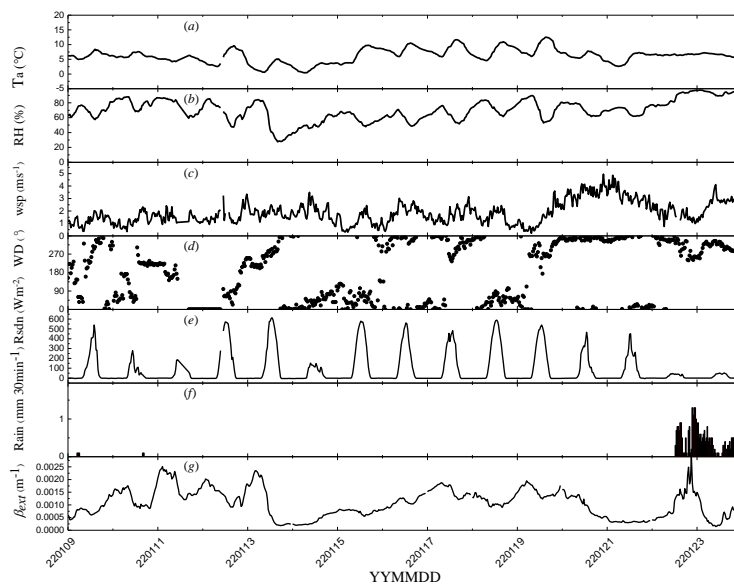


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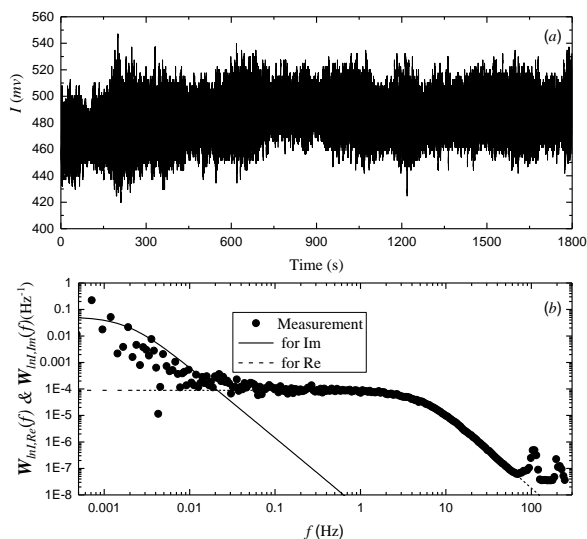
616 Figure 1. Photographs of the measurement site. (a) Map of Hefei City and (b) expanded view of the
617 measurement site on the USTC campus, which is marked as the red rectangle in (a). Points T and R in (b)
618 show the locations of the transmitter and receiver, respectively. Point P in (b) marks the meteorological
619 tower position. There are four heavy traffic roads surrounding the measurement site. Figures 1a and b @
Baidu are from website:

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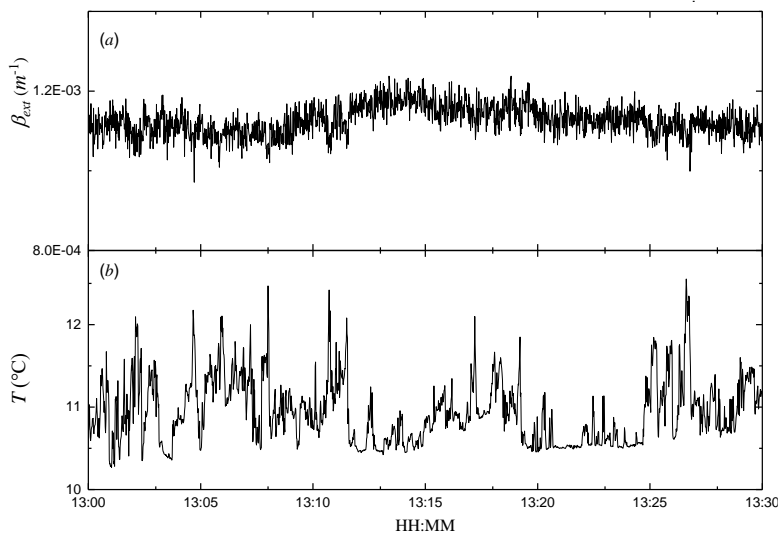
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Figure 2. Temporal variations in the (a) air temperature (T), (b) relative humidity (RH), (c) wind speed (wsp), (d) wind direction (WD), (e) total radiation (Rsdn), (f) precipitation (Rain), and (g) extinction coefficient (β_{ext}). Details can be found in the text.

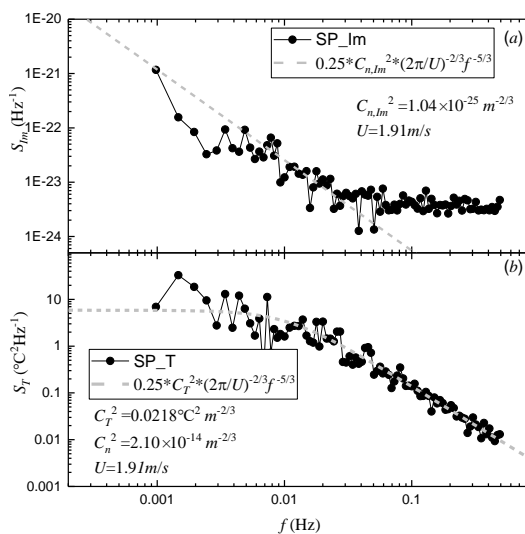


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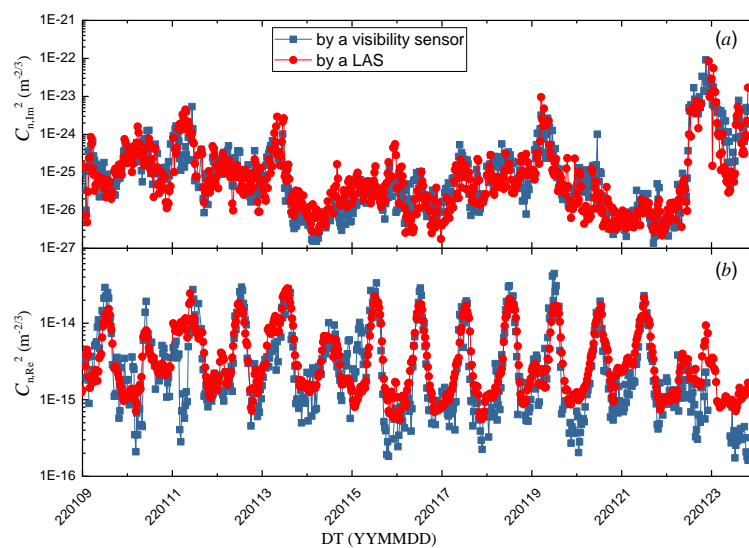
Figure 3 Temporal variations in the light intensity received by the LAS and (b) power spectral density of logarithm light intensity during 2022-01-16 13:00-13:30.



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 631 Figure 4 Temporal variations in (a) extinction coefficient and (b) air temperature during 2022-01-
 632 16 13:00-13:30.
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634
 635 Figure 5. Power spectral density of (a) extinction coefficient and (b) air temperature during 2022-
 636 01-16 13:00-13:30.

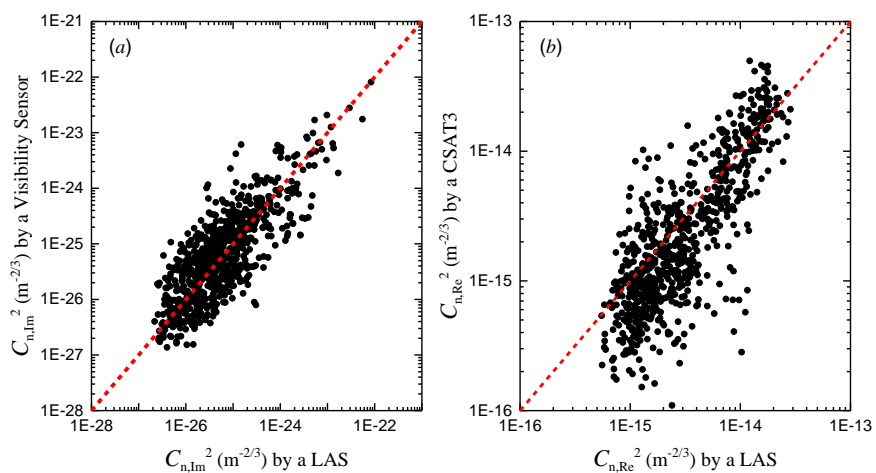


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Figure 6. Temporal variations in (a) the imaginary part and (b) real part of the AERISP during 09-23 Jan. 2022.



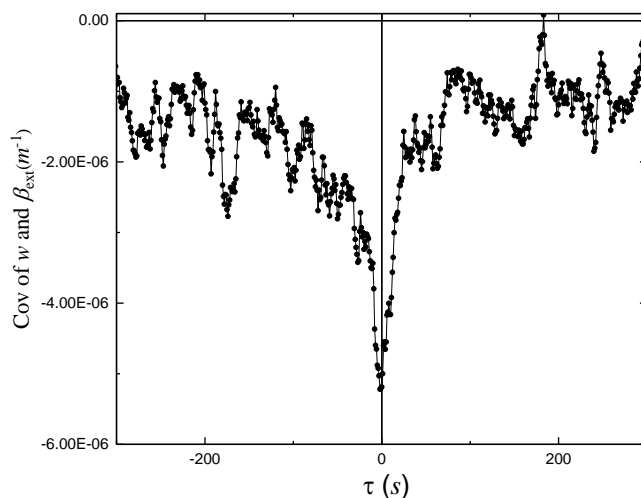
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Figure 7. Comparison of (a) the imaginary part and (b) real part of the AERISP during 09-23 Jan. 2022.

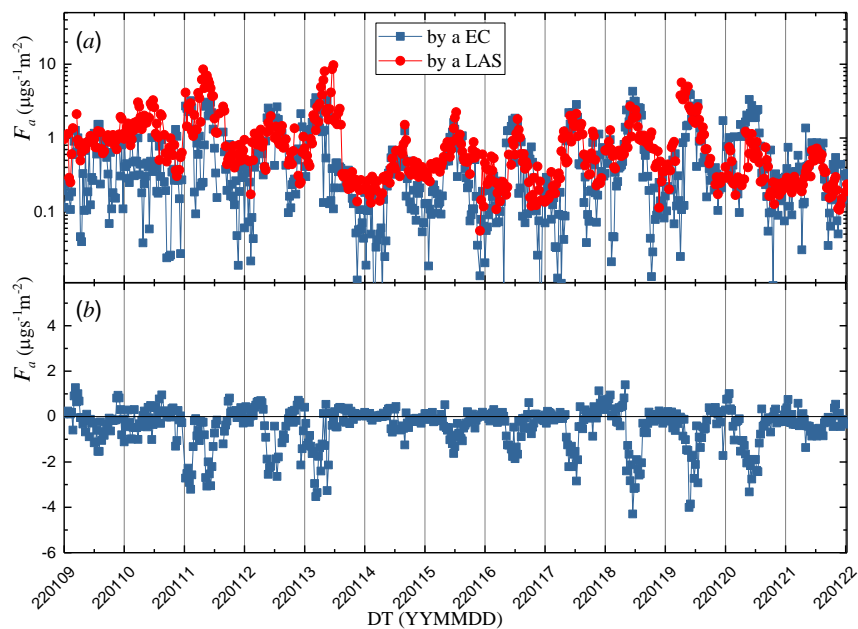


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Figure 8. (a) Delay covariance between the extinction coefficient and vertical velocity during 2022-01-16 13:00-13:30.



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Figure 9. Temporal variations in (a) absolute value of aerosol flux based on the AERISP and EC methods and (b) the imaginary part and (b) aerosol flux based on the EC methods during 09-21 Jan. 2022.