Fluorescence lidar observations of wildfire smoke inside cirrus: A contribution to smoke-cirrus - interaction research

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Abstract

A remote sensing method, based on fluorescence lidar measurements, that allows to detect and to quantify the smoke content in upper troposphere and lower stratosphere (UTLS) is presented. The unique point of this approach is that, smoke and cirrus properties are observed in the same air volume simultaneously. In this article, we provide results of fluorescence and multiwavelength Mie-Raman lidar measurements performed at ATOLL observatory from Laboratoire d'Optique Atmosphérique, University of Lille, during strong smoke episodes in the summer and autumn seasons of 2020. The aerosol fluorescence was induced by 355 nm laser radiation and the fluorescence backscattering was measured in a single spectral channel, centered at 466 nm and having 44 nm width. To estimate smoke particle properties, such as number, surface area and volume concentration, the conversion factors, which link the fluorescence backscattering and the smoke microphysical properties, are derived from the synergy of multiwavelength Mie-Raman and fluorescence lidar observations. Based on two case studies, we demonstrate that the fluorescence lidar technique provides the possibility to estimate the smoke surface area concentration within freshly formed cirrus layers. This value was used in smoke INP parameterization scheme to predict ice crystal number concentrations in cirrus generation cells.

Introduction

Aerosol particles in the upper tropospheric and lower stratospheric (UTLS) height regime play an important role in processes of heterogeneous ice formation, however our current understanding of these processes is still insufficient for a trustworthy implementation in

numerical weather and climate prediction models. The ability of aerosol particles to act as ice nucleating particles (INP) depends on meteorological factors such as temperature and ice supersaturation (as a function of vertical velocity), as well as on the aerosol type in the layer in which cirrus developed (Kanji et al., 2017). Heterogeneous ice nucleation initiated by insoluble inorganic materials such as mineral dust has been studied since a long time (e.g., DeMott et al 2010, 2015; Hoose and Möhler, 2012; Murray et al., 2012; Boose et al., 2016; Schrod et al., 2017; Ansmann et al. 2019b), while the potential of omnipresent organic particles, especially of frequently occurring aged, long-range-transported wildfire smoke particles, to act as INP is less well explored and thus not well understood (Knopf et al., 2018). Wildfire smoke can reach the lower stratosphere via pyro-cumulonimbus (pyroCb) convection (Fromm et al., 2010; Peterson et al., 2018, 2021; Hu et al., 2019; Khaykin et al., 2020) or via self-lifting processes (Boers et al., 2010,Ohneiser et al., 2021). It is widely assumed that the ability of smoke particles to serve as INP mainly depends on the organic material (OM) in the shell of the coated smoke particles (Knopf et al., 2018), but may also depend on mineral components in the smoke particles (Jahl et al., 2021). The ice nucleation efficiency may increase with increasing duration of the long-range transport as Jahl et al. (2021) suggested. Disregarding the progress made in this atmospheric research field during the last years, the link between ice nucleation efficiency and the smoke particle chemical and morphological properties is still largely unresolved (China et al., 2017; Knopf et al., 2018).

To contribute to the field of smoke-cirrus-interaction research, we present a laser remote sensing method that allows us simultaneously to detect and quantify the smoke particles amount inside of cirrus layers together with cirrus properties and to provide INP estimates in regions close to the cloud top where ice formation usually begins. The unique point of our approach is that, for the first time, smoke and cirrus properties are observed in the same air volume simultaneously. Recently, a first attempt (closure study) was performed to investigate the smoke impact on High Arctic cirrus formation (Engelmann et al. 2021). However, the aerosol measurements had to be performed outside the clouds layers, and then an assumption was needed that the estimated aerosol (and estimated INP) concentration levels also hold inside the cirrus layers. Now, we propose a method to directly determine INP-relevant smoke parameters inside the cirrus layer during ice nucleation events. This also offers the opportunity to illuminate whether an INP reservoir can be depleted in cirrus evolution processes or not. Furthermore, this

63 new lidar detection method permits a clear discrimination between, e.g., smoke and mineral dust
64 INPs.

65   Multiwavelength Mie-Raman lidars or High Spectral Resolution lidars (HSRL) are
66 favorable instruments to provide the vertical profiles of the physical properties of tropospheric
67 aerosol particles. In particular, the inversion of the so-called 3β+2α lidar observations, based on
68 the measurement of height profiles of three aerosol backscatter coefficients at 355, 532, and 1064
69 nm and two extinction coefficients at 355 and 532 nm, allows us to estimate smoke
70 microphysical properties (Müller et al., 1999, 2005; Veselovskii et al., 2002, 2015). However,
71 the aerosol content in UTLS height range can be low, so that particle extinction coefficients
72 cannot be determined with sufficient accuracy and are thus not available in the lidar inversion
73 data analysis. To resolve this issue Ansmann et al. (2019a, 2021) used the synergy of
74 polarization lidar measurements and Aerosol Robotic Network (AERONET) sunphotometer
75 observations (Holben et al., 1998) to derive conversion factors (to convert backscatter
76 coefficients into microphysical particle properties) and to estimate INP concentrations for dust
77 and smoke aerosols with the retrieved aerosol surface area concentration as aerosol input.

78   Dust particles are very efficient ice nuclei in contrast to wildfire smoke particles. In this
79 context, the question arises: How can we unambiguously discriminate smoke from dust
80 particles? This is realized by integrating a fluorescence channel into a multiwavelength aerosol
81 lidar (Reichardt et al., 2017; Richardson et al., 2019; Veselovskii et al., 2020; 2021). The
82 fluorescence capacity of smoke (ratio of fluorescence backscattering to the overall aerosol
83 backscattering), significantly exceeds corresponding values for other types of aerosol, such as
84 dust or anthropogenic particles (Veselovskii et al., 2020; 2021), and thus allows us to
85 discriminate smoke from other aerosol types. The fluorescence technique provides therefore the
86 unique opportunity to monitor ice formation in well identified wildfire smoke layers, and thus to
87 create a good basis for long-term investigations of smoke cirrus interaction.

88   In this article, we present results of fluorescence and multiwavelength Mie-Raman lidar
89 measurements performed at the ATOLL (ATmospheric Observation at liLLe) of the Laboratoire
90 d'Optique Atmosphérique, University of Lille, during strong smoke episodes in the summer and
91 autumn seasons of 2020. The results demonstrate that the fluorescence lidar is capable to monitor
92 the smoke in the UTLS height range and inside the cirrus clouds formed at or below the
93 tropopause. We start with a brief description of the experimental setup in Sect.2. In the first part

94   of the result section (Sect.3.1 and 3.2), it is explained how smoke optical properties can be

95   quantified by using fluorescence backscattering information and how we can estimate smoke

96   microphysical properties (volume, surface area, and number concentration) from measured

97   fluorescence backscatter coefficients. In this approach, multiwavelength Mie-Raman aerosol

98   lidar observations are used in addition. The retrieved values of the smoke particle surface area

99   concentration are then the aerosol input in the smoke INP estimation. A case study is discussed

100   in Sect.3.2. Two case studies are then presented in Sect.3.3 to demonstrate the capability of a

101   fluorescence lidar to monitor ice formation in extended smoke layers and to provide detailed

102   information on aerosol microphysical properties and smoke-relate INP concentration levels.

103

104   **2. Experimental setup**

105   The multiwavelength Mie-Raman lidar LILAS (LIlle Lidar AtmosphereS) is based on a

106   tripled Nd:YAG laser with a 20 Hz repetition rate and pulse energy of 70 mJ at 355 nm.

107   Backscattered light is collected by a 40 cm aperture Newtonian telescope and the lidar signals

108   are digitized with Licel transient recorders of 7.5 m range resolution, allowing simultaneous

109   detection in the analog and photon counting mode. The system is designed for simultaneous

110   detection of elastic and Raman backscattering, allowing the so called $3\beta+2\alpha+3\delta$ data

111   configuration, including three particle backscattering (β_{355}, β_{532}, β_{1064}), two extinction (α_{355}, α_{532})

112   coefficients along with three particle depolarization ratios (δ_{355}, δ_{532}, δ_{1064}). The particle

113   depolarization ratio, determined as a ratio of cross- and co-polarized components of the particle

114   backscattering coefficient, was calculated and calibrated in the same way as described in

115   Freudenthaler et al. (2009). The aerosol extinction and backscattering coefficients at 355 and 532

116   nm were calculated from Mie-Raman observations (Ansmann et al., 1992), while β_{1064} was

117   derived by the Klett method (Fernald, 1984; Klett, 1985). Additional information about

118   atmospheric parameters was available from radiosonde measurements performed at

119   Herstmonceux (UK) and Beauvechain (Belgium) stations, located 160 km and 80 km away

120   from the observation site respectively.

121   This lidar system is also capable to perform aerosol fluorescence measurements. A part of

122   the fluorescence spectrum is selected by a wideband interference filter of 44 nm width centered

123   at 466 nm (Veselovskii et al., 2020; 2021). The strong sunlight background at daytime restricts

124   the fluorescence observations to nighttime hours. To characterize the fluorescence properties of

125   aerosol, the fluorescence backscattering coefficient β_F is calculated from the ratio of

126   fluorescence and nitrogen Raman backscatters, as described in Veselovskii et al. (2020). This

127   approach allows to evaluate the absolute values of β_F, if the relative sensitivity of the channels is

128   calibrated and the nitrogen Raman scattering differential cross section σ_R is known. In our

129   research we used $\sigma_R = 2.744*10^{-30}$ cm^{2}sr^{-1} at 355 nm from Venable et al. (2011). All β_F profiles

130   presented in this work were smoothed with the Savitzky – Golay method, using second order

131   polynomial with 21 points in the window. The efficiency of fluorescence backscattering with

132   respect to elastic backscattering β_{532} is characterized by the fluorescence capacity $G_F = \dfrac{\beta_F}{\beta_{532}}$.

133   For most of atmospheric particles β_F is proportional to the volume of dry matter, while

134   dependence of β_{532} on particle size is more complicated. As a result, G_F depends not only on

135   aerosol type, but also on particle size and the relative humidity RH. Uncertainty of β_F calculation

136   depends on the chosen value of σ_R and on relative transmission of optical elements in

137   fluorescence and nitrogen channels. These system parameters do not change with time. The

138   relative sensitivity of PMTs, however, may change. Regular calibration of the channels relative

139   sensitivity (Veselovskii et al., 2020), demonstrates that corresponding uncertainty can be up to

140   10%. At high altitudes the statistical uncertainty becomes predominant. We recall also, that only

141   a part of the fluorescence spectra was selected by the interference filter in the receiver, so

142   provided values of β_F and G_F are specific for the filter used. Analyzing the fluorescence

143   measurements we should keep in mind, that the sensitivity of this technique can be limited by the

144   fluorescence of optics in the lidar receiver. The minimal value of G_F, which we measured during

145   observation in cloudy conditions in the lower troposphere was about 2×10^{-8}. Thus, at least, in the

146   measurements with G_F above this value, the contribution of optics fluorescence can be ignored.

147

148   **3. Results of the measurements**

149   *3.1. Observation of smoke particles in UTLS*

150   Smoke particles produced by intensive fires and transported across the Atlantic are

151   regularly observed in the UTLS height range over Europe (Müller et al., 2005; Hu et al., 2019;

152   Baars et al., 2019, 2021). One of such events, observed over Lille in the night of 4-5 November

153   2020, is shown in Fig.1. The figure provides height – time displays of the range corrected lidar

154   signal and the volume depolarization ratio at 1064 nm together with the fluorescence

155   backscattering coefficient. A narrow smoke layer occurred in the upper troposphere in the period
156   from 23:00 – 06:00 UTC. The smoke was detected at heights above 12 km after midnight. The
157   particles caused a low volume depolarization ratio (<5%) at 1064 nm and strong fluorescence
158   backscattering (β_F>1.2×10^{-4} Mm^{-1}sr^{-1}). The backward trajectory analysis indicated that the
159   aerosol layer was transported over the Atlantic and contained products of North American wild
160   fires.

161   Vertical profiles of aerosol β_{532} and fluorescence β_F backscattering coefficients for the
162   period from 02:00 - 05:30 UTC are shown in Fig.2a. The fluorescence capacity G_F in the center
163   of smoke layer (not shown) was about 4.5×10^{-4}. The depolarization ratio of aged smoke in the
164   UTLS height range usually shows a strong spectral dependence (Haarig et al., 2018; Hu et al.,
165   2019). For the case presented in Fig.2a the particle depolarization ratio in the center of the smoke
166   layer decreased from 16±4% at 355 nm (δ_{355}) to 4±1% at 1064 (δ_{1064}). The tropopause height H_{tr}
167   was at about 13000 m, thus the main part of the smoke layer was below the tropopause. By the
168   end of day the smoke layer became weaker (β_F<0.3×10^{-4} Mm^{-1}sr^{-1}) and ascended up to 14500 m,
169   which is above the tropopause. The corresponding vertical profiles of β_{532} and β_F are shown in
170   Fig.2b. The fluorescence capacity in the center of the layer is about 4.5×10^{-4}, which is close to
171   the value observed during 02:00 - 05:30 UTC period.

172   An important advantages of the fluorescence lidar technique is the ability to monitor
173   smoke particles inside cirrus clouds. The results of smoke observations in the presence of ice
174   clouds are shown in Fig.3. Cirrus clouds occurred during the whole night in the height range
175   from 6.0 km – 10.0 km. To quantify the fluorescence backscattering inside the cloud (which was
176   rather weak in this case), the lidar signals were averaged over the full 18:00 – 06:00 UTC time
177   interval in Fig.3a. The fluorescence backscatter coefficient shown in Fig.3c decreased from
178   β_F=0.015×10^{-4} Mm^{-1}sr^{-1} at 5000 m (near the cloud base) to a minimum value of 0.01×10^{-4} Mm^{-}
179   ^{1}sr^{-1} at 7000 m inside the cirrus layer. Above the tropopause the fluorescence backscattering
180   increased strongly and reached the maximum (about 0.3 ×10^{-4} Mm^{-1}sr^{-1}) in 11000 m -13000 m
181   height.

182   The analysis of fluorescence measurements performed during strong smoke episodes in
183   the summer and autumn of 2020, when smoke layers from North American fires frequently
184   reached Europe, demonstrates that the fluorescence capacity varied within the range of 2.5×10^{-4}
185   to 4.5×10^{-4}. The variations are a function of smoke composition, relative humidity and particle

186 size. However, in the upper troposphere, where relative humidity is low, G_F was normally close
187 to 4.5×10^{-4}. This relatively low range of G_F variations allows the estimation of the backscattering
188 coefficient attributed to the smoke particles from fluorescence measurements as:

189
$$\beta_{532}^s = \frac{\beta_F}{G_F}.$$
(1)

190   Fig.3d shows the smoke backscattering coefficient β_{532}^s, calculated from β_F for
191 $G_F = 4.5 \times 10^{-4}$, together with β_{532}. The dynamical range of β_{532} variations is high. To make smoke
192 backscattering visible above H_{Tr}, β_{532} is plotted in expanded scale in Fig.3d. The β_{532}^s values,
193 though being strongly oscillating above the tropopause, match the β_{532} indicating that the smoke
194 contribution to backscattering was predominant.

195

196 **3.2. Estimation of smoke particles content based on fluorescence measurements**
197   The possibility to detect fluorescence backscattering inside the cirrus clouds reveals also
198 the opportunity for a quantitative characterization of the smoke content. This can be realized by a
199 synergistic use of fluorescence and multiwavelength Mie – Raman lidar observations. The flow
200 chart, summarizing the main steps of this procedure, is presented in **Appendix 1**. For the smoke
201 layers with sufficient optical depth, the number N, surface area S and volume V concentrations
202 can be evaluated, by inverting the $3\beta+2\alpha$ observations consisting of three backscatter
203 coefficients (355, 532, 1064 nm) and two extinction coefficients (355, 532 nm) (Müller et al.,
204 1999; Veselovskii et al., 2002; Pérez-Ramírez et al., 2013). The conversion factors C_N, C_S, C_V,
205 introduced as

206 $$C_N = \frac{N}{\beta_F}, \; C_S = \frac{S}{\beta_F}, \; C_V = \frac{V}{\beta_F},$$
(3)

207 allow the estimation of smoke particle concentration inside the clouds from fluorescence
208 backscattering, assuming that smoke contribution to the fluorescence is predominant. Moreover,
209 it allows estimation of the particles concentration in weak smoke layers in UTLS, where $3\beta+2\alpha$
210 observations are normally not available.
211   On 23-24 June 2020, a strong smoke layer was observed in 4500 – 5500 m height during
212 the whole night (Fig.4). The vertical profiles of the aerosol backscattering and extinction
213 coefficients ($3\beta+2\alpha$) are shown in Fig.5a, while the particle depolarization ratios δ_{355}, δ_{532}, δ_{1064}

214 and the lidar ratios at 355 nm and 532 nm (LR_{355}, LR_{532}) are presented in Fig.5b. The

215 depolarization ratio decreases with wavelength from 9±1.5% at 355 nm to 1.5±0.3% at 1064 nm

216 and the lidar ratio at 532 nm significantly exceeds corresponding value at 355 nm (80±12 sr and

217 50±7.5 sr respectively), which is typical for aged smoke (Müller et al., 2005). The

218 multiwavelength observations were inverted to determine the particle effective radius r_{eff},

219 number, surface area and volume concentrations for seven height bins inside the smoke layer.

220 The effective radius r_{eff} in Fig.5c increases through the layer from 0.15 μm to 0.2 μm

221 simultaneously with the increase of the fluorescence capacity G_F from 2.8×10^{-4} to 3.6×10^{-4}.

222 Retrieved values of N, S, V were used for the calculation of the conversion factors (Eq. 3) for

223 each height bin. In the center of the smoke layer (at 4.9 km) the factors are: $C_N = 88 \times 10^4$

224 $\dfrac{cm^{-3}}{Mm^{-1}sr^{-1}}$, $C_S = 35 \times 10^4 \dfrac{\mu m^2 cm^{-3}}{Mm^{-1}sr^{-1}}$, and $C_V = 2.4 \times 10^4 \dfrac{\mu m^3 cm^{-3}}{Mm^{-1}sr^{-1}}$. Thus, when β_F is given in $Mm^{-1}sr^{-1}$,

225 the calculated values of N, S, and V are given in cm^{-3}, $\mu m^2 cm^{-3}$ and $\mu m^3 cm^{-3}$ respectively.

226 Fluorescence backscattering is proportional to the particle volume concentration, so C_V is not

227 sensitive to the effective radius variation. The conversion factors C_N and C_S, on the contrary,

228 depend on the particle size. Fig.5d shows the profiles of N, S, V obtained by inversion of $3\beta + 2\alpha$

229 observations (symbols) together with corresponding values (N^S, S^S, V^S) obtained from β_F, using

230 the mean conversion factors for seven height bins considered. The volume concentrations V and

231 V^S agree well for all seven height bins. For the surface area concentrations the agreement is still

232 good, but for N and N^S the difference is up to 30%. We need to emphasize, that the conversion

233 factors presented are specific for our lidar system (for the interference filter installed in

234 fluorescence channel). It is worthwhile to mention that the ratio V/α_{532} of the volume

235 concentration V in Fig.5d to the extinction coefficient α at 532 nm in Fig.5a, as well as the ratio

236 S/α_{532}, are very close to respective extinction-to-volume and extinction-to-surface-area-

237 concentration conversion factors presented for aged wildfire smoke by Ansmann et al. (2021).

238   The conversion factors depend on the smoke composition. To estimate the variation range

239 of C_N, C_S, C_V, several smoke episodes were analyzed and corresponding results are presented in

240 Table 1. The table provides the fluorescence capacity G_F and the conversion factors at the

241 heights, where $3\beta + 2\alpha$ data could be calculated. Mean values of $<C_N>$, $<C_S>$, $<C_V>$ derived for

242 these episodes and corresponding standard deviations are:

243 $$<C_N>=(61\pm32)\times10^4 \frac{cm^{-3}}{Mm^{-1}sr^{-1}} \; ; \; <C_S>=(28\pm6.4)\times10^4 \frac{\mu m^2 cm^{-3}}{Mm^{-1}sr^{-1}} \; ; \; <C_V>=(2.2\pm0.2)\times10^4 \frac{\mu m^3 cm^{-3}}{Mm^{-1}sr^{-1}} \quad (4)$$

244   Table 1 shows also the volume and surface area concentrations of the smoke particles obtained

245   from the inversion of $3\beta+2\alpha$ observations (V, S) and calculated from β_F (V^S, S^S) using the

246   conversion factors in Eq. (4). Standard deviations of V^S and S^S from corresponding values of

247   $V_{3\beta+2\alpha}$ and $S_{3\beta+2\alpha}$ are 10% and 25% respectively.

248   The mean conversion factors in Eq. (4) are now used to estimate the smoke

249   microphysical properties inside the cloud, assuming in addition that the predominant contribution

250   to the fluorescence is provided by the smoke. Table 2 summarizes the number, surface area, and

251   volume concentrations of smoke particles inside the ice clouds, estimated from fluorescence

252   measurements for four episodes considered in this paper. On September 12-13, 2020, the smoke

253   layer with high fluorescence and low depolarization ratio at 1064 nm (below 4%) was observed

254   during the whole night inside the 2.0 km – 5.0 km height range. The cirrus cloud occurred above

255   11000 m also during the whole night. Fig.6a presents vertical profiles of the aerosol β_{532} and

256   fluorescence β_F backscattering coefficients. Fluorescence backscattering shows a maximum at

257   3.5 km, but it is detected even inside the cloud. The smoke backscattering coefficient β_{532}^s,

258   computed from β_F for G_F=3.6×10^{-4} agrees well with β_{532} inside the 2.0 – 10.0 km height range

259   (Fig.6b). The height profile of the surface area concentration of the smoke particles S^S, calculated

260   from β_F using the respective conversion factor in Eq. (4), is shown in Fig.6c. In the smoke layer,

261   S^S is up to 60 $\mu m^2/cm^3$, while in the center of the cloud in 12 km – 13 km height the average

262   value of S^S is 1.6±0.4 $\mu m^2/cm^3$. Corresponding values of number and volume concentrations in

263   the cloud center are 3.5±1.8 cm^{-3} and 0.13±0.013 $\mu m^3/cm^3$.

264   The temperature in the cloud ranged from about -50°C to almost -70°C and was -68°C at

265   cirrus top in Fig.6b where ice nucleation usually starts. We applied the immersion freezing INP

266   parameterization of Knopf and Alpert (2013) for Leonardite (a standard humic acid surrogate

267   material) and assume that this humic compound represents the amorphous organic coating of

268   smoke particles. The INP parameterization for smoke particles is summarized for lidar

269   applications in Ansmann et al. (2021). The selected parameterization allows the estimation of the

270   INP concentration as a function of ambient air temperature (freezing temperature), ice

271   supersaturation, particle surface area, and time period for which a certain level of ice

272   supersaturation is given. We simply assume a constant ice supersaturation of around 1.45 during

273   a time period of 600 s (upwind phase of a typical gravity wave in the upper troposphere). The

274   temperature at cirrus top height is set to -68°C and the aerosol surface area concentration to 2.0

275    $\mu m^2/cm^3$ as indicated in Fig.6c. The obtained INP concentrations of 1-10 L^{-1} for these

276   meteorological and aerosol environmental conditions can be regarded as the predicted number

277   concentration of ice crystals nucleated in the cirrus top region. Ice crystal number concentration

278   of 1-10 L^{-1} are typical values in cirrus layers when heterogeneous ice nucleation dominates

279   (typical values of INP concentrations and supersaturation are discussed e.g. in Sullivan et al,

280   (2016), Ansmann et al. (2019; 2021), Engelmann et al. (2021)). It should be mentioned that the

281   required very high ice supersaturation levels of close to 1.5 (ice supersaturation of 1.1-1.2 is

282   sufficient in case of mineral dust particles) are still lower than the threshold supersaturation level

283   of >1.5 at which homogeneous freezing starts to dominate. At low updraft velocities around 10-

284   25 cm/s, as usually given in gravity waves in the upper troposphere (Barahona et al., 2017),

285   heterogeneous ice nucleation very likely dominates the ice production when cirrus evolves in

286   detected aerosol layers.

287

288   ***3.3. Ice formation inside the smoke layers***.

289   During September 2020 we observed several episodes with ice cloud formation inside of

290   smoke layers. One of such episodes occurred on 11-12 September 2020 and is shown in Fig.7.

291   The height – time display of the fluorescence backscattering coefficient reveals the smoke layer

292   in the 5.0 - 10.0 km height range. Inside this layer, we can observe a short time interval of 15

293   minutes with a strongly increased depolarization ratio around 10.5 km height (red spots),

294   indicating ice cloud formation. Fig.8 shows vertical profiles of the aerosol backscattering

295   coefficients β_{355}, β_{532}, and β_{1064} as well the particle depolarization ratios δ_{355}, δ_{532}, and δ_{1064} for

296   two temporal intervals. The first interval (23:00 – 00:30 UTC) is prior to ice cloud formation and

297   the second one (01:20 – 01:45 UTC) covers ice occurrence period. The depolarization ratios at

298   all three wavelengths were < 5% below 6 km height. Above that height δ_{355} significantly

299   increased reaching the value of 10% at 7 km (Fig.8b), which is indicative of a change of the

300   particle shape (from spherical to irregular shape). The fluorescence capacity also changed with

301   height, being about $G_F=4.5\times10^{-4}$ at 5.5 km and it decreases to 3.5×10^{-4} by 8 km. The profile of

302   β_{532}^{s} shown in Fig.8c is calculated assuming $G_F=4.0\times10^{-4}$ and it matches well the profile of β_{532}

303   for the whole height range. The aerosol layer at 10.5 km is thus a pure smoke layer. Ice

304 formation at 10.5 km (Fig.8d-f) leads to a significant increase of β_{532} while β_{532}^{s} (or the

305 respective fluorescence backscatter coefficient β_F) remains low and at the same level as observed

306 below the cirrus layer, i.e., below 10 km height. The depolarization ratios at all three

307 wavelengths increases to typical cirrus values around 40%. The temperature at 10.5 km is about -

308 50 °C, and the surface area concentration of the smoke particles inside the cloud, estimated from

309 β_F, is about 10 $\mu m^2/cm^3$ (see Fig.8f, thin blue line). For these temperature and aerosol conditions

310 we yield smoke INP concentrations of 1-10 L^{-1} for ice supersaturation values even below 1.4

311 (1.38-1.4) and updraft duration of 600 s. When comparing Fig.8c and 8f at cirrus level it seems

312 to be that ice nucleation on the smoke particles widely depleted the smoke INP reservoir.

313   Another case of ice formation in the smoke layer was observed on 17-18 September

314 2020. Strong smoke layers occurred in the 5.0 km – 9.0 km height range as shown in Fig.9.

315 During the period from 22:30 – 00:00 UTC, the depolarization increased at 8.5 km height,

316 indicating ice formation. Vertical profiles of the particle parameters prior and during ice

317 formation are shown in Fig.10. The β_{532}^{s} calculated for $G_F=3.5\times10^{-4}$ matches well with β_{532}

318 below 6.9 km and above 8.0 km (Fig.10c), but inside the 7.0 km – 8.0 km height range

319 $\beta_{532} > \beta_{532}^{s}$, meaning that G_F was decreased. The depolarization ratio in the 7.0 km – 8.0 km

320 height range shows some enhancement (Fig.10b): in particular, δ_{532} increased from 10% to 12%.

321 Cloud formation at 8.5 km (Fig.10d) led to a significantly smaller increase of the depolarization

322 ratio, compared to the case on 11-12 September. Prior to the cloud formation the values of δ_{1064},

323 δ_{532}, and δ_{355} at 8.5 km were of 3%, 10%, and 13% respectively (Fig.10b) and in the cloud

324 corresponding depolarization ratios increase up to 9%, 15%, 20%. The reason is probably that

325 the signal averaging period from 22:45 to 23:45 UTC includes cloud-free section. Three gravity

326 waves obviously crossed the lidar field site and triggered ice nucleation just before 23 UTC, 15-

327 30 minutes after 23 UTC, and around mid night (00:00 UTC). The temperature at cloud top at

328 about 8.5-8.6 km height was close to -35°C. For this high temperature and the high particle

329 surface area concentration of 200 $\mu m^2/cm^3$ (see Fig.10d, thin blue line) we yield smoke INP

330 concentrations of 1-10 L^{-1} for a relatively low ice supersaturation of 1.30-1.33 and an updraft

331 period of 600 s. Again, a depletion of the INP reservoir is visible after formation of the cirrus

332 layer (see Figs.6c and 6d around and above 8.5 km height).

333

Conclusion

The operation of a fluorescence channel in the LILAS lidar during strong smoke events in the summer and autumn seasons of 2020 has demonstrated the ability of the fluorescence lidar technique to discriminate ice from smoke particles in atmospheric layers in the UTLS height range in large detail. The fluorescence capacity G_F of smoke particles during this period varied within a relatively small range: $2.5\text{-}4.5\times10^{-4}$, thus the use of the mean value of G_F allows to estimate the contribution of smoke to the total particle backscattering coefficient. The fluorescence lidar technique makes it possible to estimate smoke parameters, such as number, surface area and volume concentration in UTLS height range in a quantitative way by applying conversion factors (C_N, C_S, C_V) which link the fluorescence backscattering and the smoke microphysical properties. These factors, derived from the synergy of multiwavelength Mie-Raman and fluorescence lidar observations, show some variation from episode to episode, however, the use of mean values $<C_N>$, $<C_S>$, $<C_V>$ allows estimation of smoke properties in UTLS height regime with reasonable accuracy. Based on two case studies, we demonstrated that the fluorescence lidar technique provides the unique possibility to characterize the smoke particles and their amount inside cirrus cloud layers. The smoke input parameter (surface area concentration) in smoke INP parameterization schemes that are used to predict ice crystal number concentrations in cirrus generation cells, can now be estimated within freshly formed cirrus layers.

The smoke parameters such as fluorescence capacity and conversion factors were derived from observations of aged wildfire smoke, transported over Atlantic in 2020. However, smoke composition, depends on many factors, such as burning materials type, flame temperature and environmental conditions, thus the smoke fluorescence properties may also vary. Hence, it is important to perform the measurements for different locations and seasons. The fluorescence backscattering in UTLS height range is quite weak, so to perform measurements with higher temporal resolution more powerful lidar systems are needed. A dedicated high power Lidar, LIFE (Laser Induced Fluorescence Explorer), will be designed and operated at ATOLL, in the frame of OBS4CLIM/ACTRIS-France .

Data availability. Lidar measurements are available upon request (philippe.goloub@univ-lille.fr).

366 *Author contributions*. IV processed the data and wrote the paper. QH and TP performed the
367 measurements. A.A. analyzed results of fluorescence measurements and ice formation in smoke
368 layers. PG supervised the project and helped with paper preparation. MK developed software for
369 data processing.
370 .
371 *Competing interests*. The authors declare that they have no conflict of interests.

372

373

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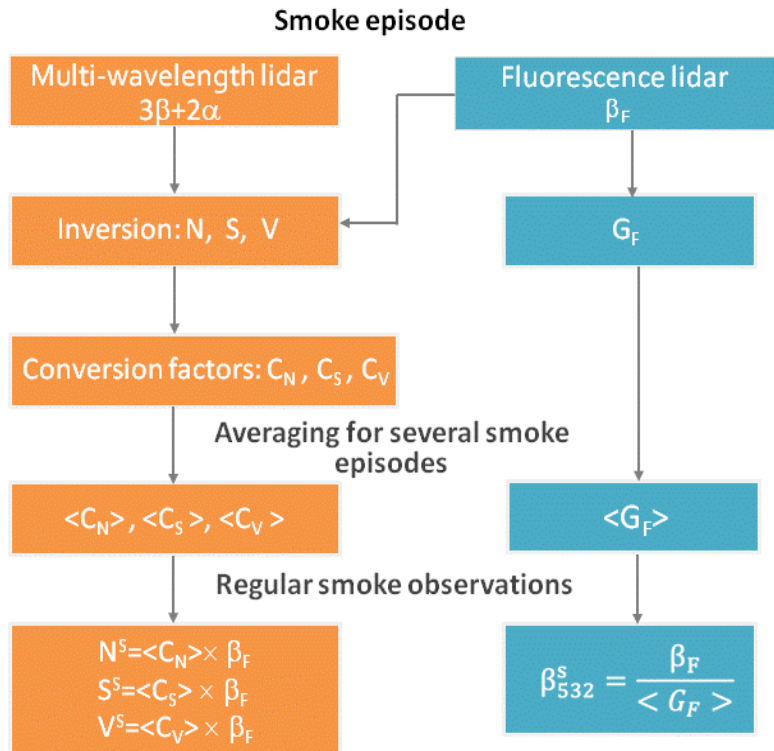
Table 1. Conversion factors C_N, C_S, and C_V, and fluorescence capacity G_F at height H for five smoke episodes. Volume and surface area concentration of smoke particles, obtained by the inversion of $3\beta+2\alpha$ lidar observations (V, S), are given together with values calculated from fluorescence measurements (V^S, S^S) and using conversion factors (Eq. 4).

Date dd/mm/yy	H, km	C_N, $10^4 \frac{cm^{-3}}{Mm^{-1}sr^{-1}}$	C_S, $10^4 \frac{\mu m^2 cm^{-3}}{Mm^{-1}sr^{-1}}$	C_V, $10^4 \frac{\mu m^3 cm^{-3}}{Mm^{-1}sr^{-1}}$	G_F, 10^{-4}	V, μm^3/cm^3		S, μm^2/cm^3	
						V	V^S	S	S^S
23/06/20	4.9	88	35	2.4	3.5	21±4	19±4	306±75	237±60
11/09/20	7.5	75	28	2.0	3.9	7.6±1.6	8.7±1.6	111±25	111±25
14/09/20	6.0	90	34	2.3	3.7	6.4±1.3	6.1±1.3	94±25	78±20
17/09/20	6.8	21	21	2.3	2.9	8.0±1.6	7.8±1.6	73±18	100±25
20/09/20	4.9	33	22	2.0	4.3	2.7±0.5	2.9±0.6	31±8	37±9

Table 2. Number N^S, surface area S^S, and volume V^S concentrations of smoke particles inside the ice cloud at height H estimated from fluorescence measurements by applying the conversion factors in Eq. (4) for four measurement sessions.

Date dd/mm/yy	Time UTC	H, km	β_F, 10^{-4} Mm^{-1}sr^{-1}	N^S, cm^{-3}	S^S, μm^2/cm^3	V^S, μm^3/cm^3
12/09/20	01:20-01:45	10.5	0.32	20±10	9±2.3	0.7±0.15
12-13/09/20	21:00-03:00	12.5	0.06	3.5±1.8	1.6±0.4	0.13±0.03
17/09/20	22:45-23:45	8.5	6.5	400±200	180±45	14±3
24-25/11/20	18:00-06:00	8.0	0.013	0.8±0.4	0.36±0.09	0.03±0.006

562   **Appendix A.** Estimation of smoke parameters from Mie-Raman and fluorescence lidar

563   measurements.

564

565   Fig.A. Flow chart showing the main steps of the procedure of smoke parameters estimation from

566   multiwavelength Mie-Raman and fluorescence lidar measurements. Procedure includes the

567   following steps. (i) For a strong smoke layer the $3\beta+2\alpha$ data set, derived from multiwavelength

568   Mie-Raman lidar observations, is inverted to the particle number N, surface S and volume V

569   density. (ii) Conversion factors C_N, C_S, C_V are calculated from Eq.(3) by using the fluorescence

570   backscattering coefficient β_F. (iii) Different smoke events are analyzed to get mean values of

571   conversion factors $<C_N>$, $<C_S>$, $<C_V>$. These mean values are used to estimate smoke

572   concentration in weak layers in UTLS and inside cirrus clouds in regular observations. The mean

573   value of smoke fluorescence capacity $<G_F>$ allows estimation of smoke contribution β_{532}^s to the

574   total backscattering coefficient β_{532}.

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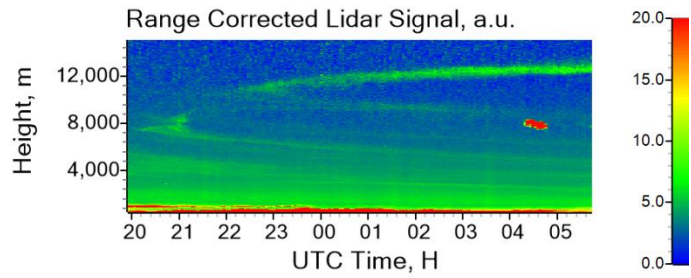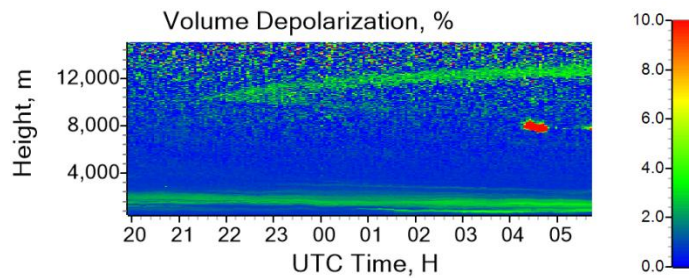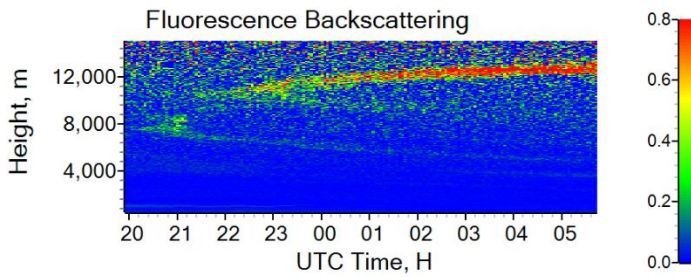
580
Fig.1. Range corrected lidar signal at 1064 nm, volume depolarization ratio at 1064 nm and
fluorescence backscattering coefficient (in 10^{-4} $Mm^{-1}sr^{-1}$) on 4-5 November 2020.

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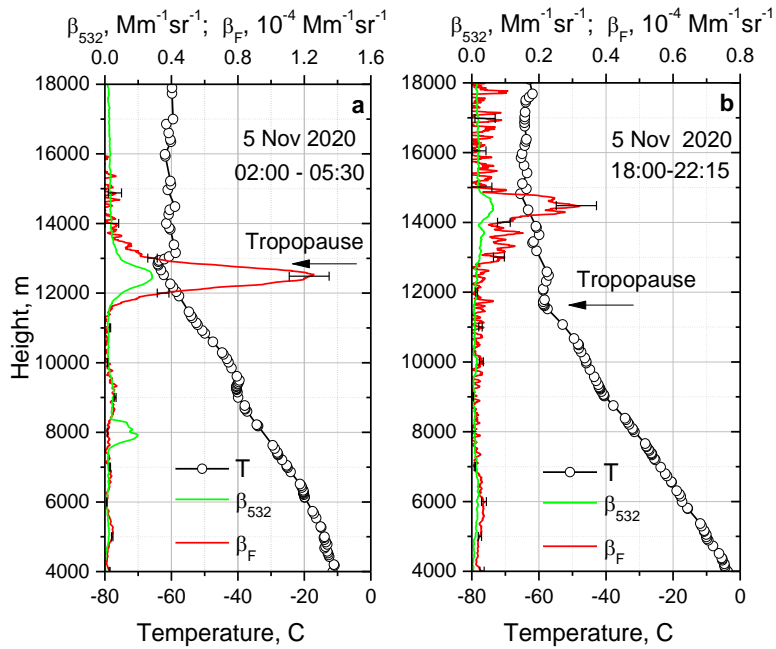
584

585
586 Fig.2. Vertical profiles of aerosol backscattering β_{532} and fluorescence backscattering β_F
587 coefficients on 5 November 2020 for the periods (a) 02:00 - 5:30 UTC and (b) 18:00 – 22:15
588 UTC. Open symbols show the temperature profile measured by the radiosonde launched at
589 Herstmonceux (UK).
590
591

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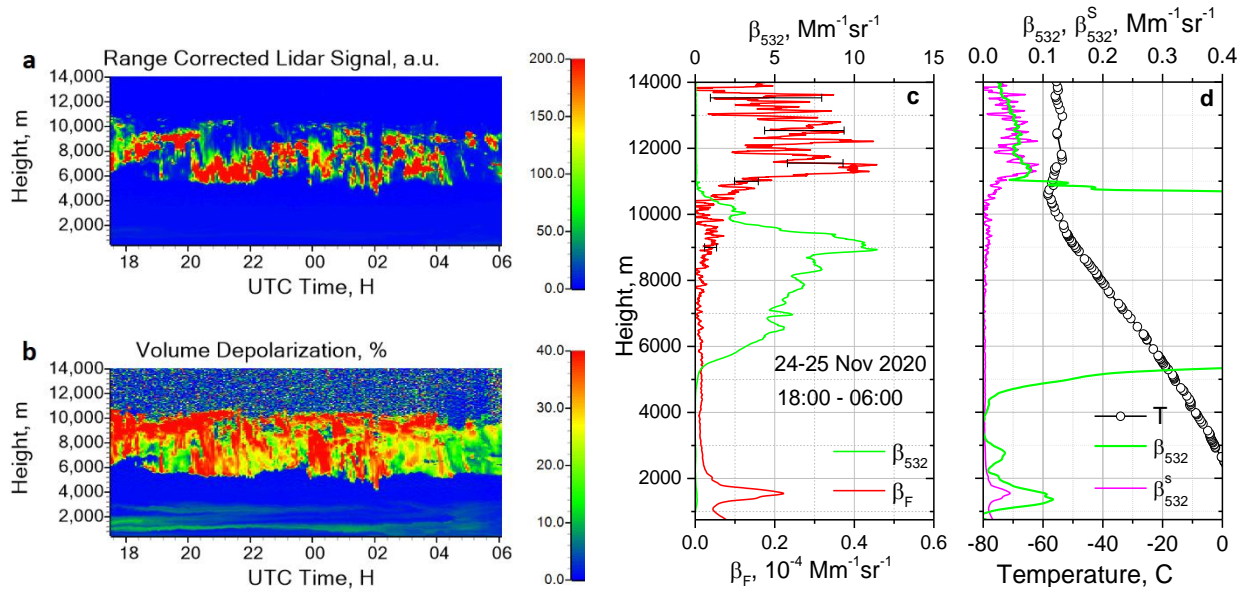
593   Fig.3. Smoke fluorescence in the presence of clouds on 24 – 25 November 2020. (a, b)
594   Spatiotemporal variations of the range corrected lidar signal and volume depolarization at 1064
595   nm. (c) Vertical profiles of the aerosol β_{532} and fluorescence β_F backscattering coefficients. (d)
596   Aerosol backscattering β_{532} together with smoke backscattering β_{532}^s coefficient, computed from
597   β_F for $G_F=4.5\times10^{-4}$. Open symbols show temperature profile measured by the radiosonde at
598   Herstmonceux.
599
600
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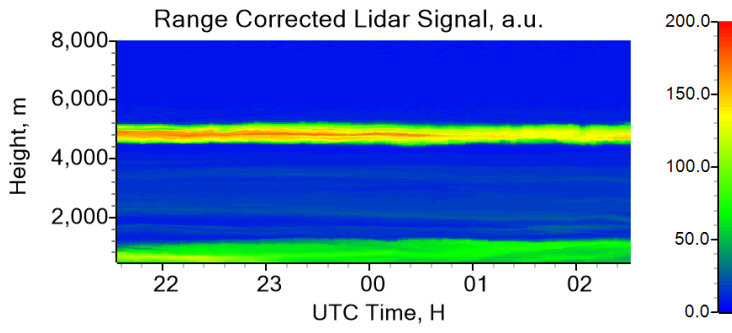
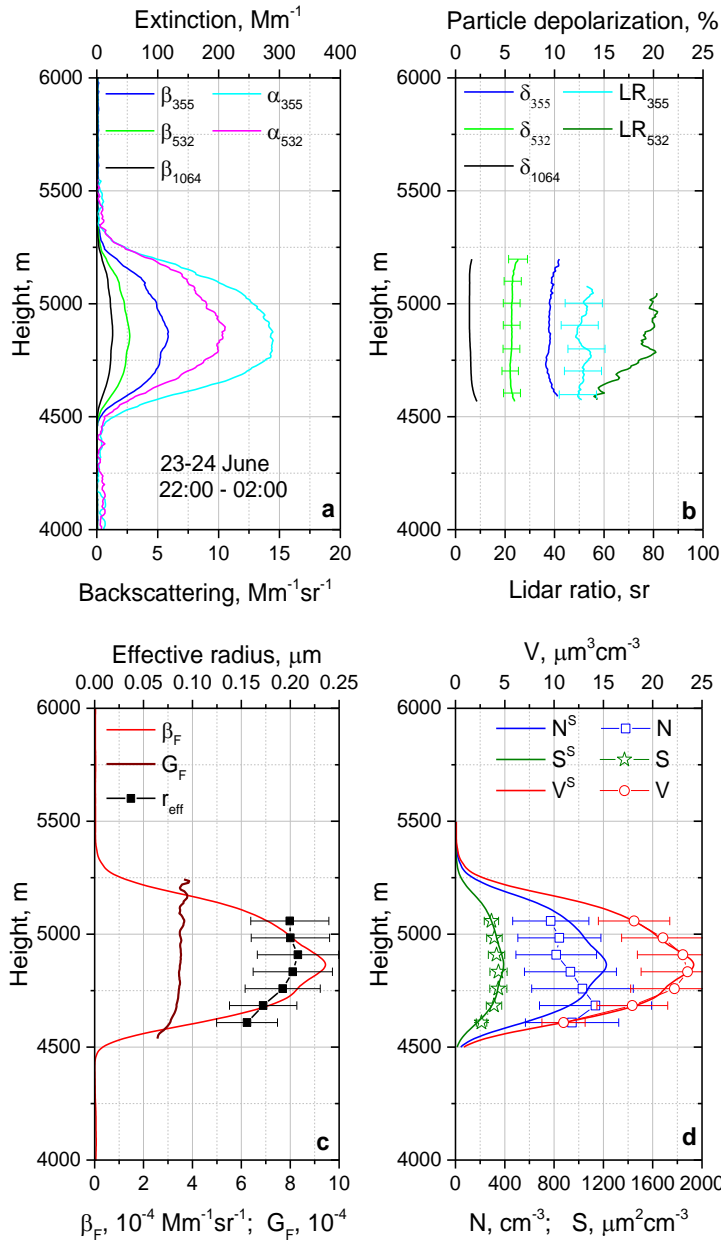
602

603
604   Fig.4. Range corrected lidar signal at 1064 nm on 23-24 June 2020, revealing a smoke layer
605   between 4500 and 5200 m height.
606

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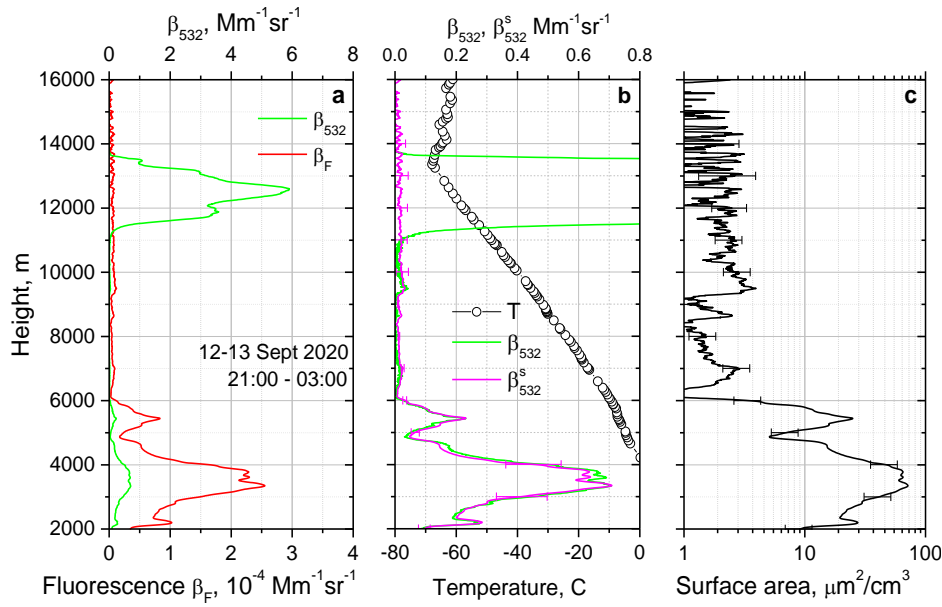
609
610 Fig.5. Smoke layer on 23-24 June 2020. (a) Vertical profiles of backscattering (β_{355}, β_{532}, β_{1064})
611 and extinction (α_{355}, α_{532}) coefficients. (b) Particle depolarization ratios (δ_{355}, δ_{532}, δ_{1064}) and lidar
612 ratios (LR$_{355}$, LR$_{532}$). (c) Fluorescence backscattering (β_F), fluorescence capacity (G_F) and the
613 particle effective radius (r_{eff}). (d) Number (N, N^S), surface area (S, S^S) and volume (V, V^S)
614 concentrations obtained by inversion of 3β+2α observations (symbols) and calculated from the
615 fluorescence backscattering (lines) by using the mean conversion factors defined in Eq. (3).
616

617
618 Fig.6. Observation of smoke fluorescence on 12-13 September 2020, 21:00 – 03:00 UTC. (a)
619 Vertical profiles of the aerosol backscattering β_{532} and fluorescence backscattering β_F
620 coefficients. (b) Aerosol backscattering β_{532} together with smoke backscattering β_{532}^s coefficient
621 computed from β_F for $G_F=3.6\times10^{-4}$. (c) Surface area concentration of the smoke particles
622 calculated from β_F by using the respective conversion factor from (4). Open symbols show the
623 temperature profile measured by the radiosonde at Herstmonceux.

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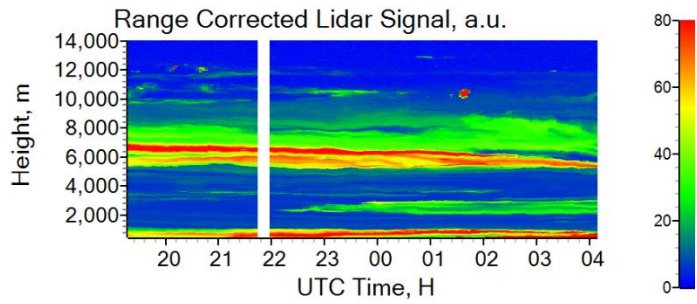
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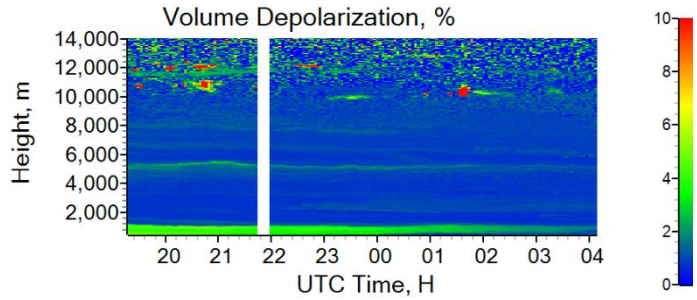
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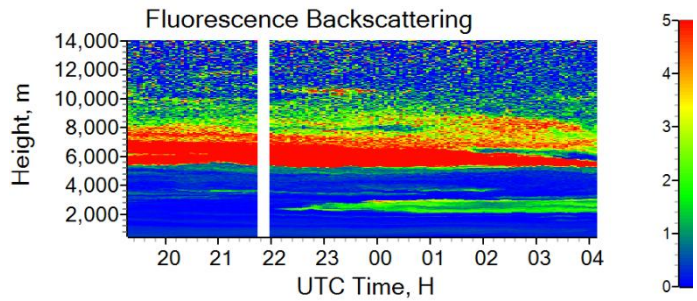
630
631 Fig.7. Formation of ice particles at heights above 10 km inside a smoke layer on 11-12 September 2020.
632 Spatiotemporal variations of range corrected lidar signal at 1064 nm, volume depolarization ratio at
633 1064 nm and fluorescence backscattering coefficient (in 10^{-4} $Mm^{-1}sr^{-1}$).

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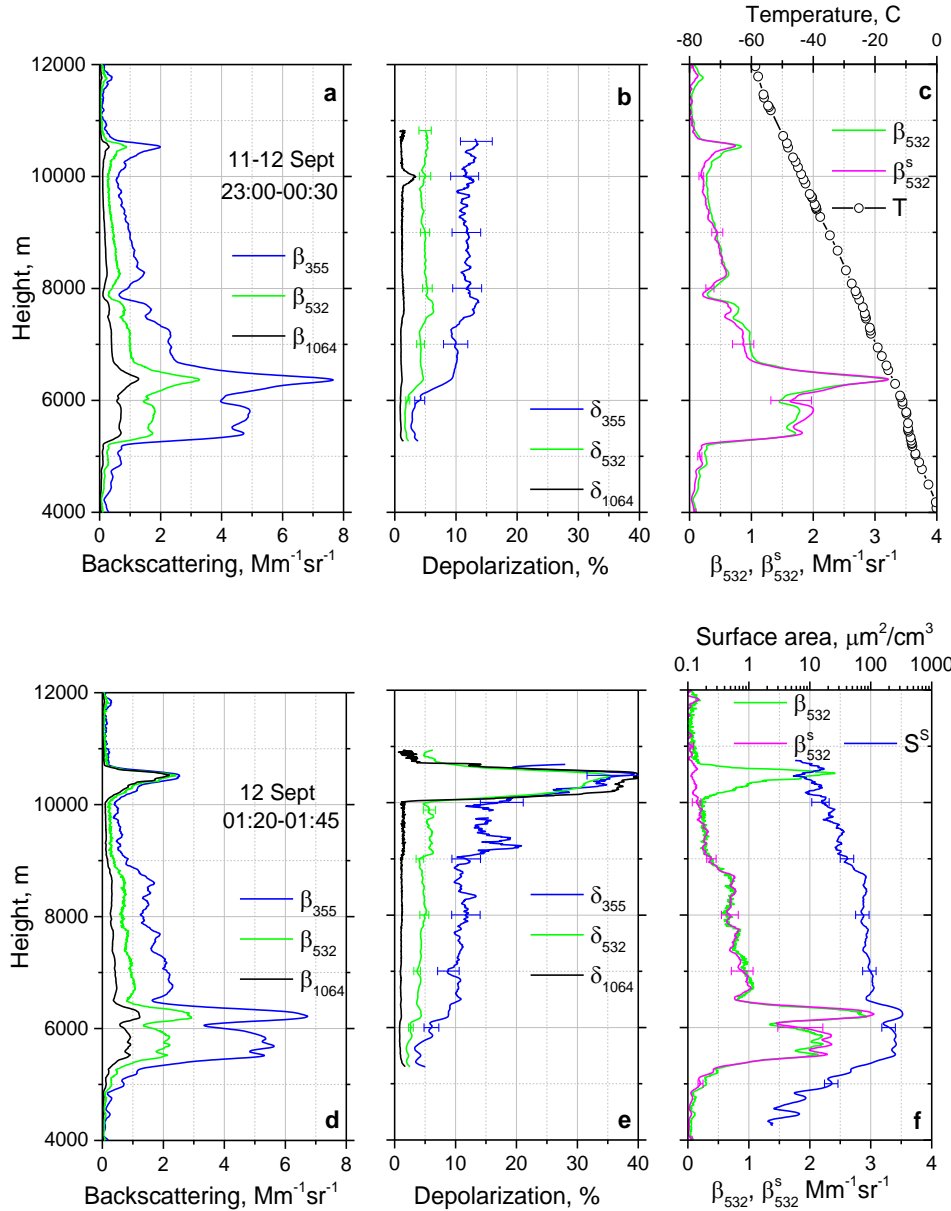
637
638 Fig.8. Formation of ice particles at 10-11 km height inside a smoke layer on 11-12 September
639 2020. Vertical profiles of (a, d) the aerosol backscattering coefficients β_{355}, β_{532}, β_{1064}; (b, e) the
640 particle depolarization ratios δ_{355}, δ_{532}, δ_{1064}; (c, f) β_{532} together with backscattering coefficient of
641 smoke β_{532}^{s}, calculated from fluorescence backscattering β_F assuming $G_F=4.0\times10^{-4}$. Plot (f)
642 shows also the smoke surface area concentration S^S of the smoke particles calculated from β_F by
643 applying the respective conversion factor in Eq. (4). Results are given for the time intervals
644 23:00 – 00:30 UTC and 01:20 – 01:45 UTC: prior and during ice cloud formation at 10.5 km
645 height. The temperature profile measured by the radiosonde at Herstmonceux is shown with open
646 symbols in panel (c).

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650 Fig.9. Formation of ice particles at heights above 8 km inside the smoke layer on 17-18 September
651 2020. Spatiotemporal variations of range corrected lidar signal at 1064 nm, volume depolarization
652 ratio at 1064 nm and fluorescence backscattering coefficient (in 10^{-4} $Mm^{-1}sr^{-1}$).

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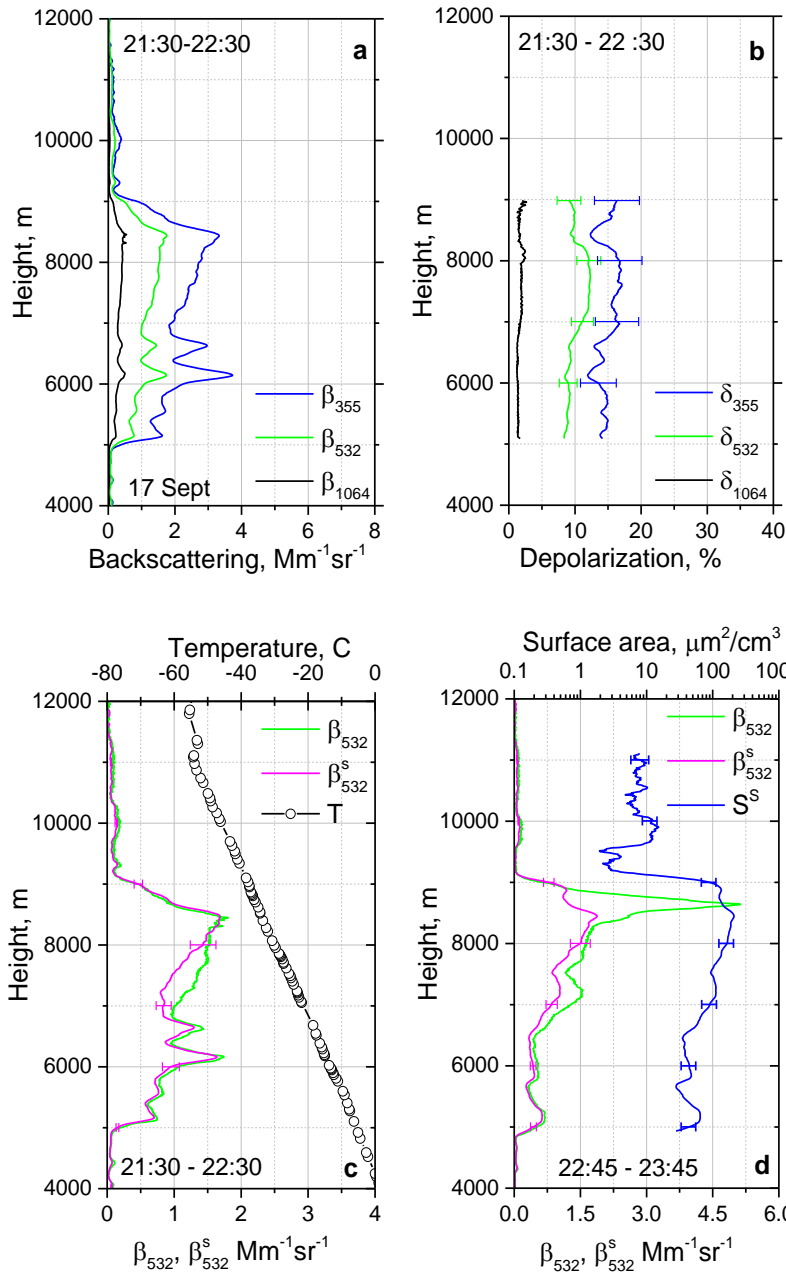
657
658 Fig.10. Formation of ice particles at 8.5-8.6 km height inside a smoke layer on 17 September
659 2020. Vertical profiles of (a) the aerosol backscattering coefficients β_{355}, β_{532}, β_{1064}; (b) the
660 particle depolarization ratios δ_{355}, δ_{532}, δ_{1064}; (c, d) β_{532} together with backscattering coefficient
661 of smoke β_{532}^{s}, calculated from fluorescence backscattering β_{F} assuming G_{F}=3.5×10^{-4}. Results are
662 given for the time intervals (a-c) 21:30 – 22:30 UTC and (d) 22:45 – 23:45 UTC: prior and
663 during ice formation at 8.5 km height. Plot (d) shows also the surface area concentration of the
664 smoke particles S^{S} calculated from β_{F} by applying the respective conversion factor from (4). The
665 temperature profile measured by the radiosonde at Herstmonceux is shown with open symbols in
666 panel (c).