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Combining of Mie-Raman and fluorescence observations: a step forward in aerosol classification with lidar technology

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Abstract

The paper presents an innovative approach to reveal variability of aerosol type at high spatio-temporal resolution, by combining fluorescence and Mie-Raman lidar observations. The multi-wavelength Mie-Raman lidar system in operation at the ATOLL platform (ATmospheric Observatory of liLLe), Laboratoire d'Optique Atmosphérique, University of Lille, includes, since 2019, a wideband fluorescence channel allowing the derivation of the fluorescence backscattering coefficient β_F. The fluorescence capacity G_F, which is the ratio of β_F to the aerosol backscattering coefficient, is an intensive particle's property, strongly changing with aerosol type, thus providing a relevant basis for aerosol classification. In this first single version of the algorithm, only two intensive properties are used for classification: the particle depolarization ratio at 532 nm, and the fluorescence capacity, G_F. We applied our new classification approach to ATOLL high performance lidar data obtained during 2020 – 2021 period, which includes strong smoke, dust and pollen episodes. It is demonstrated that separation of the main particle's types and their mixtures can be performed with height resolution about 60 m and temporal resolution better than 10 minutes for the current lidar configuration.

1. Introduction

Atmospheric aerosol is one of the key factors influencing the Earth's radiation budget through absorption and scattering of solar radiation and by affecting cloud formation. The processes of aerosol–radiation and aerosol-cloud interaction depend on aerosol size, shape, morphology, absorption, solubility, etc., thus knowledge of the chemical composition and mixing

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state of the aerosol particles is important for modeling of aerosol impact (Boucher et al., 2013). The aerosol properties may vary in a wide range, so in practice usually several main types of aerosols are separated on a base of their origin: e.g. urban, dust, marine, biomass burning (Dubovik et al., 2002). Successful remote characterization of column integrated aerosol composition from the observations of Sun – sky photometers and spaceborne multiangle polarimeters was demonstrated in numerous publications (Dubovik et al., 2002; Giles et al., 2012; Hamill et al., 2016; Schuster et al., 2016; Li et al., 2019; Zhang et al., 2020). The aerosol impacts, however, depends also on vertical variations/distributions of particle concentration and composition, which cannot be derived from these instruments.

One of the recognized remote sensing techniques for vertical profiling of aerosol properties is a lidar. Multiwavelength Mie-Raman and HSRL (High Spectral Resolution Lidar) lidar systems provide unique opportunity to derive height-resolved particle intensive properties, such as lidar ratios, Angstrom exponents and depolarization ratios at multiple wavelengths. Based on this information, particle type can be determined (Burton et al., 2012, 2013; Groß et al., 2013; Mamouri et al., 2017; Papagiannopoulos et al., 2018; Nicolae et al., 2018; Hara et al., 2018; Voudouri et al., 2019; Wang et al., 2021; Mylonaki et al., 2021 and references therein). However, there is a fundamental difference in particle classification based on the Sun – sky photometer and on lidar observations. From both direct Sun and azimuth scanning measurements of the photometer more than 100 observations are available. From this information the spectrally dependent refractive index and absorption Angstrom exponent can be determined, which is important for aerosol classification (Schuster et al., 2016; Li et al., 2019). The commonly used multiwavelength lidars are based on a tripled Nd:YAG laser and are capable of providing three backscattering (355 nm, 532 nm, 1064 nm), two extinction (355 nm, 532 nm) coefficients and up to three particle depolarization ratios (so called $3\beta+2\alpha+3\delta$ set). Thus the number of available lidar observations is eight or less which limits the performance of the aerosol typing algorithms. Nevertheless, the results obtained by different research groups demonstrate that lidar-based particle identification is possible. In publications of Burton et al. (2012, 2013) classification was performed from four intensive parameters measured by the HSRL system: the lidar ratio at 532 nm (S_{532}), the backscattering Angstrom exponent for 532/1064 nm wavelengths ($BAE_{532/1064}$), and particle depolarization ratios at 532 nm and 1064 nm (δ_{532}, and δ_{1064}). With these input

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parameters eight aerosol types: smoke, fresh smoke, urban, polluted maritime, maritime, dusty mix, pure dust and ice were discriminated.

Important information on aerosol vertical distribution comes from the EARLINET/ACTRIS lidar-network, aiming at unifying multiwavelength Mie-Raman lidar systems over Europe (Pappalardo et al., 2014). For the automation of aerosol classification, several approaches were developed in the frame of EARLINET. These approaches include the Mahalanobis distance-based typing algorithm (Papagiannopoulos et al., 2018), a neural network aerosol classification algorithm (NATALI) (Nicolae et al., 2018), and algorithm based on source classification analysis (SCAN) (Mylonaki et al., 2021). All these algorithms have demonstrated their ability for aerosol classification. In particular, the NATALI is able to identify up to 14 aerosol mixtures from $3\beta+2\alpha+1\delta$ observations.

Nevertheless, the above-mentioned algorithms have to deal with a fundamental limitation: the particle intensive properties, even for pure aerosols (generated by a single source) exhibit strong variations. For example, the lidar ratio S_{355} of smoke in publication of Nicolae et al. (2018) varies in 38 sr – 70 sr range, and in our own measurements we observed for aged smoke S_{355} as low as 25 sr (Hu et al., 2021). Strong variation of smoke lidar ratios in EARLINET/ACTRIS observations is discussed also in the recent publication of Adam et al. (2021). Such uncertainty in parameters of the aerosol model complicates the aerosol classification. Thus, it is desirable to combine the Mie-Raman observations with another range resolved technique, providing additional independent information about aerosol composition. Such information can be obtained from laser induced fluorescence emission.

Application of fluorescence lidar technique was intensively considered during the last decade to study aerosol particles. Lidar measurements of the full fluorescence spectrum with multianode photomultipliers (Sugimoto et al., 2012; Reichardt et al., 2014, 2017; Saito et al., 2022) provides an obvious advantage in particle identification. However, even a more simple fluorescence lidar with a single wideband fluorescence channel, opens new opportunities for aerosol characterization (Veselovskii et al., 2021a; 2021b; Zhang et al., 2021). Such fluorescence configuration could be implemented in existing Mie-Raman lidars, and the fluorescence backscattering coefficient β_F is calculated from the ratio of fluorescence and nitrogen Raman signals. To characterize the aerosol fluorescence properties, the fluorescence capacity G_F is introduced as the ratio of β_F to aerosol backscattering coefficient at one of laser wavelengths

(Veselovskii et al., 2020). The fluorescence capacity is an intensive particle parameter, which changes strongly with aerosol type, being the highest for smoke and the lowest for dust (Veselovskii et al., 2021a). Thus, the combination of Mie – Raman and fluorescence backscatter provides a basis to improve particle classification.

The goal of the present study is to develop a new approach for the evaluation of the spatio-temporal variations of particle types in the troposphere, thus classification should be performed with sufficient height and temporal resolutions. A Mie – Raman lidar provides several particle intensive parameters, however, the profiles of particle parameters associated with the extinction coefficient, such as lidar ratio or extinction Angstrom exponent, may contain strong noises, because the extinction coefficients are derived from the slope of Raman lidar signals, thus averaging over significant spatio-temporal intervals is demanded. Meanwhile, the particle depolarization and the fluorescence *capacity* can be calculated with high spatio-temporal resolution and in our recent publication (Veselovskii et al., 2021a) we have demonstrated that the $\delta - G_F$ diagram allows to separate several aerosol types, such as dust, pollen, urban (continental) and smoke.

In this paper, we present results of aerosol classification on a base of fluorescence and Mie-Raman lidar measurements performed at the ATOLL (ATmospheric Observation at liLLe) at Laboratoire d'Optique Atmosphérique, University of Lille, during 2020 – 2021 period, which includes strong smoke, dust and pollen episodes. We start with a description of the experimental setup and data processing scheme in Sect.2. In Sect.3 we present the algorithm for aerosol classification on a base of depolarization and fluorescence measurements. Results of the application of the developed approach to different atmospheric situations, including smoke, dust and pollen episodes are given in Sect.4.

2. Experimental setup and data analysis

2.1. Lidar system

The multiwavelength Mie-Raman lidar LILAS (LIlle Lidar AtmosphereS) is based on a tripled Nd:YAG laser with a 20 Hz repetition rate and pulse energy of 70 mJ at 355 nm. Backscattered light is collected by a 40 cm aperture Newtonian telescope and the lidar signals are digitized with Licel transient recorders with 7.5 m range resolution, allowing simultaneous detection in the analog and photon counting mode. The system is designed for the detection of elastic and Raman backscattering, allowing the so called 3β+2α+3δ data configuration, including three particle backscattering (β_{355}, β_{532}, β_{1064}), two extinction (α_{355}, α_{532}) coefficients along with three particle depolarization ratios (δ_{355}, δ_{532}, δ_{1064}). The particle depolarization ratio, determined as a ratio of cross- and co-polarized components of the particle backscattering coefficient, was calculated and calibrated in the same way as described in Freudenthaler et al. (2009). Many calibration and operation procedures have been automated for the LILAS system to improve the overall performance of the lidar in terms of observation frequency and data quality. The aerosol extinction and backscattering coefficients at 355 and 532 nm were calculated from Mie-Raman observations (Ansmann et al., 1992), while β_{1064} was derived by the Klett method (Klett, 1985). For calculation of α and β at 532 nm we use the rotational Raman scattering instead of the vibrational one (Veselovskii et al., 2015), which allows to increase the power of Raman backscatter and to decrease separation between the wavelengths of elastic and Raman components. Additional information about atmospheric parameters was available from radiosonde measurements performed at Herstmonceux (UK) and Beauvechain (Belgium) stations, located 160 km and 80 km away from the observation site respectively.

The LILAS system can also profile the laser induced fluorescence of aerosol particles. A part of the fluorescence spectrum is selected by a wideband interference filter of 44 nm width centered at 466 nm (Veselovskii et al., 2020). The strong sunlight background at daytime restricts the fluorescence observations to nighttime hours. The fluorescence backscattering coefficient β_F, is calculated from the ratio of fluorescence and nitrogen Raman backscattering signal, as described in Veselovskii et al. (2020). This approach allows us to evaluate the absolute values of β_F, if the relative sensitivity of the channels is calibrated and the nitrogen Raman scattering differential cross section is known. All β_F profiles presented in this work were smoothed with the Savitzky – Golay method, using second order polynomials with 21 points in the window. For the calculation of the fluorescence capacity G_F, in principle, backscattering coefficients at any laser wavelength can be used. In our study we always used β_{532}, because it is calculated with the use of rotational Raman component and is considered to be the most reliable, thus the fluorescence capacity is calculated as $G_F = \frac{\beta_F}{\beta_{532}}$.

2.2.Calculation of the particle backscattering coefficient from Mie-Raman measurements

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Mie – Raman lidar measurements allow independent evaluation of aerosol extinction and backscattering coefficients. Commonly used approach for β calculation was formulated in the paper of Ansmann et al. (1992). This approach includes the choice of a reference height, where the scattering is purely molecular. However, such range is not always available, for example, in the presence of the low level clouds. Moreover, when long-term spatio-temporal variations of backscattering coefficients are analyzed, the uncertainty in the choice of the reference height leads to oscillations in β profiles. To resolve this issue, we modified the Raman method as described below.

In an elastic channel, the backscattered radiative power P_L, at wavelength λ_0 and distance z is described by the lidar equation:

$$P_L = O(z)\frac{1}{z^2}C_L(\beta_L^a + \beta_L^m)\exp\left\{-2\int_0^z(\alpha_L^a + \alpha_L^m)dz'\right\} = O(z)\frac{1}{z^2}C_L(\beta_L^a + \beta_L^m)T_L^2, \quad (1)$$

while in a Raman channel, it can be written as:

$$P_R = O(z)\frac{1}{z^2}C_R\beta_R\exp\left\{-\int_0^z(\alpha_L^a + \alpha_R^a + \alpha_L^m + \alpha_R^m)dz'\right\} = O(z)\frac{1}{z^2}C_R\beta_R T_L T_R. \quad (2)$$

Here O(z) is the geometrical overlap factor, which is assumed to be the same for elastic and Raman channels. C_L and C_R are the range independent constants, including efficiency of the detection channel. T_L and T_R are one-way transmissions, describing light losses on the way from the lidar to distance z at laser λ_L and Raman λ_R wavelengths. Backscattering and extinction coefficients contain aerosol and molecular contributions: $\beta_L^a + \beta_L^m$ and $\alpha_L^a + \alpha_L^m$, where the superscripts "a" and "m" indicate aerosol and molecular scattering, respectively. Raman backscattering coefficient is:

$$\beta_R = N\sigma_R, \quad (3)$$

where N is the number of Raman scatters (per unit of volume) and σ_R is the Raman differential scattering cross section in the backward direction.

Dividing equation (1) on (2) we get:

$$\frac{P_L}{P_R} = \frac{C_L}{C_R}\frac{(\beta_L^a + \beta_L^m)}{\beta_R}\frac{T_L}{T_R} \quad (4)$$

Backscattering coefficient is calculated from (3) and (4) as:

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$$\beta_L^a = \frac{P_L}{P_R}\frac{C_R}{C_L}\sigma_R N \frac{T_R}{T_L} - \beta_L^m = \frac{P_L}{P_R} KN \frac{T_R}{T_L} - \beta_L^m \quad (5)$$

The differential transmission $\frac{T_L}{T_R}$ can be calculated the same way, as it is done for the water vapor measurements (Whiteman, 2003). For rotational Raman signal, which we use in our 532 nm channel (Veselovskii et al., 2015), $\lambda_L \approx \lambda_R$, so $\frac{T_L}{T_R}$ =1.

The calibration constant $K = \frac{C_R}{C_L}\sigma_R$ can be found by comparing β_L^a in Eq.5 with the backscattering coefficient $\tilde{\beta}_L^a$ computed with the traditional Raman method, using the reference height (Ansmann et al., 1992).

$$K = (\tilde{\beta}_L^a + \beta_L^m)\frac{P_R}{P_L}\frac{1}{N}\frac{T_L}{T_R} \quad (6)$$

For simplicity, hereinafter we will use notation β_L instead β_L^a. Thus, if during the measurement session we have a temporal interval, where the reference height is available, we can determine the calibration constant K and use it for β_L calculations from eq.5, assuming that relative sensitivity of channels during the session is not changed. Even if cloud layers occur during the whole session, we can use K from the previous cloud-free profiles (assuming, again, that the relative sensitivity of channels is the same). We will call this approach for β calculation as “modified Raman method”, to distinguish it from traditional one (Ansmann et al., 1992).

To estimate variations of the relative sensitivity of the channels, we analyzed long-term cloudless measurements when the reference height was available for every individual profile. The results demonstrate that variations of calibration constant during the session (about 8 hours) were below 3%. Fig.1 and 2 present the application of this modified Raman method to the measurements on 2 March 2021. The dust layer extended from 2 km to 8 km height and inside this layer the ice and liquid clouds were formed during the 00:00 – 05:00 UTC interval, thus β_{532} could not be calculated with traditional Raman technique. The temporal interval 19:00 – 20:00 was used to find calibration constant K. Fig.1 shows vertical profiles of backscattering coefficient $\tilde{\beta}_{532}$ calculated with traditional Raman method (with reference height), and β_{532} calculated with modified method (with the calibration constant). Profiles of $\tilde{\beta}_{532}$ and β_{532}

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coincide for the whole height range. The calibration constant *K*, shown on the same plot, does not demonstrate height dependence, though oscillations around the mean value increase with height. For computations, we choose the value of *K* at low altitudes averaged inside some height interval. Fig.2 provides spatio-temporal variations of β_{532}, particle depolarization δ_{532} and the fluorescence capacity G_F.

Depolarization measurements reveal the presence of dust (δ_{532}≈30%) and the ice cloud above 4 km (δ_{532}>40%). The liquid cloud below 4 km after midnight can be identified by a low depolarization ratio δ_{532}<3%. The fluorescence capacity of dust is low, about 0.2×10^{-4}. However, below 2 km, G_F is significantly higher, up to 1.2×10^{-4}. In combination with a high depolarization ratio (up to 20%), it can indicate the presence of pollen at low altitudes. On the fluorescence capacity panel, we can clearly see that after 01:00 UTC the dust and pollen layers are mixed below 2 km, resulting in a value of G_F about 0.5×10^{-4}. The fluorescence capacity inside ice and liquid clouds is low, below 0.01×10^{-4}. Fig.2 clearly demonstrates the advantage of simultaneous depolarization and fluorescence measurements for the study of cloud formation in the presence of aerosol. All spatio-temporal distributions of β_{532} presented in this paper were calculated from Eq.5 with a modified Raman method.

3. Aerosol classification based on fluorescence measurements

3.1. *Approach for aerosol classification*.

As was discussed in our recent publication (Veselovskii et al., 2021), the δ-G_F diagram allows to separate several aerosol types, including smoke, dust, pollen, urban, ice and liquid water particles. Smoke and urban aerosols both have a small depolarization ratio, but the fluorescence capacity of smoke is almost one order higher, so these particles can be separated. Dust and pollen both have high depolarization ratio (up to 30%), but G_F of dust is significantly lower, which again provides basis for discrimination. The depolarization ratio of some aerosol types is characterized by strong spectral dependence. For example, the depolarization ratio of aged smoke decreases with wavelength. It is below 5% at 1064 nm but at 355 nm in upper troposphere it may exceed 20% (Haarig et al., 2018; Hu et al., 2019; Veselovskii et al., 2021b), which complicates smoke and dust separation. For pollen, on the contrary, the depolarization ratio at 1064 nm can be the highest (Veselovskii et al., 2021a). Thus, choice of δ_{1064} for δ-G_F diagram could be advantageous. However, as mentioned, the backscattering coefficient at 1064

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nm is calculated with Klett method (Klett, 1985), which, besides assumption about lidar ratio, needs reference height and cannot be used in cloudy situations. This is why in our study we used the δ_{532}-G_F diagram.

In our present work, we consider a simple classification scheme since we use only two intensive parameters G_F and $\delta_{532.}$ Our goal is to demonstrate that in the δ_{532}-G_F diagram, our lidar observations form clusters and characteristic patterns which can be attributed to different aerosol types or their mixtures. We consider four aerosol types: dust, smoke, pollen and urban, and two cloud types: liquid and ice clouds. Dust and pollen are large particles of complicated shape, while smoke and urban pollution are small particles with low depolarization. In our classification "urban aerosol" includes continental aerosol, sulfates and soot. At this stage, we do not discriminate particles by their absorption.

Typical ranges of G_F and δ_{532} variations for four aerosol types are given in Table 1 and are shown in Fig.3. These ranges are based on results obtained in LOA and on particle depolarization ratios commonly used for aerosol classification (Nicolae et al., 2018; Papagiannopoulos et al., 2018, Mylonaki et al., 2021). The aerosol parameters, even for a single type, may present significant variations. Moreover, actual aerosols exist usually as mixtures. For example, the depolarization ratio δ_{532} of Saharan dust near the source regions is up to 35% (Veselovskii et al., 2020a), but after transportation and mixing with local aerosol δ_{532} can be below 20% (Rittmeister et al., 2017). In many studies, the dust with decreased depolarization ratio is classified as "polluted dust" (e.g. Burton et al., 2012, 2013). At a moment, we do not introduce the discrimination between the two subtypes and mark as "dust" the particles with 15%<δ_{532}<35%, and 0.1×10^{-4}<G_F <0.5×10^{-4}. Urban and smoke particles both have a low depolarization ratio δ_{532}<8%, but the fluorescence capacity of smoke is almost one order higher.

The pollen over north of France is usually mixed with other aerosols, and the particles, which we mark as "pollen" are actually the mixtures containing pollen. Depolarization ratio of clean pollen varies strongly for different taxa (Cao et al., 2010). For birch pollen, Cao et al. (2010) reported δ_{532}=33%, and in the measurements over Finland during birch pollination (Bohlmann et al., 2019), observed values of δ_{532} up to 26%. In our classification scheme we type as "pollen" the particles mixtures with 15%<δ_{532}<30%, and 0.8×10^{-4}<G_F <3.0×10^{-4}. Thus, the ranges of the parameters for different aerosol types chosen in Fig.3 and Table 1 include as variation of "pure" aerosol parameters as their possible "contamination" by other particle types.

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Table 1. Ranges of particle depolarization δ_{532} and fluorescence capacity G_F, which were used for classification of four types of aerosols.

Aerosol type	δ_{532} (%)	G_F, ($\times 10^{-4}$)
Dust	15 - 35	0.1 – 0.5
Pollen	15 - 30	0.8 – 3.0
Urban	1 - 8	0.1 – 0.8
Smoke	1- 8	2.0 – 6.0

The mixing of aerosols should be revealed in δ_{532}-G_F diagram. For example, pollen can be mixed with urban particles. At different heights the pollen contributes differently to β_{532}, so at δ_{532}-G_F diagram, the data points will form the pattern, which extends from location, attributed to "pure" urban aerosol to location, attributed to "pure" pollen. To estimate, how such pattern looks like, a simplified modeling for fixed particle parameters was performed. Corresponding results are shown in Fig.3 by symbols (circles). The particle depolarization ratio δ of the mixture, containing urban aerosol (u) and pollen (p), with depolarization ratios δ^u and δ^p, can be calculated as:

$$\delta = \frac{\left(\frac{\delta^p}{1+\delta^p}\right)\beta^p + \left(\frac{\delta^u}{1+\delta^u}\right)\beta^u}{\frac{\beta^p}{1+\delta^p} + \frac{\beta^u}{1+\delta^u}} \tag{7}$$

The fluorescence capacity of the mixture is given by:

$$G_F = \frac{\beta^u G_F^u + \beta^p G_F^p}{\beta} \tag{8}$$

Here total backscattering $\beta = \beta^u + \beta^p$.

We assume that the depolarization ratios of pollen and urban aerosol are δ_{532}^p=30% and δ_{532}^u=3%, while the fluorescence capacities are G_F^u=0.2×10^{-4} and G_F^p=2.5×10^{-4}. The calculations in Fig.3 were performed for values of pollen contribution $\frac{\beta_{532}^p}{\beta_{532}}$ in 0 - 1.0 range with step 0.1. In the δ_{532}-G_F diagram the computed points provide a characteristic curve, which in the next section will be compared with experimental results. The same computations were performed for a smoke (s) and dust (d) mixture, assuming δ_{532}^d=30%, δ_{532}^s=3%, G_F^d=0.2×10^{-4} and G_F^s=4.0×10$^{-4.}$ Corresponding results are shown in Fig.3 with stars. In a similar way, the characteristic curves for other mixtures can be also represented.

We are also able to identify liquid water and ice layers. Liquid water cloud layers have low fluorescence capacity (G_F<0.01×10^{-4}) and δ_{532}<3%. Ice particles also have low G_F, but at heights where ice clouds are usually observed, the signal of fluorescence backscattering is noisy. Thus at high altitudes ice particles are discriminated by a high depolarization ratio δ_{532}>40 %.

3.2. *Classification of spatio-temporal observations*

The input parameters in our classification scheme are the spatio-temporal distributions of β_{532}, δ_{532} and G_F, which are presented as matrices $\beta_{532}^{i,j}$, $\delta_{532}^{i,j}$, $G_F^{i,j}$, where i=1… N_T; j=1… N_H. Values N_T and N_H are the numbers of temporal and height intervals in the analyzed dataset. In a single measurement we accumulate 2×10^3 laser pulses, so temporal resolution of the measurements is about 100 s, while the height resolution is 7.5 m.

The particle intensive parameters cannot be evaluated reliably when the backscattering coefficient is low. Thus, we set a threshold value for β_{532} (normally 0.2 Mm^{-1}sr^{-1}); namely, when $\beta_{532}^{i,j}$<0.2 Mm^{-1}sr^{-1} the elements of the matrices $\delta_{532}^{i,j}$ and $G_F^{i,j}$, are classified as “low signal” and ignored. For the remaining elements, we determine the aerosol type, using our two-staged typing algorithm. On the first stage, a primary typing is being made for each point *(i,j)* separately, in accordance with parameter ranges given in the Table 1. The elements, which are out of all these ranges, are marked as “undefined”. We consider 6 types of the particles, respectively dust, smoke, pollen, urban, ice crystals and water droplets. Moreover, there can be two additional results of primary typing: “undefined” and “low signal”. Thus, there are altogether 8 possible results of primary typing.

The particle parameters, calculated with high spatio-temporal resolutions, contain statistical noise which influences the results of the primary typing, thus producing high frequency oscillations of non-physical character. From a physical point of view, the aerosol single-type areas should form smooth regions, so a special smoothing procedure (stage 2 of our algorithm) was developed to remove the oscillations. The smoothing procedure is based on a convolution with Gaussian kernel

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$$Z = \exp\left(-\left(\frac{t^2}{s_T^2} + \frac{h^2}{s_H^2}\right)\right) \quad (9)$$

where t and h are temporal and height coordinates. The resolution of typing is being controlled by the parameters s_T and s_H, which are set as the number of temporal and height bins. In the results presented we used s_T=3 and s_H=5, thus the temporal and height resolution of our typing procedure is estimated to be about 8 minutes and 60 m respectively.

4. *Application of classification algorithm to LILAS data*

The classification algorithm, described in the previous section, was applied to the data of the Mie-Raman- Fluorescence lidar at the ATOLL platform, located on the campus of Lille University, during 2020 – 2021 period. Here we present results of aerosol classification for several relevant atmospheric situations, to demonstrate that different aerosol types are well separated based on δ_{532}-G_F diagram.

12 September 2020: Wildfire smoke

Fig.4 presents the spatio-temporal variations of aerosol and fluorescence backscattering coefficients (β_{532} and β_F) together with the particle depolarization ratio δ_{532} and the fluorescence capacity G_F during smoke episode on the night 12-13 September 2020. The smoke layer extends from approximately 2 km to 5 km height, and it is characterized by high fluorescence capacity $G_F>3.0\times10^{-4}$ and low depolarization ratio $\delta_{532}<7\%$. The cirrus clouds occurred above 11 km height during the whole night. The smoke layer was transported from North America; detailed analysis of the layer origin and transportation is given in the recent publication of Hu et al. (2021). The results of aerosol typing for this episode are shown in Fig.5. On the δ_{532}-G_F diagram these data form two clusters. First cluster includes points in the range $2.0\times10^{-4}<G_F<6.0\times10^{-4}$ and $2\%<\delta_{532}<7\%$, such high fluorescence and low depolarization are typical for smoke particles. The second cluster consists of points localized inside $0.1\times10^{-4}<G_F<0.8\times10^{-4}$ and $1\%<\delta_{532}<3\%$ intervals, which is typical of urban particles. After cluster localization, the observations can be plotted as aerosol types, using the parameters in Table 1 and the FBC algorithm, described in section 3.2. The aerosol types in Fig.5b are spatially separated and contain no high frequency oscillations. Urban particles are localized at low heights, below 1 km. We would like to remind that, at the condition of high relative humidity (RH), the fluorescence *capacity* can decrease due to the particle's hygroscopic growth. In accordance with radiosonde data the relative humidity

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below 1 km was quite high (about 70% at 500 m), which can explain the wide range of G_F variation observed for urban particles in Fig.5a.

30 May 2020: Urban vs Pollen

Pollen grains represent a significant fraction of primary biological materials in the troposphere and fluorescence induced emission provides an opportunity for their identification. Fig.6 presents spatio-temporal variations of β_{532}, β_F, δ_{532}, G_F during pollen season on the night 30-31 May 2020. Presence of different types of pollen over Lille in Spring – Summer 2020 was discussed in our recent publication (Veselovskii et al., 2021). In accordance with radiosonde data from Herstmonceux station, the RH at midnight was about 40% at 500 m and it increased up to 70% at 2000 m. The aerosol is located inside the planetary boundary layer (PBL) below 2.5 km. At altitudes below 1 km, the depolarization ratio δ_{532} after 23:00 increases up to 20% simultaneously with an increase of the fluorescence capacity above 2.0×10^{-4}, which can be an indication of pollen presence.

On the δ_{532}-G_F diagram in Fig.7a, the data points spread from the values typical for the urban particles to the values typical for the pollen. Contribution of pollen to the total backscattering changes with height and the points form the pattern, similar to characteristic curve, calculated for urban – pollen mixture in Fig.3. The spatio–temporal distribution of aerosol types is shown in Fig.7b. The urban particles are predominant, while pollen is localized below 1 km height. The grey color corresponds to unidentified aerosol type, which in our case is the mixture of urban particles and pollen.

One indicator of pollen presence in an aerosol mixture, can be a higher value of δ_{1064} in respect to δ_{532} (Veselovskii et al., 2021). Fig.8a shows the vertical profile of the backscattering coefficient β_{532} averaged over 23:00 – 01:00 UTC period together with the particle depolarization ratios δ_{532} and δ_{1064}. The ratio $\frac{\delta_{1064}}{\delta_{532}}$ is about 1.5 at 0.75 km height, which corroborates suggestions about pollen presence. Both depolarization ratios decrease with height, simultaneously with decrease of fluorescence capacity, as follows from Fig.8b. The backscattering Angstrom exponent (BAE) $A^{\beta}_{532/1064}$ in Fig.8b is about 1.25 and it does not demonstrate significant variations in 0.7 km – 2.25 km height range. The reason can be that the

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contribution of pollen to the total backscattering is not high and the corresponding effect can be masked by other processes, such as particle hygroscopic growth.

14 September 2020: wildfire smoke vs pollen mixture

Another strong smoke episode occurred in the night 14-15 September 2020, and corresponding distributions of β_{532}, β_F, δ_{532}, and G_F are shown Fig.9. The elevated smoke layer with low depolarization ratio (δ_{532}<5%) and high fluorescence capacity (up to 4.0×10^{-4}) was observed at approximately 6 km height during the whole night. Inside the PBL the depolarization ratio is higher, up to 15%, while fluorescence capacity is low, compared to the elevated layer (about 1.0×10^{-4}). On the δ_{532}-G_F diagram in Fig.10a we can see the cluster of data points, corresponding to the smoke. The same time, a part of the points are inside the range of parameters attributed to the pollen (Table 1). The remaining points should be attributed to the mixture of pollen, smoke and urban aerosol. On the distribution of the particle types (Fig.10b) this mixture is marked with gray color. The pollen particles are localized below 1 km. Presence of pollen over Lille in September is not common, but it can be transported from other regions. The transport of pollen can be analyzed with a global-to-meso-scale dispersion model SILAM (Sofiev et al., 2006). The SILAM pollen index for this date demonstrates the transport of pollen to northern France from the southeast of France and the east Mediterranean.

Fig.11a presents profiles of δ_{532} and δ_{1064} together with β_{532} for the temporal interval 00:00 – 04:00 UTC. The relative humidity, in accordance with radiosonde data from Herstmonceux station, did not exceed 50% below 1.7 km. Above that height RH increased up to 75% at 2.5 km, thus the observed increase of β_{532} above 1.5 km can be partly related to RH growth. The relative humidity inside the smoke layer did not exceed 10%. Similarly to Fig.8, δ_{1064} exceeds δ_{532} at low heights. The ratio $\frac{\delta_{1064}}{\delta_{532}}$ is about 1.5 at 1 km and inside the smoke layer $\frac{\delta_{1064}}{\delta_{532}} \approx 0.4$. Higher values of depolarization ratio at 532 nm compared to 1064 nm are typical for aged smoke (Haarig at al., 2018; Hu et al., 2019, 2021). The BAE does not present significant height variations: $A^{\beta}_{532/1064}$ is about 1.0 inside the PBL and it increases to 1.25 inside the smoke layer (Fig.11b). Simultaneously, the fluorescence capacity in the smoke layer increases about a

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factor 4, comparing to the PBL, which demonstrates efficiency of the fluorescence technique for discriminating smoke from other aerosol types.

10 April 2020: Urban vs Pollen

In the beginning of April, we experienced several atmospheric situations, for which elevated layers were classified as urban aerosols. One of such cases, on the night 10 -11 April 2020, is shown in Fig.12. Lidar observations were performed at an angle of 45 degrees to the horizontal, so the minimum height reachable in the analysis is 350 m. The relative humidity, in accordance with radiosonde data from Herstmonceux station, increased with height from 54% at 1.0 km to 65% at 2.2 km. The layer with depolarization ratio δ_{532} below 5% was observed at about 2 km height during the night. The fluorescence capacity in the layer is low (below 0.5×10^{-4}), so it is identified as urban aerosol. For the period 21:00 – 23:00 UTC the depolarization ratio below 500 m has increased simultaneously with the fluorescence capacity, which can be an indication of pollen presence.

On the δ_{532}-G_F diagram (Fig.13a) most of the points are classified as urban aerosol. However, at low altitudes the particles have relatively high depolarization ratios δ_{532}>15%, and the points on δ_{532}-G_F diagram provide the pattern typical for urban – pollen mixture. Our algorithm identifies particles mainly as urban aerosol (Fig.12b), and the regions with grey color at low heights, correspond to urban – pollen mixture.

The presence of pollen is supported by the profiles of δ_{532} and δ_{1064} shown in Fig.14. At low heights δ_{1064} exceeds δ_{532} and the ratio $\frac{\delta_{1064}}{\delta_{532}}$ is about 1.4 at 0.5 km. However, inside the elevated layer this ratio decreases and becomes about 0.8 at 2.25 km, which indicates that mixture composition changed and the pollen contribution decreased. In the same height range the fluorescence capacity decreases from 0.38×10^{-4} to 0.28×10^{-4}, while BAE gradually increases from 0.75 to 1.25, which can be due to decrease of pollen contribution.

11 August 2021: contacting layers of smoke and urban aerosol

Separation of smoke and urban particles is a challenging task for Mie – Raman lidar, because both types have small effective radius, and similar depolarization ratios δ_{532}. However, the fluorescence capacity of smoke is about factor 4-5 higher than that of urban aerosol, which

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allows their reliable separation. The analyses of the measurements in the night 11-12 August 2021 are shown in Fig.15. The RH decreases with height from 70% to 40% inside 500 m – 2250 m range. The main part of aerosol is concentrated below 2500 m and two height intervals can be distinguished. Above approximately 1500 m the layer with high fluorescence capacity (up to 3.0×10^{-4}) is observed, while in the layer below 1500 m, the G_F is low, (below 0.8×10^{-4}).

On the δ_{532}-G_F diagram in Fig.16a one cluster of points is localized mainly inside the interval $2.0\times10^{-4}<G_F<4.0\times10^{-4}$ and $4\%<\delta_{532}<10\%$. Such properties can be attributed to smoke. The points in Fig.16a form also a pattern typical for urban – pollen mixture. From the distribution of aerosol types in Fig.16b we conclude that the points in the first cluster correspond to the upper smoke layer, while the lower layer is represented by urban particles and by their mixture with pollen. The pollen becomes predominant below 800 m during 20:00 – 22:00 UTC. The smoke and urban layers are in contact and the particle mixing occurs, which increases dispersion within the clusters.

1 April 2021: Dust

Dust layers transported from Africa are regularly observed over North Europe and especially North of France. One such dust episode took place in the night 1-2 April 2020 and the corresponding spatio-temporal variations of β_{532}, β_F, δ_{532}, and G_F are shown in Fig.17. The dust layer, with depolarization ratio exceeding 30%, and low fluorescence, extends from approximately 1.0 km to 5.0 km height. The fluorescence capacity varied inside the layer. In the center it was the lowest (about 0.1×10^{-4}), but at the bottom of the layer and near the top, G_F increased up to $(0.2\div0.3)\times10^{-4}$, probably due to the mixing with local pollution. In Fig.18a, (δ_{532}-G_F diagram), we observed a typical cluster of dust particles. There is also a second small cluster which is attributed to urban aerosols. On the distribution of particle types in Fig.18b the urban aerosol occurs below 800 m after 23:00 UTC.

Conclusion

The results presented in this study can be considered as the first important step in the combination of Mie – Raman and fluorescence lidar data. In this version of our algorithm, only two intensive parameters are used for classification: the particle depolarization ratio δ_{532} and the fluorescence capacity G_F. These parameters are chosen because they are specific for different

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types of aerosol and can be calculated with high spatio-temporal resolution. Moreover, δ_{532} and G_F can be calculated at lower altitudes, compared to extinction related parameters, such as lidar ratio and extinction Angstrom exponent. Thus classification, in principle, is possible at ranges with incomplete geometrical overlap. Finally, computation of β_F does not demand the use of reference height, only calibration of relative sensitivity of the channels is needed. In our version of algorithm for β_{532} calculation, we also use calibration constant instead of the reference height. Thus, aerosol classification is possible, even in the presence of low level clouds.

Though only two input parameters are considered in the classification algorithm, the use of fluorescence measurements provides advances in aerosol classification. Analysis of numerous observations, performed at Lille University for the period 2020 – 2021 demonstrates the possibility to separate four types of aerosols, such as dust, smoke, pollen and urban. Moreover, we are able to identify the layers containing the liquid water particles and ice. The number of determined aerosol classes can be increased, by considering the particle mixtures. In particular, "pure" dust can be considered separately from "polluted" one. Polluted dust can be discriminated by lower values of the depolarization ratio and by higher the fluorescence capacity.

Fluorescence technique is especially promising for separation of smoke and urban particles, because fluorescence capacity of smoke is about factor five higher. The important advantage of fluorescence measurements is the ability to identify the biological particles in the atmosphere, such as pollen, which are usually not included in the classification schemes, based on Mie-Raman observations. At the same time, our observations demonstrate that biological particles are frequently observed during Spring – Autumn seasons and may contribute significantly to the aerosol composition inside the PBL. The developed approach allows to identify aerosol types with high spatio-temporal resolutions, which is estimated to be 60 m for height and less than 10 minutes for time, for the current instrumental configuration. Such resolution provides an opportunity for investigating the dynamics of aerosol mixing in the troposphere.

The next step in algorithm development will be the increase of input parameters number. We plan to include the backscattering Angstrom exponents and the depolarization spectral ratios ($\delta_{355}/\delta_{532}$ and $\delta_{532}/\delta_{1064}$), which can be also calculated with high spatio-temporal resolutions. The fluorescence capacity depends on the relative humidity, due to the effects of hygroscopic growth. Thus, information about spatio-temporal distribution of RH should be included in the analysis. It

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is also important to combine our algorithm with existing classification schemes, which we plan to consider in the near future.

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

Author contributions. IV processed the data and wrote the paper. QH and TP performed the measurements. PG supervised the project and helped with paper preparation. BB prepared algorithm for aerosol classification. MK developed software for data processing.
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Competing interests. The authors declare that they have no conflict of interests.

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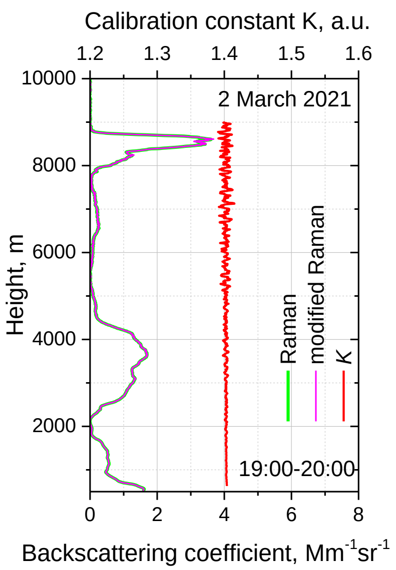

Fig.1. Backscattering coefficients at 532 nm for period 19:00 – 20:00 UTC on 2 March 2021 calculated from Mie-Raman observations using the reference height (green) and the calibration constant *K* (magenta). Calibration constant is shown with red line.

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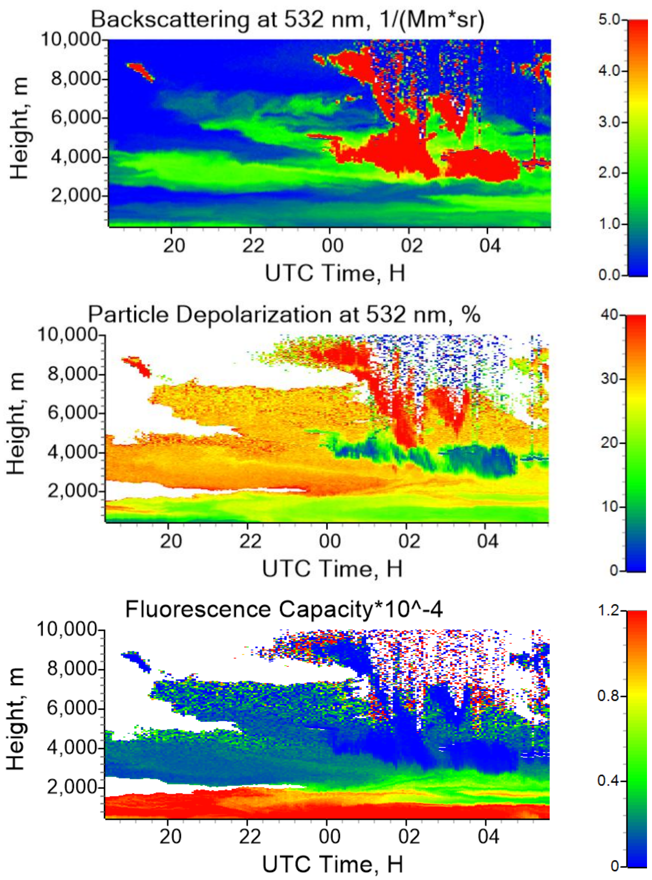

Fig.2. Spatio-temporal distributions of the backscattering coefficient β_{532}, the particle depolarization ratio δ_{532} and the fluorescence capacity G_F in the night 2-3 March 2021. The backscattering coefficient β_{532} is calculated with the modified Raman method. The values of δ_{532}, and G_F are shown for β_{532}>0.2 $Mm^{-1}sr^{-1}$.

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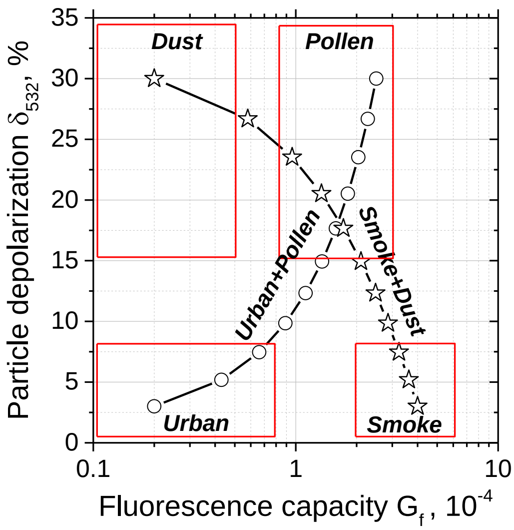

Fig.3. Aerosol typing with δ_{532}-G_F diagram. Chosen ranges of the particle parameters variation for dust, pollen, smoke and urban aerosol are shown by rectangles. The symbols show the results of simulation performed for pollen+urban (circles) and smoke + dust (stars) mixtures. Relative contribution of pollen (smoke) to the total backscattering β_{532} varied in 0 – 1.0 range with step 0.1. Particle parameters used in calculations are given in the text.

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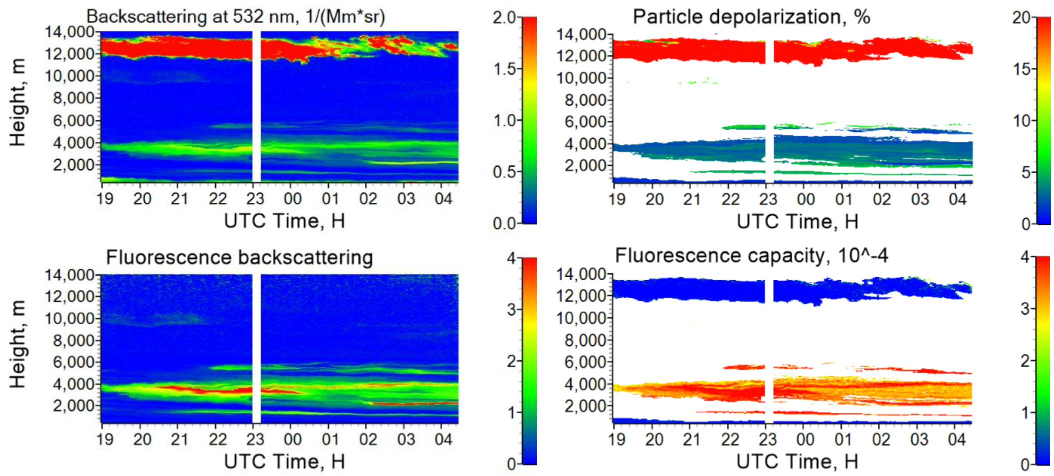

Fig.4. Spatio-temporal distributions of the backscattering coefficient β_{532}, the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$), the particle depolarization ratio δ_{532}; and the fluorescence capacity G_F in the night 12-13 September 2020. Calculation of δ_{532} and G_F was not performed for β_{532}<0.2 $Mm^{-1}sr^{-1}$.

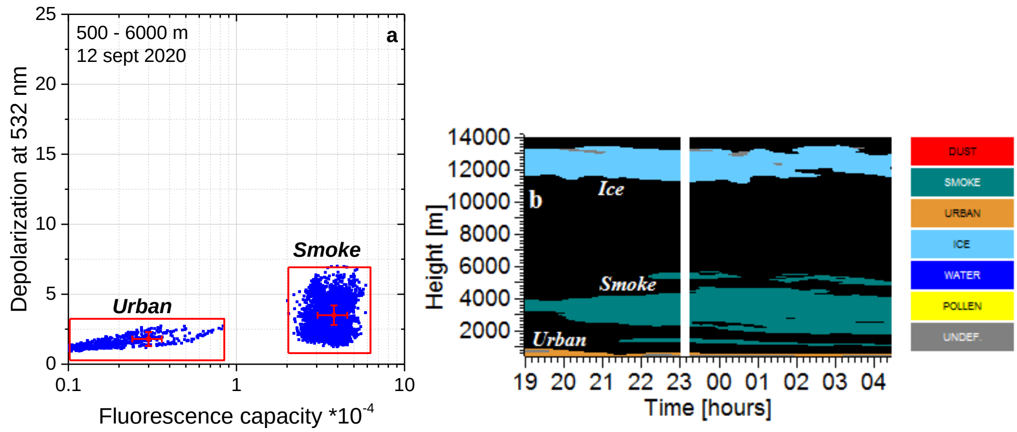

Fig.5 (a) The δ_{532}-G_F diagram for data from Fig.4 in 500 – 6000 m height range; (b) spatio-temporal distribution of aerosol types in the night 12-13 September 2020.

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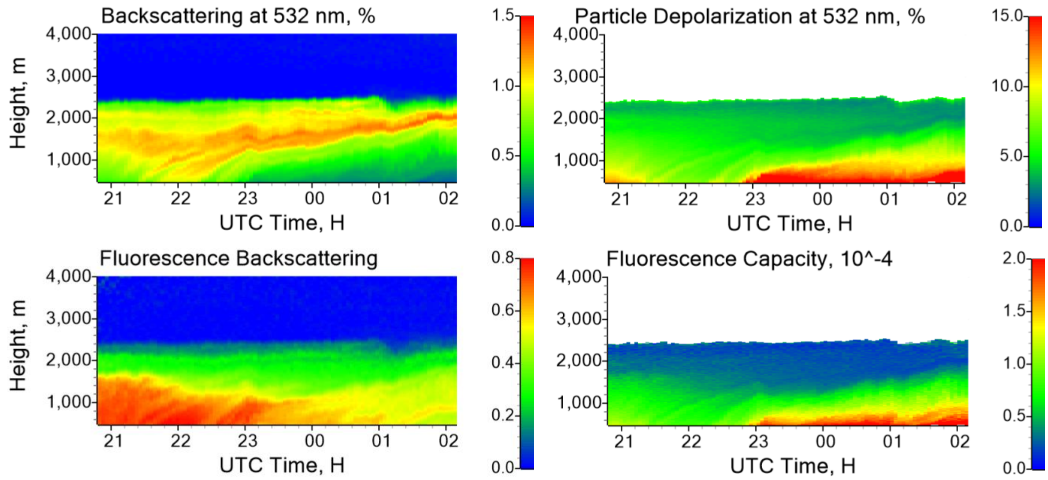

Fig.6. Spatio-temporal distributions of the backscattering coefficient β_{532}; the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$); the particle depolarization ratio δ_{532}; and the fluorescence capacity G_F in the night 30-31 May 2020.

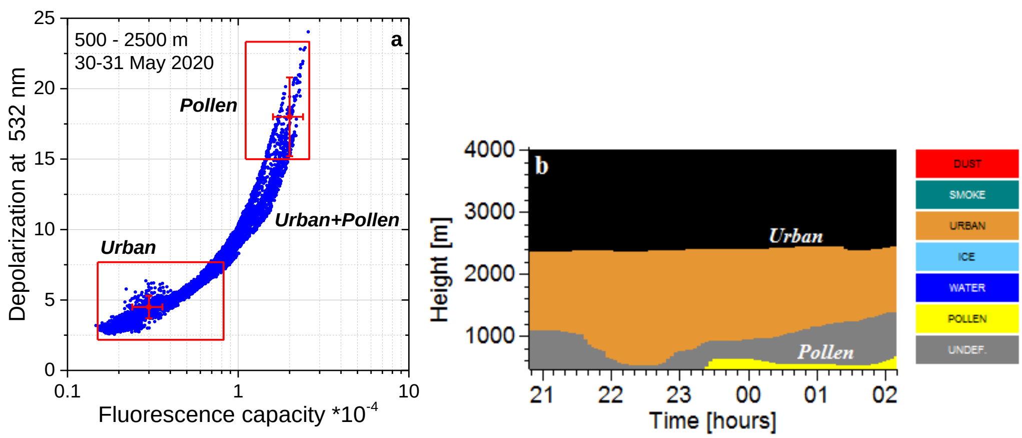

Fig.7. (a) The δ_{532}-G_F diagram for observations in 500 m – 2500 m height range and (b) spatio-temporal distribution of aerosol types on the night 30-31 May 2020.

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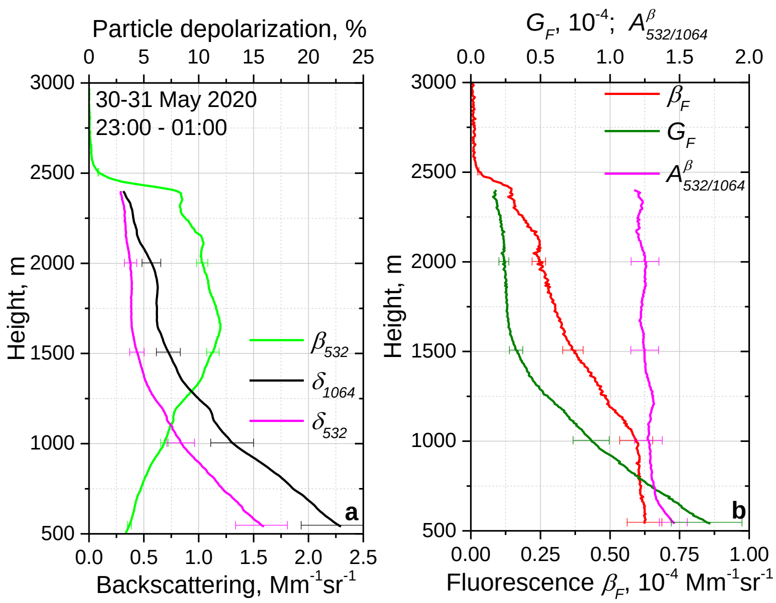

Fig.8. Vertical profiles of (a) backscattering coefficient β_{532} and particle depolarization ratios δ_{532}, δ_{1064}; (b) fluorescence backscattering $\beta_{F,}$ fluorescence capacity G_F and backscattering Angstrom exponent $A^{\beta}_{532/1064}$ on 30 May 2020 for period 21:00 – 23:00 UTC.

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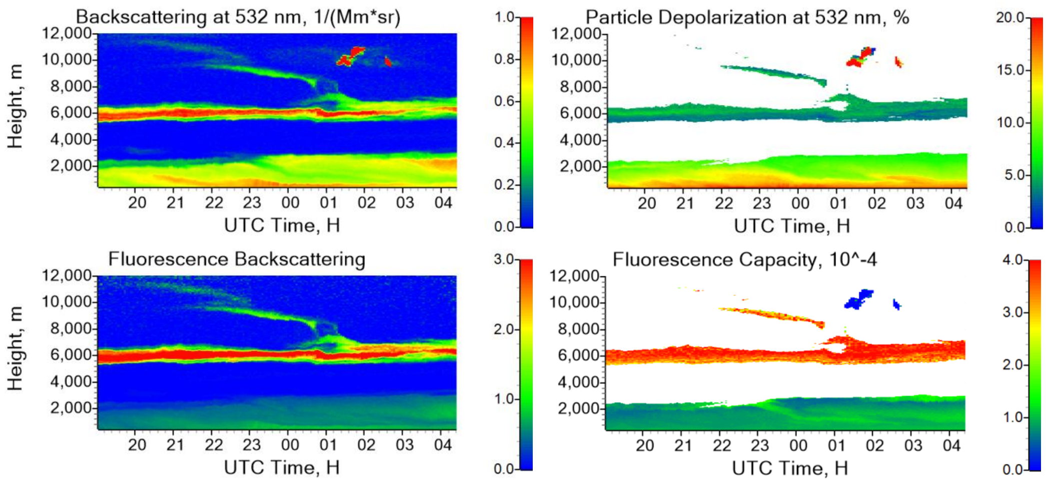

Fig.9. Spatio-temporal distributions of the backscattering coefficient β_{532}, the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$), the particle depolarization ratio δ_{532}, and the fluorescence capacity G_F in the night 14 – 15 September 2020.

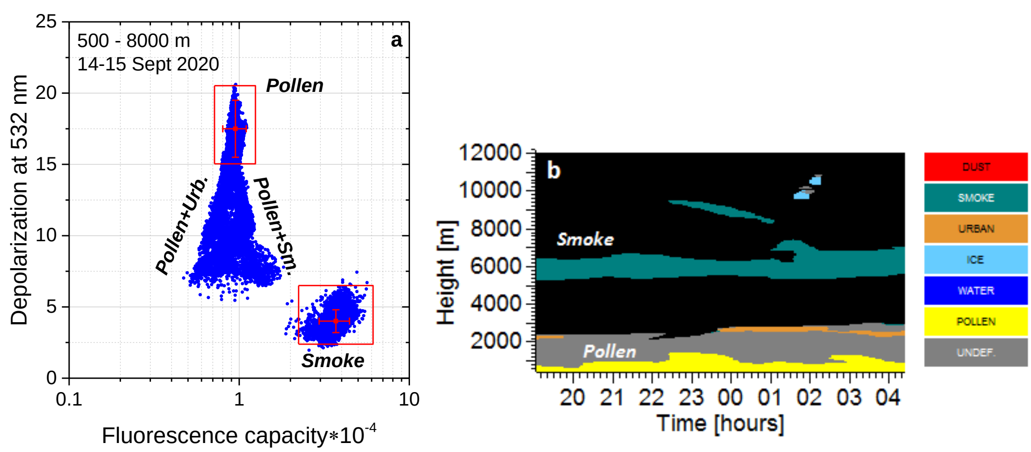

Fig.10. (a) The δ_{532}-G_F diagram for observations in 500 m – 8000 m height range and (b) spatio-temporal distribution of aerosol types in the night 14 – 15 September 2020.

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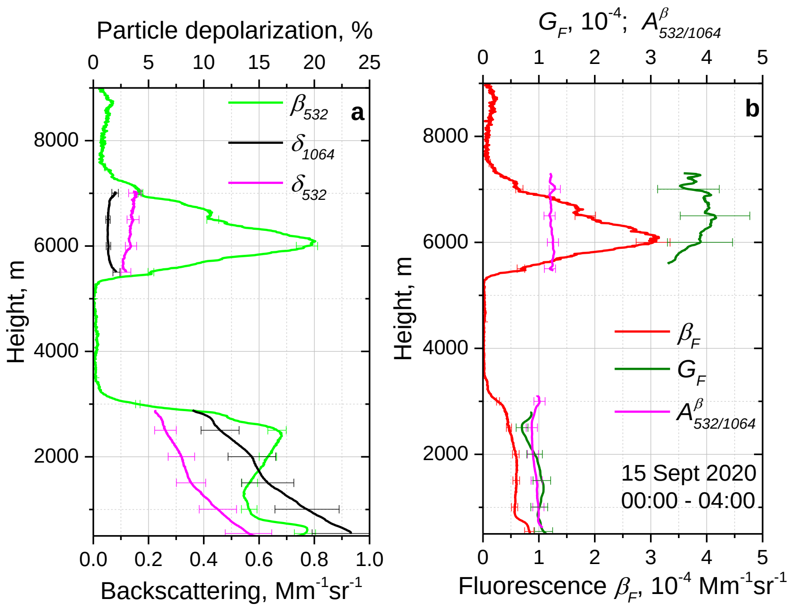

731
732 Fig.11. Vertical profiles of (a) backscattering coefficient β_{532} and particle depolarization ratios
733 δ_{532}, δ_{1064}; (b) fluorescence backscattering β_F, fluorescence capacity G_F and backscattering
734 Angstrom exponent $A^{\beta}_{532/1064}$ on 15 September 2020 for period 00:00 – 04:00 UTC.
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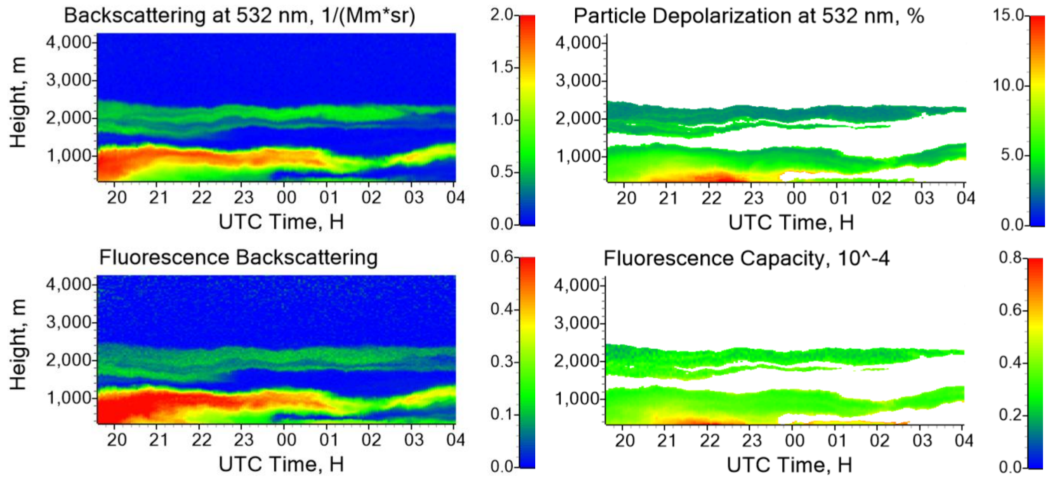

Fig.12. Spatio-temporal distributions of the backscattering coefficient β_{532}, the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$), the particle depolarization ratio δ_{532}; and the fluorescence capacity G_F in the night 10 – 11 April 2020. Measurements are performed at an angle of 45 dg to horizon.

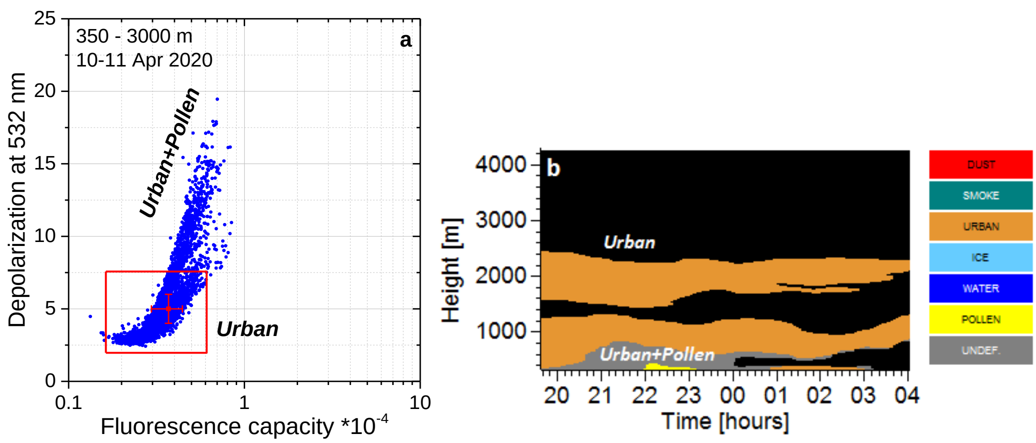

Fig.13. (a) The δ_{532}-G_F diagram for observations in 350 – 3000 m height range and (b) spatio-temporal distribution of aerosol types in the night 10 – 11 April 2020.

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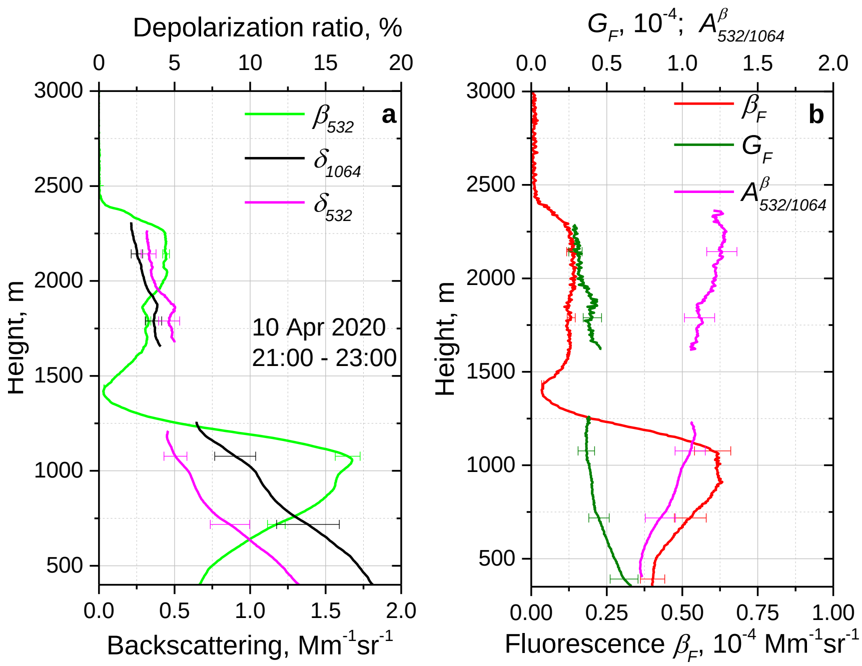

Fig.14. Vertical profiles of (a) backscattering coefficient β_{532} and particle depolarization ratios δ_{532}, δ_{1064}; (b) fluorescence backscattering β_F, fluorescence capacity G_F and backscattering Angstrom exponent $A^{\beta}_{532/1064}$ on 10 April 2020 for period 21:00 – 23:00 UTC.

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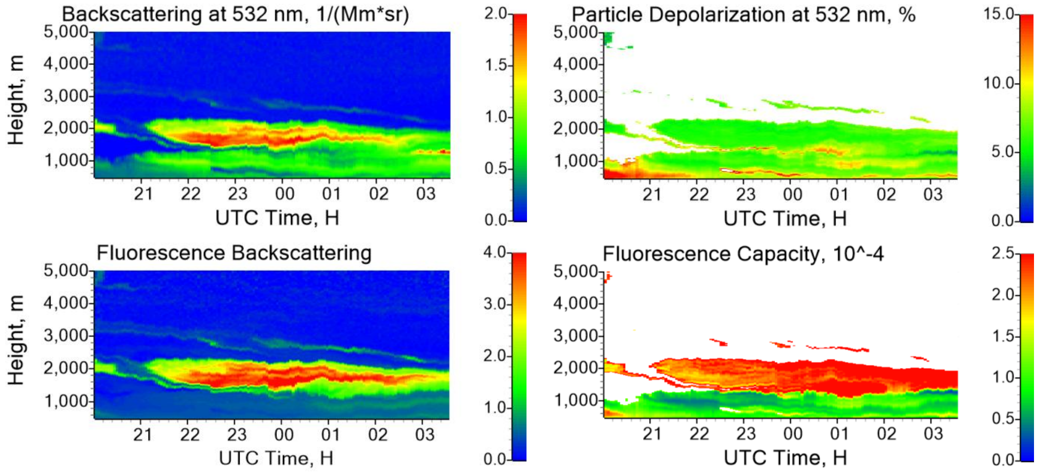

Fig.15. Spatio-temporal distributions of the backscattering coefficient β_{532}, the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$), the particle depolarization ratio δ_{532}, and the fluorescence capacity G_F in the night 11 – 12 August 2021.

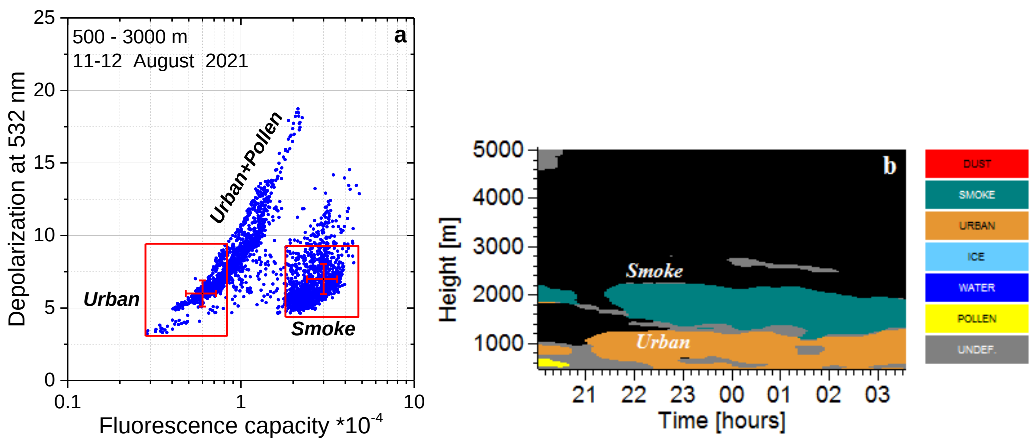

Fig.16. (a) The δ_{532}-G_F diagram for observations in 500 – 3000 m height range and (b) spatio-temporal distribution of aerosol types in the night 11-12 August 2021.

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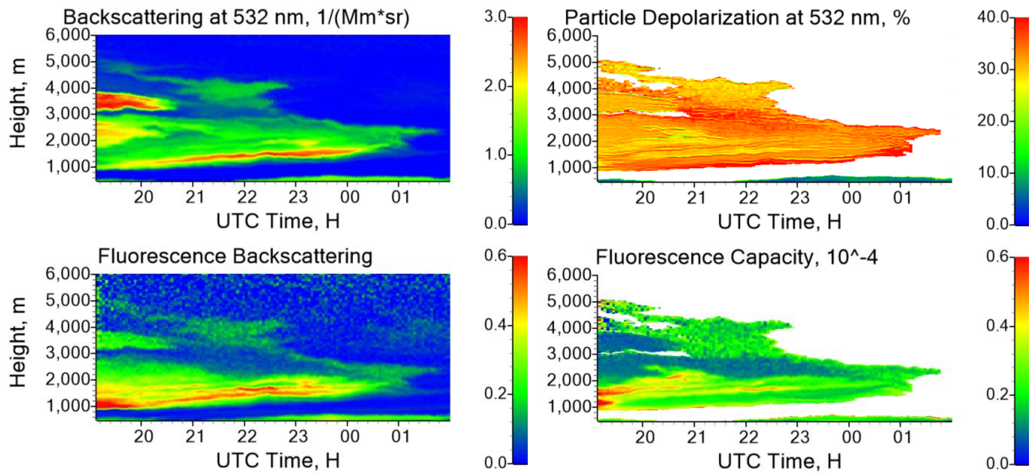

Fig.17. Height – temporal distributions of the backscattering coefficient at 532 nm β_{532}, the fluorescence backscattering coefficient β_F (in 10^{-4} $Mm^{-1}sr^{-1}$), the particle depolarization ratio at 532 nm δ_{532}, and the fluorescence capacity G_F in the night 1-2 April 2021. For the values β_{532}<0.2 $Mm^{-1}sr^{-1}$ the δ_{532} and G_F were not calculated.

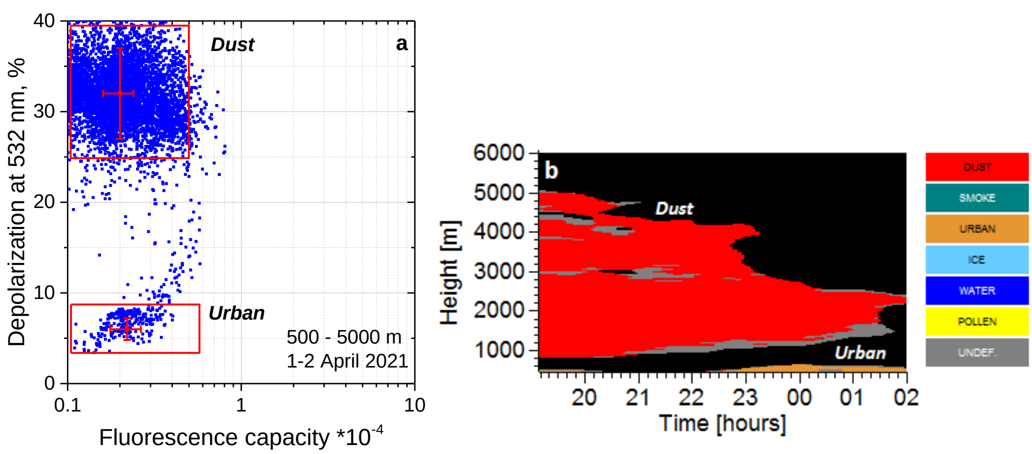

Fig.18. (a) The δ_{532}-G_F diagram for observations in 500 – 5000 m height range and (b) spatio-temporal distribution of aerosol types in the night 1-2 April 2021.