We would like to thank the two reviewers for devoting time to read our manuscript and provide valuable comments for improving it and increasing its scientific value. We have modified our manuscript following the guidelines given by the two reviewers.

Kind regards,

Giannakaki Elina, on behalf of all the co-authors

<u>Interactive comment of RC2 on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-271, 2019.</u>

We would like to thank the Anonymous Referee 2 for the constructive comments and recommendations. We rephrased the abstract and the methodology section in order to generalize the application of our methodology as suggested. More emphasis has been also given to lidar ratio selection as suggested by the two reviewers. Below we answer to 2nd reviewer's comment (RC). The **RC**s are given in **bold**, our replies in plain font and the corresponding *changes in the manuscript* are given in *italic*.

Abstract. To my understanding the focus of the paper is to present the Elastic Extinction Retrieval (EER) methodology, using data at Finokalia as a case study. Therefore the abstract should be rephrased accordingly.

Indeed, the focus of the paper is to present the Elastic Extinction Retrieval, providing the advantages and disadvantages, using a case study at Finokalia. So, we rephrased the abstract accordingly.

A new method, called ElEx, is proposed for the estimation of extinction coefficient lidar profiles using only the information provided by the elastic and polarization channels of a lidar system. The method is applicable both during day-time and night-time lidar measurements under well-defined aerosol mixtures. When the two aerosol type have different optical properties, permit the separation of the aerosol mixture. ElEx uses the particle backscatter profiles at 532 nm and the vertically resolved particle linear depolarization ratio measurements at the same wavelength. The particle linear depolarization ratio and the lidar ratio values of pure aerosol types are taken from literature. The total extinction profile is then estimated and compared well with Raman retrievals. In this study, ElEx was applied in an aerosol mixture of marine and dust particles at Finokalia station during CHARADMEx campaign. Any difference between ElEx and Raman extinction profiles indicates that the non-dust component could be probably attributed to polluted marine or polluted continental aerosols. Comparison with sun-photometric aerosol optical depth observations is performed as well during daytime. Differences in the total aerosol optical depth is varying between 1.2 and 72% and is attributed to the limited ability of the lidar to correctly represent the aerosol optical properties in the near range due to overlap problem.

Line 25. Please quantify what is reasonable difference.

We deleted the word reasonable, and instead we give information about the calculated difference.

Differences in the total aerosol optical depth is varying between 1.2 and 72% and is attributed to the limited ability of the lidar to correctly represent the aerosol optical properties in the near range due to overlap problem.

Line 33. Please replace "The types of aerosols" with "Pure aerosol types"
The phrase "The types of aerosols" has been replace by "Pure aerosol types".

Pure types of aerosols can be categorized roughly as mineral dust, sea salt, volcanic, carbonaceous, or sulfate aerosols originating from various natural and anthropogenic sources.

Line 35 Replace "is occurred" with "occur" and in the references use e.g.
The phrase "is occurred" has been replaced by "occur". We used e.g. in the references provided.

Several lidar studies have revealed that a broad variety of aerosol mixtures occur in the European continent (e.g. Balis et al., 2004, Papayannis et al. 2005)

Line 37. Add "the" before the European.
We added "the" before the European.

The mixing occurs because of the relatively long pathways of air masses across different aerosol source regions before the detection over the European continent.

Line 38. Please provide a reference for your claim that there is no mixing between maritime and desert dust.
The phrase is confusing. We deleted the phrase "For example, Saharan dust observed in South Europe is often already lifted over Africa to heights above 1-2 km, so that mixing with marine particles is almost prohibited".

Lines 52 to 64. The authors should mention here that many of the references correspond to the POLIPHON algorithm. In addition they should outline here that in this paper they aim to present a general concept algorithm approach rather than presenting cases studies as they did in their 2017 papers.
The POLIPHON is used for separation of dust and non dust backscatter contributions and the new, extended approach to separate even the fine and coarse dust backscatter fractions, while ElEx is a method to calculate the extinction profile with an Elastic lidar. However, both of them are using the separation technique and so we added a sentence about the POLIPHON algorithm. Also, more emphasis is given about the contribution of this study.

The technique is the base of POLIPHON algorithm for ground –based lidars (Ansmann et al., 2019) and has also been applied to CALIPSO aerosol profiles either on selected case studies (Giannakaki et al., 2011) or on a statistical basis (Marinou et al., 2017).
In this contribution, we propose a method to determine the extinction coefficient profile using only the elastic and polarization lidar channels at 532 nm in a well-defined aerosol mixture. The method has been first suggested by Giannakaki et al. (2017) and further applied by Ansmann et al. (2017). In this contribution, we fully outline the methodology providing an extended sensitivity analysis along with the main advantages and limitations of it.

Section 2. To my understanding the main focus of this section should be 2.4 which should be rephrased as "Description of the EER method. This section should be written in a general way and not dependent on the certain lidar system. I would suggest to expand this section, and move earlier as 2.1. In addition they authors should provide the equations they use so that the reader can reproduce their methodology. In

addition I would suggest that they should not limit the description of the methodology to mixtures of marine and dust but in general for mixtures of aerosols with distinct intensive parameters. They should also examine how sensitive is their algorithm to the a priori lidar ratio values considered. Of course then they can continue with the Finokalia case study as demonstration case study.

Thank you the reviewer for this comment. We rearranged the methodology section. It is now 2 subsections. In section 2.1 we present the general methodology that is not based either on the lidar system nor on the specific case study. The methodology is given 3 steps (2.1 Backscatter and Depolarization retrievals, 2.2 Separation of backscatters and 2.3 Estimation of Extinction). The method is not presented based on a mixture of desert dust and marine aerosol mixture but based on Type 1 and Type 2 in general. The flowchart of ElEx is given in Figure 1 and a discussion of the pure aerosol types that can be observed along with their intensive optical properties is given in subsection 2.2.3. In addition we provide the equations so that the reader can reproduce ElEx methodology. In section 2.2 we present our lidar system and data processing. Our case study is given in detail in subsection 3.2 and the sensitivity analysis of the lidar ratio is part of subsection 3.2.

2 Methodology

2.1 Elastic Extinction Retrieval: ElEx methodology

A new method, called ElEx, is proposed for the estimation of extinction coefficient lidar profiles using only the information provided by the elastic and polarization channels of a lidar system.

2.2.1 Backscatter coefficient and particle depolarization profile

At the first step, we retrieve the backscatter coefficient (β_t) at 532 nm. For the elastic backscatter signal at 532 nm the lidar equation for the backscatter coefficient at the emitted wavelengths is solved following the Klett-Fernald retrieval methods (Klett, 1981). For the calibration of the profile of the measured 532 nm elastic backscatter signal, pure Rayleigh signals are simulated based on actual temperature and pressure profiles from numerical weather forecast data or actual nearby radiosonde observations. The measured 532 nm signals are then fitted to the Rayleigh signal profile in the aerosol-free middle to upper troposphere. The solution is possible only under the assumption of a constant with height relation between aerosol extinction and backscatter coefficient (lidar ratio), The assumed lidar ratio can be selected either from climatological studies or, if available, from night time measurements. Additional information provided from dust models, backward trajectories, sunphotometers and/or satellite retrievals may be helpful for the right selection of the initial lidar ratio value. ElEx also uses the particle depolarization ratio at 532 nm. The particle depolarization ratio is computed from the volume *depolarization ratio by means of the determined particle backscatter coefficient (Freudenthaler et al., 2009).*

2.2.2 Separation of the backscatter profile

At the second step we need to decompose our profile to its aerosol components, assuming that only two aerosol types are observed. The procedure to separate the aerosol mixture into two components starts from the equation of the particle depolarization ratio:

$$\delta_p = \frac{\beta_1^{\perp}+\beta_2^{\perp}}{\beta_1^{\parallel}+\beta_2^{\perp}} \quad (1)$$

$\beta^{\perp}$ and $\beta^{\parallel}$ are so-called cross- and parallel-polarized particle backscatter coefficients that can in principle be computed from the lidar return signals detected with the cross-polarized and parallel-polarized signal channels.

As shown by Tesche et al. (2009), the separation of the two aerosol components is possible through the simple equation 2:

$$\beta_1 = \beta_t \frac{(\delta_t-\delta_2)(1+\delta_1)}{(\delta_1-\delta_2)(1+\delta_t)} \quad (2)$$

In order to perform the decomposition of the backscatter profile the two aerosol types should be distinguishable in terms of the particle depolarization ratio. This mean that the decomposition is possible using the information of the strongly depolarizing dust and a second aerosol type with less depolarizing ability, like marine, pollution or even biomass burning aerosols.

From the profile of the total particle backscatter coefficient β_t and the backscatter coefficient of the 1st aerosol type β_1 we calculate the remaining backscatter coefficient of the 2nd aerosol type.

$$\beta_2 = \beta_t - \beta_1 \quad (3)$$

2.2.3 Estimation of Elastic Extinction profile

The separation of the backscatter profile give us the opportunity to calculate the total extinction coefficient by applying the correct lidar ratio to its aerosol component.

$$a_t = a_1 + a_2 = \beta_1 \cdot lr_1 + \beta_2 \cdot lr_2 \quad (3)$$

This is only possible in case of the mixture of two aerosol types that are well known in terms of lidar ratio. ElEx is presented in the flowchart of Figure 1. Type 1 and 2 could be any types that can be distinguishable *is terms of depolarization and lidar ratio values. Since ElEx methodology is strongly dependent on the selection of pure lidar ratio and particle depolarization values we briefly present the aerosol types that are mostly observed in Europe along with their intensive optical properties.*

Continental polluted aerosols are typically small and do not significantly depolarize the backscattered light (δ_{aer}^{532}= 0.04 ± 0.04; (Heese, et al., 2016), and due to the high carbon content, these particles reveal high lidar ratios (Giannakaki, et al., 2010). On the contrary, clean continental type differentiates from the polluted continental type due to its less light absorbing properties. The clean continental type shows low depolarizing ability with values lower than 0.07 (Omar, et al., 2009) and low lidar ratio values, i.e., 20–40 sr (Ansmann, et al., 2001; Giannakaki, et al., 2010).

In the absence of significant transport of continental aerosols, particles over the remote oceans are largely of marine origin (Prospero, et. al., 1989).The sea-salt particles feature a predominant coarse mode, however, they are spherical in humid conditions and weakly absorbing, in contrast to the dust particles. Therefore, they yield low particle lidar ratio values, are almost non depolarizing and exhibit low particle depolarizing ratio values (Burton, et al., 2013; Dawson, et al., 2015). This aerosol type is mainly identifiable by the low particle lidar ratio, i.e., 15–25 sr at 532nm (Burton, et al., 2012).

Desert areas around the world emit huge quantities of dust aerosols which also actually extend considerably over adjacent regions, such as oceans (Jaenicke, et al., 1978) and can be transported over very long distances (Prospero, et al., 1989; Papayannis, et al., 2008; Mona, et al., 2012). The optical properties of dust particles are considerably different from the other types, thus making them easy to identify, especially in the absence of aerosol mixtures.

Biomass burning is a major global source of atmospheric aerosols. Generally, smoke particles are relatively small, spherical, and highly absorbing that produce low depolarization and large lidar ratios (Amiridis, et al., 2009; Baars, et al., 2012; Nicolae, et al., 2013Giannakaki, et al., 2016). The optical properties of smoke particles may vary due to the vegetation type of the emitting source, the combustion type (smouldering or flaming fires), and atmospheric conditions (Balis, et al., 2003). Furthermore, the particles are susceptible to changes of their optical properties during their lifetime in the atmosphere (Nicolae, et al., 2013). The similarities of the physical characteristics of smoke particles and continental particles result in similar optical properties, making these types difficult to distinguish.

Volcanoes are another important source of atmospheric aerosols. Volcanic eruptions eject great amounts of material in the atmosphere (tephra), while the fraction smaller than 2mm is labeled as volcanic ash. Most of these aerosols will settle only a few tens of kilometres away from the volcano but smaller particles can travel thousands of kilometres and affect wider areas (Mattis, et al., 2010; Sicard, et al., 2012; Papayannis, et al., 2012; Kokkalis, et al., 2013; Pappalardo, et al., 2013). The optical properties of volcanic ash aerosols is generally similar to the one of desert dust, as was shown by Ansmann et al. (2010) and Wiegner et al. (2012) for fresh ash with particle linear depolarization ratios reaching 0.37 and lidar ratios of 50–65 sr. Aged volcanic particles as observed by Papayannis et al. (2012) indicate higher sphericity less non-sphericity with depolarization ratio values of about 0.1–0.25 and lidar ratios for 355nm within the range 55–67 sr and for 532nm 76–89 sr.

ElEx is not limited to nighttime Raman observations, and thus is also applicable to daytime lidar measurements as long as the observed aerosol types have different particle depolarization ratios to permit for an accurate separation between them. The more the particle depolarization ratio differ the better the separation we achieve. The accuracy of the extinction coefficient is also depended on the knowledge of the correct lidar ratio. For example the intensive properties of marine and Saharan dust particles are already well defined, while smoke particles are characterized by large variability especially on lidar ratio values. In case of Raman extinction profiles availability the purity of the non-dust component can be additionally checked. In stations with more complicated aerosol mixtures within the planetary boundary layer the methodology is possible to be applied to free tropospheric aerosol layers. This is particularly useful in case of free tropospheric layers of dust or volcanic particles with smoke or pollution.

2.2 Description of the lidar system and lidar data processing

The main instrument of this paper is the Polly^{XT} lidar as it is described by Althausen et al. (2009) and Engelmann et al. (2016). An overview of PollyNET can be found in Baars et al. (2016). Polly^{XT} works with a Nd:YAG laser, emitting pulses at 1064, 532 and 355 nm, with a repetition frequency of 20 Hz. The receiver consists of a Newtonian telescope, with a diameter of 300 mm and a field of view of 1 mrad. Photomultiplier tubes are used for the detection of the elastically backscattered photons at 355, 532 and 1064 nm, as well as for the inelastically backscattered photons at 387 and 607 nm, which correspond to the Raman shift by nitrogen molecules at 355 and 532 nm, respectively. Additionally, the cross-polarized component of radiation at 355 and 532 nm is detected, allowing for the determination of the particle *linear depolarization ratio (also called depolarization ratio). In this study, the polarization channels permit us to identify non-spherical dust particles from the near spherical marine aerosols. The Polly^{XT} Arielle system used in this study has been provided by the TROPOS Institute.*

The vertical resolution of the signal profiles is 30 m and the raw data are typically stored as 30 s average values. Data were collected on the web page of PollyNET (http://polly.tropos.de) where the "quicklooks" of all measurements are available.

Lidar data was collected during CHARADMExp campaign (Characterization of Aerosol mixtures of Dust And Marine origin) that took place from the 20th of June 2014 until the 20th of July 2014. CHARADMExp was an experimental campaign of ESA, implemented by the National Observatory of Athens (NOA), aimed to characterize dust and marine particles along with their mixtures (http://charadmexp.gr). The site for the campaign is the monitoring ACTRIS station of Finokalia in Greece as presented in Fig. 1. Finokalia station is located at a remote coastal site in the northeast of the island Crete, Greece, in the Eastern Mediterranean (35.338°N, 25.670°E, 252 asl). The station is located at a top hill, facing the sea within a sector from 270° to 90°. No touristic or other human activities can be found at a distance shorter than 15 km within the aforementioned sector. The site is affected by marine and dust particles by 95% of the time (Mihalopoulos et al., 1997; Kouvarakis et al., 2000).

For the purposes of this study backscatter coefficient profiles are calculated via Fernald–Klett method (Fernald, 1984; Klett, 1981). The method requires independent information on the lidar ratio and the reference value of the particle backscatter coefficient. Afterwards, the calibrated depolarization ratio profiles at 532 nm, are calculated (Freudenthaler et al. 2009). An overlap correction was applied on the basis of a simple technique proposed by Wandinger and Ansmann (2002). Thus, the aerosol profiles are retrieved down to 500 m. Extinction coefficient profiles at 532 nm are also retrieved based on the Raman method (Ansmann et al., 1992) and are only used for validation purposes of the proposed methodology at a later stage in this study.

2.3 Models

Four-day backward trajectories are calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) to gather information about the origin of the observed aerosols and the synoptic patterns corresponding to the measurements. The HYSPLIT 4 model is a complete system for computing simple trajectories to complex dispersion and deposition simulations using either puff or *particle approaches. A discussion of the model is given by Draxler and Hess (1997) and Draxler and Hess (1998). The simulations were performed using the GDAS meteorogical data. Backward trajectories were computed for several altitudes for the CHARADMExp campaign period, confirming that the origin of the air masses arriving over our site, were from Saharan region. The BSC-DREAM8b model is additionally used to verify the presence of Saharan dust, indicated from the trajectory analysis. The BSC-DREAM8b model described the desert dust emissions and transport (Nickovic et al., 2001; Pérez et al., 2006; Basart et al., 2012). Moreover, sea salt emissions and transport are described with the atmospheric model RAMS/ICLAMS (Solomos et al., 2011). The model is a further developed version of RAMS 6.0 (Pielke et al., 1992) that allows for a fully prognostic treatment of the sea salt particles and their life cycle in the atmosphere. The simulations were used to verify the presence of marine aerosols in the atmosphere during the campaign and specifically during the day under study.*