Optical and microphysical characterization of aerosol layers over South Africa by means of multi-wavelength depolarization and Raman lidar measurements

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19   **Abstract**

20   Optical and microphysical properties of different aerosol types over South Africa

21   measured with a multi-wavelength polarization Raman lidar are presented. This study

22   could assist in bridging existing gaps relating to aerosol properties over South Africa,

23   since limited long-term data of this type is available for this region. The observations

24   were performed under the framework of the EUCAARI campaign in Elandsfontein.

25   The multi-wavelength Polly^{XT} Raman lidar system was used to determine vertical

26   profiles of the aerosol optical properties, i.e. extinction and backscatter coefficients,

27   Ångström exponents, lidar ratio and depolarization ratio. The mean microphysical

28   aerosol properties, i.e. effective radius and single scattering albedo were retrieved

29   with an advanced inversion algorithm. Clear differences were observed for the

30   intensive optical properties of atmospheric layers of biomass burning and

31   urban/industrial aerosols. Our results reveal a wide range of optical and microphysical

32   parameters for biomass burning aerosols. This indicates probable mixing of biomass

33   burning aerosols with desert dust particles, as well as the possible continuous

34   influence of urban/industrial aerosol load in the region. The lidar ratio at 355 nm, the

35   lidar ratio at 532 nm, the linear particle depolarization ratio at 355 nm and the

36   extinction-related Ångström exponent from 355 to 532 nm were 52 ± 7 sr; 41 ± 13;

37   0.9 ± 0.4 % and 2.3 ± 0.5, respectively for urban / industrial aerosols, while these

38   values were 92 ± 10 sr; 75 ± 14; 3.2 ± 1.3 % and 1.7 ± 0.3 respectively for biomass

39   burning aerosols layers. Biomass burning particles are larger and slightly less

40   absorbing compared to urban / industrial aerosols. The particle effective radius were

41   found to be 0.10 ± 0.03 μm, 0.17 ± 0.04 μm and 0.13 ± 0.03 μm for urban/industrial,

42   biomass burning, and mixed aerosols, respectively, while the single scattering albedo

43   at 532 nm were 0.87 ± 0.06, 0.90 ± 0.06, and 0.88 ± 0.07 (at 532 nm), respectively for

44   these three types of aerosols. Our results were within the same range of previously

45   reported values.

46

1. Introduction

Atmospheric aerosols of natural and anthropogenic origin contribute substantially to global climate variability (IPCC, 2013). Currently, the magnitude of the (anthropogenic) aerosol impact on climate causes the largest uncertainty on our knowledge of climate change (Forster et al., 2007). Large uncertainties exist due to the diversity, not only with respect to aerosol particle size, composition, sources and lifetime variation, but also with regard to the spatial and temporal distributions of aerosols. Thus, the impacts of aerosols on climate must be understood and quantified on a regional scale rather than on a global-average basis (Piketh et al., 2002).

High-quality aerosol measurements in the southern hemisphere are rather limited. South Africa is located at the southernmost tip of the African continent, extending from 22° S to 34° S latitude and from 16° E to 32° E longitude. Previous studies have indicated that South Africa is one of the countries in the world that is largely affected by aerosol load, due to various natural and anthropogenic sources (Piketh et al., 2000; Piketh et al., 2002; Formenti et al., 2002, 2003; Campbell et al., 2003; Eck et al., 2003; Freiman and Piketh, 2003; Ichoku et al., 2003; Ross et al., 2003; Winkler et al., 2008; Queface et al., 2011; Tesfaye et al., 2011; Venter et al., 2012; Tiitta et al., 2014). Intensive efforts have been undertaken during recent years to characterize aerosol pollution in South Africa. In general, previous studies pointed at the importance of regional circulation of air masses and seasonal pollutant variation. The optical properties of aerosols have been studied by means of sun photometers (e.g. Queface et al., 2011; Eck et al., 2003), *in situ* data (e.g. Laakso et al., 2012) and satellite observations (e.g. Tesfaye et al., 2011) in these studies, which are based on columnar aerosol optical properties. Ground-based Raman lidars provide vertically resolved information on the distribution and optical properties of aerosols. Giannakaki

72 et al. (2015) used Raman lidar data obtained over a one year period at Elandsfontein

73 in South Africa (26°15′ S, 29°26′ E, 1745 m above sea level (a.s.l.)) to study the

74 geometrical characteristics, intensive and extensive optical properties of free-

75 tropospheric aerosol layers. In addition to these characteristics that can be determined

76 with lidar data, multi-wavelength Raman lidar measurements can also be used to

77 determine profiles of microphysical particle properties by using inversion algorithms

78 (Twomey, 1977; Veselovskii et al., 2002, Müller et al., 2001). In this study we expand

79 our study of aerosols in South Africa by providing information on the microphysical

80 and optical properties of aerosol layers. This type of aerosol lidar observations are

81 valuable for spaceborne lidars such as CALIPSO (Cloud-Aerosol Lidar and Infrared

82 Pathfinder Satellite Observations) (e.g. Omar et al., 2009), since lidar ratio values for

83 different aerosol types are required for reliable aerosol extinction retrievals.

84 Therefore, this study could be useful for further improving lidar ratio selection-

85 scheme algorithms used in spaceborne lidar missions.

86 Four long-term ground-based aerosol measurements were carried out at sites in

87 economically growing countries in Asia, Africa and South America within the

88 EUCAARI project (Kulmala et al., 2011), which included Elandsfontein in South

89 Africa. The aim of EUCAARI was to characterize particles in terms of physical,

90 optical and chemical aerosol properties. Here we report lidar observations that were

91 performed at Elandsfontein. In particular, we discuss the optical and microphysical

92 properties of aerosol layers that are caused by biomass burning and urban/industrial

93 activities at the site. We present aerosol lidar ratios, particle linear depolarization

94 ratios and Ångström exponents for biomass burning and urban/industrial aerosol

95 layers measured with a multi-wavelength Raman lidar. The possible effect of desert

96 dust particles on biomass burning aerosol layers in terms of the intensive optical and

97 microphysical properties is also addressed. In addition, effective radius and single-

98 scattering albedo are calculated with an advanced inversion algorithm.

99 The paper is organized as follows: In section 2, the research site, the methodology

100 used for the retrieval of optical and microphysical properties and the aerosol typing

101 are introduced. As a case study, the arrival of a biomass burning aerosol layer over

102 Elandsfontein is discussed in section 3. Section 4 presents the main findings of the

103 optical and microphysical aerosol properties for selected biomass burning,

104 urban/industrial and mixed aerosol layers. We close our contribution with a summary

105 and conclusion in section 5.

106 **2. Location and Methodology**
107 **2.1. Measurement site**
108 The measurement site was located on a hill top at Elandsfontein ($26^{o}15´$ S, $29^{o}26´$ E,

109 1745 m a.s.l.) in the Highveld region of South Africa. The station was located

110 approximately 150 km east of the Johannesburg-Pretoria megacity, which is the

111 largest metropolitan area in South Africa with a population of more than 10 million

112 people (Lourens et al., 2012).

113 In South Africa, anthropogenic atmospheric emissions are predominantly the product

114 of industrial activities and biomass burning (Ross et al., 2003). South Africa is the

115 most industrialized country of the sub-continent – primarily due to the industrialized

116 Highveld region (Freiman, 2003; Wenig et al., 2003). This region has clusters of

117 industrial complexes and power plants between 25.5^{o} S, 27.5^{o} E and 27.0^{o} S, 30.5^{o} E

118 (Ross et al., 2003), which contributes significantly to aerosol and trace gases pollution

119 (Freiman et al., 2003). Tropospheric NO2 distributions derived with SCIAMACHY

120 (SCanning Imaging Absorption spectroMeter for AtmosphericCHartographY) from

121 August 2002 to March 2012 (Schneider et al., 2015) are presented in Figure 1. The

122 tropospheric NO2 column density of the Highveld region in South Africa is

123 comparable to that observed over central and northern Europe, eastern North America

124 and Southeast Asia (Lourens et al, 2012).

125 In addition, emissions from biomass burning (wild fires) contribute significantly to

126 regional emission loads (e.g. Giannakaki et al., 2015). Both, natural phenomena

127 (lightning) and human induced activities are responsible for biomass burning

128 (Edwards et al., 2006). The number of hotspots, with confidence levels between 80-

129 100%, (http://earthdata.nasa.gov/data/nrt-data/firms/active-fire-data), in the latitude

130 range between -40° and 40° and longitude range between -20° and 60° are plotted in

131 Figure 2. The number of hotspots is averaged in terms of 3 months for the year 2010.

132 Wild fires originate in the sub-equatorial central African region and progress

133 southward (Roy et al., 2005). In southern Africa, the fires progress along a north-west

134 to south-east track.

135 **2.2. Description of the lidar system and lidar data processing**

136 The transportable aerosol Raman lidar Polly^{XT} that was operated remotely at

137 Elandsfontain is described by Althausen et al. (2009) and Engelmann et al. (2016).

138 Polly^{XT} works with a Nd:YAG laser emitting at its primary wavelength of 1064 nm,

139 which after frequency doubling and tripling emits at the wavelengths of 532 and 355

140 nm, respectively. The receiver consists of a Newtonian telescope with a diameter of

141 300 mm and a field of view of 1 mrad. Photomultiplier tubes (PMT) are used for the

142 detection of the elastically backscattered photons at 355, 532 and 1064 nm, as well as

143 the in-elastically backscattered photons at 387 and 607 nm that correspond to the

144 Raman-shift by nitrogen molecules at 355 and 532 nm, respectively. Additionally, the

145 cross-polarized component at 355 nm is detected and consequently allows for the

146 determination of the linear particle depolarization ratio (also called depolarization

147 ratio). To retrieve the particle depolarization ratio the Rayleigh calibration method

148 was applied within the data analysis under the assumption of pure Rayleigh

149   depolarization in an aerosol-free height range (Behrendt and Nakamura, 2002). The

150   vertical resolution of the signal profiles is 30 m and the raw data are typically stored

151   as 30 s average values (20 Hz laser frequency). Data were collected on the web page

152   of PollyNet (http://polly.tropos.de) where the "quicklooks" of all measurements are

153   available.

154   Extinction and backscatter coefficient profiles at 355 and 532 nm, respectively, were

155   obtained with the Raman method (Ansmann et al., 1992). To vertically retrieve the

156   backscatter coefficient at 1064 nm we use Fernald-Klett method (Fernald, 1984; Klett,

157   1981). With this method the particle backscatter coefficient is derived applying a

158   backward iteration starting at a chosen reference height. The method requires

159   independent information on the lidar ratio and on the reference value of the particle

160   backscatter coefficient. The cases analyzed here are night-time measurements and the

161   retrieved backscatter at 1064 nm was also evaluated by the Raman method (Ansmann

162   et al., 1992) using also the signal from the Nitrogen Raman channel at the 607 nm. An

163   overlap correction was applied on the basis of a simple technique proposed by

164   Wandinger and Ansmann (2002). The depolarization ratio, i.e. the ratio of the cross-

165   polarized to the parallel-polarized component of the backscatter coefficient (particles

166   and molecules) at 355 nm was also calculated. The contribution of the molecules can

167   easily be calculated, which then provides the linear particle depolarization ratio (Cairo

168   et al., 1999; Murayama et al., 1999).

169   The uncertainties affecting the retrieval of extinction and backscatter coefficients, and

170   thus the calculation of lidar ratio and Ångström exponents are mainly due: to the

171   statistical error due to signal detection, the systematic error associated with the

172   estimation of the atmospheric molecular number density from the pressure and

173   temperature profiles, the systematic error associated with the evaluation of the aerosol

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174 scattering wavelength dependence, the systematic error for overlap function, the

175 errors introduced by operation procedure such as signal binning (smoothing) and

176 averaging accumulating lidar returns. The overall relative errors of the lidar-derived

177 aerosol properties range between 5%-15% for the backscatter coefficients, 10%-30%

178 for the extinction coefficients, 20%-40% for the Ångström exponents, 15%-40% for

179 the lidar ratios and approximately 5%-10% for the linear particle depolarization ratio

180 (Hänel et al., 2012; Baars et al., 2016, Engelmann et al., 2016). A detailed discussion

181 on the influence of aerosol optical depth errors to Ångström exponent errors can be

182 found in Wagner et al., 2008.

183 The layer identification was based on the assumption that the optical properties should

184 be relatively stable. This means that within a chosen height layer, the variability of the

185 optical data should be less than the statistical uncertainty of the individual data points.

186 In Table 1 we provide information regarding the elevated layers that were selected for

187 the optical and microphysical aerosol characterization. The characterization of aerosol

188 types will be discussed in section 2.4.

189
190 **2.3. Retrieval of microphysical properties**
191 Microphysical particle properties are derived with an inversion algorithm that has

192 been developed at the Leibniz Institute for Tropospheric Research. A detailed

193 description of the inversion code is given by Müller et al (1999a, 1999b). A minimum

194 of three backscatter coefficients (355, 532, and 1064 nm) and two extinction

195 coefficients (355 and 532 nm), with measurement errors less than 30%, are required

196 as input in order to obtain microphysical results that have reasonably low

197 uncertainties (Müller et al., 2001). The selection of the individual inversion solutions

198 is based on the concept that the back-calculated optical data should agree with the

199 original data within the limits of the measurement errors, and that a pre-selected

200   discrepancy level, which is an output parameter of the inversion algorithm (Müller et

201   al., 1999a), is not exceeded. The mean particle size in terms of the effective radius is

202   then calculated along with the standard deviation from these selected individual

203   solutions. One also obtains a range of complex refractive indexes by applying this

204   method. The complex refractive index is a wavelength-independent quantity.

205   Therefore, inversion can only provide a wavelength-independent value that represents

206   the entire range of wavelengths from 355 – 1064 nm. The single-scattering albedo can

207   then be calculated from the volume concentration distribution, which is another data

208   product of the inversion algorithm, and the associated mean complex refractive index

209   by means of a Mie scattering algorithm.

210   Uncertainties associated with the retrievals are in general <30% for effective radius.

211   The real part of the complex refractive index is derived to an accuracy better than

212   ±0.1, while the imaginary part is obtained for its correct order of magnitude if the

213   value is <0.01i (for larger values of the imaginary part the uncertainty is <50%). The

214   single-scattering albedo can be calculated with an accuracy of ±0.05, if uncertainties

215   of the input optical data are on average <10-15%. A detailed error analysis is

216   presented by Müller et al. (1999b, 2001) and Veselovskii et al., (2002, 2004).

217 **2.4. Aerosol classification**
218 The identification of the source of aerosol particles is possible with the synergetic use

219   of in-situ and satellite measurements, as well as utilizing model estimations.

220   The HYSPLIT_4 (Hybrid Single Particle Langrangian Integrated Trajectory) model

221   (Draxler and Hess, 1997) was used to compute backward air mass trajectories

222   employing the kinematic approach and by using the re-analysed National Oceanic and

223   Atmospheric Administration (NOAA) dataset with a resolution of 2.5^o x 2.5^o (latitude,

224   longitude) as input. Four-day backward trajectories were selected, because they

225   extend far enough back in time and distance to cover the main source regions

226 suspected to affect the region investigated. The trajectories were calculated for the

227 center of the layer observed and for the time of the lidar measurement.

228 The number of fire hotspots is given by Moderate Resolution Imaging

229 Spectroradiometer (MODIS) collection-5 active-fire product data (Giglio, L. et al.,

230 2010). The number of hotspots, obtained from MODIS for four days prior to each of

231 the measurements, was superimposed on the trajectory analysis map in order to detect

232 the presence of smoke particles over our site for the cases analyzed.

233 Trace gases were measured as part of routine air quality monitoring at the site by the

234 national electricity supplier, i.e. Eskom. A Thermo Electron 43C SO_2 analyser and a

235 Thermo Electron 42i NO_x analyser were used to measure SO_2 and NO_x respectively.

236 H_2S was measured with a Thermo Electron 43A SO_2 analyzer with a Thermo Electron

237 340 converter. 15-minute data were averaged for the extent of measurement time for

238 each of the measurements periods (Table 1). For instances where the combined use of

239 trajectory analysis and fire hotspots did not indicate the presence of biomass burning

240 aerosols we checked whether the measured NO_x, SO_2 or H_2S concentrations were

241 higher than the seasonal mean values of that measured for the entire period of the

242 EUCAARI campaign. These seasonal mean values are presented in Laakso et al.

243 (2012). In addition, when the trace gases concentrations were lower than the mean

244 seasonal values measured during the EUCAARI campaign and biomass-burning

245 activity or desert dust advection were absent, we checked if the daily concentration of

246 the trace gases exceeded the mean critical values.

247 There were also cases that indicated desert dust aerosol particles in addition to the

248 smoke, which originated either from the Kalahari or the Namibia desert that could

249 have additionally contributed to the aerosol loads. Therefore, the measured aerosol

250 optical properties determined for these cases were attributed to a mixing state where

251 smoke particles were possible to be mixed with desert dust aerosols. Additional

252 mixing with urban / industrial aerosols is also possible.

253 An example of a measurement of biomass burning aerosols is discussed in the

254 subsequent section in order to demonstrate the methodology used to derive the optical

255 and microphysical aerosol properties.

256

257 **3. Biomass burning aerosols on 1ˢᵗ October 2010 at Elandsfontein,**

258 **South Africa**

259 In this section we will study a geometrically deep layer that extends up to 2.1 km

260 height above ground level (AGL) as observed on the 1ˢᵗ of October 2010. The

261 atmospheric structure, in terms of range corrected signals, is quite stable which

262 indicates similar optical properties throughout the layer

263 (http://polly.tropos.de/?p=bilder&lambda=1064&Jahr=2010&Monat=10&Tag=1&Ort

264 =11#bildanker). High backscatter returns are observed on the day when the

265 measurement is conducted in relation to the previous and the next day (as can be

266 already seen in Figure 4 (a) – light green).

267 MODIS fire hotspots product reveal that several fires were active during the period

268 28ᵗʰ of September 2010 – 1ˢᵗ of October 2010 as shown in Figure 3 (a). In Figure 3

269 (b), four-day backward trajectories arriving at Elandsfontein on 1ˢᵗ of October 2010 at

270 00:00 are presented. The trajectories are computed for arrival heights of the bottom,

271 center and top of the observed layer. The trajectory analysis along with MODIS fire

272 hotspots reveals that the air masses are highly possible to carry smoke particles at

273 Elandsfontein on the day of the measurement.

274 In Figure 4 the optical lidar profiles are presented. The backscatter and extinction

275 maximum at all three wavelengths were observed within the 0.9 to 1.9 km height

276 range. High values of the lidar ratio of 96 ± 5 sr at 355 nm and 89 ± 5 sr at 532 nm

277   indicate that the smoke particles inside this layer were most likely highly light-

278   absorbing . The Ångström exponent, related to extinction between 355 and 532 nm,

279   was 1.8 ± 0.1, which points to comparably small particles and indicative of fresh

280   smoke (eg. Müller et al., 2005). A constant particle depolarization ratio in the order

281   of 4% is observed at 355 nm throughout the layer. The lack of significant vertical

282   variability of the lidar ratio, the Ångström exponent and the particle depolarization

283   ratio suggests the presence of the same type (biomass burning) of aerosols throughout

284   the layer.

285   The mean values of extinction (at 355 and 532 nm) and backscatter coefficients (at

286   355, 532 and 1064 nm) were calculated within the defined layer and were used as

287   input in the inversion algorithm. Effective radius, complex refractive index and

288   single-scattering albedo were calculated with the microphysical inversion code. An

289   effective radius of 0.15 ± 0.02 μm was determined, while the single-scattering albedo

290   was approximately 0.86 at 532 nm that indicates relatively strong-absorbing aerosols.

291   **4. Results and Discussion**

292   We performed optical lidar data analysis, microphysical retrievals and aerosol typing

293   for each of the thirty eight aerosol layers listed in Table 1 in the same way as

294   presented in the example in section 3. Each aerosol layer in Table 1 was classified

295   into one of the three aerosol types, i.e. urban/industrial, biomass burning, and mixed

296   aerosols after thorough visual inspection of the backward trajectories, MODIS

297   hotspots fires products and in-situ aerosol observations, as explained in section 2.4.

298   Table 2 summarizes the mean intensive optical properties (lidar ratio at 355 and 532

299   nm, depolarization ratio at 355 nm and Ångström exponent related to extinction

300   between 355 and 532 nm) presented together with the associated standard deviations,

301   ranges (minimum and maximum values) and medians.

302 Figure 5 presents the particle lidar ratios at 355 nm versus the extinction-related

303 Ångström exponent for urban/industrial (black), biomass burning (red) aerosol layers

304 as well as for the mixed aerosol layers (green). Different aerosol types occupy

305 different areas in the Ångström-exponent–lidar-ratio plot. Aerosols from urban and

306 industrial activities are on average characterized by larger Ångström exponents than

307 (pure or mixed) biomass burning aerosols. The lidar ratios of biomass burning

308 aerosols are among the highest compared to literature with a mean value of 92 ± 10 sr

309 (e.g. Müller et al., 2007; Nicolae et al, 2013; Amiridis et al., 2009). Urban / Industrial

310 aerosol layers were found to have lower lidar ratio values in the range between 41 and

311 59 sr at 355 nm. Our results indicate that biomass burning aerosols have lower lidar

312 ratios when they are mixed either with desert dust aerosols or with urban / industrial

313 aerosols. This might be due to the non-spherical shape of desert dust that may have a

314 significant effect on the lidar ratio. Model calculations show that a deviation from the

315 spherical shape can efficiently increase particle backscattering and thus lower the lidar

316 ratio (Mishchenko et al., 1997), which was also confirmed by Müller et al. (2003).

317 Ångström exponent values of these aerosols ranged from 1.6 to 2.5, with a mean value

318 of 2.0 ± 0.4, which is larger (smaller particles) than the mean value of 1.7 ± 0.3 we

319 observed for 'pure' biomass burning aerosols. The role that hot air close to the surface

320 of the earth plays in generating these dust size distribution is not well understood

321 (Nisantzi et al., 2014). Wind stress close to the surface may be very complex and the

322 sudden release of all the moisture in the hot soil particles may strongly influence the

323 cracking of larger particles into smaller ones and thus lead to a much more

324 complicated size distribution than observed during desert dust outbreaks (Mamouri et

325 al., 2014).

326   It is evident from Figure 5 that Ångström exponent values for the different aerosol

327   types overlap. Therefore, another intensive aerosol property, the linear particle

328   depolarization ratio, which is an indicator of non-spherical particles, was also used.

329   Figure 6 shows the lidar ratio at 355 nm versus the depolarization ratio at the same

330   wavelength for the three aerosol types. Different clusters of data pairs can be

331   identified. Lower depolarization ratio values were found for urban/industrial aerosol

332   layers. These aerosol layers are also characterized by lower lidar ratios and thus the

333   data points representing urban / industrial pollution occupy the lower left region in

334   Figure 5. Significantly larger particle linear depolarization ratios with a mean of 8.3 ±

335   0.7 % were found for mixed aerosols. Typical desert dust aerosol depolarization ratios

336   determined in field measurements performed in the northwestern corner of the Sahara

337   ranged from 30 to 35% at 532 nm with a mean value of 31 ± 3% (Freudenthaler et al.,

338   2009). In addition, particle depolarization ratios ranging between 30 to 35% were also

339   observed for Asian desert dust (Sugimoto et al., 2003, Shimizu et al., 2004, Shin et

340   al., 2015) and desert dust originating from Middle East dust sources (Mamouri et al.,

341   2013). Depolarization ratios of the mixtures of biomass burning aerosols and desert

342   dust particles determined for African biomass burning and dust mixtures ranged

343   between 8 − 26% at 532 nm (Weinzierl et al., 2011, Tesche et al., 2009). Therefore

344   depolarization values reported in this study are at the lower end of these values. This

345   observed difference can be attributed to the different contribution of desert dust

346   particles to the biomass burning plume. However, we should also note that the

347   geometrical shape of the dust particles over the Kalahari desert could be different

348   from the shape of Saharan dust. Also, the possible influence of the backsground urban

349   / industrial aerosols in the mixture should be kept in mind.

350 A wide range of (lower) depolarization ratios and lidar ratios was found for biomass

351 burning aerosols. This observed variability can be attributed to differences in the

352 chemical composition of the particles that depends on the source region, relative

353 humidity in the atmosphere, the type of fire, or the combined effect of these factors. In

354 addition, the mixing of the biomass burning aerosols with maritime or even

355 urban/industrial background aerosols cannot be excluded as a possible reason for the

356 variability of lidar ratio and depolarization ratio values.

357 Several statistics of lidar ratios and Ångström exponents for different aerosol types in

358 the world are available for comparison. Figure 7 provides some of the general

359 literature with regard to the lidar ratios values at 355 nm and Ångström exponents of

360 urban/industrial and biomass-burning aerosols, as well as for mixtures of biomass

361 burning and desert dust aerosols. To interpret the x-axis of the Figure 7 one should

362 also look the Table 4. It is evident from Figure 7 that intensive aerosol properties are

363 in good agreement with values found from other studies.

364 The lidar ratio at 355 nm, in particular, shows similar values for urban / industrial

365 aerosols in various regions of the world. Ångström exponent values found for

366 urban/industrial particles in this study are at the upper limit of results previously

367 published for this aerosol type, which indicates slightly smaller particles at

368 Elandsfontein that can most probably be ascribed by differences in the emission

369 sources. The depolarization ratio is at the lower limit indicating spherically shaped

370 anthropogenic particles.

371 The lidar ratio for biomass burning aerosol layers is within the range of previously

372 reported values, although the values tend to be more at the upper limit of the reported

373 values. The Ångström exponents are in very good agreement with previous studies.

374 Müller et al. (2007) studied the growth of free-tropospheric forest fire smoke particles

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375   and indicated that the Ångström exponent decreases with the duration of transport.

376   The Ångström exponent values found in this study (1.7 ± 0.3) corresponds to travel

377   times of the biomass burning aerosols between 1 and 3 days, which is confirmed by

378   back-ward trajectory analysis. The characteristics of biomass burning emissions in the

379   subtropical South African region vary according to the type of fuel burned (vegetation

380   type), meteorology and combustion phase (Ross et al., 2003). For example, flaming

381   grass fires produce smoke with more soot compared to smoke emitted from

382   smoldering wood and bush fires (Posfai et al., 2003). Thus differences in the chemical

383   composition of the particles might be one of the reasons for the observed large lidar

384   ratio.

385   For the mixed aerosols the lidar ratio values reported here are in very good agreement

386   with previous studies for the mixture of desert dust and biomass burning aerosols. The

387   contribution of desert dust particles within the observed biomass burning plumes is

388   probably lower, thus resulting in a lower depolarization ratio and larger Ångström

389   exponent than what has been reported in literature for biomass burning mixed with

390   dust as mentioned previously. Groβ et al. (2011) reported neutral wavelength-

391   dependence of the particle depolarization ratios for mixed dust and smoke layers for

392   which Ångström exponents varied between 0.12 and 0.16, while Tesche et al. (2011)

393   reported wavelength-independent linear particle depolarization ratios of 0.12-0.18 at

394   355, 532 and 710 nm for mixed dust and smoke layers. In that sense our results on

395   particle depolarization ratios at 355 nm are similar to results from these studies

396   reporting linear particle depolarization ratio at 532 nm.

397   In Figure 9 the effective radius against the Ångström exponent is plotted. In general

398   the plot shows the same features already noted for Figure 5. On average the largest

399   aerosols are determined for biomass burning aerosols (red) with an effective radius of

400 0.17 ± 0.04 µm. Particles from anthropogenic pollution (black) are smaller with a

401 mean effective radius of 0.1 ± 0.03 µm. Our results indicate that the influence of

402 Kalahari desert dust on biomass burning plumes leads to smaller particles compared

403 to pure biomass burning aerosols with a mean effective radius of 0.13 ± 0.03 µm.

404 Mean microphysical properties i.e. effective radius, single scattering albedo and

405 complex refractive index are listed with their associated standard deviations, ranges

406 (minimum and maximum values) and medians in Table 3. The particles in the

407 biomass-burning aerosol layers show a mean effective radius of 0.17 ± 0.04 µm,

408 which is within the range of values reported in previous studies for biomass burning

409 aerosols. Reid et al. (1998) reported count median diameter values ranging from 0.12

410 µm for fresh particles to 0.21 µm for aged particles near rain-forest fires in Brazil.

411 Radke et al. (1988) obtain values of approximately 0.22 µm for particles from forest

412 fires in North America. Wandinger et al. (2002) found larger biomass burning

413 aerosols with an effective radius of approximately 0.25 µm. Effective radii in the

414 range between 0.19 and 0.44 µm were found for biomass burning aerosol layers

415 resulting from long-range transport across Romania (Nicolae et al., 2013). Müller et

416 al, (2007) presented values ranging between 0.13 and 0.15 nm for plumes ageing

417 between one to three days.

418 The three types of aerosols cover a wide range of single-scattering albedo values as

419 shown in Table 3. The mean single-scattering albedo for biomass burning aerosol is

420 0.90 ± 0.06 (at 532 nm). Lower single scattering albedos are reported in literature for

421 fresh biomass burning particles in Europe. Nicolae et al. (2013) reported a value of

422 0.78 ± 0.02, while Reid et al. (1998) found that single scattering albedo ranges

423 between 0.74 and 0.77 for fresh smoke. Previous studies show that aged biomass

424 burning layers are characterized by larger single scattering albedos. For example,

425   Murayama et al. (2004) found a value of 0.95 ± 0.06 at 532 nm, while Noh et al.

426   (2009) reported single scattering albedos of 0.92 at the same wavelength. Therefore

427   our results indicate moderately absorbing particles resulting from fresh or medium

428   aged (less than 3 days) biomass burning aerosols.

429   For the mixed aerosols we determined lower mean scattering-albedos of 0.88 ± 0.07,

430   which is slightly higher than the mean single-scattering albedo of 0.87 ± 0.06

431   determined for urban/industrial aerosol layers. Laakso et al. (2012) reported values of

432   0.84 ± 0.08 (637 nm) at ground level at Elandsfontein, South Africa. Quaface et al.

433   (2011) determined significantly larger values of 0.91 and 0.89 at 440 nm and 670 nm,

434   respectively, from AERONET data collected at Skukuza in South Africa. Our results

435   indicate that elevated anthropogenic aerosol layers from urban and industrial activities

436   are characterized by stronger light-absorption.

437   Complex refractive indexes are also reported in Table 3. Real parts of the complex

438   refractive index of these particles are mostly > 1.5, while imaginary parts vary from

439   0.007i to 0.04i. Lower real parts of the refractive index are found for biomass burning

440   aerosols compared to the urban/industrial particulates with values ranging from 1.35

441   to 1.57. The imaginary parts of the refractive index of biomass burning aerosol layers

442   are < 0.03i (with the exception of one case that shows an imaginary refractive index

443   of 0.046i). A large variation of refractive indices for the real and imaginary parts is

444   observed for mixed aerosols. This might allude to the different levels of contribution

445   of Kalahari desert dust to biomass burning aerosol layers.

446

447   **5. Summary and Conclusions**

448   Thirty eight aerosol layers of urban/industrial, biomass burning, and mixed aerosols

449   were studied with regard to their optical and microphysical properties at

450   Elandsfontein, South Africa. The combination of Raman lidar observations with

451   backward trajectory analysis, satellite fire observations and *in situ* data allowed for

452   source identification of the elevated aerosol layers. Measurements of the lidar ratios

453   and depolarization ratios are presented in order to assist in the separation of

454   anthropogenic, biomass burning, and mixtures of aerosols.

455   A wide range of optical (lidar ratio and depolarization ratio) and microphysical (single

456   scattering albedo, complex refractive index) properties was determined for biomass

457   burning aerosols, indicating differences in chemical composition. Aerosols from

458   urban and industrial activities are on average characterized by larger Ångström

459   exponents than (pure or mixed) biomass burning aerosols. Lidar ratios for biomass

460   burning aerosols are among the highest found in literature with a mean value of $92 \pm$

461   10 sr, while the anthropogenic aerosols are characterized by lower lidar ratios in the

462   range between 41 and 59 sr at 355 nm. Ångström exponents were found to be similar

463   for all types of aerosol types under study, with slightly larger values determined for

464   anthropogenic aerosols. Mean effective radii of 0.17 ± 0.04 μm and 0.1 ± 0.03 μm

465   were calculated for biomass burning and urban / industrial aerosols, respectively. We

466   have also shown that in certain instances biomass burning aerosols may contain a

467   small amount of desert dust particles resulting in higher depolarization ratios and

468   lower lidar ratios than the values reported for pure biomass burning aerosols.

469   Moderately absorbing particles were found for biomass burning layers with a mean

470   single scattering albedo of 0.9 ± 0.06. Mixed aerosols were found more absorbing

471   with a mean single-scattering albedo of 0.88 ± 0.07. A slightly lower mean single-

472   scattering albedo of 0.87 ± 0.06 was found for urban / industrial aerosol layers.

473   However, this value was larger than the values reported for the same site from

474 ground-based in-situ measurements. Our optical and microphysical results for the

475 analyzed aerosol types agreed very well with similar studies reported in literature.

476 Ground-based lidar networks provide information on the vertical and horizontal

477 distribution of optical aerosol properties in a systematic and statistically significant

478 manner. Different lidar networks that are globally distributed observe aerosols in

479 Europe, South America, Asia and North America. The analysis of lidar measurements

480 presented here could assist in bridging existing gaps with regard to our knowledge of

481 the vertical distribution of optical and microphysical aerosols in the South African

482 atmosphere, since limited long-term data of this nature is available for this region. Our

483 results could also be useful for lidar ratio selection schemes needed for elastic-

484 backscatter lidars. In that sense our findings could be used in advancing lidar

485 algorithms used for present and/or future satellite lidar missions.

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487
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TABLES
Table 1. Aerosol type, time and altitude range of aerosol layers used for optical and microphysical aerosol characterization

aerosol source	date	time [UTC]	height [m]	Extinction Coefficient [Mm-1]	
				355 nm	532 nm
urban / industrial	25 March 2010	18:00 – 19:50	2100 – 2670	196 ± 18	75 ± 12
	25 March 2010	18:00 – 19:50	2790 – 3450	190 ± 36	68 ± 14
	25 March 2010	18:00 – 19:50	1560 – 1980	260 ± 6	78 ±12
	16 April 2010	21:20 – 23:54	1980 – 2250	147 ± 13	58 ± 9
	16 April 2010	21:20 – 23:54	2280 – 2520	129 ± 10	39 ± 4
	16 April 2010	21:20 – 23:54	2610 – 3180	196 ± 43	81 ± 14
	14 May 2010	18:00 – 00:00	930 – 1360	238 ± 37	127 ± 25
	15 May 2010	18:30 – 20:20	1380 – 1860	196 ± 26	86 ± 19
	15 May 2010	18:30 – 20:20	2250 – 2700	81 ± 7	28 ± 3
	30 November 2010	17:15 – 18:00	960 – 1300	121 ± 6	44 ±13
	30 November 2010	17:15 – 18:00	1350 – 1920	146 ± 26	50 ± 11
	30 June 2010	17:00 – 18:00	1420 – 1620	101 ± 5	34 ± 5
	30 June 2010	17:00 – 18:00	1650 – 1830	71 ± 11	37 ± 7
	10 January 2011	19:15 – 20:15	1890 – 2160	303 ± 45	146 ± 31
	13 January 2011	21:00 – 22:00	1200 – 1800	342 ± 24	163 ± 17
	13 January 2011	21:00 – 22:00	1920 – 2250	267 ± 42	158 ± 29
	13 January 2011	21:00 – 22:00	2430 – 2880	199 ± 23	68 ±12
biomass burning	1 October 2010	00:10 – 01:00	1090 – 1900	331 ± 9	158 ± 8
	5 October 2010	18:10 – 23:10	1115 – 1750	432 ± 62	227 ± 37
	5 October 2010	18:10 – 23:10	1980 – 2700	256 ± 18	132 ± 15
	6 October 2010	20:00 – 00:00	1175 – 1540	277 ± 27	142 ± 5
	6 October 2010	20:00 – 00:00	1565 – 2160	214 ± 14	111 ± 11
	6 October 2010	20:00 – 00:00	2190 – 2520	152 ± 6	85 ± 16
	6 October 2010	20:00 – 00:00	2610 – 2820	121 ± 19	80 ± 6
	21 October 2010	01:30 – 02:30	880 – 1530	261 ± 28	131 ± 20
	21 October 2010	01:30 – 02:30	1685 – 2280	168 ± 7	66 ± 16
	21 October 2010	01:30 – 02:30	2400 – 2880	171 ± 30	70 ± 14
	22 August 2010	00:00 – 01.00	1205 – 1565	340 ± 13	162 ± 8
	22 August 2010	00:00 – 01.00	1685 – 1920	354 ± 5	190 ± 8
	22 August 2010	02:00 – 03:00	1115 – 1535	335 ± 6	163 ± 10
	22 August 2010	02:00 – 03:00	1745 – 2250	331 ± 15	170 ± 4
mixed aerosols	16 August 2010	17:00 – 18:00	1115 – 1445	316 ± 24	151 ± 9
	16 August 2010	19:00 – 20:00	995 – 1265	296 ± 7	157 ± 11
	18 August 2010	19:00 – 20:00	1175 – 1355	154 ± 9	75 ± 4
	18 August 2010	19:00 – 20:00	1415 – 1715	174 ± 11	66 ± 4
	18 August 2010	19:00 – 20:00	1865 – 2160	184 ± 6	66 ± 3
	22 August 2010	17:00 – 18:00	1145 – 1505	286 ± 3	109 ±4
	22 August 2010	17:00 – 18:00	1595 – 2040	267 ± 16	119 ± 8

858 **Table 2.** Mean value ± standard deviation of aerosol lidar ratio at 355, particle depolarization ratio and
859 Ångström exponent related to extinction between 355 and 532 nm for the examined aerosol types, as
860 well as value of range and median
861

aerosol source	mean ± stdv	range	median
lidar ratio at 355 nm [sr]			
urban / industrial	52 ± 7	41 – 59	54
biomass burning	92 ± 10	81 – 119	88
mixed aerosols	74 ± 11	59 – 90	73
lidar ratio at 532 nm [sr]			
urban / industrial	41 ± 13	23 – 74	38
biomass burning	75 ± 14	47 – 92	79
mixed aerosols	46 ± 13	33 – 68	40
particle depolarization ratio at 355 nm [%]			
urban / industrial	0.9 ± 0.4	0.3 – 1.7	1.0
biomass burning	3.2 ± 1.3	1.2 – 5.7	2.7
mixed aerosols	8.3 ± 0.7	7.3 – 9.1	8.1
ångström exponent related to extinction between 355 and 532 nm			
urban / industrial	2.3 ± 0.5	1.3 – 3.0	2.4
biomass burning	1.7 ± 0.3	1.0 – 2.4	1.7
mixed aerosols	2.0 ± 0.4	1.6 – 2.5	2.0

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863 **Table 3.** Mean value ± standard deviation of effective radius and single-sccattering albedo for the
864 examined aerosol types, as well as range and median.

| Aerosol Source	mean ± stdv	range	median
effective radius [µm]			
urban / industrial	0.10 ± 0.03	0.07 – 0.16	0.09
biomass burning	0.17 ± 0.04	0.11 – 0.28	0.17
mixed aerosols	0.13 ± 0.03	0.09 – 0.19	0.13
single-scattering albedo at 532 nm			
urban / industrial	0.87 ± 0.06	0.75 – 0.96	0.88
biomass burning	0.90 ± 0.06	0.77 – 0.98	0.90
mixed aerosols	0.88 ± 0.07	0.76 – 0.95	0.89
complex refractive index			
urban / industrial	1.61 (± 0.11) + 0.021 (± 0.010)i	1.47 – 1.78 (RRI) 0.007 – 0.039 (IRI)	1.64 (RRI) 0.020 (IRI)
biomass burning	1.43 (± 0.07) + 0.016 (± 0.011)i	1.35 – 1.57 (RRI) 0.002 – 0.046 (RRI)	1.40 (RRI) 0.015 (IRI)
mixed aerosols	1.52 (± 0.15) + 0.022 (± 0.015)i	1.33 – 1.74 (RRI) 0.004 – 0.046 (IRI)	1.56 (RRI) 0.019 (IRI)

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867   **Table 4.** The code used in Figure 7 and the respective reference.

868

Code	Reference
A03	Anderson et al., 2003
A05	Ansmann et al., 2005
A09a	Ansmann et al., 2009
A09b	Amiridis et al., 2009
A11	Arboledas et al., 2011
B03	Balis et al., 2003
B12a	Baars et al., 2012
B12b	Burton et al., 2012
B13	Burton et al., 2013
G10	Giannakaki et al., 2010
G11	Groβ et al., 2011
G13	Groβ et al., 2013
G16	This study
H15	Hesse et al., 2015
I15	Illingworth et al., 2015
K14	Kanitz et al., 2014
K12	Kompulla et al., 2012
M05	Müller et al., 2005
M07	Müller et al., 2007
M04	Murayama et al., 2004
M13	Murayama et al., 2013
N13	Nicolae et al., 2013
P12	Preiβler et al., 2012
P13	Preiβler et al., 2013
R98	Reid et al., 1998
T11	Tesche et al., 2011
W02	Wandinger et al., 2002
W11	Weinzierl et al., 2011
X08	Xie et al., 2008

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875   **FIGURES**
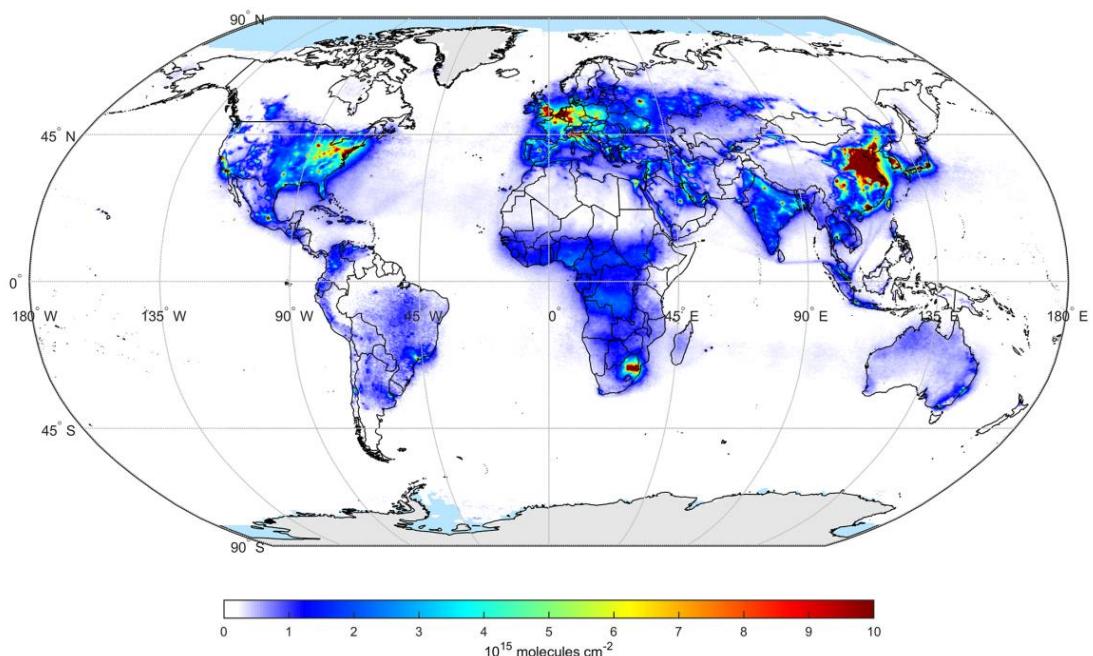
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879   **Figure 1.** Global map of long-term average tropospheric NO$_2$ column derived from
880   SCIAMACHY data from August 2002 to March 2012 (Schneider   et al., 2015)
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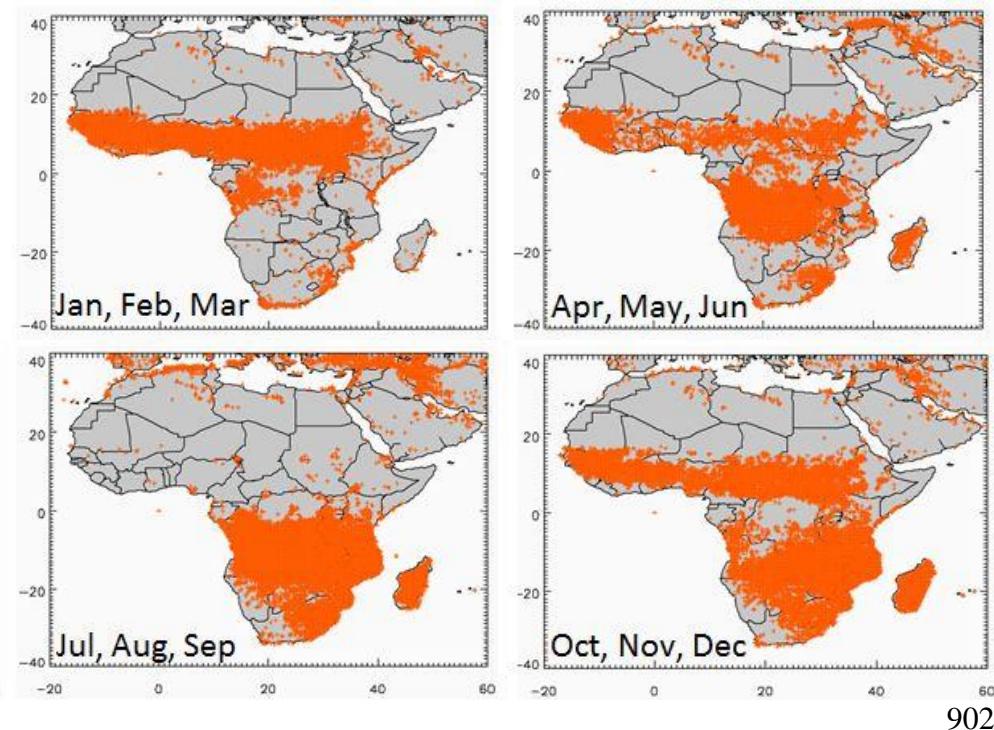
903 **Figure 2.** Number of fire hotspots with confidence levels between 80-100% averaged
904 in terms of 3 months for the year 2010 in the latitude range between -40° and 40° and
905 longitude range between -20° and 60°
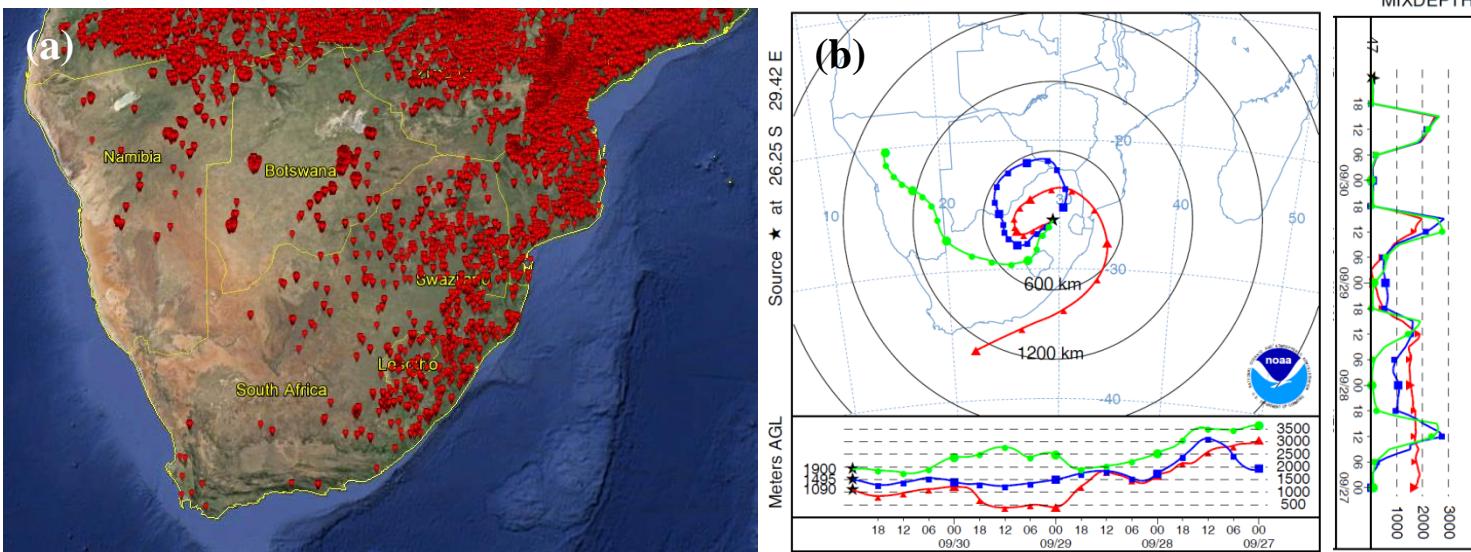
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922 **Figure 3.** MODIS fire hotspots for the period 28 September 2010 – 01 October 2010 and for the latitude range between -35° and -15° W and the
923 longitude range between 10° and 40° S (a). Four-day backward trajectories arriving at Elandsfontein on 1 October 2010 at 00:00 for arrival
924 height of the bottom (1090 m), center (1495 m) and top (1900 m) of the aerosol layer observed (b).
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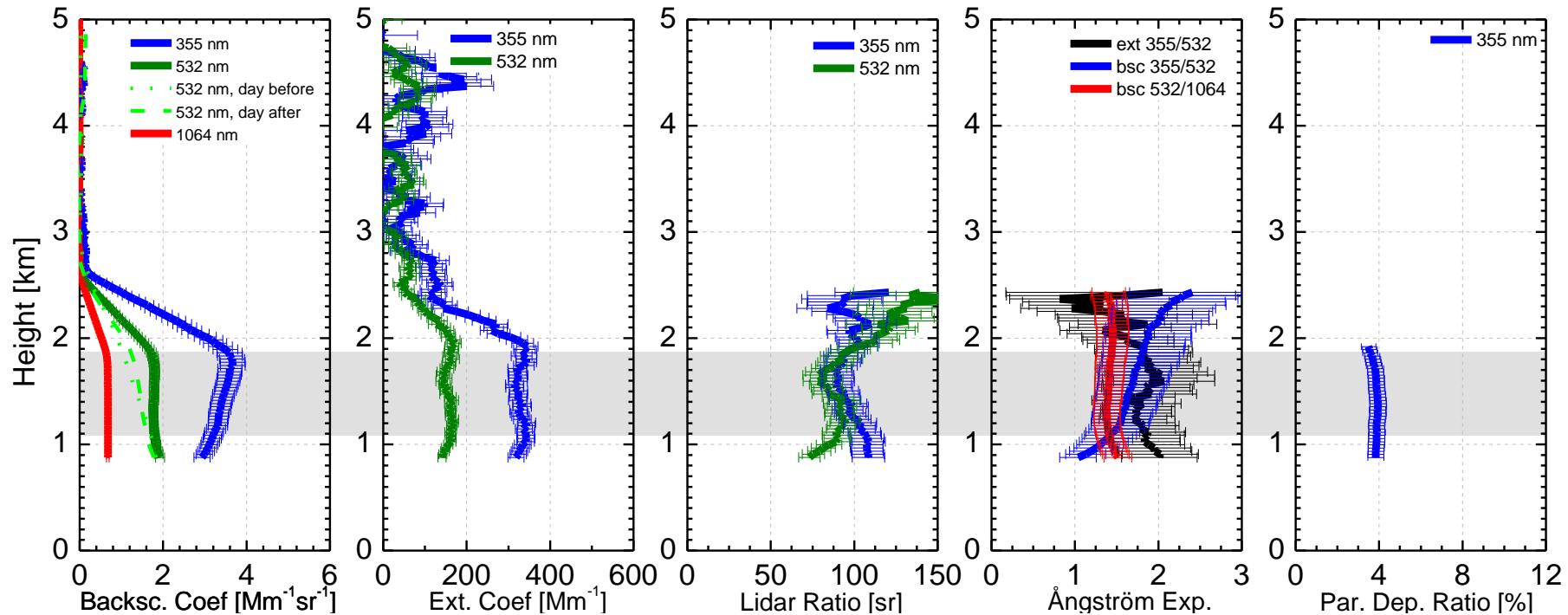
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930 **Figure 4.** Backscatter coefficients, extinction coefficients, lidar ratios, Ångstrom exponents and particle depolarization ratio at Elandsfontein on
931 1 October 2010, 00:10 – 03:59 UTC
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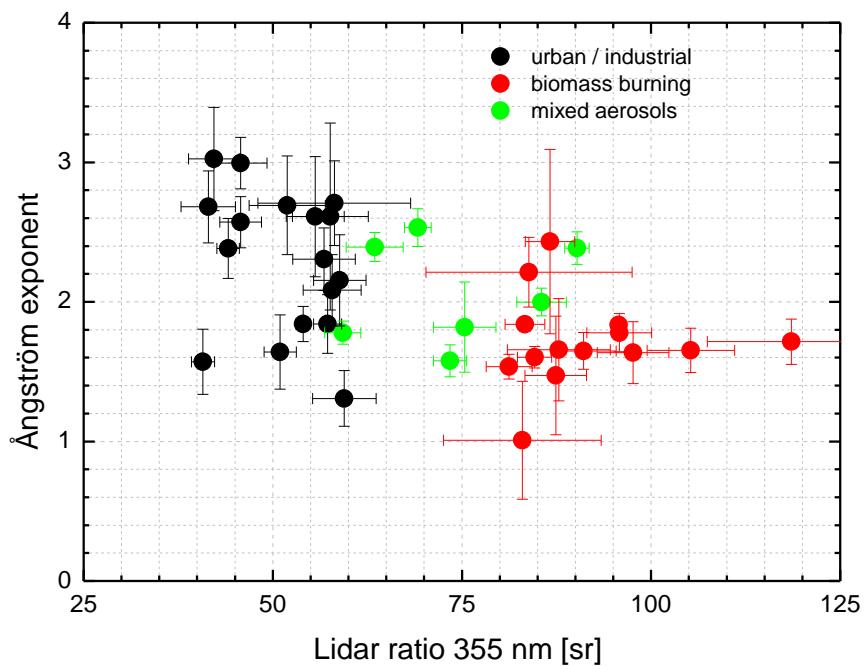
936
937 **Figure 5.** Lidar ratio at 355 nm versus the extinction-related Ångström exponent from
938 355 to 532 nm for the three aerosol types investigated in our study
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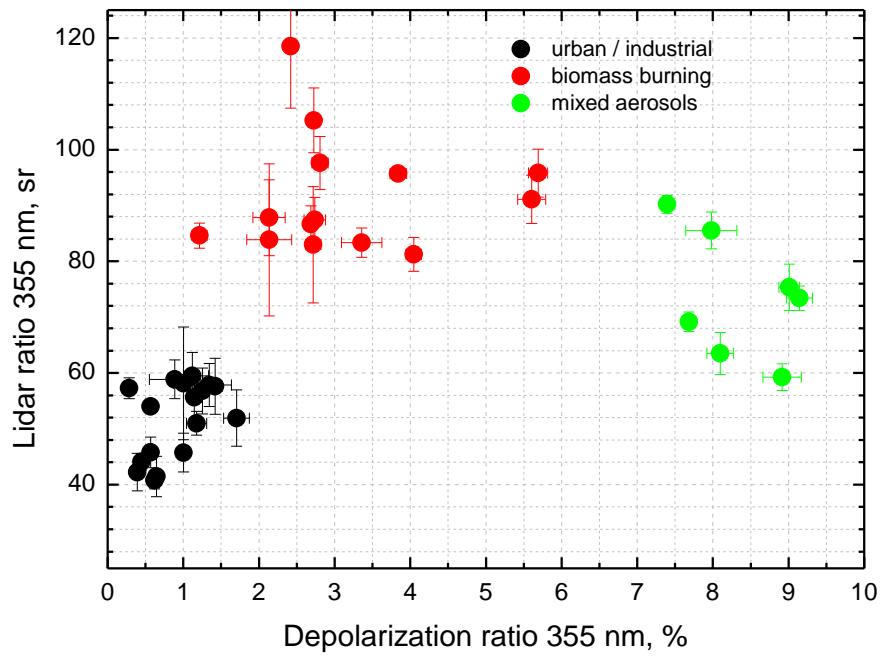
Figure 6. Lidar ratio at 355 nm versus the depolarization ratio at 355 nm for the three
aerosol types investigated in our study

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946 **Figure 7.** General literature values for lidar ratio at 355 nm, Ångström exponent and depolarization ratio (355 or 532 nm) for urban/industrial
947 (black), biomass burning (red) and for mixed biomass burning with desert dust aerosols (green). The x-axis are the studies presented in Table 4.
948 Floating columns are referring to range values while the symbols are referring to mean values with one standard deviation. The depolarization
949 values is at 355 nm except for the cases noted with asterisk (*) which are referring to visible wavelength (532 nm or 710 nm).

950
951 **Figure 7.** Effective radius versus Ångström exponent for the three aerosol types
952 investigated in our study.
953