

Article

# Smoke from Australian bushfires over Marambio, Antarctica, in January 2020

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**Abstract:** Australian smoke from the extraordinary biomass burning in December 2019 has been observed over Marambio, Antarctica, on 7th-10th January, 2020. The smoke plume was transported thousands of kilometers over the Pacific Ocean, and reached the Antarctic Peninsula at 13 km height, as determined by satellite lidar observations. The proposed origin and trajectory of the aerosol are supported by back-trajectory model analyses. Ground-based Sun-Sky-Moon photometer belonging to the Aerosol Robotic Network (AERONET) measured aerosol optical depth (500 nm wavelength) above 0.3, which is unprecedented for the site. Inversion of sky radiances provide the optical and microphysical properties of the smoke over Marambio. The AERONET data near the fire origin in Tumbarumba, Australia, was used to investigate the changes in the measured aerosol properties after transport and ageing. The analysis shows an increase in the fine mode particle radius and a reduction in absorption (increase in the single scattering albedo). The available long-term AOD data series at Marambio suggests that smoke particles could have remained over Antarctica for several weeks after the analyzed event.

**Keywords:** Antarctica; aerosol; optical properties; biomass burning, Australian fires, smoke ageing

## 1. Introduction

Antarctica is the most isolated and pristine continent. Studies about Antarctic aerosols started in the 1960's, both with remote sensing and in situ techniques [1,2]. Those first measurements indicated the presence of sulfate, sea salt and crustal aerosols. Despite the very low concentrations, the presence of extraneous particles proves that aerosol transport takes place on a global scale [3]. Coarse sea salt particles dominate the aerosol mass over Antarctica, especially in coastal areas. The main sources of secondary aerosol (fine mode) over Antarctica are the marine sulphur and organic aerosol precursors [4]. A significant fraction of fine mode of Antarctic aerosol might originate from a source not yet identified [5]. The absorption is a key parameter to determine the aerosol radiative effect [6]. Significant black carbon concentrations were measured in Antarctica, with the main sources suggested to be biomass burning aerosols transported from South-America, Africa and Australia [7,8].

In the Antarctic Peninsula, severe environmental changes in the past decades are likely linked to natural variability (changes in atmospheric circulation, local sea-ice loss), although other processes with anthropogenic origin like the ozone depletion may have played a role too. As a result, some of

29 the largest increases in near-surface air temperature in the Southern Hemisphere in the 20<sup>th</sup> century  
30 were registered in the Antarctic Peninsula [9]. Long-term observations are needed to establish whether  
31 and how these changes can affect the aerosol amount and properties in the region [8].

32 Long range transported aerosol from biomass burning affects polar regions, especially the Arctic  
33 [10–15], but also Antarctica [7,16,17]. The occurrence of bushfires is likely to increase due to climate  
34 change [18,19] and thus the occurrence of aerosol events caused by long-range transported smoke over  
35 polar regions. The aerosol optical properties of the transported aerosol, thus its radiative effects, are  
36 largely uncertain. Ageing processes during transport affect the size, shape and absorption properties  
37 of the smoke particles [20–22]. Other important aerosol effects are the deposition on snow and ice, and  
38 the interaction with clouds [3,8].

39 In January 2020, bushfires in Australia burned over 20,000 ha ( $2 \times 10^8 m^2$ ) of land and injected large  
40 amounts of smoke into the atmosphere [23]. The MetService at New Zealand reported a huge smoke  
41 plume that was transported 2,000 km across the Tasman Sea and reached the lower South Island,  
42 which reduced visibility and produced red skies. Satellite imagery revealed the plume extension,  
43 and the World Meteorological Organization reported the transport of the smoke across the Pacific  
44 Ocean to South America, and that smoke had ‘probably’ reached Antarctica (<https://public.wmo.int/en/media/news/australia-suffers-devastating-fires-after-hottest-driest-year-record>). The Australian  
45 Bureau of Meteorology reported in spring 2019 the highest fire weather danger that was measured  
46 by the Forest Fire Danger Index across the whole Australia [24]. A detailed and qualitative satellite  
47 analysis about bushfires in Australia from September 2019 to January 2020 can be found at [https://worldview.earthdata.nasa.gov/?tr=australia\\_fires\\_2019\\_2020](https://worldview.earthdata.nasa.gov/?tr=australia_fires_2019_2020) and <https://asdc.larc.nasa.gov/news/new-storymap-studying-the-2019-2020-australian-bushfires-using-nasa-data>.  
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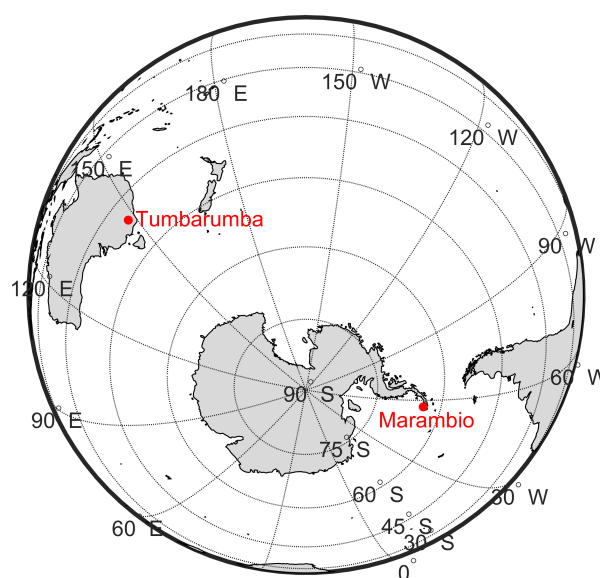
51 Recent studies have thoroughly described this smoke event [25,26]. The smoke generated  
52 Pyro-cumulonimbus (pyroCb) clouds that caused unprecedented stratospheric perturbations, similar  
53 to those caused by moderate volcanic eruptions [25]. In particular, it was observed the generation  
54 of potential vorticity and anticyclonic circulations caused by absorptive aerosol heating [26]. The  
55 paths and extension of several stratospheric smoke plumes are described in detail. The main plume  
56 generated a self-maintained anticyclonic vortex measuring 1000 km in diameter, that ascended during  
57 three months until it reached 35 km height; it lofted a confined bubble of carbonaceous aerosols and  
58 water vapour that travelled westbound round-the-world over 66,000 km, until the beginning of April  
59 2020 [25]. Both works explicitly indicate that a smoke patch was transported over Antarctica and  
60 remained there for several weeks. The smoke over Antarctica generated a weaker vortex but was  
61 subject to strong aerosol heating due to the permanent daylight and rose up to 27 km [25,26]. Another  
62 recent study based on LIDAR observations at Punta Arenas, Chile, confirms the transport of the smoke  
63 at stratospheric levels over South America, and provides optical properties such as optical depth, lidar  
64 ratio and depolarization ratio [27].

65 Ground based remote sensing instruments like the AErosol RObotic NETwork (AERONET)  
66 photometers [28] are globally deployed in order to assess aerosol properties. The Group of  
67 Atmospheric Optics at the University of Valladolid (GOA-UVa), in collaboration with the Finnish  
68 Meteorological Institute and the National Weather Service in Argentina, installed in 2018 an AERONET  
69 Sun/sky/Moon photometer at Marambio station, located in the Antarctic Peninsula. This instrument  
70 detected an extraordinary and unprecedented aerosol event over Marambio on 7th-10th January 2020,  
71 with record values of aerosol optical depth (AOD), and optical properties that indicate the presence of  
72 aged smoke aerosol particles. The aim of this paper is to assess the origin of this aerosol event and  
73 characterize the optical and microphysical properties of the aerosols.

## 74 2. Sites and instrumentation

### 75 2.1. Measurement stations

76 Marambio is an Argentinian Antarctic station located in the ice-free Seymour-Marambio island,  
77 at the north-east tip of the Antarctic Peninsula ( $64.240^{\circ}$  S,  $56.625^{\circ}$  W and 200 m a.s.l., see Figure 1).  
78 Marambio is characterized by a permafrost ecosystem. The Antarctic region has no permanent human  
79 habitation but Marambio station operates all-year round and acts as a logistic center for the scientific  
80 projects and also for other Argentinian Antarctic stations. There are around 40-70 persons in Marambio  
81 even in austral winter [8]. The temperature at Marambio varies between  $+10^{\circ}\text{C}$  in summer and  $-30^{\circ}\text{C}$   
82 during winter. However the wind speed can reach 100 km/h decreasing the apparent temperature to  
83  $-60^{\circ}\text{C}$ . The strong winds can blow snow and dust (in winter or summer) into the air. The prevailing  
84 wind directions are southwest and northwest [8].



**Figure 1.** Location of the Marambio station (Antarctica) and Tumberumba AERONET station (Australia).

85 In January 2018 the GOA-UVA installed a Sun/sky/Moon photometer CE318-T (*Cimel Electronique*  
86 *S.A.S.*) and an OMEA 3C all-sky camera (*Alcor System*) in Marambio station. This instrumentation  
87 is installed on the rooftop of the Scientific Laboratory, which is located in the middle of the station,  
88 between the the airfield and station buildings. This laboratory belongs to the Argentinian National  
89 Meteorological Service (*Servicio Meteorológico Nacional*) and is shared with the Argentinian National  
90 Direction of the Antarctic (*Dirección Nacional del Antártico*). The laboratory's rooftop is also equipped  
91 with instrumentation focused on atmospheric studies by different techniques, e.g. a Precision Filter  
92 Radiometer (PFR), a Vaisala Ceilometer, Brewer spectroradiometer, MAX-DOAS, to cite some. In  
93 addition, several aerosol in-situ instruments, which belong to the Finnish Meteorological Institute  
94 (FMI), installed since January 2013, are located in a container 300 m away from the main site [8].

95 In order to identify the bushfire aerosol properties near its source, we used an AERONET station  
96 located in the vicinity of the bushfires in Australia. The Tumberumba AERONET site ( $35.708^{\circ}$  S,  
97  $147.950^{\circ}$  E and 776 m a.s.l.), located at the south-east of Australia (see Figure 1), is equipped also with  
98 a Sun/sky/Moon photometer CE318-T photometer since July 2019. The Sun photometer is located on  
99 top of a rounded hill in cleared land used for grazing of cattle. Native eucalypt woodland is located to

100 the west and south east. The area suffered a large bush fire in December 2019-January 2020, known  
101 as the Dunns Road Bushfire. Native forest and grazing paddocks burnt within 10 km of the site. A  
102 flux tower, with CSIRO instrumentation located 20 km to the north east in the Bago State Forest was  
103 damaged when the ground infrastructure was burnt. At this site, persistent high values of aerosol  
104 optical depth were detected for several weeks (data are available in the AERONET website).

## 105 2.2. Instruments and data

106 The Cimel CE318-T is designed to measure at filter-selected wavelengths the direct Sun and  
107 Moon irradiance, and the diffuse sky radiance. These measurements are useful to retrieve different  
108 aerosol properties [29–31]. The Cimel radiometer is the reference instrument of AERONET. See  
109 Holben *et al.* [28], Giles *et al.* [32] for details about the instrument, data processing and calibration.  
110 The generated data are public and can be freely downloaded from the AERONET website (<https://aeronet.gsfc.nasa.gov>).  
111 The GOA-UVa is responsible of an AERONET calibration center located in  
112 Valladolid, Spain, in cooperation with NASA, the University of Lille and the Spanish Meteorological  
113 Agency. The CÆLIS software tool was developed for the management of this calibration center and  
114 instrument monitoring [33–35]. This tool allowed for quick detection of the event as well as assisted in  
115 the routine monitoring of the CE318-T instrument performance at Marambio. This operational aspect  
116 is crucial in polar regions.

117 In this paper we have used the AERONET version 3 level 1.5 (cloud-screened) aerosol products  
118 of: AOD at 340, 380, 440, 500, 675, 870, 1020 and 1640 nm [32], which is derived from Sun/Moon direct  
119 irradiance measurements; aerosol particle size distribution and single scattering albedo (SSA) at 440,  
120 675, 870 and 1020 nm, both retrieved by the AERONET operational inversion code using as input the  
121 multi-wavelength AOD and sky radiance measurements taken at almucantar and hybrid geometry  
122 [31,36]; and the AOD fine mode fraction at 500 nm, which was calculated by the spectral deconvolution  
123 algorithm (SDA) [37]. A set of quality criteria was applied to ensure the reliability of the inversion  
124 data, i.e. solar zenith angle  $>50^\circ$  (for almucantar scans), minimum number of symmetrical angles and  
125 retrieval error between 5% and 8% depending on AOD [38]. The Ångström exponent (AE), which  
126 represents the AOD spectral variation and contains information about the aerosol size predominance,  
127 has been calculated by a least square fit of the AERONET AOD values in the spectral range from 440 to  
128 870 nm. Concerning the inversion products, the fine and coarse modes are separated at the inflection  
129 point within the radius interval 0.439–0.992  $\mu\text{m}$ . The size distribution parameters (median and effective  
130 radius, volume concentration) are provided for each mode.

131 In order to identify the height of the aerosol layers, vertical profiles of total attenuated backscatter  
132 at 532 nm have been obtained from CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) for  
133 the Antarctica overpass of the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite  
134 Observations) satellite [39]. These data belong to the CALIPSO Lidar Level 1 V4.10 product  
135 (CAL\_LID\_L1-Standard-V4-10<sup>1</sup>).

136 The Aerosol Index -PyroCumuloNimbus- (AI-PCN) product [40], from the Ozone Mapping  
137 Profiler Suite (OMPS) instrument on board the joint NASA/NOAA Suomi National Polar-orbiting  
138 Partnership (NPP) satellite [41], has been used to track the spread of high aerosol concentrations in the  
139 atmosphere. This satellite product is based on the detection of absorbing particles in the ultraviolet  
140 region. The aerosol index is derived from normalized radiances using 2 wavelength pairs at 340 and  
141 378.5 nm. This product is linked to both the thickness and the height of the aerosol layer located in the  
142 atmosphere.

143 In addition, airmass back-trajectories at Marambio have been obtained from the National Oceanic  
144 and Atmospheric Administration (NOAA) Air Resources Laboratory's (ARL) Hybrid Single-Particle  
145 Lagrangian Integrated Trajectory (HYSPPLIT) model [42]. HYSPPLIT has been run with the trajectory

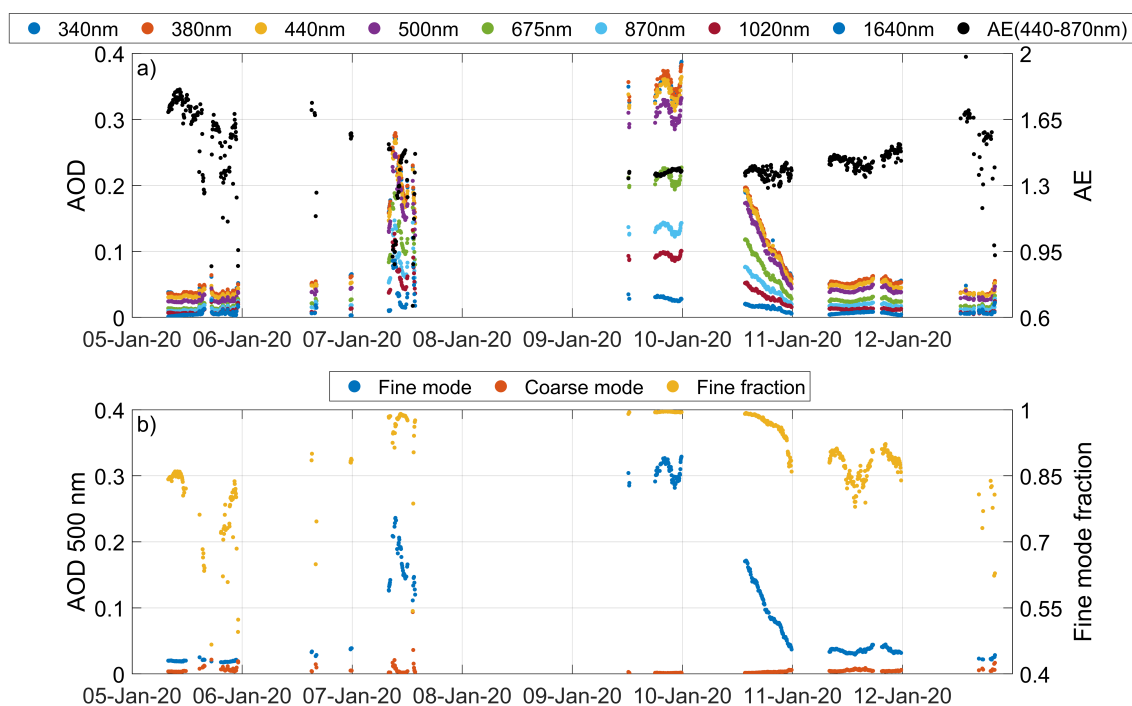
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<sup>1</sup> [https://www-calipso.larc.nasa.gov/products/lidar/browse\\_images/std\\_v4\\_index.php](https://www-calipso.larc.nasa.gov/products/lidar/browse_images/std_v4_index.php)

146 ensemble option; this option starts multiple trajectories from the starting location. Each member of the  
 147 trajectory ensemble is calculated by offsetting the meteorological data by a fixed grid factor, resulting  
 148 in 27 members for all-possible offsets in X,Y, and Z<sup>2</sup>.

### 149 3. Smoke event detection over Marambio

150 On 7th January, 2020 the aerosol optical depth at Marambio raised from values below 0.05 to  
 151 higher than 0.2 (at 500 nm), as can be observed in Figure 2a. The AOD on 8th January is not available  
 152 due to cloudiness, but the AOD on the 9th was even higher, about 0.3 at 500 nm. The AOD decreased  
 153 on 10th January to AOD values below 0.05 at 500 nm at the end of the day. The AE values from 7th to  
 154 10th January were about 1.3, i.e. it was lower than the AE observed before and after the event (about  
 155 1.6). However the fine mode AOD shown in Figure 2b considerably increased during the mentioned  
 156 event, indicating fine particle predominance, especially on 9th January, whereas the coarse mode AOD  
 157 was constant and stable for all the shown days. Figure 2b shows that the fine mode fraction of the  
 158 AOD (500 nm) was larger than 90% from 7th to 10th January and nearly 100% on 9th January. The fine  
 159 mode fraction is still above 85% on the 6th and 11st January, but below 85% on 5th and 12nd January.  
 160 These results indicate the presence of a fine particle event over Marambio from 7th to 10th January.

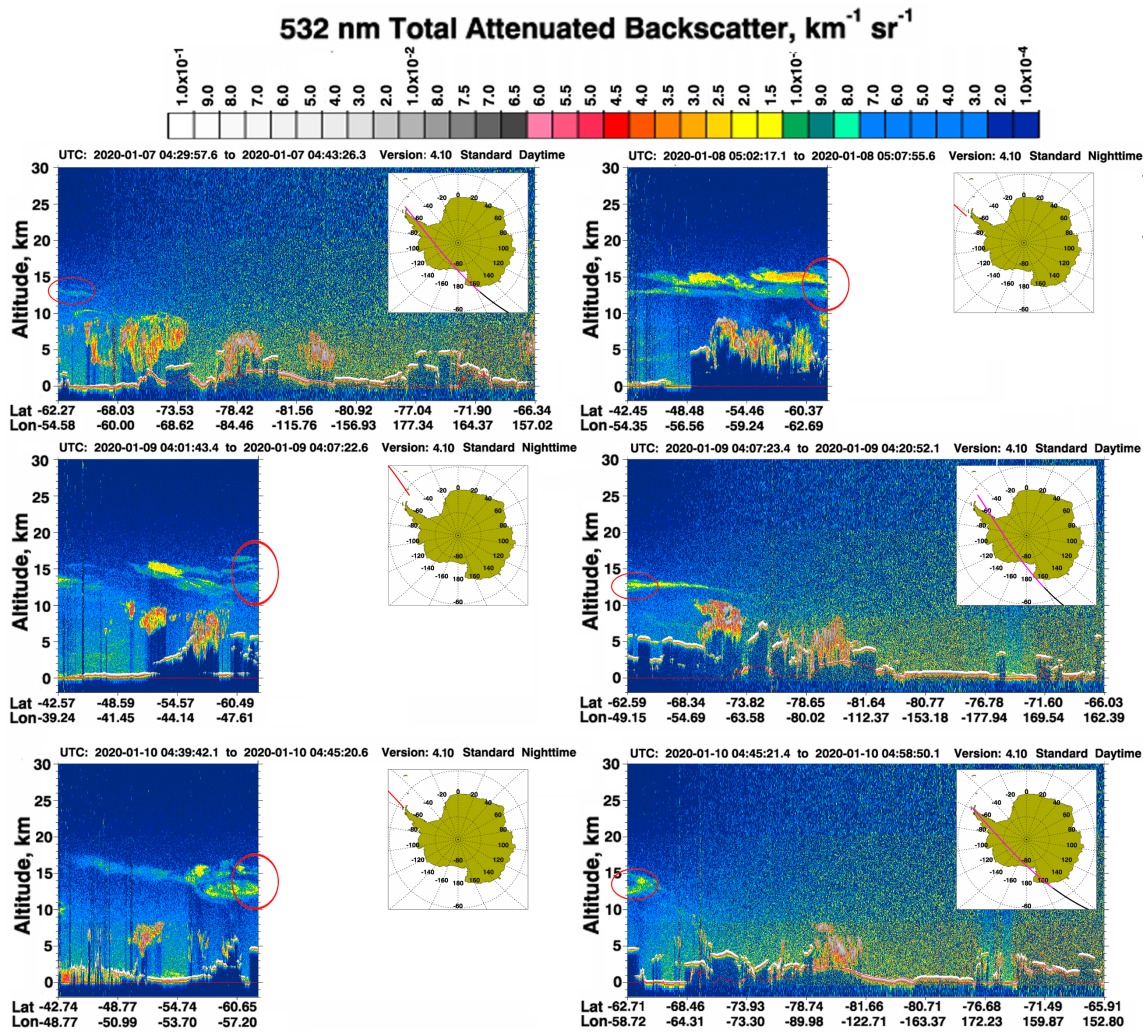


**Figure 2.** (a) Aerosol optical depth (AOD) for different wavelengths and Ångström exponent (AE) at Marambio on 5-12th January 2020. (b) Fine and coarse mode aerosol optical depth at 500 nm and the fine mode fraction at Marambio on 5-12th January 2020.

161 The height of the aerosol layer over Marambio could provide additional information about  
 162 its origin. Figure 3 shows the vertical attenuated backscatter profiles at 532 nm for selected  
 163 CALIOP/CALIPSO overpasses close to Marambio during the analysed event. A weak layer around  
 164 13 km asl was detected over Marambio at the beginning of 7th January 2020. A stronger layer can be  
 165 seen over Marambio on the 8th, 9th and 10th January 2020 at the same height. Another layer around

<sup>2</sup> <https://www.ready.noaa.gov/hypub-bin/trajtype.pl>

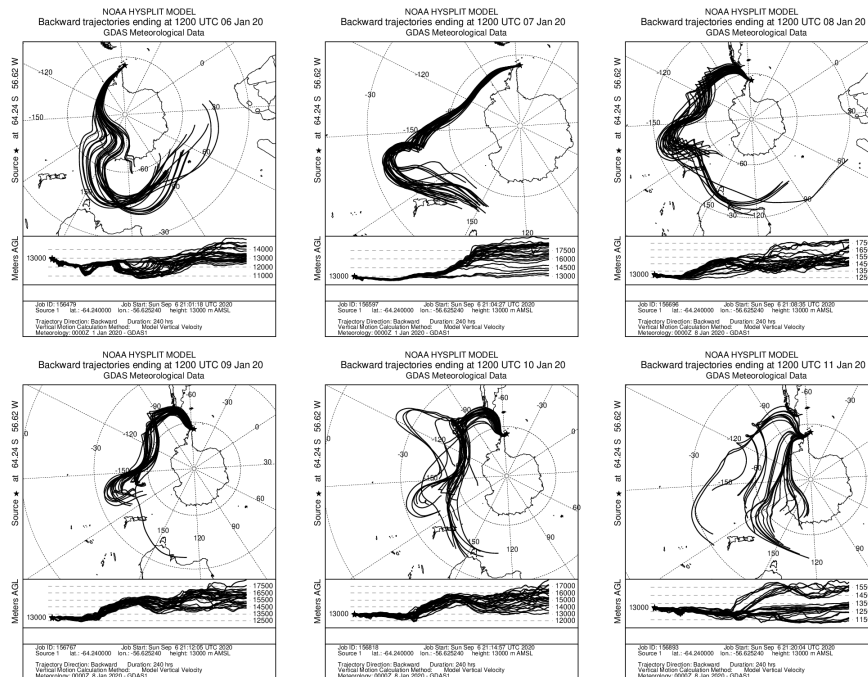
15 km asl is frequently visible on those days. All these observed high altitude layers over Marambio presented lower total attenuated backscatter at 532 nm compared to 532 nm (not shown), and an attenuated color ratio (backscatter at 1064 nm / backscatter at 532 nm) in general about 0.4-0.6. The depolarization ratio at 532 nm CALIOP product was mostly between 0.0 and 0.2 for the marked layers. They were generally classified as 'stratospheric aerosol' by the CALIOP vertical feature mask product (Version 4.20). The aerosol subtype CALIOP product (Version 4.20) identifies these layers mostly as 'elevated smoke'.



**Figure 3.** Vertical profiles of total attenuated backscatter at 532 nm from CALIOP/CALIPSO overpasses. The profiles are shown for six different overpasses from 7th to 10th January 2020. The highest aerosol layers close to Marambio location are marked with a red circle.

In order to detect the origin of the analyzed aerosol event, ensemble HYSPLIT trajectories running for 240 hours backwards and ending at 13 km asl over Marambio (the observed height of the aerosol layer) have been computed. These backtrajectories are shown in Figure 4 for the days from 6th to 11th January, at 12:00 UTC. The air masses at 13 km altitude over Marambio came mainly from the middle of South Pacific ocean area (origin at about 15 km asl) for the aerosol event days (7th-10th January 2020), with some of the ensemble computations originating in Southeastern Australia. This supports our hypothesis that the detected aerosol could be smoke that originated in the bushfires of Australia austral summer 2020. Similar layer structure and heights were found by Ohneiser *et al.* [27] over Punta Arenas, Chile, where the smoke plume was detected by means of a ground-based lidar, and HYSPLIT backtrajectories indicate that it originated in the Australian bushfires. The backtrajectories on 6th and

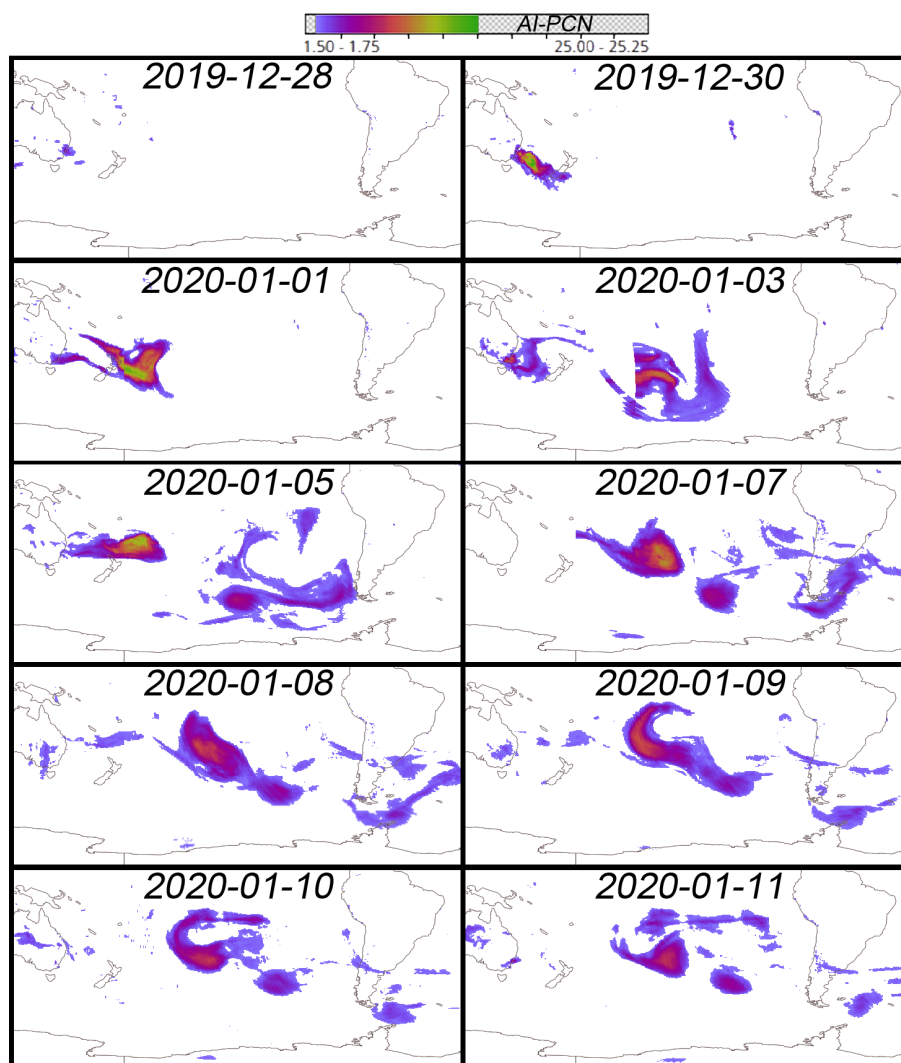
183 11th January have a different origin than they show during the event, crossing the Southern Ocean  
184 and Antarctic Peninsula areas.



**Figure 4.** Ensemble HYSPLIT 10-days back-trajectories computed from Marambio at 13 km asl from 6th to 11st January 2020.

185 The AI-PCN from OMPS/NPP satellite is shown in Figure 5 for several days between 28 December  
186 2019 and 11th January 2020 over the South Pacific Ocean. This product, as well as True Color  
187 reflectance for all days from 10th December 2019 to 13th January 2020 and corresponding animations,  
188 can be found as supplementary material to this paper. The AI-PCN in Figure 5 shows aerosol load  
189 appearing over southeastern Australia on 28th (and 29th, not shown) December, 2019, as a consequence  
190 of massive bushfires. CALIOP/CALIPSO data on 31st December (not shown) reveal that this aerosol  
191 was injected up into the stratosphere to 13-15 km height [26], similar to the case reported by [43]  
192 for wildfire-driven thunderstorms (pyro-cumulonimbus).

193 From 30th December 2019 to 1st January 2020 the aerosol plume was transported eastwards,  
194 crossing over New Zealand (see Figure 5) at around 15 km asl as indicated by CALIOP (not shown).  
195 The following days the plume was transported over the Pacific Ocean. A detailed description of  
196 this smoke plume as well as subsequent plumes is provided by Khaykin *et al.* [25], Kablick III *et al.*  
197 [26]. Some smoke patches escaped from the vortex and moved eastwards. For instance, two plumes  
198 travelled to the south and the center of South-America on 5th January 2020. The first one reached  
199 southern Argentina and Chile on 6th January, and approached to Marambio on 7th January. The  
200 aerosol plume stayed over Marambio area from 8th to 10th January and moved towards Northeast  
201 on 11th January. For details, see Aerosol Index animation provided as supplementary material to this  
202 paper, as well as the supplementary information to [26, S2 animation]. This aerosol transport from  
203 Australia to Marambio was additionally confirmed by the MODIS (Moderate Resolution Imaging  
204 Spectroradiometer) AOD data and NAAPS (Navy Aerosol Analysis and Prediction System) model (see  
205 supplementary material), providing further evidence that the aerosol plume detected over Marambio  
206 originated in the bushfires in Australia, and was transported more than 10,000 km over the Pacific  
207 Ocean.



**Figure 5.** Aerosol Index PyroCumuloNimbus (AI-PCN) maps over Pacific ocean for 10 different days during the smoke transport from Australia to Marambio station. These maps have been extracted from NASA's Earth Observing System Data and Information System (EOSDIS) worldview tool (<https://worldview.earthdata.nasa.gov>)

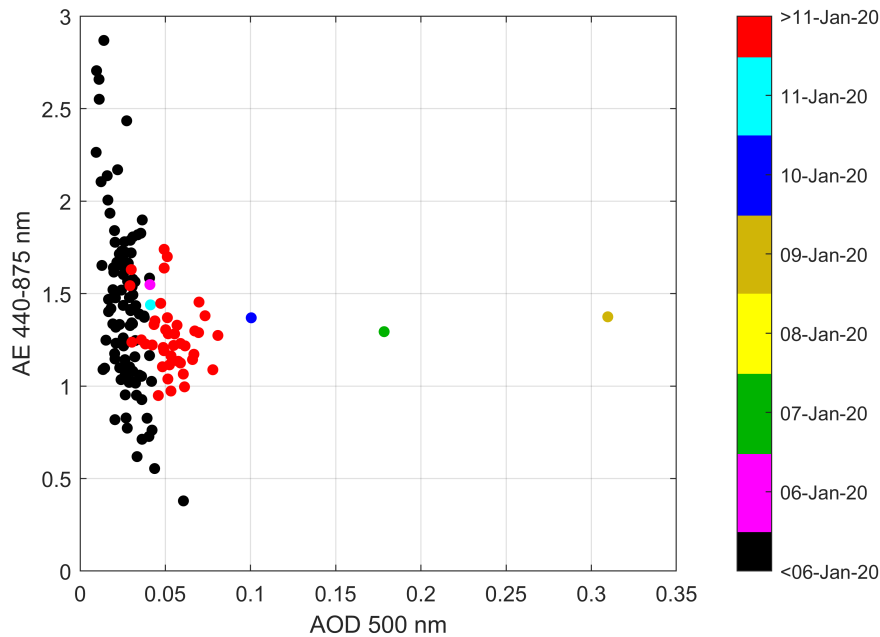
#### 208 4. Aerosol properties

209 The scatter plot between Ångström exponent and aerosol optical depth can be used to classify  
 210 the aerosol [e.g. 44,45]. In this plot, the horizontal axis indicates the daily AOD whereas the vertical  
 211 axis indicates the AE, related to the size predominance (larger size for lower AE). The AERONET  
 212 data collected at Marambio until now, are depicted in Figure 6. They include summer campaigns in  
 213 2008 and 2009 performed by CEILAP-UNIDEF (National Scientific and Technical Research Council,  
 214 Argentina) and routine measurements since January 2018. These data indicate very low AOD with  
 215 average of 0.038 at 500 nm, and relatively high and variable AE (440-870 nm) in the range 1.0 to 2.5,  
 216 with overall mean of 1.38 (median 1.33). This can be considered the background aerosol at the site.  
 217 Only occasionally (11% of the days) the AE is below 1.0, indicating in these cases the predominance  
 218 of coarser (possibly marine) particles. Similar AOD values are reported by Tomasi *et al.* [46] for the  
 219 GAW-PFR observations at Marambio in 2011-2013.

220 The three investigated days (7th, 9th and 10th January 2020) have the highest AOD daily means  
 221 of the entire data set. Until this episode, AOD had not exceeded 0.1 (at 500 nm wavelength), clearly  
 222 showing that this event was extraordinary at the site. Moreover, the AOD daily mean values of  
 223 AOD after the event (until April 2020), marked in red color in Figure 6, are consistently above the



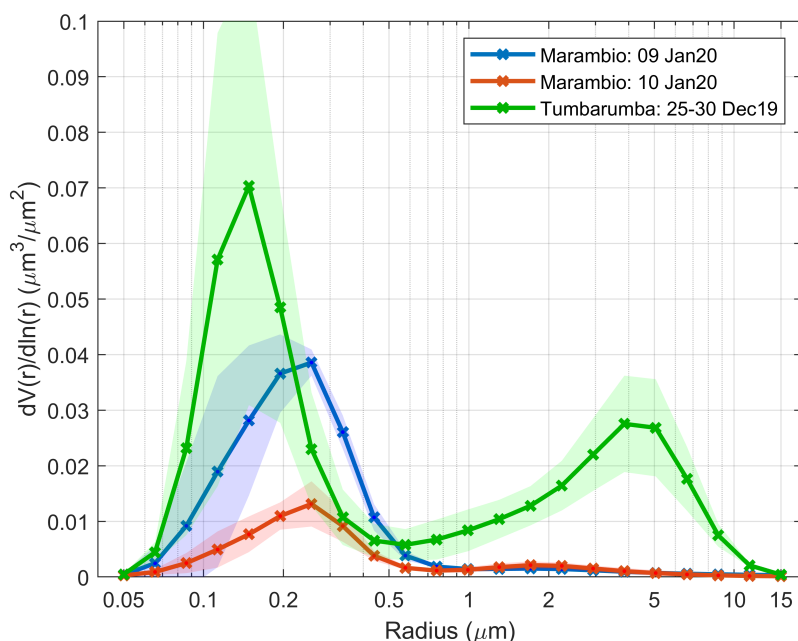
224 background values, suggesting that some residual aerosol could be present in the atmospheric column  
 225 for some weeks after the 7-10th January event. The analysis by Kablick III *et al.* [26] also indicates that  
 226 smoke could be present at Antarctic latitudes until mid-February. The AE, in the range 1.3 to 1.4 (also  
 227 observed in Figure 2a), shows intermediate values which would indicate a certain contribution of  
 228 coarse mode particles. However the fine/coarse mode separation of the AOD indicated that the coarse  
 229 mode had a minor contribution to the AOD. The analysis of the particle size distribution is necessary  
 230 to tackle this apparent contradiction.



**Figure 6.** Daily mean of Ångström exponent (AE) vs. aerosol optical depth (AOD) at 500 nm at Marambio (until April 2020). Colorbar indicates the date of each data point highlighting the analyzed days of the event.

231 The favorable sky conditions during the event on 9th and 10th January (clouds are very frequent in  
 232 this region), allowed sky radiance scans (almucantar and hybrid geometries) which could be inverted  
 233 to retrieve the optical and microphysical properties of the aerosol. Given that the AOD during the  
 234 event was much larger than the background values (0.3 on Jan. 9th vs. 0.025 on the 5th), we can assume  
 235 that the inverted properties mainly correspond to those of the elevated smoke layer. We calculated  
 236 the daily mean aerosol size distribution on the 9th and the 10th January, based on 6 and 9 inversions  
 237 respectively. The result is shown in Figure 7, together with the average aerosol size distribution  
 238 measured near the smoke plume origin, at the Tumbarumba site in Australia. The latter corresponds  
 239 to the mean of the daily averages in the period 25-30th December 2019, during which high AOD was  
 240 persistently measured due to active fires.

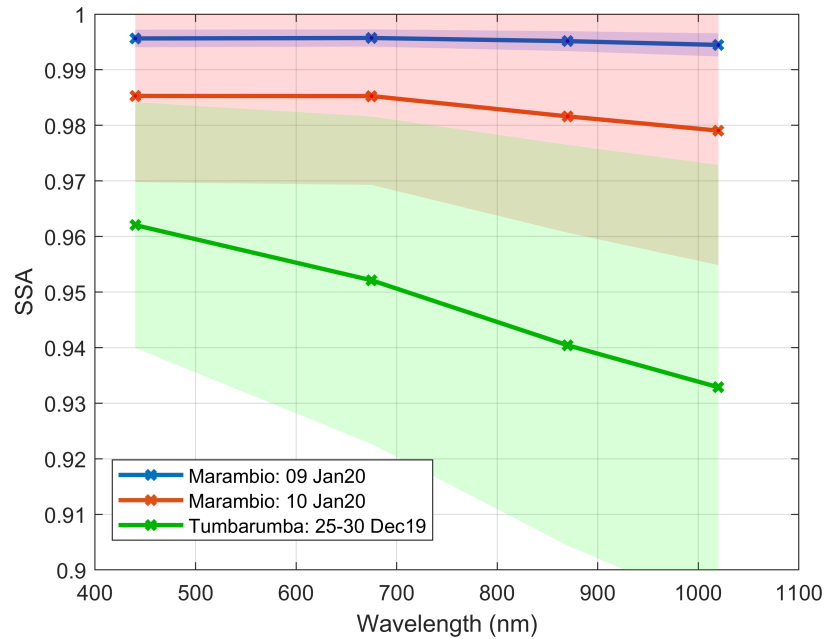
241 The size distribution at Tumbarumba was bimodal, with predominant fine mode. The average  
 242 fine mode fraction was 0.64 in particle volume. However the coarse mode played a minor role in  
 243 terms of AOD, with fine mode fraction of the AOD (500 nm) above 0.94, as derived from the spectral  
 244 deconvolution algorithm data. This minor role of the coarse mode was also noted in other studies  
 245 about biomass burning [47,48]. The lack of optically significant coarse mode is typical of biomass  
 246 burning aerosols of several source regions [49]. The mean AOD (500 nm) derived here for the fresh  
 247 biomass burning aerosol at Tumbarumba was 0.56, indicating a significant aerosol load, that peaked  
 248 on 1-3 January, with AOD (500 nm) above 5.0.



**Figure 7.** Volume particle size distribution over Marambio, Antarctica, on 9th and 10th January 2020 (daily averages), as well as the mean of the daily averaged size distribution over Tumbarumba, Australia, for the period 25-30 December 2019. Color shaded areas represent  $\pm 1$  standard deviation.

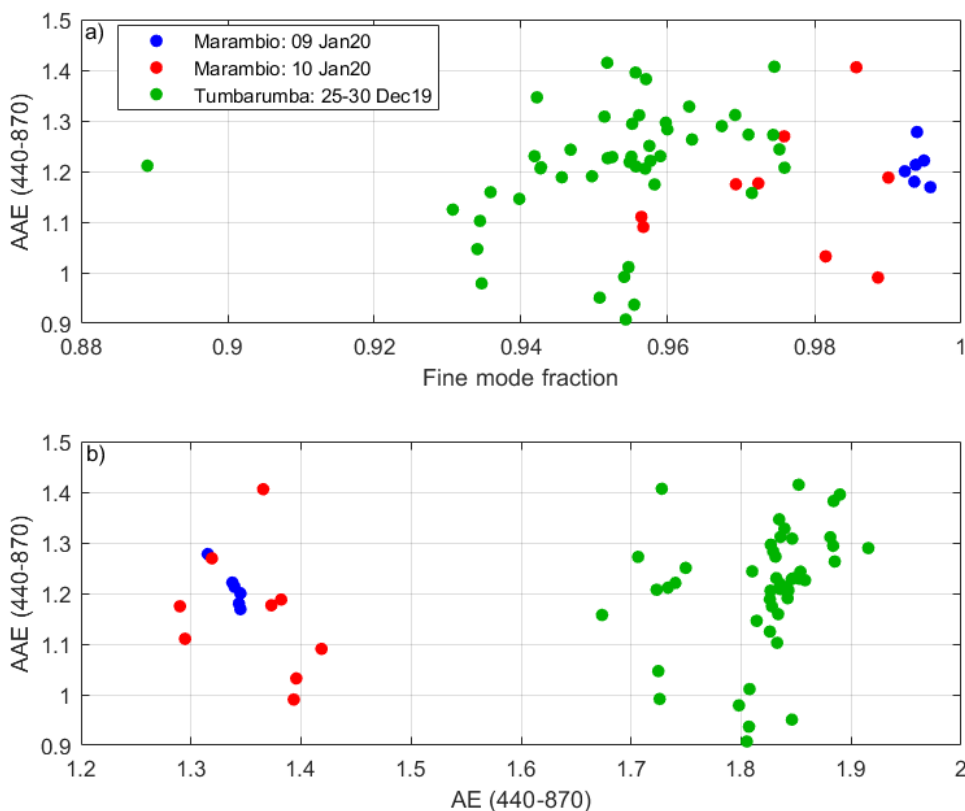
249 Two main features arise from the comparison of the mean size distributions over Marambio and  
 250 Tumbarumba. First, the coarse mode nearly disappeared in the lofted layer over Antarctica, suggesting  
 251 that the large particles were most effectively scavenged during the long-range transport. Similar  
 252 results have been observed in other aerosol events [e.g. 50]. Second, the fine mode modal radius was  
 253 larger at Marambio ( $0.22 \mu\text{m}$ ), than it was at Tumbarumba ( $0.16 \mu\text{m}$ ). These are average values for the  
 254 mentioned periods (see Figure 7). An increase in the fine mode radius is typical for aged smoke, due  
 255 to coagulation and condensation processes [21,49,51]. This increase in the fine mode modal radius is  
 256 likely the reason for the above-mentioned reduction in the AE to 1.34 at Marambio while the averaged  
 257 daily AE at Tumbarumba was 1.82 for the 25-30 Dec. period. Radiative transfer simulations have been  
 258 done using as input the AERONET averages of fine and coarse aerosol concentrations (at both sites),  
 259 varying the fine mode modal radius; the obtained results (not shown) support the hypothesis that,  
 260 for the observed aerosol fine and coarse concentrations, the AE decreases as the fine mode radius  
 261 increases, in agreement with the measured data.

262 Differences on the aerosol optical properties obtained at Marambio and Tumbarumba have also  
 263 been observed, possibly associated to ageing. The aerosol SSA and its wavelength dependence can  
 264 be linked with the aerosol type [52,53]. At Tumbarumba, the SSA was about 0.96-0.97 at 440 nm and  
 265 decreased in the longer wavelengths (0.93 at 1020 nm), see Figure 8. These values are typical in biomass  
 266 burning aerosols, e.g. in other fires reported in Australia [54] and the Amazonian forest [52]. The SSA  
 267 was much higher at Marambio, above 0.99 at all investigated wavelengths on 9th January, and a bit  
 268 lower on 10th, and exhibiting very small spectral dependence on both days. The condensation and/or  
 269 formation of secondary organic aerosol can result in enhanced values of SSA [55]. The loss of coarse  
 270 aerosols and the increase in relative humidity can be other possible factors that favor the increase of  
 271 the SSA during transport [50].



**Figure 8.** Single scattering albedo (SSA) as a function of wavelength over Marambio, Antarctica, on 9th and 10th January 2020 (daily averages), as well as the mean of the daily averaged SSA over Tumberumba, Australia, for the period 25-30 December 2019. Color shaded areas represent  $\pm 1$  standard deviation.

272 The analysis of intensive optical properties can be used to discriminate aerosol types [53]. The  
 273 approach consists of combining a size-related property (i.e. Ångström exponent, fine mode fraction  
 274 of the AOD) with a parameter representing the absorptive properties (i.e. single scattering albedo  
 275 or absorption Ångström exponent). Figure 9a shows the absorption Ångström exponent (AAE) as  
 276 function of the fine mode fraction of the AOD (500 nm) for Tumberumba and Marambio, as provided  
 277 by the AERONET inversion products. No significant change is observed in the AAE for both sites (1.21  
 278 at Tumberumba, 1.19 at Marambio). They are within the expected range for biomass burning aerosols  
 279 [53]. The highest FMF is found for the central day of the event at Marambio (10th January), where the  
 280 data show very low scatter. Figure 9b shows the AAE as a function of the AE of extinction. In this plot  
 281 the separation of the data from both sites is clearer, given the decrease of the AE of the transported  
 282 aerosol (1.34 at Marambio, 1.82 at Tumberumba). In view of the aerosol key aerosol types described in  
 283 Giles *et al.* [53], the observed properties of the smoke over Marambio (intermediate values of the AE;  
 284 very high FMF; and very high SSA), do not clearly match any of the described categories. The fact that  
 285 the smoke particles are found in the stratosphere and have experienced a long atmospheric transport,  
 286 with associated ageing processes, can be the reason behind these distinct properties.



**Figure 9.** Absorption Ångström exponent (AAE) as a function of: (a) Fine Mode Fraction of the AOD (500 nm); and (b) Extinction Ångström exponent (AE) derived in the range 440-870 nm. Data points are inversion data (almucantar and hybrid sky scans) for Marambio, Antarctica, on 9th and 10th January 2020, and Tumbarumba, Australia, for the period 25-30 December 2019.

## 287 5. Conclusions

288 An extraordinary aerosol event with AOD at 500 nm above 0.3 was detected at Marambio station in  
 289 the Antarctic Peninsula, by means of an AERONET Cimel CE318-T photometer. CALIPSO backscatter  
 290 profiles, Suomi NPP aerosol index and back trajectory analysis, were analyzed to confirm the origin  
 291 of the measured aerosol layer in the Australian fires that occurred about 10 days earlier, and injected  
 292 large amounts of smoke particles up to the stratosphere. The particles were transported over the ocean  
 293 and reached the Antarctic Peninsula at about 13 km height.

294 The fine mode particles largely dominated the aerosol size distribution, with the fine mode  
 295 fraction of the AOD ranging from 0.94 to 0.99. The AOD levels during the event (7-10 January 2020)  
 296 constitute the highest columnar values recorded in Marambio. The AOD data measured until April  
 297 2020 suggest that part of the smoke remained over Antarctica for several weeks after the analysed  
 298 event.

299 The optical and microphysical aerosol properties of the fresh smoke aerosol were measured at the  
 300 Australian AERONET site Tumbarumba. The comparison with Marambio data showed that the coarse  
 301 mode particles were lost during the long-range transport, and that ageing processes increased the  
 302 plume average aerosol particle size. In our case study, the fine mode modal radius increased from 0.16  
 303 to 0.22  $\mu\text{m}$ . This increase in the fine mode radius could be the main responsible for the lower AE values  
 304 at Marambio ( $\sim 1.3$ ) than at Tumbarumba ( $\sim 1.8$ ). The particles also became less absorbing, with single  
 305 scattering albedo (440 nm) increasing from 0.96 to 0.99 (from 0.93 to 0.99 at 1020 nm). Conversely, the  
 306 absorption Ångström exponent did not change significantly (from 1.21 to 1.19).

307 The photometers used in AERONET have proven their capability to provide aerosol monitoring  
308 in Antarctica, including direct Sun irradiance measurements for AOD and well as sky radiance scans  
309 during daytime. Direct Moon irradiances are monitored since austral winter 2020 and will help to  
310 improve the year-round aerosol property monitoring. Longer data sets and the joint analysis of all  
311 the co-located aerosol measurements are still needed to provide consistent aerosol climatology in  
312 the region, investigate possible trends and identify the frequency and intensity of future events of  
313 long-range transported aerosols.

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