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## Detection of aerosols in Antarctica from long-range transport of the 2009 Australian wildfires

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► **To cite this version:**

Julien Jumelet, A. R. Klekociuk, Alexander S. P., Slimane Bekki, Alain Hauchecorne, et al.. Detection of aerosols in Antarctica from long-range transport of the 2009 Australian wildfires. *Journal of Geophysical Research: Atmospheres*, 2020, 125 (23), pp.e2020JD032542. 10.1029/2020JD032542 . insu-02967511

**HAL Id: insu-02967511**

**<https://insu.hal.science/insu-02967511>**

Submitted on 7 Dec 2020

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1 **DETECTION OF AEROSOLS IN ANTARCTICA FROM LONG-RANGE**  
2 **TRANSPORT OF THE 2009 AUSTRALIAN WILDFIRES**

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5

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12 **Keypoints:**

- 13 1. First lidar identification of smoke aerosols originating from the 2009 wildfires above the  
14 French Antarctic Dumont d’Urville station
- 15 2. A dedicated simple microphysical and transport model uses CALIOP data as constraints to  
16 successfully model the aerosol overpass above Antarctica
- 17 3. Positive ozone anomaly is reported using lidar measurements

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22 **Plain Language Summary:**

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24 A series of bushfires ignited in the Australian state of Victoria on February 7th 2009 and ended  
25 up being the most devastating fire hazard in Australia before the recent 2019/2020 fires. Active remote  
26 sensing monitoring instruments are deployed on the French Antarctic station Dumont d’Urville. For  
27 the first time, the station recorded presence of aerosols having originated from this biomass burning  
28 event at stratospheric altitudes using the atmospheric laser sounding technique (lidar). We combine  
29 model calculations to space-borne and ground-based measurements to track the long-range transport  
30 of a small filament of the aerosol plume down to the Antarctic station to highlight the possible global  
31 impact of such events.

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## Abstract

We analyze the long-range transport to high latitudes of a smoke particle filament originating from the extra-tropics plume after the Australian wildfires colloquially known as ‘Black Saturday’ on February 7<sup>th</sup> 2009 and report the first Antarctic stratospheric lidar characterization of such aerosols. Using a high-resolution transport/microphysical model, we show that the monitoring cloud/aerosol lidar instrument operating at the French Antarctic station Dumont d’Urville (DDU-66°S-140°E) recorded a signature of those aerosols. The 532 nm scattering ratio of this filament is comparable to typical moderate stratospheric volcanic plume, with values between 1.4 and 1.6 on the 1<sup>st</sup> and 3<sup>rd</sup> days of March above DDU station at around the 14 and 16 km altitude respectively.

A dedicated model is described and its ability to track down fine optical signatures is validated against Antarctic lidar elastic aerosol and DIAL ozone measurements. Using one month of tropical CALIOP data to support a relatively simple microphysical scheme, we report modeled aerosol presence above DDU station after advection of the aerosol size distribution. In situ measurements also report associated positive ozone anomaly.

This case study provides evidence that biomass burning events injecting significant amounts of material up to stratospheric altitudes can be transported towards high latitudes. We highlight a potential imprint of smoke particles on the Antarctic atmosphere over larger time scales. Any underestimation of the global impact of such deep particle transport will lead to uncertainties in modeling the associated chemical or radiative effects, especially in polar regions where specific microphysical and chemical processes take place.

## 1. Introduction

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Aerosols are known to cause significant effects on the radiative balance of the Earth, both directly, through scattering and absorption of short-wave and long-wave radiation [Charlson et al., 1992], and indirectly, by acting as condensation nuclei [Penner et al., 2001]. As a matter of fact, long-term observations of stratospheric aerosols are needed for understanding the global atmospheric temperature and ozone layer variability [Thomasson and Peter 2006]. The role of particles, being either sulfated background/volcanic aerosols or carbon-based smokes as cloud condensation nuclei, is especially important in Antarctica. Recent studies also tend to demonstrate that polar aerosols can be characterized by large variations in their optical, physical and chemical composition parameters, causing a wide variety of particulate radiative properties throughout the year [Wang et al., 2005; Tomasi et al., 2007]. Besides, Polar Stratospheric Clouds (PSCs) grow from stratospheric sulfate aerosols under given temperature thresholds and have a crucial impact on stratospheric ozone depletion [Adriani et al., 2004; Spinhirne et al., 2005; WMO, 2018], and on radiative balance, which in turn modulate the local circulation [Liou, 1986; Ramanathan et al., 1989; Lachlan-Cope, 2010]. Polar clouds are also involved in the equilibrium of the largest freshwater reservoir in the world [Arthern et al., 2006].

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Because of the low aerosol number concentration, their direct effect was first thought to be negligible in Antarctic regions [Bodhaine, 1995]. But, above snow- or ice-covered areas, multiple scattering between these particles, cloud surface, and ground surface may still lead to some degree of warming [Randles et al., 2004; Liu et al., 2013]. Aside from the background sulfate aerosols, smoke particles also act as nucleation sites, favoring the growth of sulphuric acid droplets. Smoke particles also have a greater absorption/heating potential than sulfate particles [Burton et al. 2012, Weller et al., 2013]. Therefore, the global effect of a given stratospheric aerosol load has yet to be precisely estimated as it is as highly dependent on latitude as it is on chemical composition.

Worldwide information on stratospheric aerosols is now available from remote sensing instruments, especially with the numerous space missions [SPARC, 2006]. Nevertheless, the Antarctic particle budget remains hard to monitor due to an evident lack of instrumental capabilities beside the sparse ground stations which are mainly located at the edge of the continent. Pollution originating from trans-continental transport is increasing at high latitudes [Stohl and Sodemann, 2010]. Bullard et al. [2016] highlighted the significant impact of dust event and processes at high latitudes. Antarctica is more isolated from human activities than the Arctic and this geographical situation results in a lower density of aerosol particles relative to other regions [Ito, 1986].

Exchanges between troposphere and stratosphere have important impacts on atmospheric chemistry by mixing air masses containing potent greenhouse gases like ozone and water vapor [Gauss et al., 2003; Forster et al., 2007] and by air masses from lower latitudes entering the Antarctic circulation. Particle sedimentation and aerosol deposition then cause ice cores to act as climate proxies keeping track of past aerosol concentration levels [Wolff and Peel, 1985; McConnell et al., 2007]. Ice core samplings may also contain information on the frequency and intensity of past volcanic eruptions [Sigl et al., 2014]. Around 1.5% of stratospheric air reaches the surface after 10 days which is around the average age of air mass at the ground level in spring [Stohl and Sodemann, 2010]. However, the extent, scales and impact (either volcanic or biomass burning) occurring at lower latitudes on the Antarctic atmosphere through deep transport is still unclear. The particular processes taking place in Antarctica make the assessment of this impact a critical point in the interplaying interactions between ozone and climate [WMO, 2018].

This case study reports evidence of another source of stratospheric Antarctic aerosols related to biomass burning. Biomass burning may spread across large areas and significantly increase temperature and generate some explosive troposphere to stratosphere transport favored by intense forest fire. Convective processes associated with fire activity are referred to as pyroconvection and under specific heat and moisture conditions, fire-generated aerosol release can lead to thunderstorm-like convective cloud known as PyroCumulonimbus (PyroCb). This feature has been reported and investigated, with many events spanning from 1950 up to now [Livesey et al., 2004; Waibel et al., 1999; Fromm et al., 2000, 2005 ; Siebert et al., 2000 ; Fromm and Servranckx, 2003 ; Jost et al., 2004 ; Tupper et al., 2005 ; Fromm et al.; 2006, 2008 ; Peterson et al., 2015; Kablick et al., 2018 ]. One of the interesting features of the PyroCb events is that the microphysics and dynamics of associated plumes are most often very close to characteristics observed in the case of volcanic eruption [Peterson et al.,

118 2018]. Modeling studies have shown that tropospheric plumes above thunderstorms are very likely to  
119 enter the lower stratosphere, from which they may reach latitudes far removed from their emission  
120 location [Wang, 2003]. In the case of an underestimated frequency of such events, this process could  
121 be considered a new alternative to efficiently redistribute gases throughout the Upper Troposphere /  
122 Lower Stratosphere range (UTLS). While it is an efficient source process to inject particles up into the  
123 stratosphere, further studies are still required to better identify the convective storms responsible for  
124 this injection, with both observations and modeling required [Fromm et al., 2005]. In terms of optical  
125 measurement analysis, this also questions any misleading attribution of such carbonated aerosol  
126 signatures to volcanic activity. Discrimination of aerosols as of either volcanic or biomass burning origin  
127 requires microphysical information that can be provided by highly sensitive measurement capabilities  
128 such as those of lidar instruments.

129  
130 The transport of material over long distances, and especially smoke, is not often directly  
131 observed in the Antarctic UTLS. A few individual events have been documented, such as the transport  
132 of dust from Patagonia [Gassó et al., 2010] or smoke from biomass burning in South America  
133 [Evangelista et al., 2007; Pereira et al., 2006] to the tip of the Antarctic Peninsula. Fiebig et al. [2009]  
134 revealed transport of biomass burning aerosol from South America to an inland site on Queen Maud  
135 Land. Statistical studies of transport down to Antarctica have also been performed using trajectory  
136 calculations for single measurement stations or ice core drilling sites [Kottmeier and Fay, 1998]. Weller  
137 et al. [2013] investigated seasonal variations of black-carbon particles and albedo assessment with a  
138 maximum variability in spring time from decadal time series using absorption photometers. However,  
139 microphysical modeling and transport of particles from the lower latitudes down to Antarctica remains  
140 difficult due to scale issues within the models: large scale is needed for longer runs and small scale  
141 resolution needed to resolve the numerous fine filaments. Models have often been used in conjunction  
142 with ground-based measurements to attribute pollution in Antarctica to its sources [Stohl and  
143 Sodemann, 2010]. As for ground-based detection, the lidar instrument still remains one of the best  
144 suited to monitor and detect small changes in particle load in the UTLS, with a vertical resolution often  
145 under 100m and very high sampling frequency.

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147 This paper reports observations along with microphysical and transport simulation of a specific  
148 smoke filament originating from the particle plume which was largely confined within the tropics one  
149 month after the Australian bushfires. The overall complex structure of the different plume layers has  
150 been used as an additional constraint in our model both upon initialization and during the simulation  
151 we present here. We describe a methodology based on a combination of local and global lidar  
152 measurements along with a simple coupling of optical and transport modeling. We aim at taking  
153 advantage of local measurements to efficiently report on similar fine events (or those involving long  
154 range transport of volcanic aerosols). We also highlight the importance in having ground reference  
155 monitoring measurement stations to complement and validate global measurements, model results  
156 or even to some extent microphysical schemes through case studies like this one. Section 2 provides  
157 additional information on the event. Observations and numerical tools are described in section 3.  
158 Section 4 details the transport method and simulation setup. Section 5 discusses the model results  
159 against observations before drawing conclusions in section 6.

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## 161        **2. The 2009 Australian Bushfires**

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### 163                    **2.1. Event Description**

164                In the Southern Hemisphere, biomass burning activity is minimal in February. The major fire  
165 sources of Indonesia, Africa and South America become significant later in the year [Edwards et al.,  
166 2006]. Particle transport from these areas has a strong seasonality with maxima in winter for Southern  
167 Africa and spring from South America and Australia [Stohl and Sodemann, 2010]. A series of bushfires  
168 ignited in the Australian state of Victoria on February 7<sup>th</sup> 2009 and ended up being the most  
169 devastating fire hazard in Australia up to the recent 2019/2020 event and are now referred to as the  
170 ‘Black Saturday’ bushfires. Over 4500 km<sup>2</sup> of land burned to ashes, and more than 170 fatalities were  
171 recorded with major fire spots identified over 2 weeks from the Black Saturday [BOM, 2009]. The  
172 intensity of the Victorian bushfires was such that pyroconvection uplifted gas species along with  
173 particles up to stratospheric altitudes. Such typical transport of gas and particles has been reported by  
174 Wang [2003], Fromm et al. [2005] and Field et al. [2016]. On ‘Black Saturday’, the combination of typical  
175 meteorological synoptic features with lack of precipitation and localized convective processes rose the  
176 fire danger level to extreme and several PyroCb were identified [Reeder et al., 2015; Tosca et al., 2015;  
177 Field et al., 2016]. Recent investigations of the responses between the fires and the atmosphere lead  
178 to a better understanding of the pyroconvection processes and especially the risk of new fire ignitions  
179 using pyrogenic lightning. Dowdy et al. [2017] gathered evidence of pyroconvection in the Black  
180 Saturday events using a number of distinct electrified PyroCb clusters, therefore retro-validating the  
181 studies on the deep imprint of these plumes on the Southern Hemisphere atmosphere during several  
182 months [Siddaway and Peletina, 2011; Pumphrey et al., 2011; Glatthor et al., 2013].

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184                The main smoke plume was first observed at stratospheric altitudes the day after the outbreak  
185 and encircled the globe approximately 6 weeks after being first detected [Pumphrey et al., 2011].  
186 Similar to typical volcanic behaviour, smoke aerosols remained in the extratropical channel. From mid-  
187 February to June, it was mostly located between 18-22 km altitudes (see Section 5). Space-borne  
188 observations acquired onboard the CALIPSO satellite reported both the main part of the plume within  
189 tropical latitudes and various filaments reaching higher latitudes but backscatter values after 2 or 3  
190 months often reached the lower detection limit of the lidar instrument. Siddaway and Peletina [2011]  
191 used OSIRIS observations to report a predominant westward direction in the smoke plume transport  
192 and characterized advections from around 19 to 22km up before the end of April, and background  
193 radiance levels on OSIRIS observations by mid-June 2009.

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## 195        **3. Observations**

### 196                    **3.1. Ground-based lidar**

197                Lidar observations have been carried out at the French station Dumont d’Urville [DDU] (66°S -  
198 140°E), since April 1989 within the framework of a French (LATMOS/IPSL) Italian (ISAC/CNR)  
199 cooperation. The station is a primary site of the Network for Detection of Atmospheric Composition  
200 Changes (NDACC). Except for year 2000 and 2005, aerosols and PSCs measurements have been  
201 routinely carried out at DDU every year since 1989, from roughly mid-March to late September

202 [David et al., 1998, 2005] providing the only continuous aerosol lidar time series over 30 years in  
203 Antarctica. The DDU Rayleigh/Mie lidar was originally designed to operate in two exclusive modes, one  
204 for ozone and one for aerosol/PSC and temperature measurements. The ozone mode includes light  
205 emission and reception at wavelengths of 308 and 355 nm and additional reception at 332 and 387 nm.  
206 The aerosol mode includes emission and reception at wavelengths of 532 and 1064 nm. Lidar  
207 measurements are performed at DDU on a yearly winter campaign basis; the main focus being on polar  
208 stratospheric clouds monitoring during the winter and early spring months. For calibration purposes  
209 and additional aerosol records, a lighter measurement calendar also plans lidar acquisitions from early  
210 March, when the sky gets dark enough for the lidar to safely operate for extended measuring sessions.  
211 Within the operational frame of the NDACC, some alerts regarding specific events like volcanic  
212 eruption and specific air mass intrusions may trigger additional acquisitions.

213  
214 A complete description of both the instrumental design and inversion procedure is featured in  
215 [David et al., 2012]. The Nd:YAG laser source emits 500mJ per pulse at 1064nm at 10 Hz frequency,  
216 doubled in frequency to 532 nm. A collocated Excimer laser emits 55 mJ per pulse at 308 nm at 80 Hz  
217 frequency. Backscattered signal is collected by an 80 cm telescope. Aerosol vertical density profile is  
218 defined as the backscattering ratio or Scattering Ratio (hereafter called SR) expressed as the ratio of  
219 extra scattering of aerosols to the molecular scattering of air at the same altitude. After a  
220 preprocessing phase removing potential saturation effects and background noise level (low at this time  
221 of the year in Antarctica), lidar signals are inverted using the Klett-Fernald formalism [Klett 1981,1985;  
222 Fernald, 1984], to derive individual SR profiles. The zero-reference altitude is set between 28 and  
223 32 km depending on signal-to-noise ratio. The extinction-to-backscatter ratio is set to 50 sr which is a  
224 commonly assumed value for small aerosol loading and even periods of moderate volcanic eruptions  
225 [Ridley et al., 2014]. Cumulative uncertainties of the backscatter measurements induced by random  
226 detection processes, possible presence of aerosol at the reference altitude and the error in lidar ratio  
227 value do not exceed 10% as reported by Chazette et al., [1995]. Another major source of uncertainty  
228 is the molecular number density used in Rayleigh calculations derived from atmospheric pressure and  
229 temperature profiles. The lidar inversion is particularly sensitive to the molecular density and the clear-  
230 air reference altitude. Collocated radiosondes are used to estimate reference atmospheric density and  
231 interpolation and scaling to National Centers for Numerical Prediction (NCEP) daily meteorological  
232 data is done at balloon burst altitude.

233  
234 Ozone profiles are retrieved using the Differential Absorption Lidar (DIAL) technique. The  
235 ozone number density can be derived from two signals, one being strongly absorbed by ozone  
236 (308 nm), the other (355 nm) being only weakly absorbed. The Nd:YAG frequency is tripled to 355 nm  
237 to provide the reference signal whereas the Excimer laser directly emits at the 308 nm. Pre-processing  
238 of the lidar signal in ozone mode is similar to the aerosol mode. Ozone concentration is retrieved  
239 without external assumptions by comparing the number of laser shots between the absorbed and  
240 reference wavelengths [David et al., 2012].

241  
242 To complement the simulations presented in this paper, we also include a lidar profile acquired  
243 at the Australian Davis station (-68.6°S, 78°E). The 532nm SR is obtained with the Klett-Fernald  
244 formalism after normalization at 30-35km altitude. The molecular signal is derived from density profile

245 inferred from measurements by the AIRS instrument onboard the Aqua satellite. As for DDU, the lidar  
246 ratio of the background aerosol is assumed to be 50 sr. For observations used here, a mechanical  
247 chopper reduced signal levels below approximately 11 km altitude. Further details of the system and  
248 analysis procedure are provided by Alexander et al. [2011]. Observations were made with the Davis  
249 lidar system throughout the autumn during quiescent tropospheric weather, however only data  
250 collected on February 26<sup>th</sup> are relevant for comparisons with the simulations discussed below.

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### 3.2. Satellite aerosol sounders

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The Atmospheric Chemistry Experiment (ACE) was launched in August 2003 onboard the Canadian scientific satellite SCISAT-I. The high-resolution spectrometer measures during sunrise and sunset infrared absorption to provide vertical profiles of atmospheric compounds (gases and particles). Aerosols and clouds are monitored using the solar radiation extinction measured by two filtered imagers at 527 (VIS) and 1020 nm (NIR) [Dodion et al., 2007]. The satellite operates in a circular orbit at an altitude of 650 km. Aerosol extinction profiles are obtained by subtracting the gas extinction contribution (air, O<sub>3</sub>, NO<sub>2</sub>) derived from the Fourier Transform spectrometer from the total atmospheric extinction profiles. On average, the NIR ACE imager gives results that are 20% higher than SAGE II instrument [Gilbert et al., 2007].

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## 4. Numerical simulations

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### 4.1 The aerosol microphysical and transport model

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The Modèle Isentropique de transport Mésoéchelle de l'Ozone Stratosphérique par Advection (MIMOSA) model [Hauchecorne et al., 2001] is originally a Potential Vorticity (PV) advection model running on isentropic surfaces (surfaces of constant potential temperature). The advection scheme is semi-Lagrangian with a time step of 1 hour. Regridding onto the original orthonormal grid is performed every 6 hours. The model resolution is 0.5°x0.5°. The advection is driven by European Centre for Medium Range Weather Forecast (ECMWF) meteorological analyses at a resolution of 0.5°x0.5°. The accuracy and mass conservation of the model have been evaluated by Hauchecorne et al. [2001] and



288 validated against airborne lidar ozone measurements using the strong correlation between PV and  
289 ozone, a quasi-conserved chemical tracer on a week timescale within most of the lower stratosphere  
290 [Heese et al., 2001].

291  
292 A sectional aerosol microphysical module has been coupled to MIMOSA to simulate the  
293 formation, transport and microphysical evolution of stratospheric aerosol particles [Jumelet et al.,  
294 2009]. The size distribution is discretized into a number of size bins using a geometrically increasing  
295 volume scale. The number of size bins is 50 with the bin particle radius ranging from 0.01 to 5  $\mu\text{m}$ . The  
296 model prognostic variables are the number density of each size bin and chemical composition. The  
297 module also includes an optical routine that calculates theoretical properties such as volume  
298 backscatter and/or extinction coefficients at given wavelengths (in the visible and near-infrared  
299 domains e.g. the 532 nm aerosol wavelength) from the model-simulated particle size distribution and  
300 the composition-dependent refractive index.

301  
302 We consider here two different aerosol types within the model, one being the background  
303 sulfate stratospheric aerosols and the other one being the injected smoke particles. Since background  
304 sulfate aerosols are not relevant to our long-range transport case study, only the results for smoke  
305 particles are presented and discussed here. In order to simulate smoke particles, the original aerosol  
306 scheme is extended. Smoke particles can be complex objects, i.e. a homogeneous mixing of organic  
307 and inorganic aerosol components with black carbon coating in some more or less complex core/shell  
308 structure often modelled using fractal aggregates [Smith and Grainger, 2014]. However, our approach  
309 is simple because the objective is to calculate the aerosol optical parameters (properties integrated  
310 over the entire distribution) instead of the detailed behaviour of the size distribution and the 532nm  
311 SR will be driven by observations. Dählkotter et al. [2014] have shown that the optical properties of  
312 aged forest fire smokes (having experienced intercontinental transport) can be estimated with Mie  
313 Theory without any major uncertainty. The smoke particles are treated here as a mixture of  
314 sulfate/black carbon with overall real refractive index at 1.85 [Smith and Grainger, 2014, Mischenko et  
315 al., 2014]. The smoke SR is calculated using the Mie scattering code with aspherical aspect ratio at 1.50  
316 [Scarnato et al., 2015]. There are still computational uncertainties in the optical calculations due to the  
317 limited number of bins covering the size range that drive the overall uncertainty estimated around 20%  
318 on the backscatter coefficient.

## 319 320 **4.2 Initialisation & CALIOP SR insertion into the model**

321  
322 The initialization of aerosol size distributions is based on measurements. There was no  
323 significant volcanic influence on aerosol loading at the time of the Australian Bushfires in the southern  
324 hemisphere. For this reason, the sulfuric acid aerosol size distribution was initialized over the entire  
325 model domain with a typical background (non-volcanic) lognormal distribution:  $N_0=10\text{ cm}^{-3}$ ,  
326  $r_m=0.07\text{ }\mu\text{m}$  and  $\sigma=1.8$  [Pinnick et al., 1976; Larsen et al., 2000]. To our best knowledge, no information  
327 on the Australian smoke particle size distribution are available. The smoke size distribution is therefore  
328 initialized with a lognormal distribution based on data collected in the northern hemisphere on a  
329 comparable PyroCb particle plume after a similar event of smoke long-range transport ( $N=50\text{ cm}^{-3}$ ,

330  $r_m=0.12 \mu\text{m}$  and  $\sigma=1.3$ ) [Dahlkötter et al., 2014]. This size distribution can also be typical of the fine  
331 mode of volcanic aerosol plumes.

332

333 The initialization of the stratospheric aerosol spatial distribution is based on the insertion of  
334 CALIOP SR measurements in the model. The insertion is straightforward as compared to other  
335 approaches where CALIOP backscatter products have already been successfully assimilated in several  
336 studies using full assimilation schemes [Sekiyama et al., 2010; Campbell et al., 2010]. From a data  
337 assimilation point of view, our insertion scheme is equivalent to a sequential assimilation assuming  
338 model errors to be much greater than observational errors and using our optical model as  
339 observational operator. Our scheme remains local because the insertion of one single observation only  
340 impacts one model grid cell, meaning the length-scale of the spatial correlation is taken to be less than  
341 a size of a grid cell, i.e. less than  $0.5^\circ$ .

342

343 The insertion procedure is the following: one variable of the aerosol size distribution  
344 parameter (the so-called control variable) at each grid cell is modified iteratively in order for the model-  
345 calculated SR to match the CALIOP-observed SR. The choice of the control variable depends on the  
346 value of the observation, controlling the aerosol type. When CALIOP SR is smaller than the background  
347 value (taken as 1.025, from simple Mie Calculations on  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  droplets), one can assume that the  
348 measurements correspond to stratospheric sulfuric acid aerosols and the control variable is the total  
349 aerosol concentration  $N$ . The median radius and standard deviation remain unchanged ( $r_m=0.12 \mu\text{m}$   
350 and  $\sigma=1.3$ ). Note that, in that case, the optical module uses the aerosol optical index of sulfuric  
351 acid/water solution in equilibrium with the gas phase [Jumelet et al., 2009]. When CALIOP SR is greater  
352 than 1.025, the measurement is taken as indicative of the presence of smoke particles and the control  
353 variable is the mode radius.  $N$  and standard deviation remain unchanged. In that case, the optical  
354 module uses the smoke refractive index. This choice of control variable for smoke type is based on the  
355 fact that there is little information on smoke particle size and, within the range of smoke size  
356 distribution parameters, model-calculated SR is assumed to be much more sensitive to the mode  
357 radius than to the total number concentration, the latter variable displaying a linear behavior regarding  
358 the aerosol backscatter coefficient.

359

360 The first 2 weeks of the simulation in January, before the fire outbreak, define the spin-up  
361 phase. In this phase, continuous high-frequency CALIOP SR insertion on the whole grid slowly shifts  
362 modelled stratospheric aerosol SR towards CALIOP measurements. After the spin-up, the average  
363 aerosol levels do not change much beyond sedimentation, and the model reaches some degree of  
364 equilibrium regarding the forcing of satellite observations.

365

366

### 4.3 Model set-up and simulations

367

368 A key aim of the present study is to test the model ability to forecast the position of plumes  
369 of either volcanic or biomass origin. For this reason, simulating these long-range transport events use  
370 aerosols as tracers. After the initialization/spin-up phase, CALIOP SR data are inserted in the model  
371 during the run, but only outside a large free-running model domain. The CALIOP insertion carries on in  
372 order to maintain realistic boundary conditions on this free running domain. The sensitivity of the

373 model results has been tested by varying the latitude of the free running domain boundary beyond  
374 which the aerosol particles are freely advected (no CALIOP SR insertion). Stable results were obtained  
375 when the CALIOP insertion is confined to the region equatorward of 50°S latitude. Of course, the  
376 CALIOP insertion domain covers the Australian source region. We keep the latitude of the limit  
377 between the free running insertion zones relatively low to highlight the efficiency of the transport  
378 model.

379

380 The model is integrated from January 1<sup>st</sup> to May 1<sup>st</sup>, with an advection step of 1 hour, and a  
381 regrided output is dumped every 6 hours. The model is run at several isentropic levels every 10 K over  
382 the 400-450 K potential temperature range on a southern hemispheric grid. It is forced with ECMWF  
383 temperature and pressure fields at a 0.5°x0.5° resolution. The spin-up phase covers the first 2 weeks  
384 of January. After that, CALIOP SR data are only inserted outside the large free-running model domain,  
385 equatorwards of 50°S. The signature of the Australian fire plumes at stratospheric altitudes is clearly  
386 detected after 2 days after the outburst on February 7<sup>th</sup> above the 410 K level.

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## 388 **5. Results**

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### 390 **5.1 CALIOP and lidar observations**

391

392 Two weeks of 532 nm CALIOP SR averaged data are presented on Figure 1. The figure shows  
393 the evolution of smoke plume distribution between February 1<sup>st</sup> and 16<sup>th</sup>. Recall that the fires started  
394 on February 7<sup>th</sup>. To avoid any false detection, only SR above 1.1 (clearly indicative in this study of smoke  
395 particles) are considered. The SR averaged over the 15-25 km altitude range show the plume in the  
396 South Pacific (Figure 1a). The SR vertical distribution as a function of longitude (data averaged over the  
397 25°S-50°S latitudinal band) also confirm the presence of smoke particles in the 15-22 km altitude range  
398 (cf. Figure 1b). Figure 1c shows the zonal mean SR distribution (data averaged along latitude circles)  
399 with isentropic levels superimposed; smoke particles remain mostly confined between the 380 K and  
400 450 K isentropic levels during the first 2 weeks of February, although an analysis at higher time  
401 resolution revealed that few smoke particles can be detected up to the 500 K level a few days after the  
402 fire outbreak (not shown).

403

404 The plume is identified above DDU station during a short time span in early March, with  
405 unambiguous signatures on the 1<sup>st</sup> and 3<sup>rd</sup>. At that time, no direct comparison with CALIOP is available.  
406 CALIOP data and results of model simulations (described in the next section) indicate more overpasses  
407 of the smoke plume above DDU, but no lidar measurements were available due to bad local weather  
408 conditions. Both vertical ground-based SR profiles obtained on the 1<sup>st</sup> (Figure 2a) and 3<sup>rd</sup> of March  
409 (Figure 2b) reveal clear signals of smoke particles at 13-14 and 16-17 km respectively. SR peaks at 410 K  
410 and 450 K on the March 1<sup>st</sup> (Figure 2a) and March 3<sup>rd</sup> respectively. Note that the lidar data are averaged  
411 over ~160 min on March 1<sup>st</sup> and ~210 min on March 3<sup>rd</sup>. SR peaks above 1.4 in both cases which is  
412 higher than SR signals of minor volcanic eruptions [Vernier et al., 2011]. An additional weaker layer  
413 can also be seen just above 15km (Figure 2a). It is worth pointing out that measurements at this time  
414 of the year (at the end of continuous Antarctic daylight) were performed for calibration purposes in  
415 March, related to nighttime sky conditions.

416

## 417 **5.2 Model simulations versus DDU lidar detection**

418

419 Figure 3a shows the plume filament on February 26<sup>th</sup>, a few days prior detection above DDU on  
420 March 3<sup>rd</sup>, near the location of the Davis station. Figure 3b shows the associated lidar profile on the  
421 same day, with a broad but weak peak of about 1.05 around 20km. This small SR value may be  
422 indicative of background aerosols, especially as 2009 is a volcanic quiescent time period. Besides, lidar  
423 measurements have been integrated on several hours. Even if the plume position is at the Davis  
424 longitude, identification is not clear looking at the lidar profile, especially considering that for small SR  
425 values, the molecular scattering derived from the pressure and temperature profiles is a sensitive  
426 parameter. Still, aerosol loading is present and there is a strong agreement between Figure 3a and 3b,  
427 in that the simulated plume does not reach Davis with scattering ratios greater than 1.1.

428

429 Figure 4 shows modeled SR fields between February 10<sup>th</sup> (Figure 4a) and March 10<sup>th</sup> (Figure 4i)  
430 on the 410 K level corresponding to the altitude of the ground-based SR peak on March 1<sup>st</sup>. The highest  
431 CALIOP SR values are detected during the first half of February, with values peaking slightly above 1.4  
432 and gradually decreasing below the 1.1 threshold during the first half of April (not shown). A weak  
433 plume spanning both sides of South America is visible in the modeled SR field in February. This is not  
434 related to the Australian fires and will not be discussed here. A week after the fire outbreak, a small  
435 patch of high SR values appears on the 10<sup>th</sup> of February east of Australia (Figure 4a). This patch,  
436 indicative of the smoke plume, carries on being transported eastward. At the same time, it is very  
437 strongly stretched by wind shear, forming a large filament extending over thousands of kilometers.  
438 Filamentary structures quickly form at the subtropical barriers. Between the 20<sup>th</sup> and 25<sup>th</sup> of February,  
439 the plume filament split into two main parts with the main part remaining in the tropics and a smaller  
440 filament being transported towards higher latitudes.

441

442 Around the time of the fires and during the ascension between Feb 8<sup>th</sup> and 16<sup>th</sup>, part of the  
443 CALIOP data may have been misflagged so the overall aerosol load near the tropics may not be  
444 accurately depicted. This issue is still out of focus as we aim at identifying the small filament  
445 transported at high latitudes and clearly separated from the main bulk (visible from Figure 4d). Besides,  
446 the consistency of SR values (excluding high cloud scattering) has been checked prior to integration  
447 within the model.

448

449 Overall, several smoke plume filaments were observed over a wide altitude range (13-22km)  
450 with CALIOP till the end of May. Even though the bulk of the smoke plume did not cross the subtropical  
451 barrier, smaller filaments occurred throughout the extratropics due to efficient quasi-horizontal  
452 transport (along isentropic surfaces) in the lower stratosphere. After one month of long-range  
453 transport, the plume is detected above DDU on the 1<sup>st</sup> of March during the time of the measurements,  
454 between 13:33 and 16:12 UT (see Figure 2). The model indicates the passing of a thin filament just  
455 above the station on the same day (see Figure 4f); model outputs are only available every 6 hours. In  
456 order to investigate the passing of the filaments above DDU, Figure 5 displays a close-up of the  
457 simulation at 410 K and 450 K, respectively on March 1<sup>st</sup> and 3<sup>rd</sup> when the DDU lidar clearly detected

458 the plume. There is a reasonably good agreement in terms of timing. The ground-based measurements  
459 in Figure 1a and 1b identified 2 layers peaking at 410 K and 450 K. The lower layer is detected on March  
460 1<sup>st</sup>, the higher layer is detected on March 3<sup>rd</sup>. This is consistent with the model simulation which shows  
461 a DDU overpass at 410 K level only on March 1<sup>st</sup> (Figure 5c) but not on March 3<sup>rd</sup> (Figure 5d); it is very  
462 close to the time of the afternoon lidar plume measurements whereas the model output is set at  
463 12:00 UT. The model simulation also appears to be in agreement with the DDU lidar at 450 K level with  
464 a DDU overpass only on March 3<sup>rd</sup> (Figure 5b) but not on March 1<sup>st</sup> (Figure 5a).  
465

466 As expected, the modeled SR values are in excellent agreement with CALIOP SR. However, the  
467 peak modeled SR is slightly higher than the corresponding maximum values of the DDU ground-based  
468 lidar. The instantaneous model outputs (every 6 hrs) show the maximum SR values within the smoke  
469 filament whereas the ground-based lidar data correspond to around a 3 hr time average of the passing  
470 filament as seen from a local atmospheric column above the station. Model fields show that there are  
471 sharp SR gradients within the plume. It is unlikely for the ground-based instrument to probe only within  
472 the most dense parts of the filament. Furthermore, space-borne and ground-based lidars have  
473 different acquisition fields of view and different data processing (calibration, inversion, spatio-  
474 temporal averaging), and small discrepancies cannot be ruled out. In view of these caveats, the  
475 agreement between the ground-based lidar measurements and the CALIOP-driven model simulation  
476 can be viewed as good. It indicates that the setup is able to accurately model long-range transport of  
477 the information provided by CALIOP measurements from 50°S (the edge of the free-running model  
478 domain) down to Antarctica. This also suggests that the transport scheme and the ERA-Interim winds  
479 driving the advection are accurate enough if the model is used as a forecasting tool. After the DDU  
480 overpass, during the first 10 days of March, the plume is stretched and breaks into smaller parts that  
481 are scattered throughout the extratropics and high latitudes.  
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### 484 **5.3 Model simulations versus ACE measurements**

485 ACE imager measurements of extinction ratio on March 4<sup>th</sup> were used as a comparison to the  
486 modeled SR field. Figure 6. displays the model SR with crosshairs at the location of the ACE  
487 measurements. Overall, SR values peak at slightly above 1.6 as forced by CALIOP measurements. The  
488 main bulk of the plume remains confined within tropical latitudes, even though the largest SR values  
489 are found in a filament crossing out of the tropics at the square location. Figure 7 displays ACE imager  
490 measurements of extinction ratio on the same March 4<sup>th</sup>, when the plume has been clearly identified,  
491 peaking respectively at 19 and 21 km. Figure 7b displays associated VIS and NIR imager extinction  
492 coefficients. In early March, the total AOD fell to around 0.002 and 0.006 for the NIR and VIS  
493 respectively whereas it was estimated around 0.03 in February. Once again, the SR values on Figure 6  
494 follow the shape of very fine dynamic structures that can be difficult to model without dedicated  
495 microphysics and resolution but a large amount of particles seems to be able to cross the tropics.  
496 Burton et al. [2012] provides an assessment of the 532 nm extinction to scattering coefficient (lidar  
497 ratio) for smoke particles of around 60-80 sr and associated IR/Visible color ratio of around 1.5-3. Using  
498 reference stratospheric values of temperature and pressure around 200-210K and 80-100 hPa  
499 respectively, we calculate a Rayleigh contribution to the scattering ratio of around  $1.7-1.9E-4 \text{ km}^{-1} \cdot \text{sr}^{-1}$   
500 <sup>1</sup> and using the 60-80 sr lidar ratio, we derive a 1.3-1.5 532 nm scattering ratio from the ACE

501 measurements of Figure 7b which is in good agreement with the modelled 1.4-1.6 values of Figure 6.  
502 This agreement between ACE measurements of Figure 6 and 7 suggests that present plume modeling  
503 is accurate for identifying particles at high latitudes.

504

#### 505 **5.4 Local impact of the smoke plume on stratospheric ozone**

506 The DDU lidar was also used for measurements of ozone vertical profiles up to 2012. Such  
507 measurements are available on the 3<sup>rd</sup> of March, but not on the 1<sup>st</sup> of March. Figure 8a presents the  
508 lidar ozone profile measured on the same day, a few hours after the measurement of the aerosol  
509 profile. The dotted black line indicates the 3-year (2006-2008) average lidar ozone profile for this same  
510 period of the year. This background averaged profile agrees with a typical zonal mean climatology  
511 [Bencherif et al., 2011] for the relevant latitudinal range [75°S-85°S] which is indicated by the dotted  
512 red line. The interesting feature is the presence of a 1 km-thick enhanced ozone layer at about the  
513 altitude of the smoke layer.

514

515 However, similar ozone enhancements have been reported in smoke layers in previous  
516 studies. Fromm et al. [2005] studied a PyroCb injection of smoke to the stratosphere in August 1998  
517 above Northwestern Canada using balloon profiles. They reported the first highly enhanced ozone  
518 concentration at the smoke altitude. These reports remain rare because simultaneous smoke and  
519 ozone measurements in the stratosphere are not common. It would be worthwhile to consider more  
520 cases to establish unequivocally a link between smoke and ozone enhancements in the stratosphere.  
521 It has been shown in tropospheric biomass burning plumes that ozone production can be expected  
522 and is observed [Pickering et al., 1996; Andreae et al., 2001] but at stratospheric altitudes, no clear  
523 process is identified to our knowledge to explain this positive correlation between ozone and smoke  
524 particles. Yu et al. [2019] used photochemistry modeling to assess the impact of smokes on O<sub>3</sub> on the  
525 Canadian 2017 fire event, only attributing negative anomalies observed by satellite to transport of air  
526 masses from the troposphere, with no in situ chemistry.

527

528 Figure 9 shows the ozone mixing ratio on the 395 K and 430 K isentropes from ERA-Interim. A  
529 filament of ozone-rich air from higher latitude appears to be lying close to DDU. It is possible that the  
530 ozone feature, which is mainly below and inside the smoke layer, could simply show the demarcation  
531 between air that has originated from further south (having higher ozone, and being below the smoke  
532 layer) from air that is more representative of the zonal average. Further studies combining  
533 measurements and chemistry-transport model on the air mass composition inside and outside the  
534 plume would be needed to state on this positive correlation because different mechanisms could lead  
535 to opposite ozone variations (like decrease from photolysis on water vapor and increase from  
536 CO concentrations). Besides, investigations using the observations like ACE or OSIRIS at high latitudes  
537 is difficult as the extinction coefficients associated to these aerosols are relatively small (compare with  
538 Figure 7).

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541 **6. Summary and concluding remarks**

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An event of stratospheric biomass burning aerosols was detected in the lower stratosphere on the coast of Antarctica using lidar measurements at the end of the Southern Hemisphere summer. Our investigations show that these aerosols originate from the 2009 Australian ‘Black Saturday’ smoke plume. From this detection, we develop a transport/microphysic modeling tool taking advantage of space-borne measurements to be a useful complement to the groundbased measurements in a region where instrumental capabilities are low as compared to lower latitudes. Due to this low instrumental coverage and fine optical signature of such events, we therefore highlight the underestimated occurrences of such polar detections in the many reported PyroCb events, and therefore link the PyroCb impacts to scientific questions more specific to high latitude regions.

Our global simulations of the microphysical/transport model indicated that following the February 7<sup>th</sup> Black Saturday event, particles reached the stratosphere within 2 days after the fire outbreak and crossed the tropical dynamical barriers down to the high latitudes in less than a week. Lidar measurements are routinely performed from the early days of March at the French Dumont d’Urville station, around one month after the fires in Australia. We identified a clear optical signature. An average scattering ratio slightly above 1.4 is identified at the 532nm wavelength, which is comparable to the optical response of moderate volcanic plume after several months of stratospheric transport.

A first step towards a more in-depth parametrization of the different particle types present in the UTLS has been developed and applied to this case study. The modeling approach allows for high resolution hemispheric transport of a smoke plume proxy to which optical properties are fit at the microphysical level to the optical properties measured by the CALIOP space-borne lidar. The dedicated MIMOSA- $\mu\phi$  model was run over a 5-month time period, as the overall global signature of the fire plume is reported up to June.

Without implementation of a full data assimilation scheme, this simulation is built on a 2-step procedure. A first spin-up step where the model grid is filled with particles, being essentially background sulfate aerosols with an initial carbonated smoke cluster, using the CALIOP 532nm SR as the variable controlling the evolution of the bin-resolved size distribution from its reference background parameters. This first step runs through the first month of the run time and leads to a realistic pattern of the global particle stratospheric load on the southern hemisphere. In the second step, the model runs on both a forced and free regime, considering the 50°S parallel as the latitude limit beyond which the advection is freely performed. After more than one month of long-range transport, our proxy of the smoke plume is found to be at a time and place globally consistent with the lidar measurements on both March the 1<sup>st</sup> and 3<sup>rd</sup>. This simulation highlights the opportunity for air masses coming from lower latitudes to deeply imprint the Antarctic stratosphere, as only some polar cloud types and similar transport of any strong volcanic plume would feature similar SR values. The quantitative assessment of the carbonated aerosol mass and smoke composition will need further microphysical investigations and many improvements on the model are necessary.

584 This work reports to our knowledge the first lidar identification of stratospheric smoke aerosols,  
585 the observed signature observed at the French Dumont d'Urville station originates from the 2009  
586 wildfire event. We highlight the benefits of an efficient yet simple model-instrumental coupling.  
587 Accurate plume characterization can be made from a small sample of measurements and relatively  
588 simple modeling tools. Thus, we see three different outlines in this study: first, the scientific and  
589 confirmed pathway to a global impact (yet to be estimated) of long-range troposphere to stratosphere  
590 transport through pyroconvection and deep stratospheric transport for smoke. Second, the need for  
591 better characterization of the microphysical properties of such smoke, as their trapping above the  
592 Antarctic area is likely to have an impact on the ground level once deposited on ice shelves, especially  
593 over longer time scales. Third, the needed effort towards a better coupling of models and observations  
594 at the global scale. This can be achieved through assimilation of diagnostic variables such as the  
595 scattering ratio, especially when considering the overall success of space-borne lidar instruments and  
596 the need for maintaining routine lidar operations in polar stations.

597  
598 In this study few compromises are made with the model grid size, resolution, and level of  
599 microphysical detail. This highlights the importance of dedicated data assimilation tools. Satellite  
600 measurements combined with global and regional models can be an efficient complementary data  
601 source to document transport of fine volcanic or biomass burning plumes, especially considering the  
602 2019/2020 Australian wildfire event, which strength and amplitude is already acknowledged to be  
603 much greater than the 2009 event.

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## 607 **Acknowledgments and data availability**

608  
609 Operations on the Dumont D'Urville station are supported by the French Polar Institute (Institut polaire français Paul-Emile  
610 Victor), science project 209. This work was also performed within the EECLAT project (CNES-INSU). The lidar instrument is  
611 part of the NDACC international network and data are publicly available online at the NDACC/NOAA data archive  
612 (<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/dumont/ames/>). The Davis lidar observations and data analysis were supported  
613 by Australian Antarctic Science projects 737, 3140 and 4292 and these data are publically available through the Australian  
614 Antarctic Data Centre (<http://aad.aad.gov.au>).

615  
616 The MIMOSA transport model outputs are available online at the french ESPRI data archive :  
617 <https://cds-espri.ipsl.upmc.fr/etherTypo/index.php?id=1663&L=1>.

618 Measurements from the ESA SCISAT-1/ACE instrument are available online at:  
619 <https://database.scisat.ca/level2/>

620 Measurements from the CALIPSO/CALIOP space-borne lidar are available online at:  
621 [https://www-calipso.larc.nasa.gov/tools/data\\_avail/](https://www-calipso.larc.nasa.gov/tools/data_avail/)

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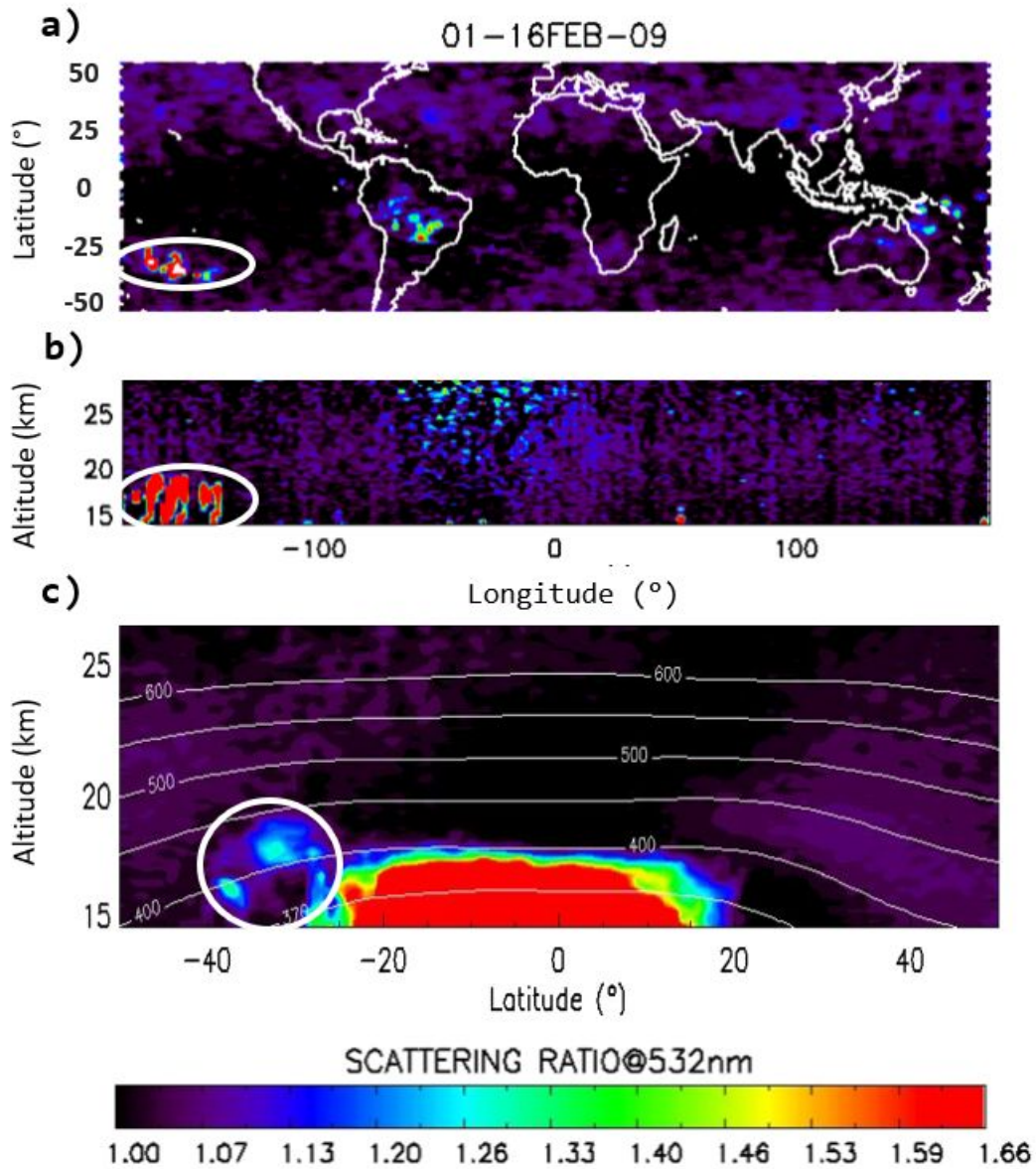
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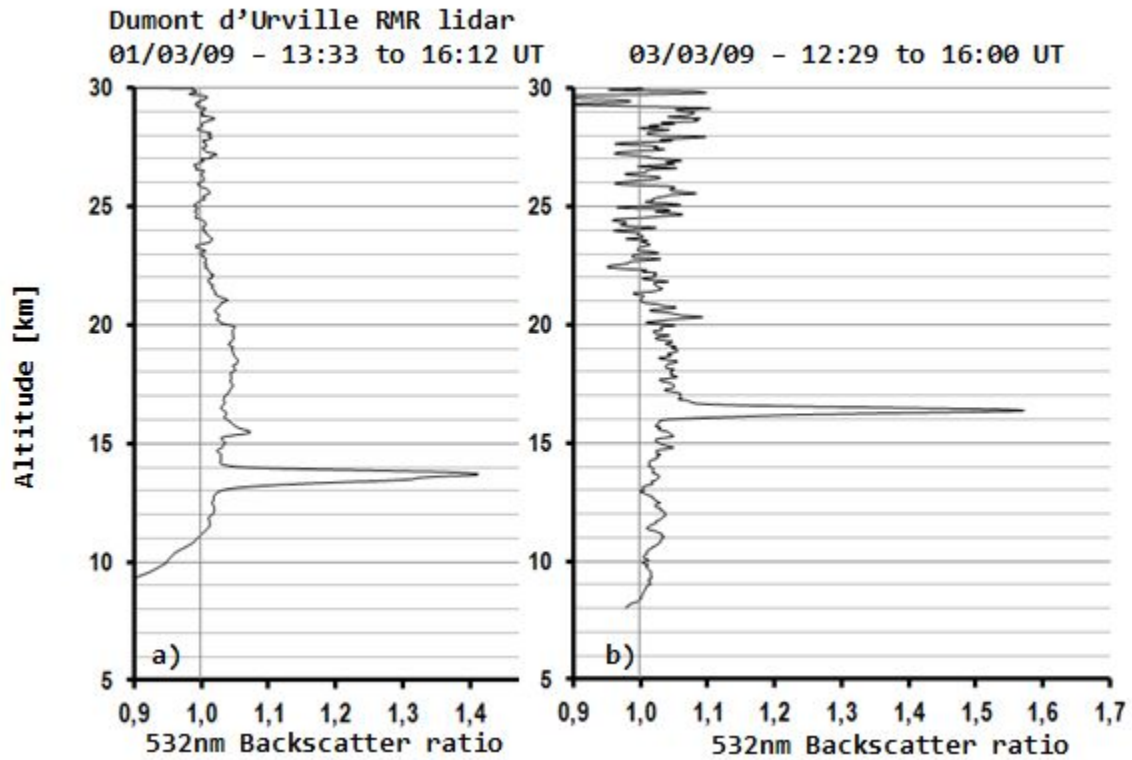
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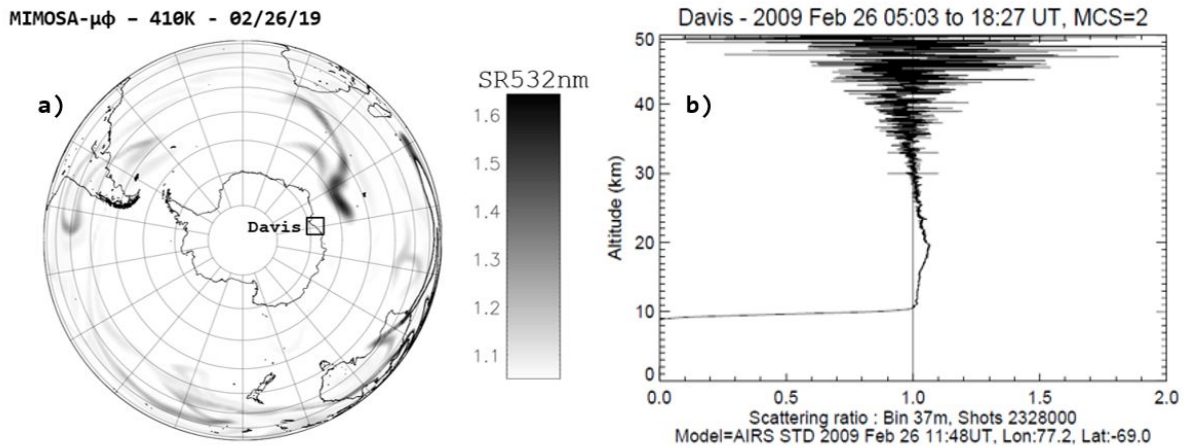
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836 **Fig. 1.** 532nm CALIOP Scattering Ratio in the first 16 days of February plotted as longitude vs  
 837 latitude, 15-20 km average (a), longitude vs altitude, averaged in the  $-25^{\circ}$  to  $-50^{\circ}$  latitudinal  
 838 band (b), and latitude vs altitude (c). The smoke plume cluster is encircled in white and is  
 839 clearly separated from the global tropospheric cloud signature. Contours of constant potential  
 840 temperature (K) are shown.



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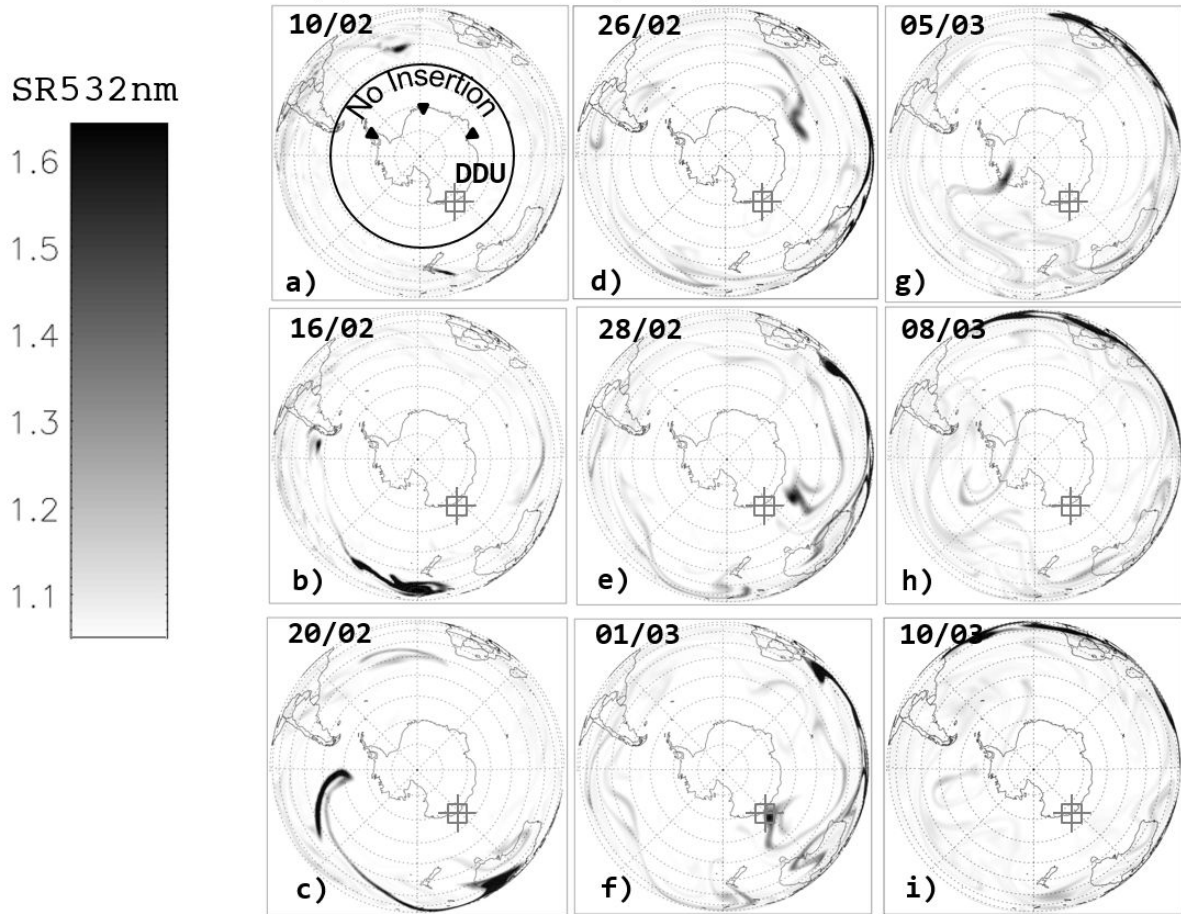
**Fig. 2.** Dumont d'Urville 532nm Scattering Ratio on March 1<sup>st</sup> (a), and 3<sup>rd</sup> (b) with smoke layers respectively visible at 13.5 km and 16.4 km.



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**Fig. 3.** (a) Model-simulated 532nm Scattering Ratio on February 26<sup>th</sup>. (b) 532nm lidar profile acquired at Davis Station.

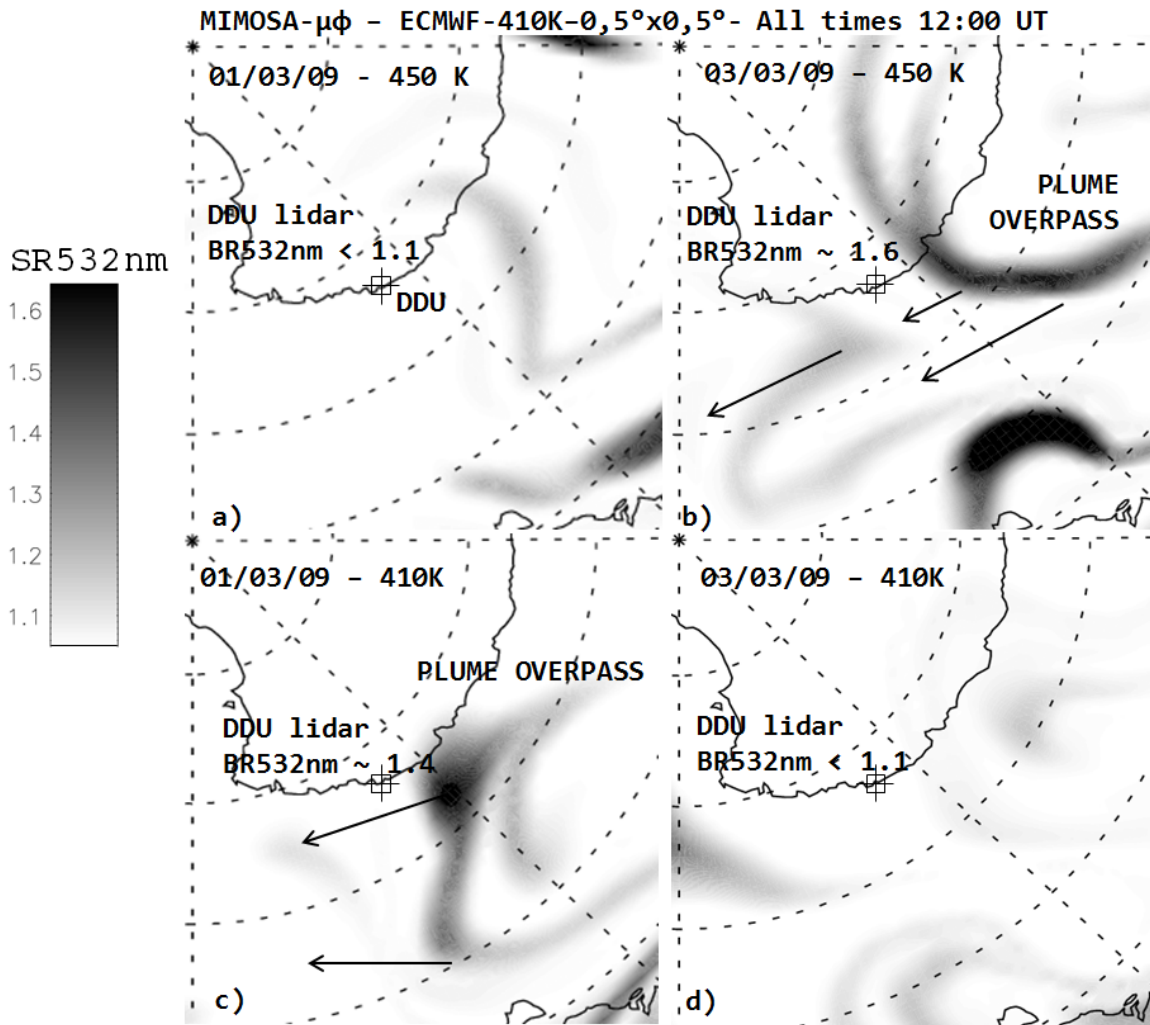
MIMOSA- $\mu\phi$  - ECMWF-410K- $0,5^\circ \times 0,5^\circ$  - All times 12:00 UT



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**Fig. 4.** 532nm MIMOSA- $\mu\phi$  SR calculations from February 10<sup>th</sup> to March 10<sup>th</sup>. Spatial resolution is  $0.5^\circ \times 0.5^\circ$  and the potential temperature level is 410 K. CALIOP SR is integrated only up to the 16<sup>th</sup> of February and in the  $(-50^\circ; 50^\circ)$  latitudinal range. The location of Dumont d'Urville station is indicated by the square.

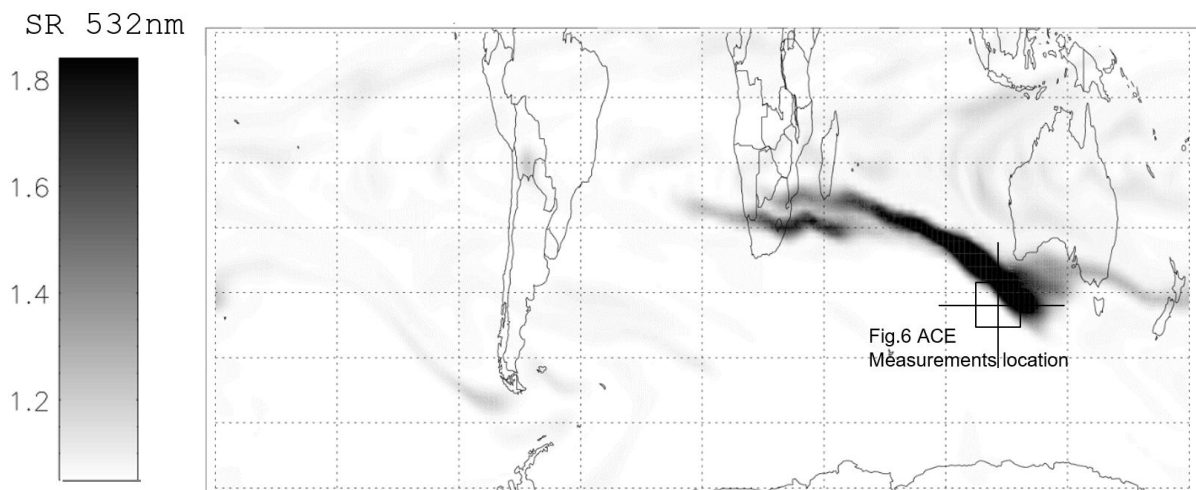




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**Fig. 5.** 532nm MIMOSA- $\mu\phi$  Scattering Ratio calculations and detailed outputs above the vicinity of DDU on March 1<sup>st</sup> at 450 K (a), 410 K (c) potential temperature levels and March 3<sup>rd</sup> at 450 K (b), 410 K (d) potential temperature levels. Plume overpass is detected in Figure 4b and 4c consistently with lidar detections in Figure 1.

MIMOSA- $\mu\phi$  - 532nm SR - 550K - 03/04/09

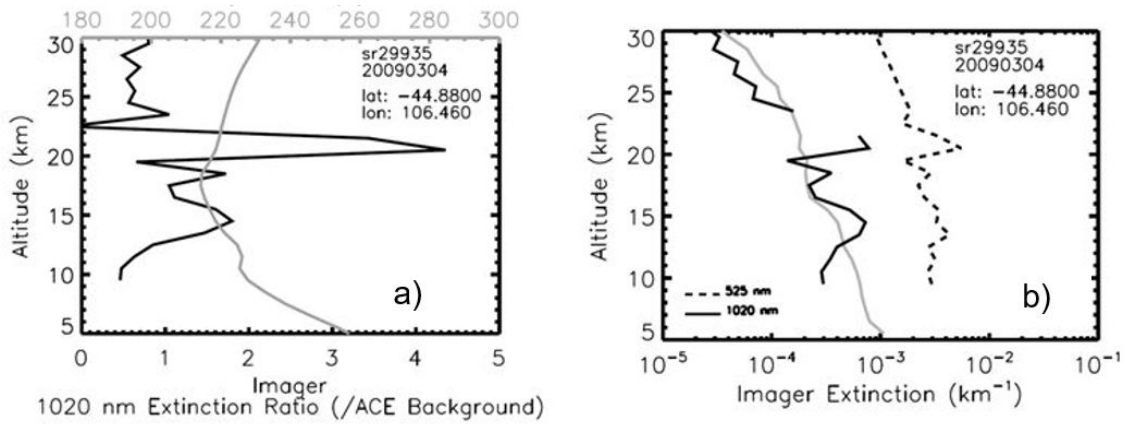


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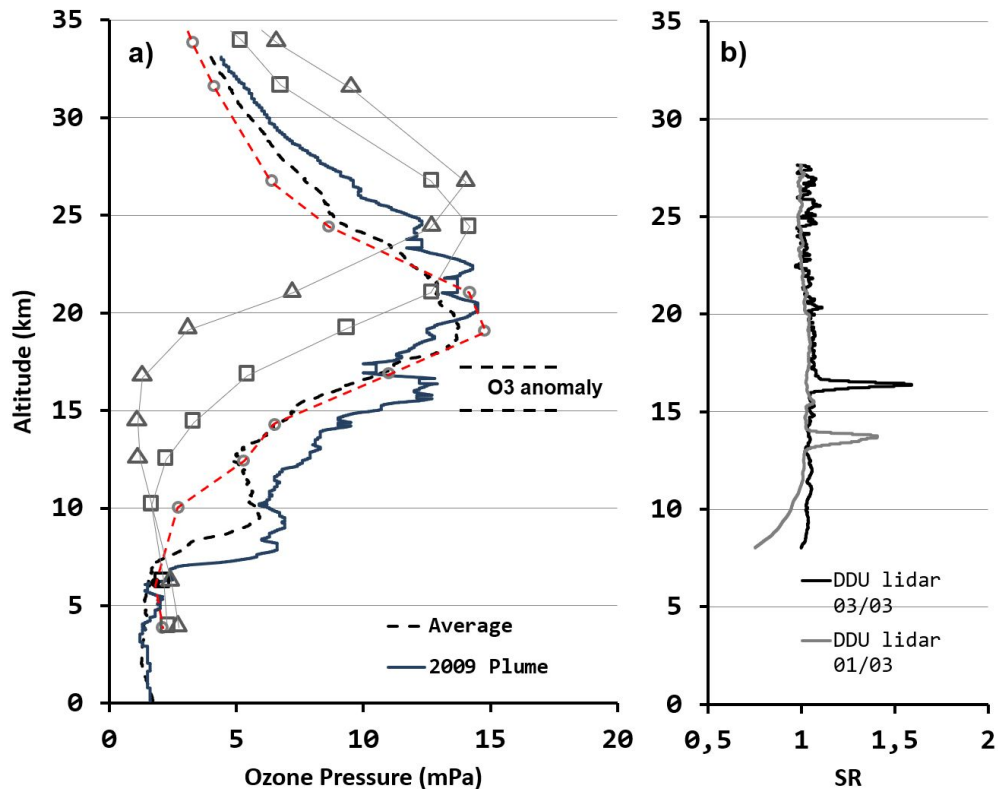


863 **Fig. 6.** 532nm MIMOSA- $\mu\phi$  SR calculations on March 4<sup>th</sup> (b). Crosshairs locate the ACE  
864 measurements displayed in Figure 7.  
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ACE extinction measurements - 03/04/09

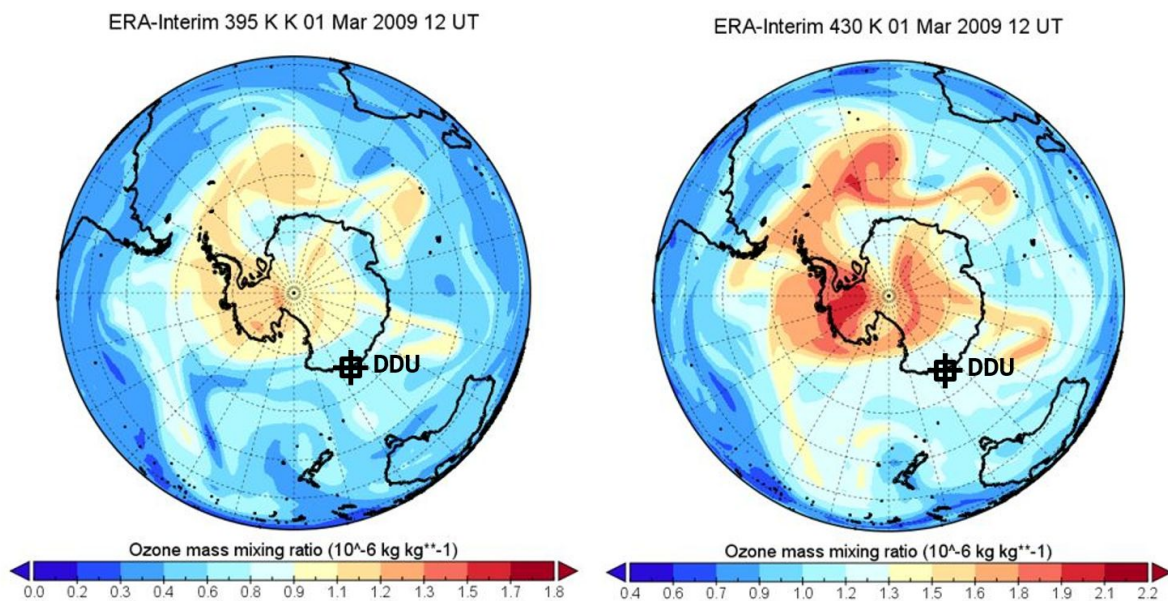


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867 **Fig. 7.** Visible and Near Infra Red ACE imager measurements on March 3<sup>rd</sup>. (a) Extinction ratio  
868 is reported as compared to the ACE reference background. Imager extinction is in (b).  
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**Fig. 8.** (a) DDU Ozone lidar measurements as a vertical pressure profile on March 1<sup>st</sup> 2009 (blue line), together with a 3-year average at the same period (black dotted line) and latitudinal typical climatology: Triangle, square and circle marked lines refer to ozone profiles recorded from balloon sondes on 2008 at the Kerguelen Islands respectively on the [15°S-25°S], [45°S-55°S] and [75°S-85°S] latitudinal bands [Bencherif et al., 2011]. Excess ozone concentration is detected at the smoke altitude range above 15 km. For reference, aerosol lidar profiles are reported in (b).



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**Fig. 9.** Ozone mass mixing ratio from ERA-Interim at 12:00 UT on 01 March 2009 on the isentropic surfaces of (left) 395 K and (right) 430 K.