

Faustine Mascaut, Olivier Pujol, Bert Verreyken, Raphaël Peroni, Jean Marc Metzger, Luc Blarel, Thierry Podvin, Philippe Goloub, Karine Sellegri, Troy Thornberry, et al.

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- ¹ Graphical Abstract
- 2 Aerosol characterization in an oceanic context around Reunion Island (AEROMARINE field cam-
- 3 paign)
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- Highlights
- 7 Aerosol characterization in an oceanic context around Reunion Island (AEROMARINE field cam-
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- 10 Sellegri, Troy Thornberry, Valentin Duflot, Pierre Tulet, Jérôme Brioude
- Optical properties, vertical distribution and transport pathways, from the marine boundary layer to the free troposphere,
- of marine aerosols in a pristine environment are examined.
- Aerosol size does not exceed the accumulation mode.
- A sketch is proposed as a characterization of marine aerosols distribution. Oceanic and insular influences in the aerosol
- 15 content are separated.
- It is argued that the AERONET station at St Denis (Reunion Island) is well representative of marine conditions.
- With the thermodynamics given by a microwave radiometer, the results will be useful for water vapor-aerosol-cloud
- ¹⁸ physical processes modeling in a pristine ocean.

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ABSTRACT

This article presents the results of the AEROMARINE field campaign which took place between February and April 2019 off the coast of Reunion Island in the South West Indian Ocean basin. The Southern Indian Ocean is of major interest for the study of marine aerosols, their distribution and variability. Six instrumented light plane flights and a ground-based microwave radiometer were used during the field campaign. These measurements were compared with the long-term measurements of the AERONET sun-photometer (based in St Denis, Reunion Island) and various instruments of the high altitude Maïdo Observatory (2.2 km above sea level, Reunion Island). These results were also analyzed using different model outputs: (i) the AROME mesoscale weather forecast model to work on the thermodynamics of the boundary layer, (ii) the FLEXPART-AROME Lagrangian particle dispersion model to assess the geographical and vertical origin of air masses, and (iii) the chemical transport model CAMS (Copernicus Atmosphere Monitoring Service) to work on the aerosol chemical composition of air masses. This allowed to highlight two points: (1) the atmospheric layer above 1.5 km is mainly composed of aerosols from the regional background; (2) the local environment (ocean or island) has little impact on the measured concentrations. Marine aerosols emitted locally are mostly measured below 0.5 km. The daytime marine aerosol distributions in the free troposphere measured by the aircraft were compared to the aerosol distribution measured at the high altitude Maïdo Observatory at night when the Observatory is located in the free troposphere. The results indicate that the high altitude site measurements are representative of the concentration of marine aerosols in the free troposphere. We also found that the CAMS reanalyses overestimated the aerosol optical depth in this region. Finally, our study strongly suggests that the AERONET station in St Denis (Reunion Island) can be considered as a representative marine station under the Tropics.

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olivier.pujol@univ-lille.fr (O. Pujol); bert.verreyken@aeronomie.be (B.	20	Because of their direct and indirect radiative forcings,
Verreyken); raphael.peroni@univ-lille.fr (R. Peroni);	21	atmospheric aerosols have a major impact on the climate.
${\tt jean-marc.metzger@univ-reunion.fr} (J.M.\ Metzger);$	22	These forcings are still poorly understood and lead to uncer-
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tainties that have persisted in models since the 1990s (Myhre 52 a region with frequent pristine conditions (where land and 23 et al., 2013). One of the largest uncertainties of the aerosol-53 human activities have little impact) that can reasonably be cloud system is the background concentration of natural aerosols, considered to be close to the preindustrial conditions. Few 25 especially over clean marine regions (Andreae and Rosen-55 26 feld, 2008). Oceans cover about 70% of the Earth's surface 56 27 and are an important reservoir of marine aerosols (mainly 57 28 sea salt and organic aerosols). Sea Salt Aerosols (SSA) are 58 29 one of the largest contributors to global aerosol loading and 59 30 therefore they play an important role in global climate. Also, 60 31 they were proposed to be a major component of primary 61 32 marine aerosol mass over the regions where wind speeds 62 33 are high and/or other aerosol sources are weak (Gantt and 63 34 Meskhidze, 2013; O'Dowd and de Leeuw, 2007). Luo et al. 64 35 (2014) showed that the AOD at 532 nm of SSA in the Ma-65 36 rine Boundary Layer (MBL) is governed by different physi- 66 37 cal factors: the surface wind speed, Sea Surface Temperature 67 38 (SST), MBL height, lower troposphere stability, and relative 68 39 humidity (impact on size and optical properties of sea salt 69 40 particles). However, there is a notable lack of data, in par-70 41 ticular over the oceans and in the southern hemisphere, de-71 42 scribing the characteristics of aerosols such as optical prop-72 erties, size distribution, temporal and spatial variabilities... 73 (Ramachandran, 2004). Pant et al. (2009) carried out mea-74 surements of the total number concentration and the size dis-75 tribution of aerosols over the Indian Ocean in 2004. They 76 observe that the aerosol concentration-wind speed correla-77 tion coefficient depends on the latitude and has a maximum 78 value where the winds are the strongest. 79 50

The southwestern Indian Ocean has been identified as 51

data have been collected in this region (Pant et al., 2009). However, it is a crucial reference point to quantify the background concentration of natural aerosols and the contribution of natural emissions to the changing climate. In pristine regions, SSA are dominant and concentrations are relatively low (e.g. Mallet et al., 2018).

Reunion Island in the southwestern Indian Ocean can be considered as a background aerosol pristine environment under trade wind conditions (Koren et al., 2014), mostly during the wet season from December to April. For details about wind circulation in the Southern Indian Ocean, including Reunion Island, see the statistical study by Mallet et al. (2018) and refs therein. Reunion Island is also a unique site in the southern hemisphere for making aerosol observations. Indeed, being in an oceanic environment and far from continents, the island is in a strategic location for carrying out measurements in a clean region, and also for the validation of spatial measurements. In addition, the Maïdo Observatory (located at 2.2 km above sea level $(a.s.l)^1$) allows: (i) to take measurements directly in the free troposphere at night (Guilpart et al., 2017; Foucart et al., 2018) and (ii) to perform long-term in-situ observations including detailed profiles of wind, temperature and water, as well as concentration, size and chemical composition of aerosols collected by ground instruments (Baray et al., 2013).

¹In this paper, altitudes are given above sea level (a.s.l)

The AEROMARINE project, which took place between109 80 February and April 2019, aimed at collecting data on ma-110 81 rine aerosol emissions, their optical properties, their trans-111 82 port and distribution off the coast of the Reunion Island. Fon12 83 this, ground-based instruments and instruments on board ul-113 84 tra light plane were used to measure concentration, size dis-114 85 tribution and optical thickness of marine aerosols over the15 86 Indian Ocean, on the western side of Reunion Island. In ad-116 87 dition, in order to characterize the thermodynamics of the17 88 MBL and the exchanges between the MBL and the free tro-118 89 posphere, a MicroWave Radiometer Profiler (MWRP) was19 90 set up in St Denis (in the north of the island). Aerosol data 91 have been complemented by the measurements of the AErosol²⁰ 92 RObotic NETwork (AERONET) sun-photometer (St Denis),21 93 and the various instruments of the Maïdo Observatory. Mod-122 94 els, such as FLEXPART-MesoNH or AROME as well as123 95 Copernicus Atmosphere Monitoring Service (CAMS) reanaliza 96 yses allowed to compare and/or support in-situ measurements 97 The instruments (on board and on the ground) and the mod₁₂₆ **o**s els used are briefly presented in the following section. 127 The overall objectives of the AEROMARINE project in 128 100 clude: 101 129

1) To characterize marine aerosol optical properties and their vertical distribution. The instruments on board the light plane helped characterize the marine aerosol optical proper ties, number concentration, and size distribution within the MBL and the free troposphere. Those results were compared with aerosol measurements at the Maïdo Observatory. Fur thermore, *AOD*-measurements were compared with those of the AERONET station at St Denis.

2) To examine the transport pathways of marine aerosols from the boundary layer to the free troposphere. Hence, it is important to estimate accurately the vertical distribution of the marine aerosols. Indeed, the MBL dynamics affect marine emission mechanisms, vertical dilution, while shallow convection is important for exchange and mixing of aerosols with the free troposphere.

The aim of this paper is to present the results obtained (section 3) during AEROMARINE that answer the above objectives. Section 4 is the conclusion of this work.

2. Instruments and models used

2.1. On-board instruments

2.1.1. PLASMA

Photomètre Léger Aéroporté pour la Surveillance des Masses d'Air (PLASMA) is a sun-tracking photometer developed by LOA and SNO PHOTONS (Karol et al., 2013). Compact (23 cm) and light (3 kg), it can be put on different mobile platforms (Popovici et al., 2018). Sun tracking is performed by means of elevation (0 – 88 °) and azimuth (0 – 360 °) rotations. Aerosol Optical Depths (*AODs*) at various wavelengths² (λ) are derived from extinction measurements of the solar radiation by molecular and aerosol scattering and absorption processes. The instrument provides *AODs* over a wide spectral range ($\lambda = 0.34 - 2.25\mu$ m) with an accuracy ΔAOD ranging from 0.005 to 0.01 according to λ . Aerosol size distribution is retrieved from the *AOD* spectral

²In this text, wavelengths are given with respect to the vacuum

17:

dependence (Karol et al., 2013). The Angström exponent @164 136 is determined from the law of Angström: $AOD_1/AOD_2 = 165$ 13 $(\lambda_1/\lambda_2)^{-\alpha}$ where AOD i is the aerosol optical depth at the action optical depth at the second depth 138 wavelength λ_i (AngstrÖm, 1961). This exponent describes 67 139 how the AOD varies with λ and so provides information on 140 168 the size distribution of the aerosols (Kusmierczyk-Michulec 141 169 et al., 2002). 142

143 2.1.2. Particle counters

The Portable Optical Particle Counter (POPS) is a 900 g₇₂ 144 in situ instrument designed by US laboratories NOAA and 173 145 CIRES that provides aerosol number size distribution (in the 146 size range $132 \text{ nm} - 3 \mu \text{m}$) using single-particle light scat-147 tering (Gao et al., 2016). POPS is a prototype made by a $3D_{176}$ 148 printer to reduce weight. It flew on board a light plane dur-177 149 ing the AEROMARINE intensive field campaign within the 150 MBL and the free troposphere. 151 179

Two Condensable Particle Counters (CPCs) (accuracy:180 152 \pm 20 %) are used simultaneously to measure the total con-181 153 centration of particles larger than 2 nm (CPC-MAGIC200) 154 and particles larger than 10 nm (CPC TSI model 3007). The183 155 difference of concentration between the two CPCs gives the 156 184 particle concentration in the size range 2 - 10 nm, which 157 185 is indicative of the recent formation of nanometric particle, 158 186 i.e. nucleation. The combination of two CPCs to investigate 159 187 nucleation was proven to be adequate in past airborne stud-160 188 ies (Crumeyrolle et al., 2010). Additionally, in synergy with 161 189 the POPS, the CPC TSI 3007 concentration enables to mea-162 190 sure the aerosol concentration in the 10-150 nm size range. 163 191

which is indicative of grown nucleated particles, and fine marine primary aerosols (size range 50 - 100 nm) that dominate the primary marine aerosol size distribution (Schwier et al., 2017).

2.2. Ground-based instruments

2.2.1. AERONET stations

The AERONET collaboration provides globally distributed observations of spectral *AOD*, inversion products, and precipitable water in diverse aerosol regimes. Aerosol optical properties are measured at multiple wavelengths ranging from the UV to shortwave infrared. *AOD* data (accuracy: \pm 0.02) are computed for three data quality levels: Level 1.0 (un-screened), Level 1.5 (cloud-screened), and Level 2.0 (cloud screened and quality-assured) (Holben et al., 1998). For comparison with PLASMA measurements, only Level 2.0 data quality for *AOD* and the Angström exponent ($\alpha_{440/870}$) are used. The 2020 data from the AERONET station (St Denis) is Level 1.5. The sun-photometer we used is located on the university campus, at St Denis located in the north of the island.

2.2.2. Microwave radiometric profiler (MWRP)

The microwave profiler RPG-HATPRO G5 gives us measurements of the microwave radiation emitted by the troposphere which provides tropospheric vertical profiles (0 - 10 km) of absolute humidity and temperature, with a special focus on the MBL. It allows to monitor with a high temporal resolution (1 min) the thermodynamic state of the atmosphere and to investigate fruitfully a wide variety of weather

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phenomena related to water vapour (Louf et al., 2015). A220 192 zenith-looking infrared ceilometer provides, together with21 193 the temperature profile retrieved from the MWRP, an esti-222 194 mate of the cloud-base height. The MWRP is also equipped 23 195 with in situ sensors for ground level measurement of temper-224 196 ature, water vapour and pressure (Louf et al., 2015). It was25 197 on the university campus between December 12th, 2018 and 26 198 March 11th, 2019. 227 199

200 2.3. Models and reanalyses

201 2.3.1. AROME

AROME-Indian Ocean (Bousquet et al., 2020) is used inp30 202 this study in order to obtain the horizontal wind fields at dif.231 203 ferent altitudes, at the places and dates where the flights were32 20 performed. This model is an adaptation of Météo-France' 933 205 operational model AROME (Seity et al., 2011) to the Indian²³⁴ 206 Ocean. AROME-IO has a horizontal resolution of 2.5 km²³⁵ 207 and is initialized and coupled to the lateral limits by Inte-236 208 grated Forecasting System (Inness et al., 2013) operationa²³⁷ 209 analyzes (ECMWF, https://www.ecmwf.int). It is also equipped 210 with a 1D coupling with the ocean in order to better representers 211 the ocean-atmosphere exchanges (Bielli et al., 2021). 240 212

213 2.3.2. Meso-NH

Meso-NH is a non-hydrostatic mesoscale model which 214 243 was developed in partnership by Centre National de Recherches 215 244 Météorologiques (CNRM) and Laboratoire d'Aérologie (LA) 216 245 and whose equations are described by Lafore et al. (1998) 217 246 and Lac et al. (2018). This multidimensional model (1D, 2D 218 247 or 3D) integrates a system of anelastic equations which al-219

lows simulations of a wide range of meteorological phenomena from the sub-synoptic scale (a few hundred kilometers) to the microscopic scale (a few meters). In this study, the resolution used is 500 m. Meso-NH takes into account different physical aspects such as turbulence, radiation, surface processes, microphysics ... It is also coupled with gaseous, aqueous chemistry and aerosol modules which provide a privileged dynamic framework for any numerical study of atmospheric physico-chemistry.

2.3.3. FLEXPART

The FLEXPART Lagrangian Particle Dispersion Model is a comprehensive community tool for atmospheric transport modeling and analysis. It is a Lagrangian particle dispersion model that simulates the transport, diffusion, dry and wet deposition and radioactive decay of tracers released from point, line, surface or volume sources. FLEXPART can be used forward in time to simulate the dispersion of tracers from their sources, or backward in time to determine their potential source contributions (Stohl et al., 2015). In our study, FLEXPART was used to determine the back-trajectories of particles in order to know their origin. Lagrangian particle models calculate the trajectories of a large number of so-called particles (which do not necessarily represent real particles, but infinitely small patches of air) to describe the transport and diffusion of tracers in the atmosphere. FLEX-PART's source code and a manual are freely available from the internet page https://www.flexpart.eu/. Recently, FLEX-PART has been coupled to the Eulerian models AROME

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248 (Verreyken et al., 2019). In this paper, we also use a ver-275
249 sion of FLEXPART that has been coupled to meteorologicab76
250 output from Meso-NH.

251 2.3.4. CAMS reanalyses

279 Copernicus Atmosphere Monitoring Service (CAMS, pre-252 280 viously MACC, https://atmosphere.copernicus.eu/) is an at-253 281 mospheric model that simulates the mixing ratios of various 254 282 aerosols, AODs or thermodynamic parameters (for example 255 283 humidity, wind, temperature ...) on a large scale and regional 256 284 scale. The data assimilation system used for CAMS is based 257 285 on the ECMWF' Integrated Forecast System (IFS). Satellite 258 286 observations are implemented in this model and allow the 259 287 study of the atmospheric composition (chemically reactive 260 288 gases, aerosols, greenhouse gases) at global scale (Morcrette 261 289 et al., 2009). CAMS allows in particular to differentiate vari-262 ous types of aerosols such as: (i) Sea Salt Aerosols (0.03-0.5 263 291 μ m; 0.5–5 μ m and 5–20 μ m) and Dust aerosols (0.03–0.55 264 292 μ m; 0.55 – 5 μ m and 5 – 20 μ m) divided into three size 265 293 ranges, (ii) Black Carbon and Organic Matter divided into 266 two modes (hydrophobic and hydrophylic) and (iii) Sulfate. 267 295 In our study, we use the mixing ratios of these different 268 296 species (Mallet et al., 2018) as well as the AOD. Mixing ra-269 297 tios and aerosol concentrations are directly related, allowing 270 comparison between in-situ measurements and CAMS data. 271

272 **3. Results**

273 3.1. From global to local scale

Mallet et al. (2018) investigated statistically, over a 8-

years period, the distribution and variability of marine aerosols in the southern Indian Ocean $\{10^{\circ}S - 40^{\circ}S; 50^{\circ}E - 110^{\circ}E\}$, by means of satellite data (POLDER and CALIOP), CAMS reanalyses, and *AOD* measurements from the AERONET sun-photometer located in St Denis (Reunion Island). They found that aerosols are mainly located below 2 km a.s.l and they estimated that SSA represents 60% to 80% of the total *AOD*, while sulphate and Organic Matter (OM) aerosols have low contributions.

For example, Figure 1 shows the CAMS mixing ratios for March 22th, 2019 at 850 hPa ($z \approx 1.5$ km a.s.l). We observe, in agreement with Mallet et al. (2018), that SSA dominate the aerosol loading in the southwestern Indian Ocean region, while sulfates and OM (hydrophilic) appear in smaller amounts, and dust aerosols are negligible. The large-scale situation for this date is representative of the different days of the AEROMARINE field campaign.

Figure 2 shows the CAMS mixing ratios η_{SSA} (for three ranges of different SSA sizes $R_{SSA} = 0.03, 0.5, 5, \text{ and } 50 \,\mu\text{m}$) corresponding to the days and location of (or around) the flights of the AEROMARINE field campaign. These mixing ratios have been converted from kg/kg into a SSA number concentration #/cm³ (Table1) in order to better compare with further *in situ* (POPS, MAGIC and TSI) measurements.

A simple enough but realistic way to perform this conversion is as follows. We first calculated the mass of SSA (in kg) for each radius R_{SSA} , $m_{SSA} = N_{SSA} \rho_{SSA} (4\pi R_{SSA}^3/3)$ where $\rho_{SSA} \approx 1183 \text{ g/cm}^3$ is a typical mass density of SSA (Bozzo et al., 2020) and N_{SSA} is the number of SSA par-



Figure 1: CAMS reanalysis $(0.25^{\circ}/0.25^{\circ})$: mixing ratio of SSA 0.03-20 μ m (top left), sulfate (top right), Organic Matter and Black Carbon (bottom left) and Dust Aerosols 0.03-20 μ m (bottom right) at 850hPa. Date: 03/22/2019 at 06:00:00 UTC.

ticles. Then, the ideal gas law gave us the volume occu-319 30 pied by 1 kg of dry air at standard temperature T and pres-320 305 sure p, i.e. $V = m_{air} RT/(pM)$, where $M \approx 29 \,\mathrm{g \, mol^{-1}}_{321}$ 306 $R \approx 8.314 \,\mathrm{J}\,\mathrm{K}^{-1}\,\mathrm{mol}^{-1}$. Since $\eta = m_{SSA}/m_{air}$, it ensues 22 307 directly $N_{SSA}/V = 3pM \eta_{SSA}/(4\pi R_{SSA}^3 RT \rho_{SSA})$. 323 308 The results of this conversion are summarized in Table 309 1. These orders of magnitude are realistic values. Another 310 precise approch would be to consider a size distribution if 311 it were fully available. The POPS, TSI and MAGIC size 312 ranges of measurements (respectively $132 \text{ nm} - 3\mu m$, $< 10 \text{ nm}_{7}$ 313 and < 2 nm) correspond to the two smallest size ranges of the₃₂₈ 314 CAMS reanalyses (SSA between 0.03 μ m and 5 μ m). The 315 CAMS concentrations retrieved (SSA_{0.03-0.5µm} and SSA_{0.5-530m}) 316 are of the same order of magnitude as the concentrations₃₃₁ 31 measured by POPS, TSI and MAGIC (see Tab.1). The largest 31

SSA concentrations (SSA_{5-20 μm}) are negligible compared to the other two CAMS size ranges.

We note for these flights that the SSA are not located on the same side of Reunion Island depending on the day. We will see later that this is explained by the wind regimes.

3.2. Thermodynamic parameters

Figure 3 present the averaged profiles (for March 2019) of the relative humidity and the temperature resulting from the measurements made by the MWRP and by the CAMS reanalyses.

The radiometer and CAMS reanalysis are in agreement on the relative humidity values between 0 and 1 km a.s.l, with a surface value of 70% and a maximum of 80% around



Aerosol characterization in an oceanic context around Reunion Island (AEROMARINE field campaign)

Figure 2: CAMS reanalysis (0.25°/0.25°): mixing ratio of SSA 0.03-0.5 μm, SSA 0.5-5 μm, and SSA 5-20 μm (from top to bottom) on 03/13/2019, 03/15/2019 (at 900 hPa), 03/22/2019 (at 850 hPa), 03/27/2019 (at 900 hPa), 04/11/2019 (at 850 hPa) and 04/18/2019 (at 1000 hPa) from left to right.

Table 1

Range of concentrations measured by POPS, TSI and MAGIC and range of SSA concentrations (size ranges: 0.03-0.5 μ m; 0.5 – 5 μ m and 5 – 20 μ m) from CAMS reanalyses in the pixel where the flight took place, for the six flight dates.

Date	MAGIC (#/cm ³)	TSI (#/cm ³)	POPS (#/cm ³)	SSA _{0.03-0.5µm} (#/cm ³)	$SSA_{0.5-5\mu m}$ (#/cm ³)	$SSA_{5-20\mu m}$ (#/cm ³)
03/13	200 to 2.10 ³	100 to 10^{3}	5 to 100	0.3 to 1.4.10 ³	$1.2.10^{-3}$ to 0.1	4.6.10 ⁻⁴ to 0.03
03/15	200 to 300	100 to 200	50 to 100	$2.3 \text{ to } 1.0.10^4$	0.06 to 60	$3.7.10^{-3}$ to 0.2
03/22	-	10 to 300	32 to 100	3 to 1.3.10 ⁴	0.08 to 76.3	$2.8.10^{-3}$ to 0.2
03/27	500 to 2.10^3	200 to 10^3	30 to 130	4.1 to $1.9.10^4$	0.01 to 12.1	$1.8.10^{-3}$ to 0.1
04/11	800 to 8.10^3	-	74 to 130	2.4 to 1.1.10 ⁶	0.06 to 58.7	$5.5.10^{-3}$ to 0.4
04/18	200 to 2.10^4	-	7 to 132	6.0 to $2.7.10^4$	0.2 to 180	0.02 to 1.2

333 0.5 to 1 km in altitude. However, above 1 km a.s.l, CAMS

reanalyses and radiometer measurements are very close to

reanalyses underestimate the relative humidity from 5% ab37 each other.

³³⁵ 2 km to 20% at 5 km altitude. For the temperature, CAMS³³⁸

From the radiometer measurements, we observe that rel-

Table 2

Availability of instruments during flights (F1 to 6).

AVAILABLE INSTRUMENTS								
Flight	PLASMA	MAGIC	POPS	TSI	34			
F1	\checkmark	\checkmark	\checkmark	\checkmark	34			
F2	\checkmark	\checkmark	\checkmark	\checkmark	34			
F3	\checkmark	\checkmark	\checkmark	\checkmark	35			
F4	\checkmark	\checkmark	\checkmark	x	35			
F5	\checkmark	\checkmark	\checkmark	x	35			
F6					35			

Relative humidity & Temperature profiles from Microwave Radiometer (MWR) & CAMS model



Figure 3: Average profiles of temperature and relative hu-364 midity from microwave radiometer and from CAMS reanalysis (20.8°S; 55.2°E) for March 2019.

ative humidity varies between 50% and 100% and tempera³⁶⁷
ture between 283 K and 300 K in the 0 – 3 km atmospheric³⁶⁸
layer. In addition, the averaged height of the boundary layer³⁶⁹
for March 2019 is 0.532 km. These values are representa³⁷⁰
tive of the thermodynamic situation during field campaign⁸⁷¹
flights. These data will be helpful to examine cloud forma³⁷²

45 tion under marine conditions.

3.3. The AEROMARINE field campaign

The AEROMARINE field campaign allowed to better understand the 3D distribution of marine aerosols around Reunion Island and how it is influenced by the dynamics of the MBL thanks to an instrumental synergy: PLASMA, POPS, Tandem CPC TSI3007 and MAGIC200 (Table 2). Six flights, of a duration of about ninety minutes, allowed to sample the aerosols from an altitude of 100 m up to 4 km and up to about 2 km off the west coast. The flight paths are shown on Fig. 4. They were designed to have most of the time a vertical profile above the ocean, and to measure air masses above the Maïdo mountain on the way back to the airport. Since focus is put on marine aerosols, we are going to divide these flights into two groups: (I) when only the ascending part is over the ocean (F1, F2, F3) and (II) when the whole flight is over the ocean (F4, F5). Finally, flight F6 is treated separately since the plane flew over the city of St Denis (red box on Fig 4). We will see that this flight is interesting to segregate data between land/ocean conditions.

3.3.1. Optical properties from PLASMA measurements

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Figure 5 displays the *AOD* and α vertical profiles measured by PLASMA during each flight. For group II, we have separated the ascending phase from the descending phase. For each flight and for each wavelength, the *AOD* is lower than 0.1 during the ascending phase, with values below 0.05 between 500 m and 1 km, and up to 1.5 km for flight F5.

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Figure 4: Flight trajectories and altitudes on 03/13/2019, ³⁹⁷ 03/15/2019, 03/27/2019 (Group I) and 04/11/2019, ⁰⁴/18/2019 (Group II). The sixth flight is a particular case (see text for details).

It is in agreement with other published results on marine 37 402 air masses (e.g., Horowitz et al., 2017 shows as multi-374 403 year daily average $AOD = 0.06 \pm 0.04$ and $\alpha = 0.7 \pm$ 375 404 0.36) Higher in altitude, the AOD changes little with the 376 405 altitude, and is constant with altitude above 2 km. It means 377 406 that most of the aerosols that contribute to AOD (i.e. the 378 407 largest ones) are located below 2 km, which is in agreement 379 408 with previous works by Lesouëf et al. (2013) and Duflot et al. 380 409 (2019). For group II, the AOD-profiles during the descent 381 410 differ from the ascent, especially below around 1 km of alti-382 411 tude, where AODs can reach 0.15 during the descent phase. 383 412 For flight F4, the AOD peaks at 500 m, in particular at 500 384 413 and 650 nm. Beside the AOD values below 1km altitude 385 414 during the descent phase, the behaviour of the AOD with 386 415 the wavelength does not depend on the altitude: the short-387 416

est wavelengths (380, 440 nm) show the highest *AOD*. This suggests that the sampled atmosphere was made of particles with size of the order of, or comparable to, these wavelengths, so particles rather in the accumulation mode, with the exception just mentioned which could indicate the presence of larger particles.

 α (Angström exponent) is a good qualitative indicator of the mean size of the sampled aerosols. The PLASMA measurements reveal that α is lower than 1.2, independently of the altitude. This clearly suggests the presence of marine aerosols like sea salt (Schuster et al., 2006), since the CAMS retrievals (Fig. 1) indicate that the contribution of dust aerosols were negligible. For group I, α presents in general a maximum around 1.0 within the 0.5-1.5 km a.s.l layer. Regardless of local maxima in α that corresponds to sudden and localized changes in the corresponding AOD, the overall behaviour of α with altitude suggests that the larger particles are situated between 0.5 - 1.5 km. For flight F1, α reaches a maximum of 1.5 at about 800 m in altitude, in agreement with the peak in AOD at 440 nm and the almost null value of AOD at 1020 nm, indicating the presence of smaller particles at this altitude.

For group II, the ascent profiles share similar characteristic to those from group I with an increase of α with the altitude up to 1.5 km and a slight decrease above. The values of α vary between 0.6 and 1.2. However, larger particles seems to occupy the altitudes above 1.5 km compared to group I. Below 1.5 km, a decrease of 0.5 can be found in α values between the descent (< 0.5) and ascent phases, which could



Figure 5: *AOD* and Angström exponent (*α*) vertical profiles (PLASMA measurements) for flights F1, F2, F3 (Group I) and flights F4, F5 (Group II).

indicate a depletion of small particles in favor to larger ones 424

418 3.3.2. Aerosol concentration measurements

426 Figure 6 presents the aerosol concentration measurements 41 427 by MAGIC (particle size larger than 2.5 nm), TSI (particle 420 428 size larger than 10 nm) and POPS (particle size larger than 42: 429 132 nm). POPS concentration profiles show that the sampled 42 430 particles (accumulation and coarse modes) have a maximum 423 431

number concentrations of around 10^2 cm^{-3} between 0.5 and 1.5 km, and around 10 to $3 \times 10^2 \text{ cm}^{-3}$ above. It confirms that larger particles are found between 0.5 and 1.5 km in altitude, in agreement with the conclusions deduced from the optical measurements. Below 0.5 km a.s.l, the POPS concentrations increase with altitude during the ascent phase.

The shape of the measured concentration profile can show a maximum (group I) or a minimum (group II) in the layer



Figure 6: Particle concentration profiles (from POPS,TSI3007 and MAGIC200 instruments) during F1, F2, F3 (Group I) and F4, F5 (Group II).

below 0.5 km. During the descent (group II), the POPS con-438
centrations decrease with altitude between the surface and 439
0.5 km. Furthermore, the aerosols concentrations are highered
between 1 and 1.5 km a.s.l compared to the ascent phase, up 441
to a factor of 10 for flight F4 at 1 km a.s.l. Above 1.5 km a.s.l 442
the POPS profiles during the ascent and descent phases are443

in agreement, with similar shapes and concentrations. POPS counts more particles during the descending phase of the flight. This is tempting to explain this increase by physicochemical processes that would modify the aerosol number or the aerosol size so that small particles became large enough to be counted by POPS. However, we have to keep in mind

time to develop a bit more.

that the trajectory of the plane during the descent is the sameara 444 as the ascending trajectory, so we cannot exclude an influ-474 445 ence of the plane in the aerosol content when coming back 475 446 Also, we cannot totally exclude that the air mass has changed 176 44 by advection between the ascent and the descent of the plane A77 448 Comparing the aerosol concentrations measured by POPS₇₈ 449 to the measurements from MAGIC and TSI will help fur-479 450 ther evaluate the vertical distribution of aerosols according 451 to their size. Differences in TSI and MAGIC concentra-481 452 tions are minimum above 1.5 km a.s.l. In particular, their482 453 values are almost identical above 1.5 km a.s.l for flight F2483 454 and above 2. km a.s.l for flight F1. Below those altitudes484 455 the MAGIC concentrations are around twice those of TSIA85 456 This means that small particles (nucleation mode) are twice486 457 (in concentration) than larger particles (Aïtken mode) below487 458 1.5 km while above, there is no small particles (MAGIC-TSLass 459 ratio close to one). Such values of number concentration in 460 the nucleation mode, between 10^3 and 10^4 cm⁻³, have beenego 461

measured also in the Mediterranean (Eleftheriadis et al., 2006) 462 For the flights in group I, the concentration vertical pro-492 463 files obtained from MAGIC and TSI present similar shapes 493 46 On average, MAGIC concentrations are 2 to 3 times higherage 465 than TSI concentrations. The highest concentrations are founds 466 in the marine boundary layer below 0.7 km a.s.l for flightese 467 F1, and below 0.5 km a.s.l for flight F2 and F3, with val-497 46 ues ranging from 2000 to $10000 \,\mathrm{cm}^{-3}$. This difference of 498 469 200 m in the marine boundary layer may be due to the factage 470 that flight F1 occurred one hour later in the morning com-500 47 pared to flights F2 and F3, so the boundary layer may haveo1 472

For flight F1, the MAGIC and TSI concentration profiles are rather constant at 2.10^2 cm⁻³ above 1 km. For flight F2, the concentrations are constant above 1.5 km at 4.10^2 cm⁻³. For flight F3, a value of 4.10^2 cm⁻³ is found above 2 km, and a value of 10^3 cm⁻³ is found in the layer between 0.8 and 1.6 km.

For group II, only MAGIC was available. Similar features can be found in the profiles of flights F4 and F5 compared to group I. Furthermore, contrary to the aerosol concentration measured by POPS, the aerosol concentrations are rather similar between the ascent and descent phases.

Other important information are obtained when comparing the POPS and MAGIC profiles. Indeed, the concentration profile differences "MAGIC - POPS" (not shown) present relatively smaller values above 0.5 km. More significant differences are obtained below this altitude. An exception occurs for flight F1 since the critical altitude is 1 km because of a more developed boundary layer, as already suggested above. Another exception is for flight F5 where significant differences are between 0.5 and 1.5 km and small differences elsewhere. For group II, no significant differences are noted between the ascent and the descent *i.e.* same shape and order of magnitudes in the concentration profile differences.

In summary, the measurements indicate that the boundary layer below 0.5 km - 0.7 km is much richer in aerosols in the nucleation or Aïtken modes (size lower than 132 nm). Above the boundary layer, and up to 1.5 km to 2 km depend-

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ing on the flight, these modes have a lower concentrations30 502 and larger particles (size greater than 132 nm - accumulation₃₁ 503 mode) are dominant. The exception in the marine boundary₅₃₂ 504 layer for flight F5, *i.e.* below 0.5 km, means that this layer isa3 505 poorer in terms of small aerosols. 534 506

3.3.3. Origin of the air masses 50

536 To further evaluate the origin of the air masses identi-508 537 fied in the previous section, we used FLEXPART mesoscale 509 538 backtrajectories to distinguish air masses influenced by local 510 539 terrestrial emissions from those with marine origin or repre-511 540 sentative of the regional background. Here, we have chosen 512 541 a backtime of 12 h because it corresponds to the domain of 513 542 AROME. 514

543 The tables A1 and A2 in the appendix presents statistical 515 544 products from the FLEXPART output. 516

545 The scientific meaning of the FLEXPART outputs needs 517 546 to be briefly reminded in order to avoid misunderstandings 518 547 about the result presented in those tables. When we release 519 **5**/1 8 an air mass at a given altitude z_r , FLEXPART is able to trace 520 549 back its probable trajectories, over a geographical grid, dur-521 ing an user-decided time interval. From that, it is then possi-522 551 ble to identify (and reckon) the oceanic pixels, viz. the trajec-523 552 tory grid-points located over the ocean, and similarly for the 524 553 island pixels which are trajectory grid-points located over 525 Reunion Island. In other words, we can quantify how much 526 555 could the ocean contribute to, or impact, the aerosol content 527 of the considered air mass. 528 557 Beside the geographic origin, it is possible to determine

The results from the previous sections helped us identify three layers: (L1) below 0.5 km, (L2) between 0.5 and 1.5 km, and (L3) above 1.5 km. For instance, in Table A1, the results after 12h of backward simulations for an altitude release at 200m for flight F1 show that 88% (first row, last column) of the air mass originated from grid cells over the ocean, and 12% from grid cells over Reunion island. In addition, the origin of the air mass can be analysed in terms of geographic location and height. Hence, 43% of the air mass originated from 0-500m in altitude (layer L1), 38% from 500 to 1500m in altitude (laver L2), and 7% above 1500m (laver L3) and above the ocean. Therefore, the origin of this air mass is mainly oceanic.

the tropospheric layer from where an air mass originates.

We have thus used FLEXPART to calculate the origin of air masses from different altitudes. For group I, we can see that the origin of the air masses is mainly oceanic since more than 73% of the backtrajectories originate from grid cells above the ocean.

In contrast, 28% of the air mass was located above Reunion island, and within the layer L2 and L3. For group II, the results are different since the air masses present a significant origin above Reunion island. In particular, the air masses released at the 200 m and 1 km altitudes for flight F4 have a dominant origin above Reunion island.

Comparing the altitude where the backtrajectories are released, and the distribution in the vertical of the backtrajectories give additional information on the vertical transport between layers. Let us first look at oceanic pixels for group

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I. First, we note that the air masses with a dominant mariness 55 origin stayed, for a large part, within the same layer they 560 originated from. For example, for a release at 200 m, 43% 590 561 of the air mass with marine origin were in the layer L1. Ex-591 562 cepted for some rare exceptions (underlined in tables), these 563 layer that presents the highest percentages (in **bold** fonts in_{**b**93} 564 tables) includes the altitude of release. However, exchanges 565 of air masses (in italic fonts in tables) between layers are 566 not negligible, even if they are not dominant. For instance 567 (flight F1), for a release at 1 km, 29% of the air mass with 568 marine origin come from the layer L1. This means that the 569 aerosol content in the lower troposphere is also impacted by 570 the mixing between contiguous layers. This result holds for 571 the air masses originating from Reunion island, although the 572 terrestrial component in the lower troposphere is mostly sig-573 nificant in the layer L2 (percentages greater than 10%). For 574 the group II flights, we have the same results although we 575 recognize that the insular influence is more present, espe-576 cially at low level for flight F4. This may be correlated with 577 MAGIC and PLASMA measurements. Indeed, for flight F4 57 the maximum MAGIC concentrations are measured belowers 57 0.5 km a.s.l, which corresponds to a majority of island pix-580 els in the Table A1. It is also at this altitude that PLASMA 581 measures the most important AODs (during the descending phase). 58 600

We are summarized with typical orders of magnitude the₆₀₁ results given in Tables A1 and A2. These conclusions are_{602} summarized on Figure 7. The vertical layering we found₈₀₃ and in particular the predominance of the oceanic aerosols₈₀₄ are explained by the interaction between the wind field and Reunion island's complex terrain. The AROME model outputs (not shown) reveal a dominant south-west and south wind weather regime occurred over the sea at the time of the flights, with sometimes strong recirculation on the lee side, off the west coast of Reunion Island.



Figure 7: Descriptive diagram of aerosols inputs and the exchanges between the atmospheric layers L1 (below 0.5 km), L2 (0.5 - 1.5 km) and L3 (above 1.5 km).

3.3.4. Special case: flight of 22 March 2019 (F6)

This flight (F6) is of particular interest since the plane flew over St Denis between 05:20 and 05:30 UTC at an altitude of about 900 m (red box on Fig.4), then ascended in spirals above the ocean up to 2.5 km (blue box on Fig.4). It finally flew over St Paul before landing.

The measured *AODs* and α over St Denis (grey boxed area on Figure 8) have a local maximum of 0.05 and minimum of 0.5 respectively, while over the ocean (blue box in fig.8), the measured *AODs* are constant (*AOD* < 0.05) and α is around 0.8 – 1. This suggests that smaller particles are

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sampled when the plane is over St Denis.



Figure 8: Particle concentration profiles from POPS, TSI3007₆₃₀ and MAGIC200 instruments (left), *AOD* and Angström exponent ($\alpha_{440/870}$) profile from PLASMA sun-photometer (right) 632 during flight on 03/22/2019 (F6).

The difference in concentration between MAGIC and POPS⁶³⁴ 60 635 is much larger over St Denis than over the ocean. This means 60 636 that, as indicated by the PLASMA optical measurements, 608 637 small particles (size lower than 132 nm) are the dominant 609 638 mode over St Denis. The MAGIC - TSI differences in con-610 639 centration further indicates that the smallest particles are those 611 640 that dominate. In contrary, the MAGIC - TSI difference 612 641 is close to zero and confirm that larger particles are found 613 642 above 1.5 km a.s.l over the ocean north east of the island. 614 643 The wind direction given by AROME (not shown here) 615 indicate (i) at 1 km (overflight of St Denis) a southeast wind 616 645 of around 10 m s⁻¹ coming from the island and (ii) at 1.5 km, 617 646 when the plane begins these spirals above the ocean, a south-618 647 east wind from the ocean with a speed of about 8 m s^{-1} . 619 The FLEXPART backtrajectory results (table A2 in the 620

appendix section) indicate that for a release at 1.5 km, the air mass origin was purely marine and stayed above 500m over the past 12 hours. This is due to a southeasterly wind regime, according to AROME wind fields (not shown). Hence, the layer at 1.5km in altitude is representative of the regional background.

3.4. Comparisons with other databases

3.4.1. Comparison with AERONET measurements

Assuming that the *AOD* is mostly influenced by marine aerosols on the north and west shore, one can compare *AOD* measured by the AERONET station at St Denis and the *AOD* measured by PLASMA on the runway.

Table 3 brings together the mean values of the *AOD* ^{500nm} on the runway (before takeoff) measured by PLASMA and the *AOD* ^{500nm} measured by the AERONET sun-photometer (St Denis) at the same time. Both the *AOD* and α from AERONET and PLASMA are in agreement for flights F2, F3 and F4. For the flight F1, the AERONET sun-photometer measures a larger Angström exponent while for flights F5 and F6 it is the *AODs* measured on the runway by PLASMA which are larger. However, these differences are consistent with the accuracy of the two photometers (AERONET: ΔAOD = \pm 0.02 and PLASMA: $\Delta AOD = \pm$ 0.005 - 0.01 according to λ). The exception of the flight over St Denis (flight F6) where *AOD* _{AERONET} > *AOD* _{PLASMA} can be explained by the difference in altitude of the two measurements (0.9 km for PLASMA and ground-based for AERONET).

So AERONET measurements are generally in agreement

Table 3

AOD at 500 nm and Angström exponent (α) on the runway from the PLASMA sun-photometer and from the AERONET sunphotometer (St Denis) for each flight date

		PLASM	A	AERONET		
Flight	AO.	D 500nm	α (AE)	AOD 500nm	α (AE)	
F1	(0.06	1.0	0.06	1.7	
F2	().07	0.6	0.07	0.7	
F3	().07	0.5	0.06	0.4	
F4	().05	0.6	0.04	0.7	
F5	().09	0.6	0.06	0.6	
F6	().12	0.4	0.06	0.4	
Over St D.	(F6) ().04	0.5	0.07	0.2	

with those carried out during flights. If one relies on the AERONET retrievals, the general agreement with the PLASMA retrievals suggests that PLASMA offered a representative 65 sampling of the aerosol content around Reunion Island. We 65 further evaluated this by comparing the AOD AERONET 65 measurements before and during the 2020-lockdown due to the Covid19 pandemic, assuming that the terrestrial sources 65 were similar between both periods. Reunion Island experienced a lockdown between 17 March 2020 and 17 May 2020 and hence mobile traffic and anthropogenic activities were 65 reduced. The three plots presented in Fig 9 show that no 65 statistically significant change in the AOD or Angström ex-660 ponent can be clearly attributed to the lockdown. This tends 66 to indicate that indeed the AERONET station at St Denis is 662 not significantly impacted by local anthropogenic aerosols 663 and confirms the results of Hamill et al. (2016) that it is a 664

marine station.



Figure 9: Mean *AOD*, mean α and number of data points from AERONET station in St Denis (Level 1.5) for the period from March 17 to May 17 (2010 - 2020).

3.4.2. Comparison with CAMS reanalyses

Figure 10 presents the daily *AOD* averages of the CAMS model (model grid point: 20.8°S; 55.2°E) and of the AERONET sun-photometer (St Denis) for March and April 2019. Over this period, CAMS overestimates the *AOD* by 0.03.

Daily differences between CAMS reanalyses and AERONET measurements are presented in Table 4 (values rounded to the hundredth). The monthly averages of the *AODs* given by the CAMS reanalyses is 0.11, for March and April 2019. The monthly averages of the *AODs* measured by the AERONET sun-photometer is 0.08 for March and April 2019. This shows an overestimate of CAMS of 0.03 for March and April 2019. In particular, the CAMS reanalyses overestimate the *AODs* from 0.03 to 0.09 for flights F1, F3, F4 and F6. The difference between CAMS and the PLASMA AERONET sunphotometer is statistically insignificant for flights F2 and F5. These results agree with those obtained by Mallet et al.



Figure 10: *AOD* measurements from AERONET sun-photometer (Level 1.5) and from CAMS reanalysis (20.8°S; 55.2°E) for March 2019 and April 2019.

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Table 4

Differences in daily AOD averages of								
the CAMS model (20.8°S; 55.2°E) and								
of the Al	ERONET sun-photometer (St							
Denis) fo	r each flight							
Flight	Flight AOD CAMS-AOD AERONET							
F1	+ 0.03							
F2	- 0.01							
F3	+ 0.04							
F4	+ 0.09							
F5	+ 0.01							
F6	+ 0.06							

(2018) who determined that the *AODs* given by CAMS over-⁶⁹⁷
estimate by about 0.05 the local AERONET measurements.⁶⁹⁸

3.4.3. Comparison with Maïdo measurements

The *in situ* measurements on board the light plane in the free troposphere are further compared to the measurements from the high altitude Maïdo Observatory (21.08 ° S, 55.38 ° E; 2.2 km a.s.l). We use the measurements made by a Scanotron particle counter described in Foucart et al. (2018) (concerning aerosols of size 10 - 600 nm) and a CPC TSI (aerosols greater than 10 nm) from the Maïdo Observatory for comparison with the in-flight data (POPS, TSI and MAGIC). Due to a complex interplay between land-sea breeze, catabatic wind and complex terrain, only the in-situ measurements Maïdo taken between 21:00 and 03:00 UTC can be considered as free tropospheric (Verreyken et al., 2021).

Figure 11 display the mensual averages for March and April 2018 for the Scanotron and the TSI located at Maïdo. The time series (POPS, TSI and MAGIC measurements) dur-

Table 5

Comparison of average nighttime concentrations (Maïdo Observatory) and concentrations in the free troposphere during flights.

	Mean nighttime concentration		Concentrations during flights			Mean nighttime concentration			
	at Maïdo D	at Maïdo D-1 (21:00-03:00 UTC)		(in free troposphere)			at Maïdo D+1 (21:00-03:00 UTC)		
	Scanotron	TSI	POPS	TSI	MAGIC	Scanotron	TSI		
Date	(#/cm ³)	(#/cm ³)	(#/cm ³)	(#/cm ³)	(#/cm ³)	(#/cm³)	(#/cm ³)		
03/13	129.7	x	0 - 21	102 - 180	101 - 146	x	x		
03/15	189.7	x	3 - 97	226 - 542	244 - 542	210.8	247.0		
03/22	321.4	x	0 - 29	147 - 746	x	143.2	x		
03/27	268.0	x	0 - 12	126 - 221	213 - 302	190.6	x		
04/11	129.1	376.4	0 - 50	x	236 - 347	109.7	346.8		
04/18	x	548.6	1 - 54	x	379 - 771	x	671.0		

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Figure 11: March and April 2018 averages of measurements¹³ at the Maïdo Observatory by the Scanotron and the CPC TSI_{714} and time series of concentrations measured during the flights₇₁₅ of 03/22/2019 (F6) and 04/11/2019 (F4).

ing the flights of March 22, 2019 and April 11, 2019 are also
 presented for comparison.

For March, the average TSI measurements at Maïdo and during the flight have the same order of magnitude (between 40 and 4.10^3 #.cm^{-3}). However, the POPS measurements during the flight (March and April) are much lower than those of the Scanotron at 2.2 km a.s.l. The Scanotron (10 – 600 nm) measures aerosols smaller than the POPS (132 nm - 3 μ m). This would mean that the majority of the aerosols measured above 2.2 km a.s.l are less than 132 nm (in agreement with CAMS section 3.1).

Table 5 presents the measurements made during flights in the free troposphere (measurements for an altitude higher than 2 km) and the average concentrations measured at the Maïdo Observatory (2.2 km a.s.l) during the nights before and after the flights, when the observatory is in the free troposphere.

Overall, the MAGIC and TSI measurements made dur-

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ing flight in the free troposphere are of the same order of748 720 magnitude as the Scanotron night measurements made at the49 721 Maïdo Observatory, in the free troposphere (between 1×10^{2} 722 and 7×10^2 cm⁻³). The two TSI CPCs (at the Maïdo Obser-751 723 vatory and on the plane) also measure identical concentra-752 724 tions above 2 km (around 10^2 cm^{-3}). 753 725 We can conclude that the nighttime measurements at thesa 726 Maïdo Observatory (at 2.2 km a.s.l) are representative of thebs 727 daytime measurements (during flights) in the free tropospheres 728 and allow sampling of purely marine aerosols. 729 757

730 4. Conclusion

In this paper, we have presented the AEROMARINE field₆₀
campaign which took place between February and April 2019₆₁
off the coast of Reunion Island (southwestern Indian Ocean).
This area, identified as a pristine region, is of major interest
for the study of marine aerosols, their vertical distribution
and their optical properties.

During this campaign, a MWRP was deployed in St De-737 nis (93 m in the north of the Island) between mid-December 738 767 2018 and mid-March 2019. This made it possible to deter-739 mine that, during the austral summer in this region, the ther-740 769 modynamic situation (humidity, temperature, and height of 741 770 the boundary layer) is relatively stable. 742 In addition, six instrumented flights allowed the aerosols⁷⁷¹ 743

to be sampled from an altitude of 100 m up to 4 km by spiraling above the ocean thanks to an instrumental synergy.
The optical properties of the aerosols were measured by

the PLASMA photometer and three particle counters (POPS₉₇₅

TSI and MAGIC) measured the aerosol concentrations for different size ranges (accumulation, coarse and Aïtken modes). POPS analysis indicates that almost all of the particles are in the accumulation mode, centered around a particle size of 132 nm.

The results obtained show *AODs* less than 0.1 (with some exceptions), which is representative of a pristine region. The various measurements (*AOD*, Angström exponents, and concentrations) also indicate that the aerosols are in the accumulation and coarse modes, and mainly below 2 km of altitude.

The FLEXPART simulations enabled to determine the most probable origin of the aerosols measured during the flights. As a result, the aerosols follow the following vertical distribution:

- Above an altitude of 1.5 km, the sampled aerosols are not substantially impacted by the surface (layer L1 has a relatively little contribution). This is interesting since it allows to quantify the background aerosol concentration. For all the flights, we have estimated that the number concentrations (in cm⁻³) are 300 (MAGIC), 230 (TSI) and 15 (POPS). Also, the assessed AOD_{550nm} and α are respectively 0.01 and 0.75.
- Below 0.5 km (in the MBL), aerosols come essentially from the surface. The origin can be oceanic (33%) or insular (8%). Insular influence are nonetheless due to special events depending on the wind regime (*e.g.*, the Cap La Houssaye may bring sometimes dust aerosols).
- The intermediate layer, *i.e.* between 1.5 and 0.5 km

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a.s.l, is a layer of mixture: aerosols are mixed witheos those coming from the lower or upper atmosphericson layers. 807 778

These results meet the initial objectives of the AERO-77 MARINE campaign: (i) to characterize marine aerosol optical properties and their vertical distribution and (ii) to ex-781 amine the transport pathways of marine aerosols from the 782 812 MBL to the free troposphere. It is worth mentioning that 783 the flights were carried out between 04:00 and 07:00 UTC,⁸¹³ 784 814 *viz.* during the transient convection regime between noctur-785 nal and diurnal conditions. Further observational studies and 786 field campaign may be necessary to examine aerosol distri-787 817 butions during purely diurnal and nocturnal regimes, *i.e.* for 788 818 well established regimes. The AEROMARINE campaign 789 819 presented here is interesting in the sense that it documents a 790 transient regime, namely a more complex regime in terms of 791 821 thermodynamics compared to established ones. 792 The measurements taken during the flights were com-793 822 pared with the CAMS reanalyses. They showed that, like in-794 823 flight measurements, SSAs are predominant around Reunion 795 824 Island and that aerosols are mainly located below 2 km. It 796 825 was also shown that CAMS overestimates the AODs (from 797 826 0.01 to 0.09) in this region in agreement with results from 798 827 Mallet et al. (2018). In addition, a comparison between PLASMA 799 828 measurements (on the runway) and the AERONET sun-photometer 800 (located in St Denis) as well as a study on the impact of 2020-801 829 lockdown due to the Covid-19 pandemic on AERONET mea-802

830 surements were carried out. The results strongly suggested 80

AODs Aerosol Optical Depths. that the AERONET station is a marine station. In other831 804

words, measurements are not impacted by local anthropogenic activities and the station can be considered as representative of marine conditions. The AEROMARINE campaign occurred around Reunion Island. A field campaign, Marion Dufresne Atmospheric Program - Indian Ocean (MAP-IO), aboard the Marion Dufresne around the Terres Australes Françaises (TAF) was planned for January 2021. Among the objectives, one of them is to better document the exchanges between the pristine Southern Indian Ocean and the atmosphere. For our topic, this campaign will allow the results presented in this paper to be deepened, since it will provide data about marine aerosol emissions and of aerosol and humidity exchanges between the pristine ocean and the MBL far from any land. All of these data (from AEROMARINE and then MAP-IO) will be helpful to feed models of water vapour-aerosols-clouds interactions. Such features will be the topic of future research.

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Acronyms

sis.

AERONET AErosol RObotic NETwork.

- **CAMS** Copernicus Atmosphere Monitoring Service. 845
- **833 CPC** Condensable Particle Counter.
- 834 MAP-IO Marion Dufresne Atmospheric Program Indian
- 835 Ocean.
- 836 MBL Marine Boundary Layer.
- 837 MWRP MicroWave Radiometer Profiler.
- **838 OM** Organic Matter.
- **PLASMA** Photomètre Léger Aéroporté pour la Surveillance
- des Masses d'Air.
- **POPS** Portable Optical Particle Counter.
- 842 SSA Sea Salt Aerosols.
- 843 SST Sea Surface Temperature.
- **TAF** Terres Australes Françaises.

Appendix

FLEXPART simulations. Flights: F1, F2, F3 and F4

Table A1

Origin of air masses for L1, L2 and L3 according to a 12 hours-simulation of the FLEXPART

model for F1, F2, F3 and F4.

Group I: F1 and F2									
FLEXPART simulation. Flight: F1. 12h-simulation			FLEXPART simulation. Flight: F2. 12h-simulation				ation		
Altitude	0-500 m	500-1500 m	> 1500 m	Total	Altitude 0-500 m 500-1500 m > 1500 m			Total	
Altitude release: 200 m				Alt	tude release: 2	00 m			
Ocean	43%	38%	7%	88%	Ocean	25%	<u>44%</u>	24%	93%
Island	2%	<u>6%</u>	4%	12%	Island	3%	3%	0.0%	7%
	Alti	tude release: 10	000 m			Alti	tude release: 10	000 m	
Ocean	29%	37%	11%	77%	Ocean	14%	51%	33%	98%
Island	4%	17%	2%	23%	Island	0	0	0	0
Altitude release: 1500 m				Alti	tude release: 15	500 m			
Ocean	13%	29%	31%	73%	Ocean	2%	37%	61%	100%
Island	3%	<u>19%</u>	15%	27%	Island	0	0	0	0
				Group II:	F4 and F5				
FLEX	KPART simu	ılation. Flight: I	F4. 12h-simul	ation	FLEXPART simulation. Flight: F5. 12h-simulation				ation
Altitude	0-500 m	500-1500 m	> 1500 m	Total	Altitude	0-500 m	500-1500 m	> 1500 m	Total
	Alt	itude release: 2	00 m		Altitude release: 200 m				
Ocean	23%	15%	4%	43%	Ocean	33%	28%	6%	67%
Island	28%	21%	8%	57%	Island	4%	<u>14%</u>	13%	33%
	Alti	tude release: 10)00 m		Altitude release: 1000 m				
Ocean	7%	21%	11%	39%	Ocean	12%	27%	11%	50%
Island	18%	33%	10%	61%	Island	6%	31%	13%	50%
	Alti	tude release: 15	i00 m			Alti	tude release: 15	500 m	
Ocean	1%	20%	20%	61%	Ocean	1%	15%	23%	41%
Island	0	22%	17%	39%	Island	5%	<u>27%</u>	24%	59%

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FLEXPART simulation. Flights: F3 and F6

Table A2

Same as Table A1 but for F3 and F6.

F3 (Group I) and F6							
FLEXPART simulation. Flight: F3							
Altitude	0-500 m 500-1500 m > 1500 m Tota						
Altitude release: 200 m							
Ocean	43%	38%	15%	96%			
Island	2%	1%	1%	4%			
	Alti	tude release: 10	000 m	1			
Ocean	19%	42%	27%	88%			
Island	2%	7%	4%	12%			
	Alti	tude release: 15	600 m	8			
Ocean	8%	41%	39%	88%			
Island	2%	<u>8%</u>	3%	13%			
FLEX	(PART simi	ulation. Flight:	F6 12h-simul	ation a			
	Alti	tude release: 15	i00 m				
Ocean	0.0%	35%	65%	100%			
Island	0.0%	0.0%	0.0%	0.0%			

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