

# High occurrence of new particle formation events at the Maïdo high altitude observatory (2150 m), Reunion Island (Indian Ocean)

Brice Foucart<sup>1, 2</sup>, Karine Sellegri<sup>2</sup>, Pierre Tulet<sup>1</sup>, Clémence Rose<sup>2</sup>, Jean-Marc Metzger<sup>3</sup>, and David Picard<sup>2</sup>

<sup>1</sup>Laboratoire de l'Atmosphère et des Cyclones (LACy-UMR 8015, CNRS, Université de La Réunion, Météo-France), 97744 Saint Denis de La Réunion, France.

<sup>2</sup>Laboratoire de Météorologie Physique (LaMP-UMR 6016, CNRS, Université Blaise Pascal), 63178, Aubière, France.

<sup>3</sup>Observatoire des Sciences de l'Univers de La Réunion, UMS 3365 (CNRS, Université de La Réunion), 97744 Saint Denis de La Réunion, France.

Correspondence to: Brice Foucart ([brice.foucart@univ-reunion.fr](mailto:brice.foucart@univ-reunion.fr)) and Karine Sellegri ([k.sellegri@opgc.univ-bpclermont.fr](mailto:k.sellegri@opgc.univ-bpclermont.fr))

**Abstract.** This study aims to report and characterize the frequent new particle formation (NPF) events observed at the Maïdo observatory, Reunion Island, a Southern Hemisphere site located at 2150 m (a.s.l.) and surrounded by the Indian Ocean. In 2014 and 2015, continuous aerosol measurements were made using both a Differential Mobility Particle Sizer (DMPS) and an Air Ion Spectrometer (AIS) to characterize the NPF events down to the lowest particle size scale. Carbon monoxide (CO) and Black Carbon (BC) concentrations were monitored, as well as meteorological parameters, in order to identify the conditions that were favourable to the occurrence of nucleation in this specific environment. We point out that the annual NPF frequency average (65%) is one of the highest reported so far. Monthly averages show a bimodal variation of the NPF frequency, with a maximum observed during transitional periods (autumn and spring). A high yearly median particle Growth Rate (GR) of 15.16 nm.h<sup>-1</sup> is also measured, occasionally peaking at values of the order of 100 nm.h<sup>-1</sup> and showing a bimodal seasonal variation with maxima observed in July and November. Yearly medians of 2 and 12 nm particle formation rates ( $J_2$  and  $J_{12}$ ) are 0.858 and 0.508 cm<sup>-3</sup>.s<sup>-1</sup> respectively, with a seasonal variation showing a maximum during winter, that correspond to low temperature and RH typical of the dry season, but also to high BC concentrations. We show that the condensation sink exceeds a threshold value ( $1.04 \times 10^{-3} \text{ s}^{-1}$ ) with a similar seasonal variation than the one of the NPF event frequency, suggesting that the occurrence of the NPF process might be determined by the availability of condensable vapours, which are likely to be transported together with pre-existing particles from lower altitudes.

## 1 Introduction

Aerosol concentrations in the atmosphere influence the Earth's radiative balance, and the formation and lifetime of clouds (Seinfeld and Pandis, 2016; Makkonen et al., 2012). Unlike the primary sources of aerosols, such as soil erosion, sea salt, and volcanic ash, nucleation is a gas-to-particle conversion process leading to the formation of new secondary aerosol particles. Nucleation and subsequent growth are responsible for New Particle Formation (NPF) events, observed in various environments around the world (Kulmala et al., 2004) but still rarely in the southern hemisphere. The frequency, intensity and duration of NPF events is highly variable according to the location where they are observed. The occurrence and characteristics of NPF episodes depend on various factors,

including the emission strength of precursors, the number concentration of the pre-existing aerosol population, and meteorological parameters (in particular solar radiation, temperature and relative humidity), which directly influence photo-chemical processes (Kulmala, 2003; Martin et al., 2010; Hallar et al., 2016). However, the relationship between these environmental parameters and the characteristics of NPF events is not fully understood and it is still a challenge to predict when an NPF event will take place and how intense it will be (Kulmala et al., 2004; Yu et al., 2008). Consequently, there is still a need to report and describe NPF in environments that have not yet been investigated, notably in the Southern Hemisphere in general and more particularly for both marine and high altitude tropical sites. These kinds of environments present true specificity in terms of RH variability, emission types (Volatile Organic Compounds (VOCs) and Marine sources) and atmospheric dynamics. Previous studies that have been conducted in the Southern Hemisphere relate to South Africa (Hirsikko et al., 2012), which is both a low latitude and a medium altitude (1400 m) site, South America (Rose et al., 2015b) and Australia (Bates et al., 1998), which are medium latitude, and Antarctica (Koponen et al., 2003) as a high latitude area. Hirsikko et al. (2012) published the highest NPF frequency ever reported, of about 86%, explaining that both the local sources (strong mining pollution) and regional conditions affected the NPF variation. Recently, Rose et al. (2015b) proposed a low-latitude zone analysis of NPF at Chacaltaya (CHC) in Bolivia, which is one of the highest in situ measurement sites in the world (5240 m). They, too, found a very high NPF frequency, of about 63.9%. This value has been partly explained by lower concentrations of pre-existing particles than at lower altitudes, leading to smaller loss of gaseous precursors, while photochemical activity is enhanced by higher radiation. In addition, at high altitude stations, turbulence at the interface between the Boundary Layer (BL) and the Free Troposphere (FT) might promote nucleation and growth processes (Hamburger et al., 2011). In general, high altitude sites report relatively high NPF event frequencies, such as 35% at Nepal Climate Observatory (5079 m; Venzac et al., 2008) or 35.9% at the Puy de Dôme station (1465 m; Boulon et al., 2011a). Reunion Island, which is located at low latitude (21 °S) in an inter-tropical area surrounded by the Indian Ocean, is still poorly documented. The island, which was partly shaped by the active basaltic volcano of Piton de la Fournaise (PdF), is characterized by angular landforms and steep slopes. The interaction of the high mountainous terrain with the synoptic flow induces large variability in wind fields at local scale. The maritime and tropical location of the island combined with the complexity of the terrain and wind exposure implies a multitude of local circulations and weather, marked by large variations in temperature and precipitation. This complex atmospheric dynamics added to a large variety of primary and secondary NPF sources (marine, organic and anthropogenic) gives special interest to this study. The Maïdo observatory is located at 2150 m a.s.l., under the influence of the marine boundary layer during daytime and of the free troposphere during nighttime. The main objective of this study is to reinforce the observations of NPF events in the Southern Hemisphere and more particularly for a site that is both marine and at high altitude. We first describe how NPF was observed at the site by DMPS and AIS interpolation (Sect. 4.1.). Based on a multiyear data set of clusters and aerosol size distributions, we secondly report the frequency (Sect. 4.2.), the intensity (Sect. 4.3.) and the characteristics of the events, and describe their seasonality. Thirdly, we analyse their annual variations with respect to the meteorological parameters (Sect. 4.4) and the pre-existing particle concentration (Sect. 4.5.).

## 2 Characteristics of the Maïdo observatory

### 2.1 Geographical location and networks

Maïdo observatory (21.080° S 55.383° E) is situated on La Reunion Island in the Indian Ocean. There are very few multi-instrumented stations in the tropics, and particularly in the southern hemisphere (Baray et al., 2013), so the Maïdo observatory was built in 2012 to respond to the needs of major international networks like NDACC (Network for the Detection of Atmospheric Composition Change, <http://www.ndacc.org>) and ACTRIS (Aerosols, Clouds, and Trace gases Research Infra-Structure network). It is a high-altitude station (2150 m), which opens up new perspectives in upper troposphere and lower stratosphere studies. Belonging to the Global Atmosphere Watch regional network (GAW), it also conducts in situ measurements to characterize the atmospheric composition of the lower troposphere. The facility dominates, in the east, the natural amphitheatre of Mafate, characterized by lush tropical vegetation, and, in the west, the highland tamarind forests. The nearest urban areas are the coastal cities of Saint Paul and Le Port with 105.000 and 40.000 inhabitants respectively, located 13 and 15 km away from the Maïdo observatory (Fig. 1).

### 2.2 Large and local scale atmospheric dynamics

At a large scale, the island is located in the descending part of the South Hadley cell (Baldy et al., 1996). It is subject to the intertropical zone atmospheric circulation, which is characterized by a trade wind flow from the south-east in this lower layer, induced by the Hadley cell and accentuated by more zonal driving by the Walker circulation. This lower layer flow is limited in altitude (about 3 km) by westerly winds (westerlies), which constitute the return of the Hadley – Walker circulation. In terms of rainfall, Reunion Island is characterized by two seasons: the hot, wet season from January to March (southern summer) and the cold, dry season (southern winter), which is longer, lasting from May to November (Baray et al., 2013). April and December are transition months that can be rainy or dry. In the southern summer, the Inter-Tropical Convergence Zone (ITCZ) is situated in the southern hemisphere and sometimes reaches Reunion Island. Over the inversion layer, the westerly flow (between 30°N and 30°S) and west winds weaken and are strongly affected by the context of heavy rains. During the southern winter, the subtropical high pressures are more powerful than during the southern summer and maintain the synoptic subsidence (descending branch of the Hadley cell) that generates and feeds both the stream of faster moving trade winds on the Mascarene region and the subtropical jet at high altitudes.

On a local scale, it is possible to observe two major atmospheric phenomena on both sides of the island. Trade winds coming from the south-east are separated (Fig.1) by the high topography of Reunion, which acts as an obstacle (Lesouëf et al., 2011). They are confined under the inversion layer and forced to bypass the island, forming two branches (Soler, 2000). During the day, a returning loop (Fig. 1) forms in the north-east when the winds converge. The inversion of the circulation then brings the winds under the cities of Saint Paul and Le Port up to the heights towards the Maïdo station. Moreover, the interaction between the trade winds and the abrupt relief of Reunion creates strong climate asymmetry and many microclimates. The warming of mountain slopes by solar radiation during the day, or radiative cooling overnight, is transmitted to the surrounding air layers and creates a complex local circulation. The nocturnal surface radiative cooling induces a cold katabatic wind on the slopes and clears the atmosphere at the Maïdo station, leaving the observatory in the free troposphere, disconnected from the anthropogenic pollution. Thus there are few clouds during the nighttime. After midday, the sea breeze cumulated

115 in the returning loop wind direction generates upward winds on the slopes which transport particles to the high station, accompanied by orographic and slope cloud formation. The surface radiative warming tends to create convection and then form vertical clouds located at the top of the relief. The number of nights with clear sky is then very large in comparison with the coastal site of Saint Denis, where LIDARs were operated from 1994 to 2011 (Baray et al., 2013).

### 2.3 Potential gas-phase precursor sources

120 The different environments that are present at local scale (Fig. 1) can correspond to different types of gas-phase precursor sources, each having different seasonal variations that can be investigated.

#### 2.3.1 Sulfur dioxide (SO<sub>2</sub>) and sulfuric acid (H<sub>2</sub>SO<sub>4</sub>)

Several studies have provided evidence that high SO<sub>2</sub> concentrations and high radiation levels favour the formation of large amounts of H<sub>2</sub>SO<sub>4</sub>, which in turn contribute to particle formation (Hyvönen et al., 2005; Mikkonen et al., 2006; Petäjä et al., 2009) and growth (Boy et al., 2005; Sihto et al., 2006; Mikkonen et al., 2011). Except for some altitude cases where the role of sulfuric acid in nucleation is limited (Boulon et al., 2011a; Rose et al., 2015b; Bianchi et al., 2016), H<sub>2</sub>SO<sub>4</sub> is thought to be among the major precursors of NPF due to its low saturating vapour pressure under conventional atmospheric temperature conditions (Kulmala and Kerminen, 2008). The Maïdo observatory can be on the pathway of sporadic SO<sub>2</sub> volcanic plumes emitted from the Piton de la Fournaise (PdF) volcano, located in the south of the island (Fig 1). In 2015, four eruptions were observed (Peltier et al., 2016) and multidisciplinary tracking of a volcanic gas and aerosol plume was conducted by Tulet et al., 2017. Unfortunately, H<sub>2</sub>SO<sub>4</sub> was not measured but their results indicated that the Maïdo station was reached by the plume several times, as evidenced by the detection of SO<sub>2</sub> concentration peaks (Fig. A1). Specifically in a volcanic plume environment, Boulon et al. (2011b) directly observed NPF events within the Eyjafjallajökull volcanic plume that reached the Puy de Dôme station, and related them to the presence of high H<sub>2</sub>SO<sub>4</sub> concentrations. During the PdF eruption that took place in April 2007, Tulet and Villeneuve (2011) used OMI and CALIOP space sensor data to estimate a total SO<sub>2</sub> release of 230 kt, 60 kt of which was transformed into H<sub>2</sub>SO<sub>4</sub>, mostly above the Indian ocean, at 6 km a.s.l. As a first analysis, we focus on the parameters influencing new particle formation (NPF) processes at the Maïdo station outside the very specific conditions encountered during a volcanic plume advection, which will be the topic of a separate study. Data were therefore screened for the presence of the Piton de la Fournaise plume at the station. The volcanic plume was considered to be present when the SO<sub>2</sub> concentration reached values higher than 1 ppb (hourly average), which is the 97th percentile estimated on non-eruptive days. According to Boulon et al. (2011b), this threshold also represented a lower limit of the presence of the plume at the Puy de Dôme station. Consequently, 47 daytime plume days that occurred during three eruptive periods (Fig. A1) were listed and removed from the 2015 data set. In addition to volcano eruptions, the major causes of SO<sub>2</sub> emissions are connected with human activities: agriculture, power plants, sugar exploitation and road traffic. Because of the relief of the island, urbanized areas are mostly located on the coast (Fig. 1). Reunion has a relatively developed urban fabric with eight cities having over 30.000 residents. For several decades population growth has been strong and urban pollution has increased in consequence.

### 150 2.3.2 Ammonia (NH<sub>3</sub>) and amines

Ammonia can act as a stabilizing chemical base for H<sub>2</sub>SO<sub>4</sub> in ternary nucleation theory. NH<sub>3</sub> can be derived from chemical fertilizers, industrial (Ge et al., 2011) and especially bovine waste (Hutchinson et al., 1982; Schade and Crutzen, 1995), decomposition of organic matter in the soil by certain bacteria (Gale, 1940) and biomass combustion. Agriculture occupies 10% of the working population of Reunion and agricultural territory covers 20%  
155 of the island (National Institute of Statistical Economic Studies, INSEE, 2017) mostly on gentle slopes (Fig. 1). Consequently, there are potential sources of ammonia that could influence the observations at the Maïdo observatory but we are not able to verify their existence because no direct measurements have been made. Amines originating from the alkylation of ammonia can be found in the atmosphere in the primary, secondary or tertiary state. For a time, significant concentrations of dimethylamine detected during the formation of nanoparticles in a  
160 natural atmosphere suggested that alkylamines could contribute to the formation and growth of new particles (Sellegri et al., 2006; Smith et al., 2010). Recently, it has been shown that amines have some weight in the calculation of nucleation rates since they act as strong bases and are at the origin of clusters that are more stable and more resistant to evaporation (Almeida et al., 2013).

### 2.3.3 Volatile Organic Components (VOCs)

165 Several studies have shown that oxidized monoterpenes can act as precursors of NPF events (Kulmala et al., 2013; Schobesberger et al., 2013; Ehn et al., 2014). In 2014, the CLOUD experiment performed at CERN confirmed that oxidation products of biogenic emissions could contribute to the nucleation of atmospheric particles (Riccobono et al., 2014). Plants are able to emit biosynthesized Volatile Organic Compounds (VOCs) into the atmosphere (Kesselmeier and Staudt, 1999), such as terpenes, which govern the correlation between sulfuric acid concentration  
170 and new particle formations (Bonn et al., 2008). The H<sub>2</sub>SO<sub>4</sub>-Organics nucleation strongly depends on temperature variation according to Yu et al. (2017). Furthermore, Kirkby et al. (2016) also show that  $\alpha$ -Pinene oxidation products can nucleate without sulfuric acid. As mentioned earlier, the Maïdo observatory is located above the caldera and at the top of the steep slopes characterized by dense, specific tropical vegetation (Fig. 1). In order to study the interactions between forests, gases, aerosols and clouds, the OCTAVE campaign will take place in 2018  
175 to obtain further information about the transport and chemical transformation of biogenic compounds at the Maïdo observatory.

### 2.3.4 Phytoplankton

Using semi controlled seawater-air enclosures, Sellegri et al. (2016) presented evidence that nucleation may occur from marine biological emissions in the open Mediterranean Sea. They identified iodine-containing species as  
180 major precursors for the formation of new particle clusters. They also found a significant correlation between iodine-containing species and some phytoplanktonic pigments (peridinin, chlorophyll b, and zeaxanthin) but not with the total autotrophic biomass (Chl a). There is seasonal and spatial variability in phytoplankton biomass in the north-western Indian Ocean (Veldhuis et al., 1997) and more particularly on the Somalian coast during a strong upwelling that occurred between 7°N and 11°N in July. Although the Indian Ocean is an oligotrophic ocean, the  
185 coasts of Reunion are sometimes subject to small phytoplanktonic blooms after severe rainfall periods, which warm the water and produce upwellings. It also appears that, during the southern winter, the island has already

been on the pathway of the nutrient plume of the Indian Ocean gyre, which is composed of both the South Equatorial and the West Australian Currents (Turquet et al., 2008)

#### 2.3.4 Biomass burning

190 Due to its location, Reunion Island is seasonally exposed to biomass burning plumes from the African continent and Madagascar, which can significantly affect the free tropospheric concentrations of ozone (Clain et al., 2009) and other pollutants like carbon monoxide and several volatile organic compounds (Dufлот et al., 2010; Vigouroux et al., 2012). This kind of event can either inhibit or stimulate the nucleation process. Primary particulate pollutants are compounded by the emission of high levels of secondary aerosol precursors, including oxides of nitrogen and  
195 sulfur, volatile carbon species, and ammonia, resulting in the production of large amounts of secondary inorganic and organic aerosols (Rastogi et al., 2014). The organic ones could be oxidized and act as a gas precursor whereas pollutants can be considered as pre-existing particles and can prevent new particles from forming. In 2015, the carbon monoxide variation seems to show a typical signature of such pollution events at Maïdo observatory (Fig. 2). The annual average concentration is about 63.7 ppb and the lowest value is 27.7 ppb, recorded on 02 February,  
200 whereas the highest is 141.4 ppb attributed to 04 September (daily averaged). It clearly appears that there is an increasing trend from May to December. Concentration peaks exceeding 0.8 ppm are visible in August, September, October and November and can be compared to the tropospheric ozone (O<sub>3</sub>) content variation observed by Taupin et al. (1999), who recorded high values from August to December with peaks in October as observed by Randriambelo et al. (1997), which correspond to biomass burning plumes from Madagascar or from South Africa.

#### 205 2.4 Instrumentation used

The aerosol and ion size distributions used in the present study were measured continuously from 01 January to 31 December 2015 at the Maïdo observatory. The size distribution of the 10-500 nm aerosol particles was measured with a Differential Mobility Particle Sizer (DMPS) while the size distribution of the 0.8-42 nm ions was measured with an Air Ion Spectrometer (AIS). Here we use ion size distributions below 10 nm as tracers for the  
210 presence of neutral sub-10 nm particles that could not be detected directly. Additional DMPS measurements conducted between May and December 2014 will also be discussed briefly in section 4.1 to evaluate the inter-annual variability of the nucleation frequency.

The DMPS was custom-built with a TSI-type Differential Mobility Analyzer (DMA) operating in a closed loop and a Condensation Particle Counter (CPC, TSI model 3010). Particles were charged to equilibrium using an Ni-63 bipolar charger at 95 MBq. The quality of the DMPS measurements was checked for flow rates and RH according to the ACTRIS recommendations (Wiedensohler et al., 2012). DMPS measurements were performed  
215 down a Whole Air Inlet with a higher size cut-off of 25 µm (under average wind speed conditions of 4 m.s<sup>-1</sup>).

The AIS was developed by Airel, Estonia, for in situ high time resolution measurements of ions and charged particles (Mäkelä et al., 1996 and Mirme et al., 2007). The device consists of two DMA arranged in parallel, which  
220 allows the simultaneous measurement of both negatively and positively charged particles. Each of the two analysers operates with a total flow of 90 litres per minute (lpm): 30 lpm of air to be sampled and 60 lpm of clean air (or carrier gas) circulating in closed loop. The AIS was directly connected to ambient air through a 30 cm long copper inlet 2.5 cm in diameter, to limit cluster ion losses along the sampling line.

225 The global radiation was measured using a Sunshine Pyranometer (SPN1, Delta-T Devices Ltd.) with a resolution  
of 0.6 W.m<sup>2</sup>. The auxiliary measurements used in the present study were the wind direction, the wind speed, the  
230 air temperature, the barometric pressure and the relative humidity. They were measured using the Vaisala Weather  
Transmitter WXT510 (<http://www.vaisala.com>).

The analyser used to measure sulfur dioxide (SO<sub>2</sub>) concentrations uses the ultraviolet fluorescence method,  
standard NF EN 14212. The molecules are excited under the action of intense, constant UV radiation (214 nm).  
235 Sulfur dioxide then de-energizes very quickly by emitting higher wavelength radiation (between 320 and 380 nm)  
than the excitation step. SO<sub>2</sub> concentration was finally calculated by means of a photomultiplier. Datasets were  
provided by the Observatoire Réunionnais de l'Air (ORA). The SO<sub>2</sub> analyser resolution was about 0.5 ppb and,  
outside eruptive periods, it never exceeded this threshold (Fig. A1). The corresponding data were used only to list  
days that were characterized by the presence of the volcanic plume at the Maïdo observatory.

235 CO monitoring was performed using a PICARRO G2401 analyzer which is compliant with international ambient  
atmospheric monitoring networks, including the World Meteorological Organization (WMO) and the Integrated  
Carbon Observation System (ICOS, <https://www.icos-ri.eu/>). It was the property of BIRA-IASB (Belgian Institute  
for Space Aeronomy).

240 Figure 3 shows the availability of data for the main aerosol and gas-phase parameters used in this study. The best  
instrument synchronization period was from May to November.

### 3 Calculations

The classification of event days was achieved visually using the contour plot of the DMPS size distribution. The  
positive and negative ion size distributions provided by the AIS confirmed the status of the event when available.  
Days were classified and separated into three main groups: undefined, (*UN*) non-event (*NE*) and event (*E*) days  
245 according to Dal Maso et al. (2005).

The monthly event frequency,  $f_m$ , was calculated as the ratio of event to non-event days, after having excluded  
undefined, missing and plume days (*PD*), according to Eq. (1):

$$f_m = \frac{\text{number of } E}{\text{number of days in month} - (\text{missing data days} + PD)} \times 100, \quad (1)$$

250 The condensation sink (*CS*; s<sup>-1</sup>), which represents the loss rate of vapours on pre-existing particles was calculated  
from the DMPS size distributions according to **Kulmala et al.'s (2001a)** Equation (2):

$$CS = 4\pi D_{vap} \int_0^{\infty} r \beta(r) N(r) dr \quad (2)$$

where  $D_{vap}$  is the condensable vapour diffusion coefficient,  $r$  the particle radius and  $N(r)$  the concentration of  
particles of radius  $r$ . Coefficient  $\beta(r)$  was calculated from the Knudsen number and is given by Eq. (3):

$$\beta(r) = \frac{1 + Kn(r)}{1 + Kn(r) \left( \frac{4}{3\alpha} + 0.337 \right) + \frac{4}{3\alpha} Kn(r)^2} \quad (3)$$

255 where  $Kn(r)$  is the Knudsen number given by  $Kn(r) = \lambda / r$ , with  $\lambda$  corresponding to the particle free path  
(depending on pressure and temperature) and the accommodation coefficient  $\alpha$ , usually set at 1. The condensation  
sink was calculated with a five minute time resolution. As the particles are **dried** in the DMPS, we are aware that  
the *CS*, which depends on the diameter of wet particles, was underestimated.

The particle growth rate (GR; nm.h<sup>-1</sup>) was determined using the "maxima" method of Hirsikko et al. (2005). The method searches, usually over the AIS channel size, for the time that corresponds to the maximum concentration in each size channel. As the DMPS offers a much more extended data set than the AIS, we applied the method to the DMPS 12-19 nm size range for which the lower limit (12) was defined by the smallest channel of the device. In order to detect the concentration maximum, a normal distribution was fitted to the time evolution of the concentration for each channel. GR corresponded to the slope of the linear regression on the time-diameter pairs. The formation rate of 12 nm particles,  $J_{12}$ , (cm<sup>-3</sup>.s<sup>-1</sup>) was calculated using the following Equation (4) given by Kulmala et al. (2007):

$$J_{12} = \frac{dN_{12-19}}{dt} + CoagS_{12} \times N_{12-19} + \frac{GR_{12-19}}{7 \text{ nm}} \times N_{12-19} \quad (4)$$

where  $N_{12-19}$  is the concentration corresponding to 12 to 19 nm particle diameters,  $CoagS_{12}$  represents the coagulation of 12 nm particles on pre-existing larger diameter particles and  $GR_{12-19}$  corresponds to the growth rate estimated between 12 and 19 nm. It was then possible to derive the nucleation rates of particles 2 nm in size,  $J_2$ , from the  $J_{12}$  previously calculated from DMPS and the growth rate of particles between 12 and 19 nm, following the method first introduced by Kerminen and Kulmala (2002) and improved by Lehtinen et al. (2007) with Eq. (5):

$$J_2 = \frac{J_{12}}{\exp\left(-\gamma \times d_2 \times \frac{CoagS(d_2)}{GR_{12-19}}\right)} \quad (5)$$

where,

$$\gamma = \frac{1}{m+1} \left[ \left( \frac{d_{12}}{d_2} \right)^{m+1} - 1 \right] \quad (6)$$

and,

$$m = \frac{\log(CoagS(d_{12})) - \log(CoagS(d_2))}{\log(d_{12}) - \log(d_2)} \quad (7)$$

## 4 Results and discussion

### 4.1 Dynamics of the NPF events at Maïdo observatory

The origin of the newly formed particles at a given site can be attributed to the local environment only if newly formed particles are detected at the smallest size range, when clusters just formed and had no time yet to grow. Thus, we used both the AIS and DMPS size distribution to investigate the evolution of NPF event at Maïdo. The AIS covers the size range between 0.90 and 46.2 nm while the DMPS covers the size range between 11.78 and 706.77 nm. Temporal interpolation was first performed to harmonize the DMPS and the AIS data sets to a 5 min resolution. For visual inspection of the consistency of the two data sets, hybrid plots were drawn up showing the AIS negative ion concentration up to 12 nm and then the particle concentration from the DMPS for larger sizes. Figure 4a shows an example of an NPF event followed by the two devices on 06 July, 2015. Typically, NPF events observed at the Maïdo observatory show an increase of small ion concentrations (2-5 nm) at dawn (06:00 UTC, corresponding to 10:00 LT). These small ions are tracers for small particles of the same size that rapidly grow to the first DMPS size classes within the next hour. The initiation of the formation of new particles at 06:00 UTC (10:00 LT) is followed by the appearance of accumulation mode particles. Further growth of the newly formed

particles is generally accompanied by the simultaneous growth of the accumulation mode particles, starting around 07:00 UTC (11:00 LT), that are likely representative of the updraft of boundary layer air to the station. We computed the diurnal variation of black carbon (BC), a good indicator of any anthropogenic, hence boundary layer, influence. The corresponding diurnal variations of BC ( $\text{ng.m}^{-3}$ ) is shown on Figure 4b. BC concentration clearly increases from 06:30 UTC (when ignoring early sharp peaks that may due to local contamination), which is half an hour later than the occurrence of the cluster mode particles. Hence we can hypothesise that boundary air convection to the site is a trigger for NPF events, most particularly when the interface BL/FT is sampled. At 07:00 UTC, as the accumulation particle concentration increase from 2000 to 8000  $\text{part.cm}^{-3}$ , the BC concentration also increase to reach 630  $\text{ng.m}^{-3}$  at 09:00, when the BL is fully sampled at the site. At the end of the afternoon, the accumulation mode particles concentrations drop to less than 1000  $\text{particle.cm}^{-3}$  and BC concentrations drop to very low values. Most high altitude stations are strongly influenced by free troposphere air during nighttime regardless the season, but mostly during wintertime (Venzac et al., 2008; Rose et al., 2015a). This is also true for stations located in complex terrains such as Jungfrauoch station in the Swiss Alps (Herrmann et al., 2015) and at the Chacaltaya station in the Andes (Rose et al., 2015b). These are indicators that the station lays in the free troposphere at night. The Aitken mode particles present during nighttime at the station are hence likely present in the free troposphere and are sampled at the site in subsiding air masses (Tulet et al., 2017). These features can also be observed on average, both for the summer and winter seasons. BC average diurnal profiles (a), together with the average diurnal variation of the number concentrations of particles larger than 100 nm ( $N_{100}$ ) (b), and the number concentration of nucleation mode particles with a diameter smaller than 30 nm ( $N_{30}$ ) (c) are shown on Figure 5. We observe that, on average, BC concentrations increase in the morning at the same time as  $N_{100}$  and  $N_{30}$ , confirming the influence of the BL on the occurrence of NPF events at the scale of the season. Moreover, we can note that during winter, BC concentrations are higher during nighttime (from 16:00 to 06:00 UTC) than during summer. This observation is also true for  $N_{100}$  with higher values from 17:00 to 02:00 UTC during winter compared to summer. We assume that during winter, trade winds favour the large scale remote primary particles transport to the Maïdo station.

#### 4.2 Nucleation and frequency analysis

Over the measurement period in 2015, 47 volcanic plume days were excluded and data was missing on 61 days. Among the 257 remaining days, 167 days (65%) were classified as event days, 55 (21%) as non-event days and 35 (14%) as undefined. As a result, the event frequency was high for the Maïdo station, with an annual average of 65% (med: 65.2%; 25ile: 52.0%; 75ile: 80.0%) for 2015. This frequency is one of the highest values reported so far, with the exceptions of the South African plateau, where NPF was reported to occur 86% of the time according to (Hirsikko et al., 2012), and savannah, with 69% of the time (Vakkari et al., 2011). Figure 6 shows the seasonal variation of the monthly event frequency,  $f_m$ . As shown in Figure 6, similar seasonal variations were observed for the nucleation frequency in 2014 and part of 2015. High NPF frequencies were observed during the transition months being on average 72.5% for October and November (spring), and even slightly higher, 89.4%, for March to May (autumn). During the southern winter and summer seasons (from June to August and from January to February), NPF was lower. As examples of high and low frequency NPF months, continuous DMPS spectra of April (93.1% occ.) and June (46.7% occ.) are available in the supplementary section though (Figure A4).

### 4.3 Particle formation, growth and nucleation rates

The yearly average particle growth rate for 12-19 nm particles was  $19.4 \pm 12.69 \text{ nm.h}^{-1}$  (Table 1), which is above the typical range of GRs reported in the literature for a quite-similar size class of 7-20 nm ( $\text{GR}_{7-20}$ ). We are aware that the different sizes that describe these classes can skew the comparison and partly explain a higher GR value at Maïdo compare to the other stations. The review by Yli-Juuti et al. (2011) of  $\text{GR}_{7-20}$ , obtained at different measurement sites located in various environments, reports a yearly average of  $6.66 \pm 3.41 \text{ nm.h}^{-1}$  (19 values). However, higher GRs have been observed for a coastal environment in Australia, with an average  $\text{GR}_{7-20}$  of  $19 \text{ nm.h}^{-1}$ , (Modini et al., 2009) and for a polluted urban environment in Tecamac, with an  $18 \text{ nm.h}^{-1}$  average  $\text{GR}_{3,7-25}$  (Iida et al., 2008). It is noteworthy that, at high altitudes, the conditions of spatially homogeneous air masses and a steady state, necessary to calculate a realistic growth rate, are not verified since air masses are progressively advected to the site from lower altitudes. Thus the GRs that are reported here are “apparent” growth rates that may be overestimated due to the transport of particles that have already nucleated and grown at lower altitudes at the same time. Nevertheless, the particle GR calculated for the Maïdo station is higher than the average GRs reported by Rose et al. (2015b) for the CHC station ( $7.62 \text{ nm.h}^{-1}$ ), Boulon et al. (2011a) for the Puy-de-Dôme station ( $6.20 \text{ nm.h}^{-1}$ ), and Venzac et al. (2008) for Nepal ( $1.8 \text{ nm.h}^{-1}$ ).

In our calculation, 19 events were not taken into account because of the special characteristics of the extreme value of GR. The beginning of the NPF was characterized by a clear verticality in the spectrum during the first hours of the event and the corresponding GR was generally very high ( $100 \text{ to } 150 \text{ nm.h}^{-1}$ ). As a supplementary, shows two examples of DMPS spectra belonging to this special class of growth rates are shown on Figure A2. Most of them were observed in December (8 cases). Dal Maso (2002) and O’Dowd and De Leeuw (2007) obtained such values ( $100 \text{ nm.h}^{-1}$ ) at Mace Head, a coastal site in western Ireland. They can be explained by the simultaneous transport of nucleated and already grown particles to the sampling site, from seaweed fields. In the case of coastal marine NPF events, the spatial homogeneity of the emission field is not verified, as for high altitude sites.

Figure 7 highlights a clear seasonal variation of  $\text{GR}_{12-19}$ , with the highest monthly averages in August ( $35 \text{ nm.h}^{-1}$ ) and the lowest in May ( $8.9 \text{ nm.h}^{-1}$ ). These variations of the GR differ from those reported in the literature for other high altitude sites. Boulon et al. (2011a), did not find a significant seasonal pattern in the GR variation at the Puy de Dôme. In Chacaltaya, Rose et al. (2015b) showed that, on average, the GRs were enhanced during the wet period, which is not in agreement with the present study, as we find high medians during the dry period ( $22.82 \text{ nm.h}^{-1}$  averaged from July to November). Moreover, the Table 2 shows that there is a good correlation between GR and CO of  $R = 0.638$  which is above the limit value of dependence that has been given by the Pearson table.

Formation rates were calculated for 12 and 2 nm particles when the  $\text{GR}_{12-19}$  was available. The yearly average formation rates  $J_{12}$  and  $J_2$  in Table 1 are respectively  $0.931 \pm 1.15$  and  $1.53 \pm 2.06 \text{ cm}^{-3}.\text{s}^{-1}$ . These formation rates are in the upper range of the values reported by Kulmala et al. (2004) from measurements performed in more than 100 locations in the boundary layer ( $J_3 = 0.01\text{-}10 \text{ cm}^{-3}.\text{s}^{-1}$ ). They are of the same order of magnitude as the ones reported for the CHC ( $1.02$  and  $1.90 \text{ cm}^{-3}.\text{s}^{-1}$  for the wet and dry seasons respectively, (Rose et al., 2015b).

$J_2$  seasonal variation follows the  $J_{12}$  seasonal variation (Fig. 8) but with higher values due to losses by coagulation during the growth process. We observe a clear seasonal cycle with maximum values during the dry season, particularly between July and September ( $J_{12} = 1.60 \text{ cm}^{-3}.\text{s}^{-1}$  and  $J_2 = 2.39 \text{ cm}^{-3}.\text{s}^{-1}$  respectively, averaged over 3 months). These observations are consistent with those reported for CHC, where  $J_2$  were reported to be twice as high during the dry season as in the wet season (Rose et al., 2015b). The lowest values are obtained around the

transition months of December, with  $J_{12} = 0.32 \text{ cm}^{-3} \cdot \text{s}^{-1}$  and  $J_2 = 0.52 \text{ cm}^{-3} \cdot \text{s}^{-1}$  (averaged between November and January), and April, with  $J_{12} = 0.44 \text{ cm}^{-3} \cdot \text{s}^{-1}$  and  $J_2 = 0.66 \text{ cm}^{-3} \cdot \text{s}^{-1}$  (averaged between April to June). The seasonal variation of the growth rate reported in Figure 7 also shows highest values around August but the seasonal variation of nucleation rates shows features different from those of the GR. Formation rates reach their maxima slightly earlier in the dry season (July) than the growth rates, and the contrast between July-August-Sept and the rest of the year is also stronger. This indicates that the condensable vapours necessary for nucleating new particles might not have exactly the same seasonal variation as the ones required for growing the newly formed particles. In addition, high particle formation and growth rates obtained in July-August do not coincide with the highest nucleation frequencies, suggesting that, during these months, NPF might be less frequent but occur in the form of stronger events.

Several factors have previously been reported to influence the seasonal variation of the NPF event frequency, GR, and nucleation rates; they include (i) the availability of condensable gases involved in the formation of new particles, (ii) the number concentration of pre-existing particles transported to the site and (iii) thermo-dynamical properties of the atmosphere, such as radiation, temperature, and relative humidity. In the following sections, we will explore the seasonal variation of the last two factors (ii and iii).

#### 4.4 Meteorological parameters and onset of NPF

A summary of incidental radiation, relative humidity, temperature and pressure monthly averages is available in Table A1. Austral seasons are reflected regarding both the daily averaged temperature and radiation represented on Figure 9a. We also computed in Table 2 the existing relationships between the monthly average meteorological parameters and the ones of the main characteristics of the NPF events. We observe that radiation is highest between September and November ( $272.19 \text{ W} \cdot \text{m}^{-2}$  on average), coinciding with one period of high NPF frequency (Fig. 5), but not with the maximum frequency of occurrence (March to May), nor with any high values of the GR or  $J_2/J_{12}$  (Figs. 7 and 8). As a consequence, no correlation is observed between radiation and the NPF variables. Hence, the availability of light for photochemistry is not the only parameter influencing the NPF frequency, nor the formation rates or growth. The temperature averages are higher from November to April ( $14.02 \text{ }^\circ\text{C}$ ). We find a significant (at the 95% confidence level) anti-correlation between temperature and the nucleation rate and GR. As mentioned earlier, this parameter would partly influence the VOC emissions (Yu et al., 2017) since it is one of the conditions for vegetation development and the decomposition of organic matter. Thus, the anti-correlation would indicate little influence of biogenic precursors from the vegetation on the intensity of the NPF and on the growth of the nucleated particles. However, the seasonal temperature variations are similar to the seasonal variation of the NPF event frequency even if the correlation is not significant. The relative humidity values are typical of an inter-tropical island with peaks in summer, between December and March (76.79% on average), and the lowest values obtained in July and September. Cloudy conditions were previously shown to inhibit formation of new particles, by scavenging newly formed clusters (Venzac et al., 2007). They might also decrease photochemical processes at the origin of the formation of condensable species contributing to the growth of clusters to stable particles. At Chacaltaya, Bolivia (5200 m a.s.l), Rose et al. (2015b) reported high frequencies during the southern winter, which coincide with the dry season. For the Maïdo station, frequency variations are not fully synchronized with the dry or wet periods as defined in Fig 9b. However, there is some uncertainty both in the dry/wet season segregation and with the exact identification of maxima/minima in the seasonal variation of the NPF frequency. When considering

410 relative humidity, we do not find any link between RH and the nucleation frequency (Table 2) but a significant  
anti-correlation with the formation rate: low RH values correspond to the July-August-September nucleation peak.  
Figure 9b shows that the appearance time of the ultrafine particles seasonal variation is well correlated to the  
sunrise. During the southern summer, NPF starts between approximately 08:00 and 10:00 LT and between 09:00  
and 10:00 during the southern winter. This correlation may be due to the need for sunlight to be available to start  
415 photochemical processes, or/and to the start of advection of precursor gases from lower altitude sources in the BL.

#### 4.5 Condensation sink

In addition to the meteorological parameters, the seasonal variation of the NPF characteristics might also be  
influenced by the presence of pre-existing particles, known to inhibit the NPF processes by increasing the  
competition for available condensable gases. We averaged the CS for two hours before the nucleation started (CS<sub>2</sub>)  
420 to properly characterize its influence on the occurrence of an NPF event. In Figure 9b, nucleation onset times are  
averaged for each season (08:00 LT for southern summer and 09:00 LT for southern winter). The yearly average  
condensation sink has been calculated to be  $2.43 \times 10^{-3} \text{ s}^{-1}$  and  $1.86 \times 10^{-3} \text{ s}^{-1}$  for CS<sub>2</sub>. These values are similar to  
the ones reported for the altitude station of Chacaltaya (Rose et al., 2015b) and Nepal (Venzac et al., 2008) which  
are  $2.4 \times 10^{-3} \text{ s}^{-1}$  and  $2.1 \times 10^{-3} \text{ s}^{-1}$  respectively and also for the Mace Head coastal station (Dal Maso, 2002) which  
425 is about  $2 \times 10^{-3} \text{ s}^{-1}$ .

Monthly averages of the CS<sub>2</sub> were calculated for event days and non-event days and are shown for 2015 on Figure  
10a, together with the NPF event monthly frequency  $f_m$ . This representation highlights monthly averaged CS<sub>2</sub>  
peaks for February ( $2.65 \times 10^{-3} \text{ s}^{-1}$ ), May ( $3.74 \times 10^{-3} \text{ s}^{-1}$ ) and September ( $4.72 \times 10^{-3} \text{ s}^{-1}$ ). The September value is  
similar to the South African savannah yearly average (Vakkari et al., 2011), which was about  $4.3 \times 10^{-3} \text{ s}^{-1}$ . It attests  
430 to a considerable presence of pre-existing particles at the Maïdo station for this period. However, the NPF  
frequency seasonal pattern does not match that of low CS<sub>2</sub>. May and September CS<sub>2</sub> peaks are associated with  $f_m$   
peak values while January and June CS<sub>2</sub> low averages ( $0.89$  and  $0.96 \times 10^{-3} \text{ s}^{-1}$  respectively) are correlated to weak  
NPF occurrence.

Moreover, we calculated that the annual CS<sub>2ev</sub> (event) median ( $1.2 \times 10^{-3} \text{ s}^{-1}$ ) was significantly higher than the  
435 annual CS<sub>2noev</sub> (no event) median ( $8.5 \times 10^{-4} \text{ s}^{-1}$ ). The previous observations thus suggest that the condensation sink  
does not inhibit NPF at Maïdo, as previously reported for other high altitude stations (Manninen et al., 2010;  
Boulon et al., 2010; Rose et al., 2015a). At these sites, the occurrence of the NPF process might be determined  
rather by the availability of condensable vapours, which are likely to be transported together with pre-existing  
particles from lower altitudes. In order to further investigate this aspect, we evaluated whether the frequency of  
440 nucleation was correlated to a frequency of exceeding a CS threshold. Hence, we calculated a monthly average  
frequency for which the CS<sub>2</sub> exceeded a threshold value of  $1.04 \times 10^{-3} \text{ s}^{-1}$ . The threshold was chosen arbitrarily as  
a value intermediate between the annual CS<sub>2ev</sub> and CS<sub>2noev</sub> medians. We started our investigation by choosing the  
median CS over the whole year as a threshold value. We then used an iteration process to fix the threshold so we  
have a reasonable seasonal variation of days exceeding this threshold values. Choosing other threshold values  
445 within CS<sub>2ev</sub> and CS<sub>2noev</sub> medians range would have led to less pronounced variability but would not have changed  
the shape of the seasonal trend. The resulting frequency at which CS<sub>2</sub> exceeded the threshold (Fig. 10b) had a clear  
seasonal variation with maxima during the transitional periods. For April-May, and September-November, more  
than 60% of the CS<sub>2</sub> were higher than  $1.04 \times 10^{-3} \text{ s}^{-1}$ , while for January, June and July the frequency was lowest.

Hence, we actually find a similar seasonal variation between the frequency of CS exceeding a threshold value and the frequency of occurrence of NPF events (also shown by Table 2). This strengthens the hypothesis that there are precursors potentially transported simultaneous to aerosols from lower altitudes.

#### 4.6 Black Carbon as a tracer of the anthropogenic contribution

Potential precursors may be of anthropogenic origin and we investigated their potential contribution by using BC as a tracer. Figure 11 shows the monthly mean concentrations of BC over the period January-October 215. The best similarity between the seasonal variations of BC and the ones of NPF parameters is found for the nucleation rate (Table 2). This may indicate that air masses influenced by a larger contribution of anthropogenic compounds are favourable to more intense NPF events, but not necessarily more frequent. Indeed, moderately high NPF frequencies are observed for high BC monthly mean values during the spring period, but the high frequencies of NPF event occurrence observed during the autumn period are not associated to very large BC monthly mean concentrations, and thus other contributors are expected during autumn. The growth rate of newly formed particles is best correlated to CO concentrations (Table 2). Condensable species necessary to form new particles are hence likely different from the ones responsible for their further growth.

#### 5 Conclusions

In the present study, we provide a new NPF observations for a high altitude site of the southern hemisphere, the Maïdo observatory. We report a remarkably high frequency of occurrence of NPF events at the Maïdo observatory (65%) with a bimodal seasonal variation of this frequency, characterized by high values during spring and autumn. We show that the condensation sink exceeds a threshold value ( $1.04 \times 10^{-3} \text{ s}^{-1}$ ) with a similar seasonal variation than the one of the NPF event frequency, suggesting that, similarly to other altitude sites, the condensation sink does not inhibit NPF at Maïdo, but the occurrence of the NPF process might be determined rather by the availability of condensable vapours, which are likely to be transported together with pre-existing particles from lower altitudes during the day. This indicates an influence of the boundary layer dynamics, confirmed by daily variations of boundary layer tracers ( $N_{100}$  and BC). The monthly average BC concentrations, taken as one anthropogenic tracer, has a main peak during winter and hence they match only a fraction of NPF frequency. Thus, anthropogenic precursors may contribute to some of the springtime NPF occurrence, but the high NPF occurrence during the autumn season is likely initiated by other contributors. The seasonal variations of the formation rate and growth rate are not correlated to the NPF frequency seasonal variation. High formation rates correspond to high BC concentrations, low relative humidity conditions and low temperatures. While annual average  $J_{12}$  and  $J_2$  are in the typical ranges found in the literature ( $9.31 \times 10^{-2} \pm 1.15$  and  $1.53 \pm 2.06 \text{ cm}^{-3} \cdot \text{s}^{-1}$  respectively),  $GR_{12-19}$  values are higher than the typical range of GRs reported in the literature ( $19.4 \pm 12.69 \text{ nm} \cdot \text{h}^{-1}$ ). At Reunion Island, the secondary emissions assessment is not established and sources are not well-identified. To complete this work, it would be valuable to have direct measures of the cluster ion composition that would provide indication of the anthropogenic, vegetation or marine sources contributions to nucleation at the site. In addition and although they are complex, modelling methods such as a detailed back-trajectory analysis should be used to understand the origin of the local air masses and source contributions at the Maïdo observatory.

## 485 Acknowledgments

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 654109 (ACTRIS-2), from the French programme SNO-CLAP, and from the OMNCG/OSU-R programme of La Réunion University. We also wish to thank the ORA (Observatoire Réunionnais de l'Air) for providing the SO<sub>2</sub> dataset as Jean SCIARE who provides the processed data from the aethalometer.

## References

- Almeida, J., Downard, A. J., Flagan, R. C. and Seinfeld, J. H.: Molecular understanding of sulphuric acid–amine particle nucleation in the atmosphere, *Nature*, 502(7471), 359–363, 2013.
- 495 Baldy, S., Ancellet, G., Bessafi, M., Badr, A. and Luk, D. L. S.: Field observations of the vertical distribution of tropospheric ozone at the island of Reunion (southern tropics), *J. Geophys. Res. Atmospheres*, 101(D19), 23835–23849, doi:10.1029/95JD02929, 1996.
- Baray, J.-L., Courcoux, Y., Keckhut, P., Portafaix, T., Tulet, P., Cammas, J.-P., Hauchecorne, A., Godin-Beekmann, S., De Mazière, M., Hermans, C. and others: Maïdo observatory: a new high-altitude station facility at Reunion Island (21° S, 55° E) for long-term atmospheric remote sensing and in situ measurements, *Atmospheric Meas. Tech.*, 6(10), 2865–2877, 2013.
- 500 Bates, T. S., Kapustin, V. N., Quinn, P. K., Covert, D. S., Coffman, D. J., Mari, C., Durkee, P. A., De Bruyn, W. J. and Saltzman, E. S.: Processes controlling the distribution of aerosol particles in the lower marine boundary layer during the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res. Atmospheres*, 103(D13), 16369–16383, doi:10.1029/97JD03720, 1998.
- 505 Bianchi, F., Tröstl, J., Junninen, H., Frege, C., Henne, S., Hoyle, C. R., Molteni, U., Herrmann, E., Adamov, A., Bukowiecki, N. and others: New particle formation in the free troposphere: A question of chemistry and timing, *Science*, 352(6289), 1109–1112, 2016.
- Bonn, B., Kulmala, M., Riipinen, I., Sihto, S.-L. and Ruuskanen, T. M.: How biogenic terpenes govern the correlation between sulfuric acid concentrations and new particle formation, *J. Geophys. Res. Atmospheres*, 113(D12) [online] Available from: <http://onlinelibrary.wiley.com/doi/10.1029/2007JD009327/full> (Accessed 24 August 2017), 2008.
- 510 Boulon, J., Sellegri, K., Venzac, H., Picard, D., Weingartner, E., Wehrle, G., Collaud Coen, M., Bütikofer, R., Flückiger, E., Baltensperger, U. and Laj, P.: New particle formation and ultrafine charged aerosol climatology at a high altitude site in the Alps (Jungfrauoch, 3580 m a.s.l., Switzerland), *Atmospheric Chem. Phys.*, 10(19), 9333–9349, doi:10.5194/acp-10-9333-2010, 2010.
- 515 Boulon, J., Sellegri, K., Hervo, M., Picard, D., Pichon, J.-M., Fréville, P. and Laj, P.: Investigation of nucleation events vertical extent: a long term study at two different altitude sites, *Atmospheric Chem. Phys.*, 11(12), 5625–5639, doi:10.5194/acp-11-5625-2011, 2011a.
- Boulon, J., Sellegri, K., Hervo, M. and Laj, P.: Observations of nucleation of new particles in a volcanic plume, *Proc. Natl. Acad. Sci.*, 108(30), 12223–12226, 2011b.
- 520 Boy, M., Kulmala, M., Ruuskanen, T. M., Pihlatie, M., Reissell, A., Aalto, P. P., Keronen, P., Maso, M. D., Hellen, H., Hakola, H. and others: Sulphuric acid closure and contribution to nucleation mode particle growth, *Atmospheric Chem. Phys.*, 5(4), 863–878, 2005.

- 525 Clain, G., Baray, J.-L., Delmas, R., Diab, R., Leclair De Bellevue, J., Keckhut, P., Posny, F., Metzger, J.-M. and Cammas, J.-P.: Tropospheric ozone climatology at two Southern Hemisphere tropical/subtropical sites, (Reunion Island and Irene, South Africa) from ozonesondes, LIDAR, and in situ aircraft measurements, *Atmospheric Chem. Phys.*, 9(5), 1723–1734, doi:10.5194/acp-9-1723-2009, 2009.
- D O’Dowd, C. and De Leeuw, G.: Marine aerosol production: a review of the current knowledge, *Philos. Trans. R. Soc. Lond. Math. Phys. Eng. Sci.*, 365(1856), 1753–1774, 2007.
- 530 Dal Maso, M.: Condensation and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers, *J. Geophys. Res.*, 107(D19), doi:10.1029/2001JD001053, 2002.
- 535 Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P. and Lehtinen, K. E.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Environ. Res.*, 10(5), 323, 2005.
- Duflot, V., Dils, B., Baray, J. L., De Mazière, M., Attié, J. L., Vanhaelewyn, G., Senten, C., Vigouroux, C., Clain, G. and Delmas, R.: Analysis of the origin of the distribution of CO in the subtropical southern Indian Ocean in 2007, *J. Geophys. Res. Atmospheres*, 115(D22), D22106, doi:10.1029/2010JD013994, 2010.
- 540 Ehn, M., Thornton, J. A., Kleist, E., Sipilä, M. and Junninen, H.: A large source of low-volatility secondary organic aerosol, *Nature*, 27th February, 20, 2014.
- Gale, E. F.: The production of amines by bacteria: The decarboxylation of amino-acids by strains of *Bacterium coli*, *Biochem. J.*, 34(3), 392, 1940.
- 545 Ge, X., Wexler, A. S. and Clegg, S. L.: Atmospheric amines – Part II. Thermodynamic properties and gas/particle partitioning, *Atmos. Environ.*, 45(3), 561–577, doi:10.1016/j.atmosenv.2010.10.013, 2011.
- Hallar, A. G., Petersen, R., McCubbin, I. B., Lowenthal, D., Lee, S., Andrews, E. and Yu, F.: Climatology of New Particle Formation and Corresponding Precursors at Storm Peak Laboratory, *Aerosol Air Qual. Res.*, 16(3), 816–826, doi:10.4209/aaqr.2015.05.0341, 2016.
- 550 Hamburger, T., McMeeking, G., Minikin, A., Birmili, W., Dall’Osto, M., O’Dowd, C., Flentje, H., Henzing, B., Junninen, H., Kristensson, A., de Leeuw, G., Stohl, A., Burkhardt, J. F., Coe, H., Krejci, R. and Petzold, A.: Overview of the synoptic and pollution situation over Europe during the EUCAARI-LONGREX field campaign, *Atmospheric Chem. Phys.*, 11(3), 1065–1082, doi:10.5194/acp-11-1065-2011, 2011.
- 555 Herrmann, E., Weingartner, E., Henne, S., Vuilleumier, L., Bukowiecki, N., Steinbacher, M., Conen, F., Collaud Coen, M., Hammer, E. and Jurányi, Z.: Analysis of long-term aerosol size distribution data from Jungfraujoch with emphasis on free tropospheric conditions, cloud influence, and air mass transport, *J. Geophys. Res. Atmospheres*, 120(18), 9459–9480, 2015.
- 560 Hirsikko, A., Laakso, L., Hörrak, U., Aalto, P. P., Kerminen, V. and Kulmala, M.: Annual and size dependent variation of growth rates and ion concentrations in boreal forest, *Boreal Environ. Res.*, 10(5), 357, 2005.
- Hirsikko, A., Vakkari, V., Tiitta, P., Manninen, H. E., Gagné, S., Laakso, H., Kulmala, M., Mirme, A., Mirme, S., Mabaso, D., Beukes, J. P. and Laakso, L.: Characterisation of sub-micron particle number

- 565 concentrations and formation events in the western Bushveld Igneous Complex, South Africa, *Atmospheric Chem. Phys.*, 12(9), 3951–3967, doi:10.5194/acp-12-3951-2012, 2012.
- Hutchinson, G. L., Mosier, A. R. and Andre, C. E.: Ammonia and Amine Emissions from a Large Cattle Feedlot, *J. Environ. Qual.*, 11(2), 288–293, doi:10.2134/jeq1982.00472425001100020028x, 1982.
- 570 Hyvönen, S., Junninen, H., Laakso, L., Dal Maso, M., Grönholm, T., Bonn, B., Keronen, P., Aalto, P., Hiltunen, V., Pohja, T., Launiainen, S., Hari, P., Mannila, H. and Kulmala, M.: A look at aerosol formation using data mining techniques, *Atmos Chem Phys*, 5(12), 3345–3356, doi:10.5194/acp-5-3345-2005, 2005.
- 575 Iida, K., Stolzenburg, M. R., McMurry, P. H. and Smith, J. N.: Estimating nanoparticle growth rates from size-dependent charged fractions: Analysis of new particle formation events in Mexico City, *J. Geophys. Res. Atmospheres*, 113(D5) [online] Available from: <http://onlinelibrary.wiley.com/doi/10.1029/2007JD009260/full> (Accessed 25 August 2017), 2008.
- Kerminen, V.-M. and Kulmala, M.: Analytical formulae connecting the “real” and the “apparent” nucleation rate and the nuclei number concentration for atmospheric nucleation events, *J. Aerosol Sci.*, 33(4), 609–622, doi:10.1016/S0021-8502(01)00194-X, 2002.
- 580 Kesselmeier, J. and Staudt, M.: Biogenic Volatile Organic Compounds (VOC): An Overview on Emission, Physiology and Ecology, *J. Atmospheric Chem.*, 33(1), 23–88, doi:10.1023/A:1006127516791, 1999.
- 585 Kirkby, J., Duplissy, J., Sengupta, K., Frege, C., Gordon, H., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., Tröstl, J., Nieminen, T., Ortega, I. K., Wagner, R., Adamov, A., Amorim, A., Bernhammer, A.-K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X., Craven, J., Dias, A., Ehrhart, S., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Krapf, M., Kürten, A., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Onnela, A., Peräkylä, O., Piel, F., Petäjä, T., Praplan, A. P., Pringle, K., Rap, A., Richards, N. A. D., Riipinen, I., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Scott, C. E., Seinfeld, J. H., Sipilä, M., Steiner, G., Stozhkov, Y., Stratmann, F., Tomé, A., Virtanen, A., Vogel, A. L., Wagner, A. C., Wagner, P. E., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M., Worsnop, D. R., Baltensperger, U., Kulmala, M., Carslaw, K. S. and Curtius, J.: Ion-induced nucleation of pure biogenic particles, *Nature*, 533(7604), 521–526, doi:10.1038/nature17953, 2016.
- 590 Koponen, I. K., Virkkula, A., Hillamo, R., Kerminen, V.-M. and Kulmala, M.: Number size distributions and concentrations of the continental summer aerosols in Queen Maud Land, Antarctica, *J. Geophys. Res. Atmospheres*, 108(D18) [online] Available from: <http://onlinelibrary.wiley.com/doi/10.1029/2003JD003614/full> (Accessed 24 August 2017), 2003.
- Kulmala, M.: ATMOSPHERIC SCIENCE: How Particles Nucleate and Grow, *Science*, 302(5647), 1000–1001, doi:10.1126/science.1090848, 2003.
- 600 Kulmala, M. and Kerminen, V.-M.: On the formation and growth of atmospheric nanoparticles, *Atmospheric Res.*, 90(2–4), 132–150, doi:10.1016/j.atmosres.2008.01.005, 2008.
- Kulmala, M., Maso, M. D., Mäkelä, J. M., Pirjola, L., Väkevää, M., Aalto, P., Miikkulainen, P., Hämeri, K. and O’ Dowd, C. D.: On the formation, growth and composition of nucleation mode particles, *Tellus B Chem. Phys. Meteorol.*, 53(4), 479–490, doi:10.3402/tellusb.v53i4.16622, 2001.

- 605 Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W. and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: a review of observations, *J. Aerosol Sci.*, 35(2), 143–176, doi:10.1016/j.jaerosci.2003.10.003, 2004.
- Kulmala, M., Riipinen, I., Sipilä, M., Manninen, H. E., Petäjä, T., Junninen, H., Maso, M. D., Mordas, G., Mirme, A., Vana, M., Hirsikko, A., Laakso, L., Harrison, R. M., Hanson, I., Leung, C., Lehtinen, K. E. J. and Kerminen, V.-M.: Toward Direct Measurement of Atmospheric Nucleation, *Science*, 318(5847), 89–92, doi:10.1126/science.1144124, 2007.
- 610 Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H. E., Nieminen, T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T., Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P. P., Paasonen, P., Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T., Mauldin, R. L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen, I., Kurtén, T., Johnston, M. V., Smith, J. N., Ehn, M., Mentel, T. F., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M. and Worsnop, D. R.: Direct Observations of Atmospheric Aerosol Nucleation, *Science*, 339(6122), 943–946, doi:10.1126/science.1227385, 2013.
- 615 Lehtinen, K. E. J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M.: Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen–Kulmala equation, *J. Aerosol Sci.*, 38(9), 988–994, doi:10.1016/j.jaerosci.2007.06.009, 2007.
- 620 Lesouëf, D., Gheusi, F., Delmas, R. and Escobar, J.: Numerical simulations of local circulations and pollution transport over Reunion Island, *Ann. Geophys.*, 29(1), 53–69, doi:10.5194/angeo-29-53-2011, 2011.
- 625 Mäkelä, J. M., Riihelä, M., Ukkonen, A., Jokinen, V. and Keskinen, J.: Comparison of mobility equivalent diameter with Kelvin-Thomson diameter using ion mobility data, *J. Chem. Phys.*, 105(4), 1562–1571, doi:10.1063/1.472017, 1996.
- Makkonen, R., Asmi, A., Kerminen, V.-M., Boy, M., Arneth, A., Hari, P. and Kulmala, M.: Air pollution control and decreasing new particle formation lead to strong climate warming, *Atmospheric Chem. Phys.*, 12(3), 1515–1524, doi:10.5194/acp-12-1515-2012, 2012.
- 630 Manninen, H. E., Nieminen, T., Asmi, E., Gagné, S., Häkkinen, S., Lehtipalo, K., Aalto, P., Vana, M., Mirme, A., Mirme, S., Hörrak, U., Plass-Dülmer, C., Stange, G., Kiss, G., Hoffer, A., Törő, N., Moerman, M., Henzing, B., de Leeuw, G., Brinkenberg, M., Kouvarakis, G. N., Bougiatioti, A., Mihalopoulos, N., O’Dowd, C., Ceburnis, D., Arneth, A., Svenningsson, B., Swietlicki, E., Tarozzi, L., Decesari, S., Facchini, M. C., Birmili, W., Sonntag, A., Wiedensohler, A., Boulon, J., Sellegri, K., Laj, P., Gysel, M., Bukowiecki, N., Weingartner, E., Wehrle, G., Laaksonen, A., Hamed, A., Joutsensaari, J., Petäjä, T., Kerminen, V.-M. and Kulmala, M.: EUCAARI ion spectrometer measurements at 12 European sites – analysis of new particle formation events, *Atmos Chem Phys*, 10(16), 7907–7927, doi:10.5194/acp-10-7907-2010, 2010.
- 635 Martin, S. T., Andreae, M. O., Artaxo, P., Baumgardner, D., Chen, Q., Goldstein, A. H., Guenther, A., Heald, C. L., Mayol-Bracero, O. L., McMurry, P. H., Pauliquevis, T., Pöschl, U., Prather, K. A., Roberts, G. C., Saleska, S. R., Silva Dias, M. A., Spracklen, D. V., Swietlicki, E. and Trebs, I.: Sources and properties of Amazonian aerosol particles, *Rev. Geophys.*, 48(2), RG2002, doi:10.1029/2008RG000280, 2010.
- 640 Mikkonen, S., Lehtinen, K. E., Hamed, A., Joutsensaari, J., Facchini, M. C. and Laaksonen, A.: Using discriminant analysis as a nucleation event classification method, *Atmospheric Chem. Phys.*, 6(12), 5549–5557, 2006.
- 645

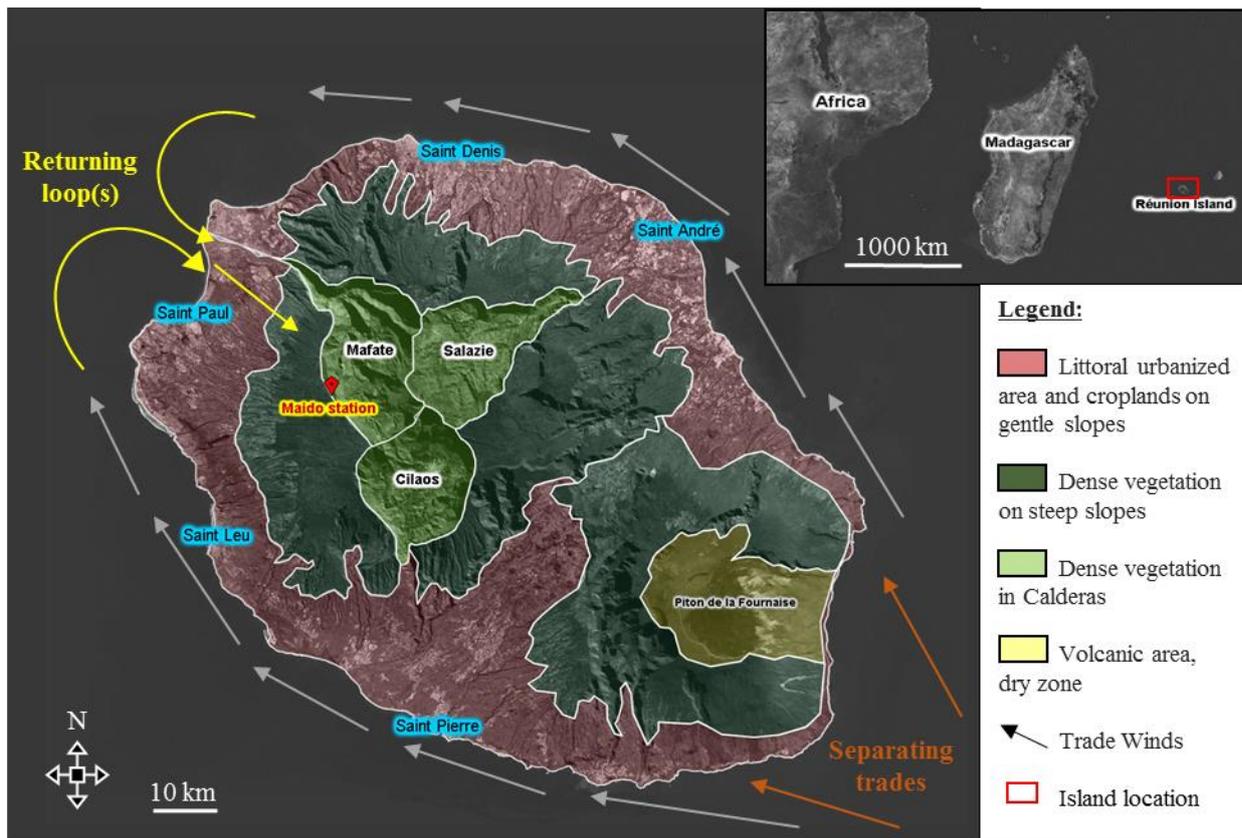
- 650 Mikkonen, S., Romakkaniemi, S., Smith, J. N., Korhonen, H., Petäjä, T., Plass-Duelmer, C., Boy, M., McMurry, P. H., Lehtinen, K. E. J., Joutsensaari, J., Hamed, A., Mauldin III, R. L., Birmili, W., Spindler, G., Arnold, F., Kulmala, M. and Laaksonen, A.: A statistical proxy for sulphuric acid concentration, *Atmospheric Chem. Phys.*, 11(21), 11319–11334, doi:10.5194/acp-11-11319-2011, 2011.
- Mirme, A., Tamm, E., Mordas, G., Vana, M., Uin, J., Mirme, S., Bernotas, T., Laakso, L., Hirsikko, A. and Kulmala, M.: A wide-range multi-channel Air Ion Spectrometer, *Boreal Environ.*, 12, 247–264, 2007.
- 655 Modini, R. L., Ristovski, Z. D., Johnson, G. R., He, C., Surawski, N., Morawska, L., Suni, T. and Kulmala, M.: New particle formation and growth at a remote, sub-tropical coastal location, *Atmospheric Chem. Phys.*, 9(19), 7607–7621, 2009.
- National Institute of Statistical Economic Studies, INSEE: INSEE Conjoncture Réunion: Bilan économique 2016, 40p, [online] Available from: <https://www.insee.fr/fr/statistiques/2856269>, 2017.
- 660 Peltier, A., Beauducel, F., Villeneuve, N., Ferrazzini, V., Di Muro, A., Aiuppa, A., Derrien, A., Jourde, K. and Taisne, B.: Deep fluid transfer evidenced by surface deformation during the 2014–2015 unrest at Piton de la Fournaise volcano, *J. Volcanol. Geotherm. Res.*, 321, 140–148, doi:10.1016/j.jvolgeores.2016.04.031, 2016.
- 665 Petäjä, T., Mauldin III, R. L., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M., Adamov, A., Kotiaho, T. and Kulmala, M.: Sulfuric acid and OH concentrations in a boreal forest site, *Atmospheric Chem. Phys.*, 9(19), 7435–7448, 2009.
- Randriambelo, T., Baldy, S. and Bessafi, M.: The seasonality of ozone profile at Reunion Island: the role of biomass burning and of transport, in *Geoscience and Remote Sensing, 1997. IGARSS '97. Remote Sensing - A Scientific Vision for Sustainable Development.*, 1997 IEEE International, vol. 1, pp. 6–8 vol.1., 1997.
- 670 Rastogi, N., Singh, A., Singh, D. and Sarin, M. M.: Chemical characteristics of PM<sub>2.5</sub> at a source region of biomass burning emissions: Evidence for secondary aerosol formation, *Environ. Pollut.*, 184, 563–569, doi:10.1016/j.envpol.2013.09.037, 2014.
- 675 Riccobono, F., Schobesberger, S., Scott, C. E., Dommen, J., Ortega, I. K., Rondo, L., Almeida, J., Amorim, A., Bianchi, F., Breitenlechner, M. and others: Oxidation products of biogenic emissions contribute to nucleation of atmospheric particles, *Science*, 344(6185), 717–721, 2014.
- Rose, C., Sellegri, K., Velarde, F., Moreno, I., Ramonet, M., Weinhold, K., Krejci, R., Ginot, P., Andrade, M., Wiedensohler, A. and Laj, P.: Frequent nucleation events at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia, *Atmos. Environ.*, 102, 18–29, doi:10.1016/j.atmosenv.2014.11.015, 2015a.
- 680 Rose, C., Sellegri, K., Velarde, F., Moreno, I., Ramonet, M., Weinhold, K., Krejci, R., Ginot, P., Andrade, M., Wiedensohler, A. and Laj, P.: Frequent nucleation events at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia, *Atmos. Environ.*, 102, 18–29, doi:10.1016/j.atmosenv.2014.11.015, 2015b.
- 685 Rose, C., Sellegri, K., Asmi, E., Hervo, M., Freney, E., Colomb, A., Junninen, H., Duplissy, J., Sipilä, M., Kontkanen, J., Lehtipalo, K. and Kulmala, M.: Major contribution of neutral clusters to new particle formation at the interface between the boundary layer and the free troposphere, *Atmos Chem Phys*, 15(6), 3413–3428, doi:10.5194/acp-15-3413-2015, 2015c.

- Schade, G. W. and Crutzen, P. J.: Emission of aliphatic amines from animal husbandry and their reactions: Potential source of N<sub>2</sub>O and HCN, *J. Atmospheric Chem.*, 22(3), 319–346, 1995.
- 690 Schobesberger, S., Junninen, H., Bianchi, F., Lönn, G., Ehn, M., Lehtipalo, K., Dommen, J., Ehrhart, S., Ortega, I. K., Franchin, A., Nieminen, T., Riccobono, F., Hutterli, M., Duplissy, J., Almeida, J., Amorim, A., Breitenlechner, M., Downard, A. J., Dunne, E. M., Flagan, R. C., Kajos, M., Keskinen, H., Kirkby, J., Kupc, A., Kürten, A., Kurtén, T., Laaksonen, A., Mathot, S., Onnela, A., Praplan, A. P., Rondo, L., Santos, F. D., Schallhart, S., Schnitzhofer, R., Sipilä, M., Tomé, A., Tsagkogeorgas, G., Vehkamäki, H.,
- 695 Wimmer, D., Baltensperger, U., Carslaw, K. S., Curtius, J., Hansel, A., Petäjä, T., Kulmala, M., Donahue, N. M. and Worsnop, D. R.: Molecular understanding of atmospheric particle formation from sulfuric acid and large oxidized organic molecules, *Proc. Natl. Acad. Sci.*, 110(43), 17223–17228, doi:10.1073/pnas.1306973110, 2013.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, Third edition., John Wiley & Sons, New York., 2016.
- 700 Sellegri, K., Yoon, Y. J., Jennings, S. G., O'Dowd, C. D., Pirjola, L., Cautenet, S., Chen, H. and Hoffmann, T.: Quantification of Coastal New Ultra-Fine Particles Formation from In situ and Chamber Measurements during the BIOFLUX Campaign, *Environ. Chem.*, 2(4), 260–270, doi:10.1071/EN05074, 2006.
- 705 Sellegri, K., Pey, J., Rose, C., Culot, A., DeWitt, H. L., Mas, S., Schwier, A. N., Temime-Roussel, B., Charriere, B., Saiz-Lopez, A., Mahajan, A. S., Parin, D., Kukui, A., Sempere, R., D'Anna, B. and Marchand, N.: Evidence of atmospheric nanoparticle formation from emissions of marine microorganisms, *Geophys. Res. Lett.*, 43(12), 2016GL069389, doi:10.1002/2016GL069389, 2016.
- 710 Sihto, S.-L., Kulmala, M., Kerminen, V.-M., Maso, M. D., Petäjä, T., Riipinen, I., Korhonen, H., Arnold, F., Janson, R., Boy, M. and others: Atmospheric sulphuric acid and aerosol formation: implications from atmospheric measurements for nucleation and early growth mechanisms, *Atmospheric Chem. Phys.*, 6(12), 4079–4091, 2006.
- Smith, J. N., Barsanti, K. C., Friedli, H. R., Ehn, M., Kulmala, M., Collins, D. R., Scheckman, J. H., Williams, B. J. and McMurry, P. H.: Observations of aminium salts in atmospheric nanoparticles and possible climatic implications, *Proc. Natl. Acad. Sci.*, 107(15), 6634–6639, 2010.
- 715 Soler, O.: *Atlas climatique de La Réunion (study report, 77p).*, 2000.
- Taupin, F. G., Bessafi, M., Baldy, S. and Bremaud, P. J.: Tropospheric ozone above the southwestern Indian Ocean is strongly linked to dynamical conditions prevailing in the tropics, *J. Geophys. Res. Atmospheres*, 104(D7), 8057–8066, doi:10.1029/98JD02456, 1999.
- 720 Tulet, P. and Villeneuve, N.: Large scale modeling of the transport, chemical transformation and mass budget of the sulfur emitted during the April 2007 eruption of Piton de la Fournaise, *Atmospheric Chem. Phys.*, 11(9), 4533–4546, doi:10.5194/acp-11-4533-2011, 2011.
- 725 Tulet, P., Di Muro, A., Colomb, A., Denjean, C., DufLOT, V., Arellano, S., Foucart, B., Brioude, J., Sellegri, K., Peltier, A., Aiuppa, A., Barthe, C., Bhugwant, C., Bielli, S., Boissier, P., Boudoire, G., Bourriane, T., Brunet, C., Burnet, F., Cammas, J.-P., Gabarrot, F., Galle, B., Giudice, G., Guadagno, C., Jeamblu, F., Kowalski, P., Leclair de Bellevue, J., Marquestaut, N., Mékies, D., Metzger, J.-M., Pianezze, J., Portafaix, T., Sciare, J., Tournigand, A. and Villeneuve, N.: First results of the Piton de la Fournaise STRAP 2015 experiment: multidisciplinary tracking of a volcanic gas and aerosol plume, *Atmos Chem Phys*, 17(8), 5355–5378, doi:10.5194/acp-17-5355-2017, 2017a.

- 730 Turquet, J., Delesalle, B., Denis, M. and Blanchot, J.: Structure et dynamique du Phytoplancton côtier de La Réunion. Résultats de l'étude complémentaire (Programme PHYTO RUN.), 2008.
- Vakkari, V., Laakso, H., Kulmala, M., Laaksonen, A., Mabaso, D., Molefe, M., Kgabi, N. and Laakso, L.: New particle formation events in semi-clean South African savannah, *Atmospheric Chem. Phys.*, 11(7), 3333–3346, doi:10.5194/acp-11-3333-2011, 2011.
- 735 Veldhuis, M. J., Kraay, G. W., Van Bleijswijk, J. D. and Baars, M. A.: Seasonal and spatial variability in phytoplankton biomass, productivity and growth in the northwestern Indian Ocean: the southwest and northeast monsoon, 1992–1993, *Deep Sea Res. Part Oceanogr. Res. Pap.*, 44(3), 425–449, 1997.
- Venzac, H., Sellegri, K., Laj, P., Villani, P., Bonasoni, P., Marinoni, A., Cristofanelli, P., Calzolari, F., Fuzzi, S., Decesari, S. and others: High frequency new particle formation in the Himalayas, *Proc. Natl. Acad. Sci.*, 105(41), 15666–15671, 2008.
- 740 Vigouroux, C., Stavrakou, T., Whaley, C., Dils, B., Duflot, V., Hermans, C., Kumps, N., Metzger, J.-M., Scolas, F., Vanhalewyn, G., Müller, J.-F., Jones, D. B. A., Li, Q. and De Mazière, M.: FTIR time-series of biomass burning products (HCN, C<sub>2</sub>H<sub>6</sub>, C<sub>2</sub>H<sub>2</sub>, CH<sub>3</sub>OH, and HCOOH) at Reunion Island (21° S, 55° E) and comparisons with model data, *Atmospheric Chem. Phys.*, 12(21), 10367–10385, doi:10.5194/acp-12-10367-2012, 2012.
- 745 Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjåraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Gruning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P. H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., De Leeuw, G., Löschau, G. and Bastian, S.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, *Atmospheric Meas. Tech.*, 5, 657–685, doi:10.5194/amt-5-657-2012, 2012.
- 750 Yli-Juuti, T., Nieminen, T., Hirsikko, A., Aalto, P. P., Asmi, E., Hörrak, U., Manninen, H. E., Patokoski, J., Dal Maso, M., Petäjä, T., Rinne, J., Kulmala, M. and Riipinen, I.: Growth rates of nucleation mode particles in Hyytiälä during 2003–2009: variation with particle size, season, data analysis method and ambient conditions, *Atmospheric Chem. Phys.*, 11(24), 12865–12886, doi:10.5194/acp-11-12865-2011, 2011.
- 760 Yu, F., Wang, Z., Luo, G. and Turco, R.: Ion-mediated nucleation as an important global source of tropospheric aerosols, *Atmospheric Chem. Phys.*, 8(9), 2537–2554, 2008.
- Yu, F., Luo, G., Nadykto, A. B. and Herb, J.: Impact of temperature dependence on the possible contribution of organics to new particle formation in the atmosphere, *Atmos Chem Phys*, 17(8), 4997–5005, doi:10.5194/acp-17-4997-2017, 2017.
- 765

## Figures

**Figure 1: Map of Reunion Island and its location. Different terrain types are represented as well as a simplification of the local atmospheric dynamic pattern around the island.**



770 Figure 2: The annual variation of CO concentrations (ppb) in 2015 (daily averages).

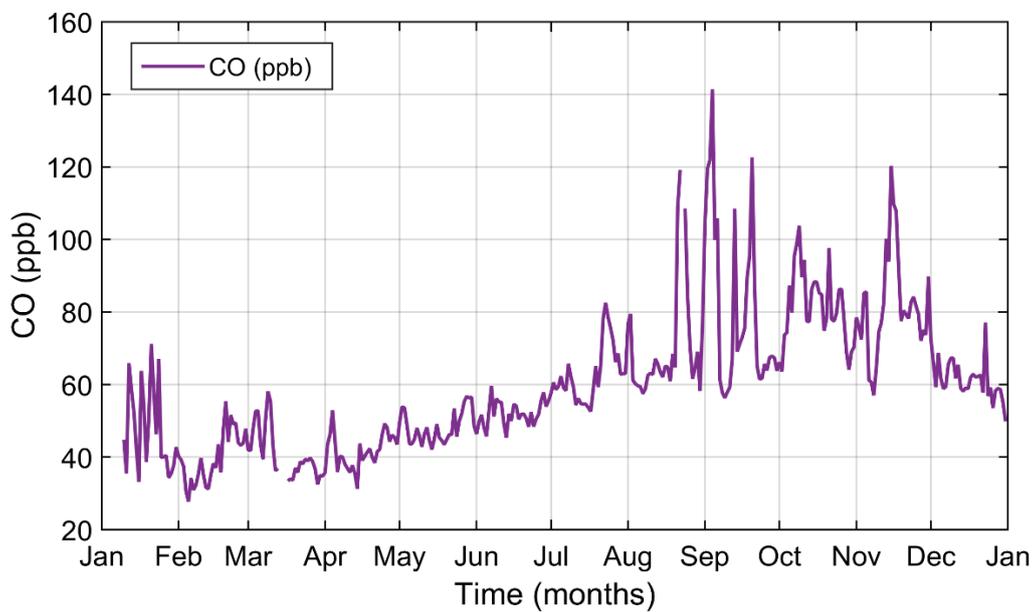
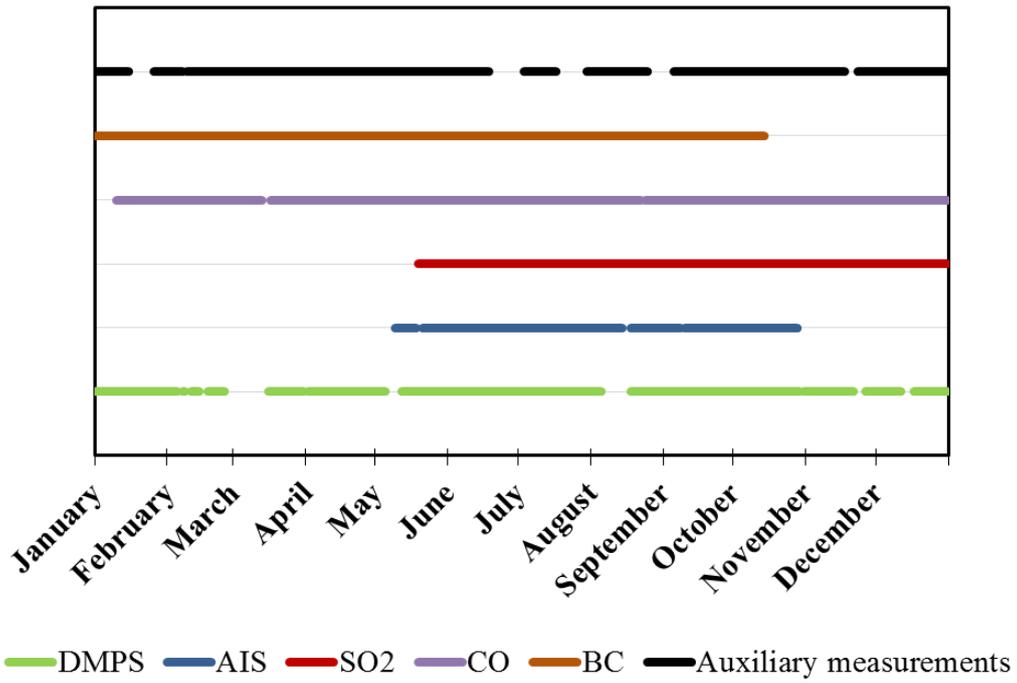
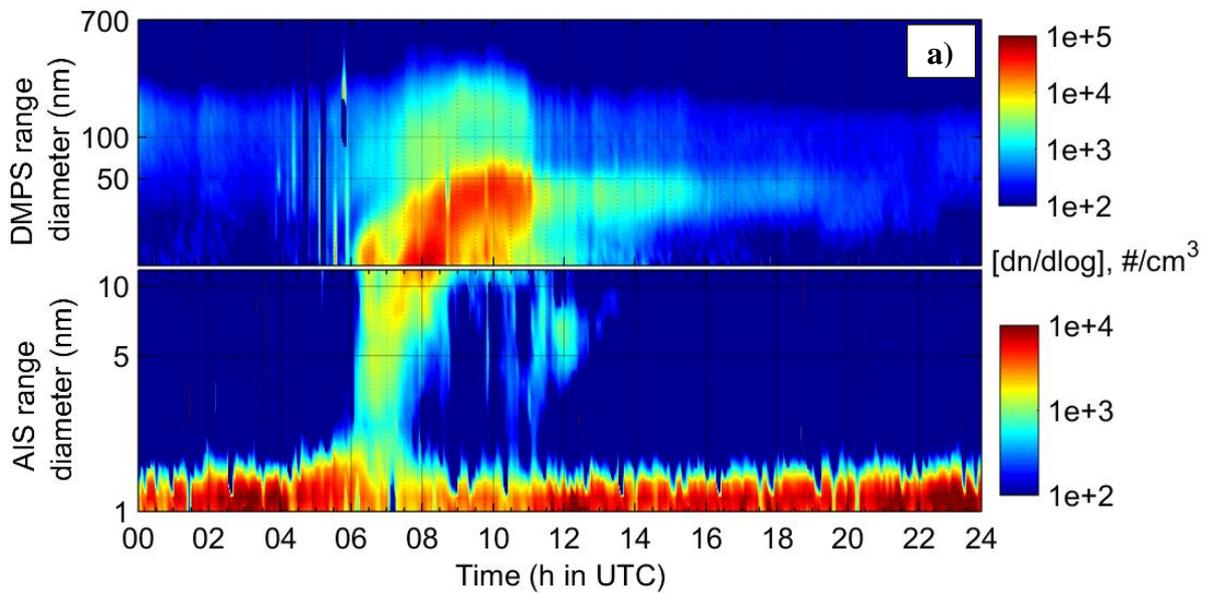
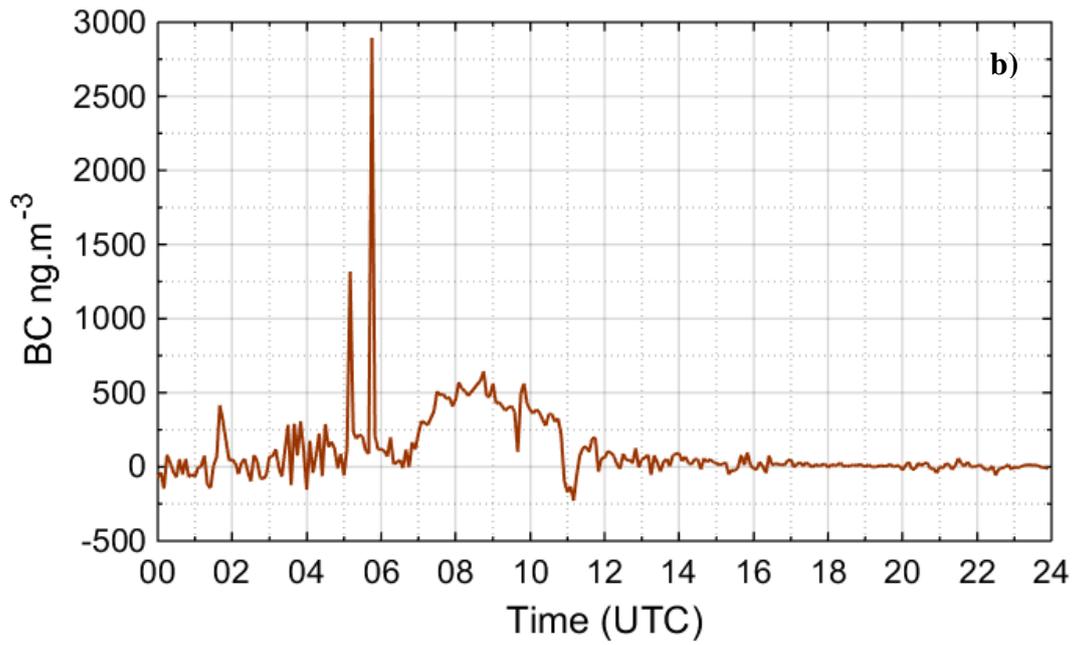


Figure 3: Dataset from 2015 Maïdo campaign. Devices recorded data simultaneously from May to November.



775 **Figure 4: July 6<sup>th</sup> 2015 diurnal variation of (a) negative ions (1-10 nm) and (10-700 nm) aerosol particle size distribution (note the different concentration scales for ion number and particle concentrations), and (b) the BC concentration variation in ng.m<sup>-3</sup>**





780

**Figure 5: Average diurnal variation during winter and summer of the (a) BC concentration, (b) number concentration of particles which diameter is larger than 100 nm ( $N_{100}$ ) and (c) number concentration of particles which diameter is smaller than 30 nm ( $N_{30}$ ).**

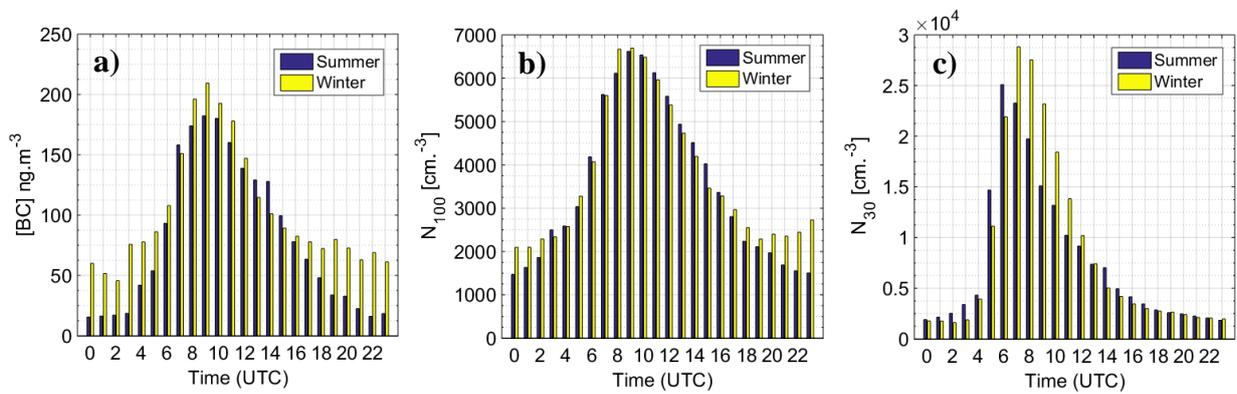
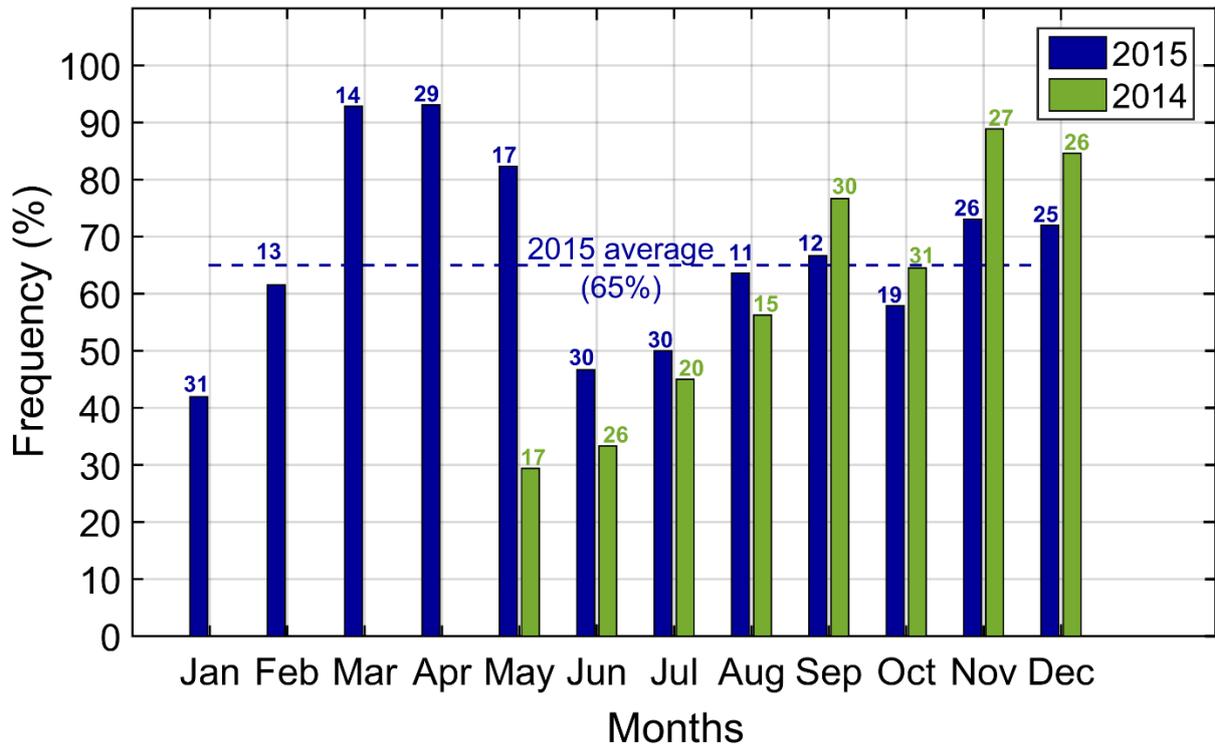
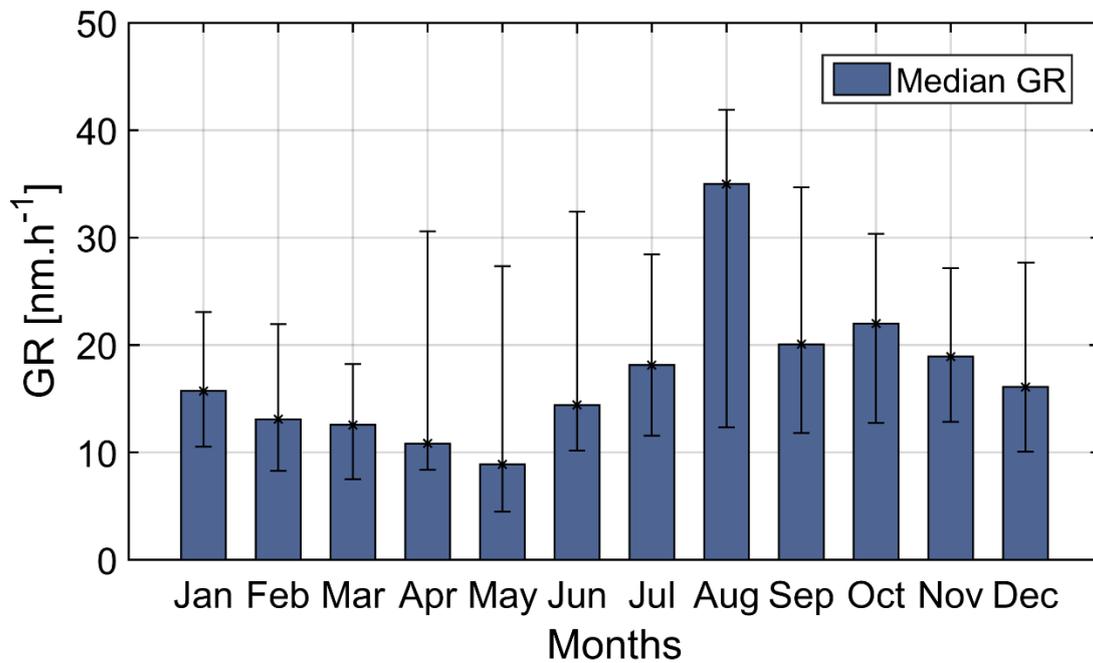


Figure 6: Monthly event frequency (%) variation during 2014 (green) and 2015 (blue). Values at the top of the bars correspond to the number of days that were taken in account for calculation.

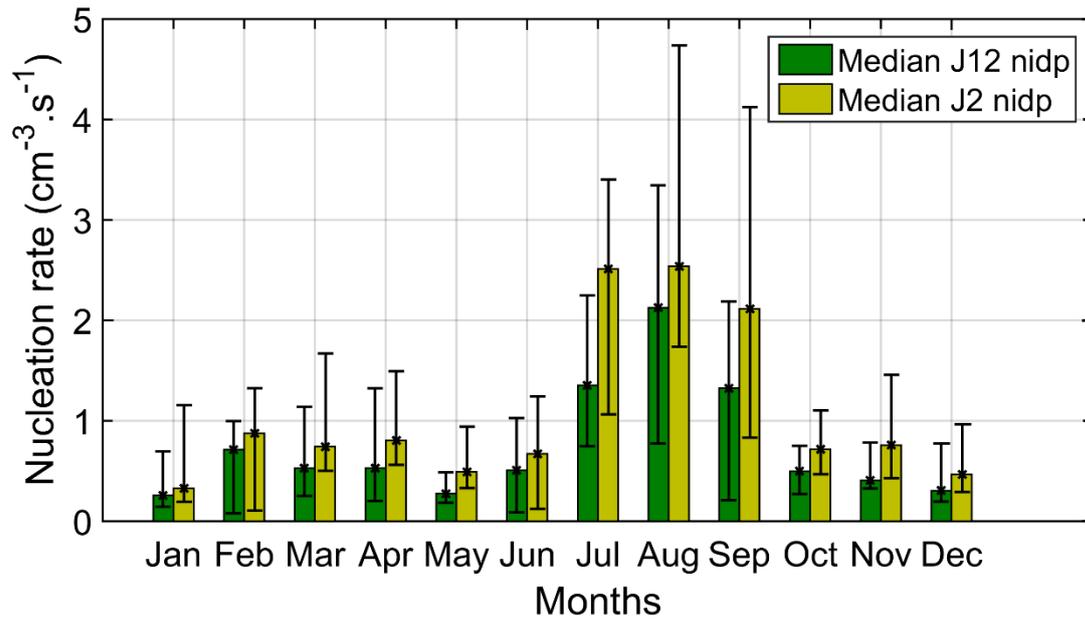


785

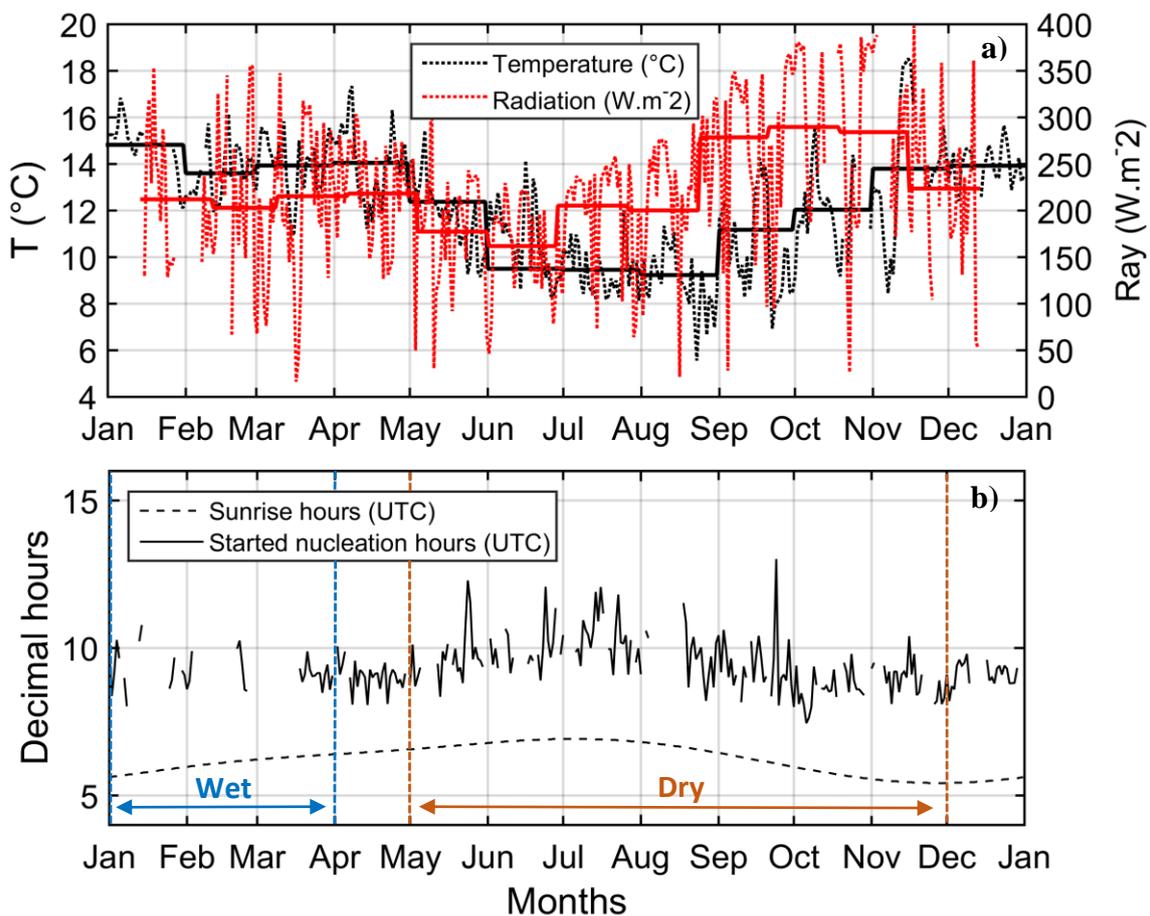
Figure 7: Monthly median growth rates in 2015. Error bars in black represent 25ile (top) and 75ile (base). To build these representation, 146 growth rates were estimated on 167 days classified as event days.



790 **Figure 8: Monthly median nucleation rates in 2015 for 2 and 12 nm sizes. Error bars in black represent 25ile (top) and 75ile (base).**

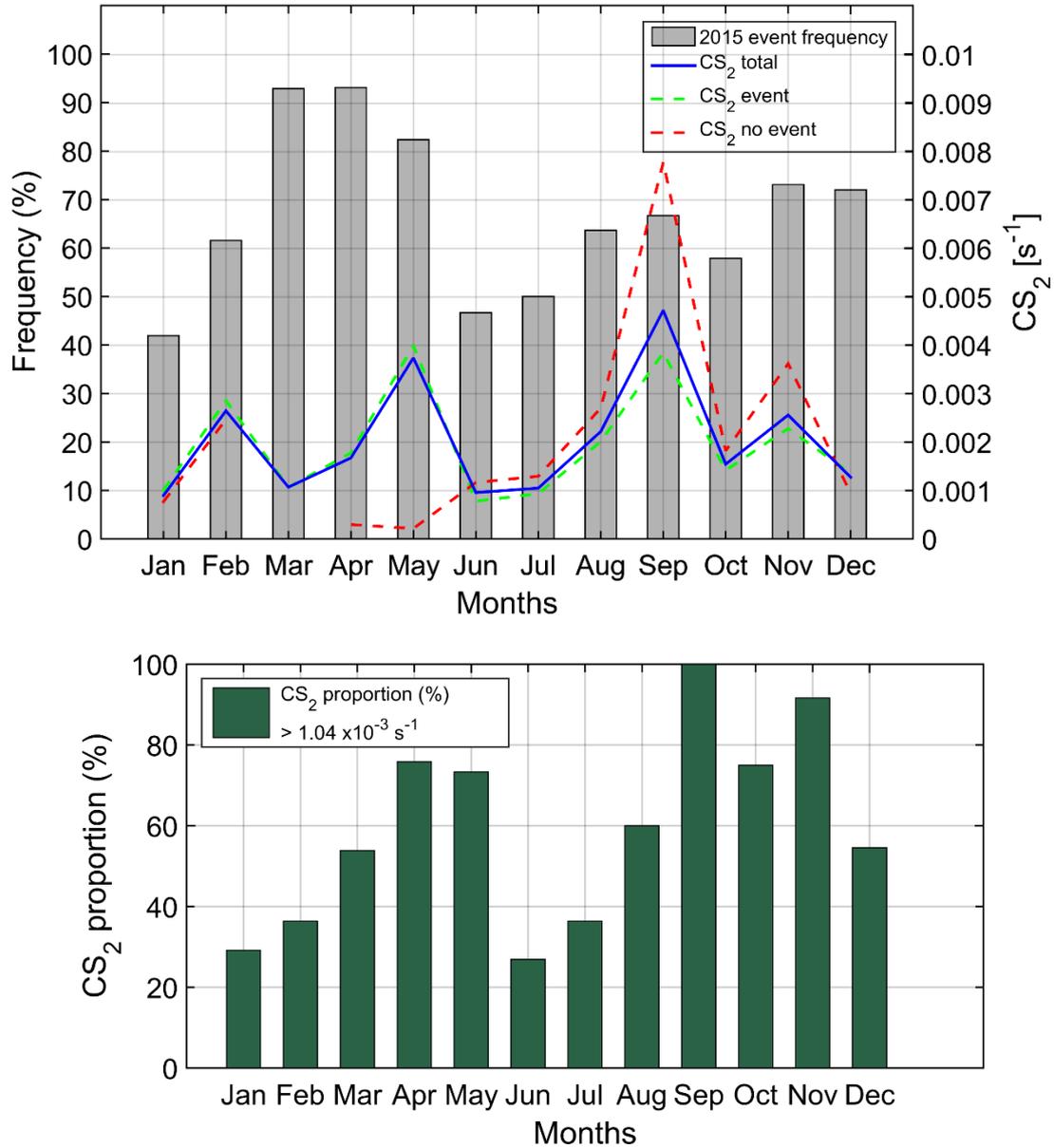


**Figure 9: a) temperature (°C) and incidental radiation (W.m<sup>-2</sup>) variations during 2015 at Maida station given by daily UTC (fine) and monthly (bold) averages; b) seasonal variation of sunrise and time of NPF onset in UTC. The dry and the wet seasons are also delimited in orange and blue respectively.**



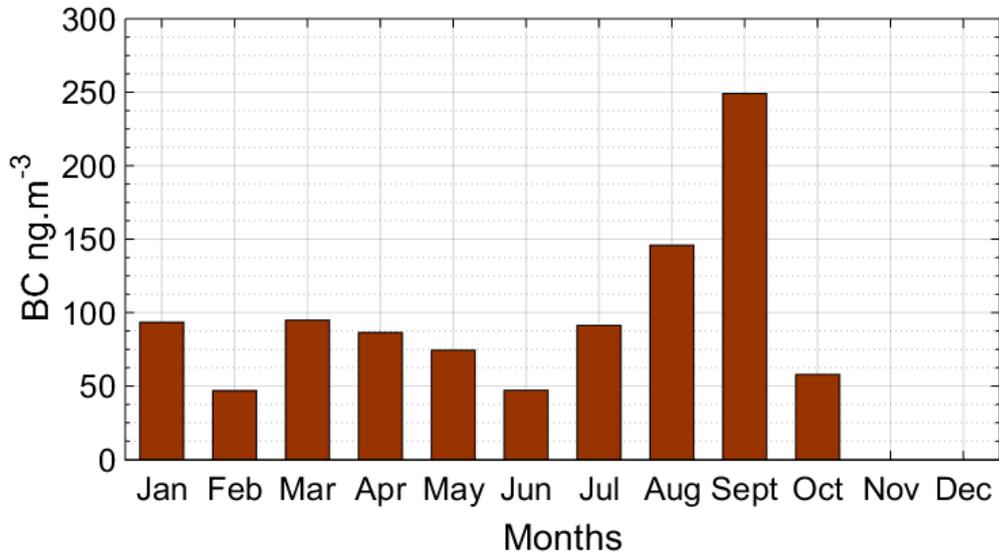
795

Figure 10: (a) Monthly CS<sub>2</sub> (blue) calculated two hours before NPF onset (scale at the right in s<sup>-1</sup>) and monthly event frequency f<sub>m</sub> (scale at the left in percentage). Averages have been calculated for event days (green), non-event days (red) and all days (blue). (b) Monthly CS<sub>2</sub> proportion exceeding the average of CS<sub>2ev</sub> and CS<sub>2noev</sub> medians ( $1.04 \times 10^{-3} \text{ s}^{-1}$ ).



800

**Figure 11: Monthly averages of the BC concentration during the year 2015**



**Table 1: Annual statistical values for G<sub>12-19</sub>; J<sub>12</sub>; J<sub>2</sub>; CS and CS<sub>2</sub> calculated on daily averages.**

	GR <sub>12-19</sub> (nm.h <sup>-1</sup> )	J <sub>12</sub> (cm <sup>-3</sup> .s <sup>-1</sup> )	J <sub>2</sub> (cm <sup>-3</sup> .s <sup>-1</sup> )	CS (s <sup>-1</sup> )	CS <sub>2</sub> (s <sup>-1</sup> )
<b>Averages</b>	<b>19.455</b>	<b>0.931</b>	<b>1.531</b>	<b>2.43 × 10<sup>-3</sup></b>	<b>1.86 × 10<sup>-3</sup></b>
Standard dev.	12.689	1.153	0.920	2.06 × 10 <sup>-3</sup>	2.53 × 10 <sup>-3</sup>
<b>Medians</b>	<b>15.16</b>	<b>0.508</b>	<b>0.858</b>	<b>1.97 × 10<sup>-3</sup></b>	<b>1.15 × 10<sup>-3</sup></b>
25ile	9.58	0.223	0.385	1.19 × 10 <sup>-3</sup>	6.55 × 10 <sup>-4</sup>
75ile	27.69	1.131	1.756	2.96 × 10 <sup>-3</sup>	2.00 × 10 <sup>-3</sup>

805

**Table 2: R correlation coefficients giving the relationships between NPF parameters (occurrence, GR and Js) and influencing factors (Ray, RH, T, P, CS<sub>2</sub>, CS<sub>2prop</sub>, CO and BC).**

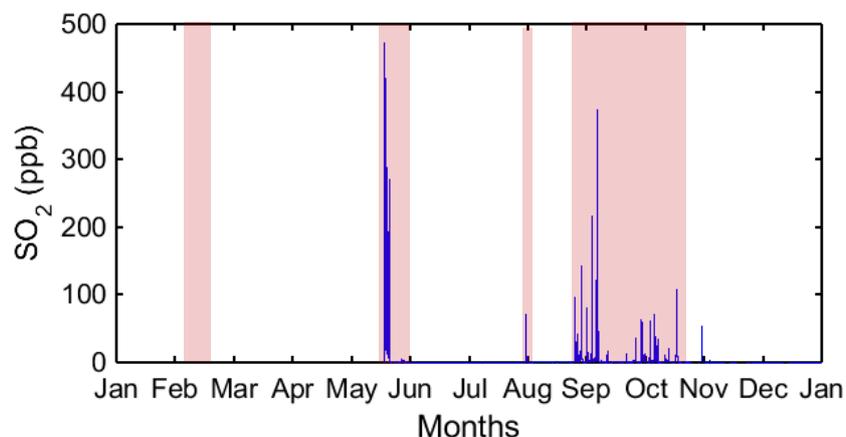
R	NPF %	GR <sub>12-19</sub>	J <sub>12</sub>	J <sub>2</sub>
Ray	0,096	0,284	0,012	0,062
RH	0,056	0,406	0,528	0,689
T	0,411	0,536	0,717	0,732
P	0,051	0,426	0,524	0,597
CS <sub>2</sub>	0,217	0,020	0,179	0,167
CS <sub>2prop</sub>	0,546	0,152	0,079	0,124
CO	0,234	0,638	0,326	0,389
BC	0,103	0,418	0,576	0,601

Correlations have been calculated with the twelve 2015 monthly averages for each parameters. Taking a degrees-of-freedom value of 10 and a risk  $\alpha = 0.05$  (95% of confidence), we obtained a lower limit value of 0.576 according to the Pearson table. The highlighted R in the table are above the limit value meaning that the variables are significantly dependent.

810

## Appendices

815 **Figure A1: SO<sub>2</sub> concentration (ppb) at Maïdo station in 2015, showing the occurrence of three eruptive periods in red (17 to 30/05, 31/07 to 02/08 and 24/08 to 18/10)**



820 Sulfur dioxide concentrations allow us to distinguish days when NPF can be affected by the presence of the volcanic plume. Based on a 1 ppb threshold, which was the 97th percentile of the series, we considered that 47 days were plume days at the Maïdo station and removed them. The eruptive periods are clearly visible on this figure.

**Table A1: Meteorological parameter averages.**

	<b>Ray (W.m<sup>-2</sup>)</b>	<b>RH (%)</b>	<b>T (°C)</b>	<b>P (hPa)</b>
January	212.07	78.77	14.82	787.21
February	202.98	77.17	13.60	788.85
March	215.42	74.11	13.93	789.45
April	218.26	66.70	14.05	789.41
May	177.43	72.20	12.38	791.51
June	161.91	70.00	9.50	790.90
July	205.30	53.78	9.46	792.52
August	200.18	64.46	9.23	791.93
September	278.64	50.02	11.17	791.02
October	289.73	63.55	12.04	791.92
November	248.20	56.26	13.80	789.91
December	223.63	77.10	13.92	790.16
<b>Yearly average</b>	<b>222.48</b>	<b>67.01</b>	<b>12.32</b>	<b>790.40</b>

**Table A2: Comparison of NPF frequency, GR, J and CS values**

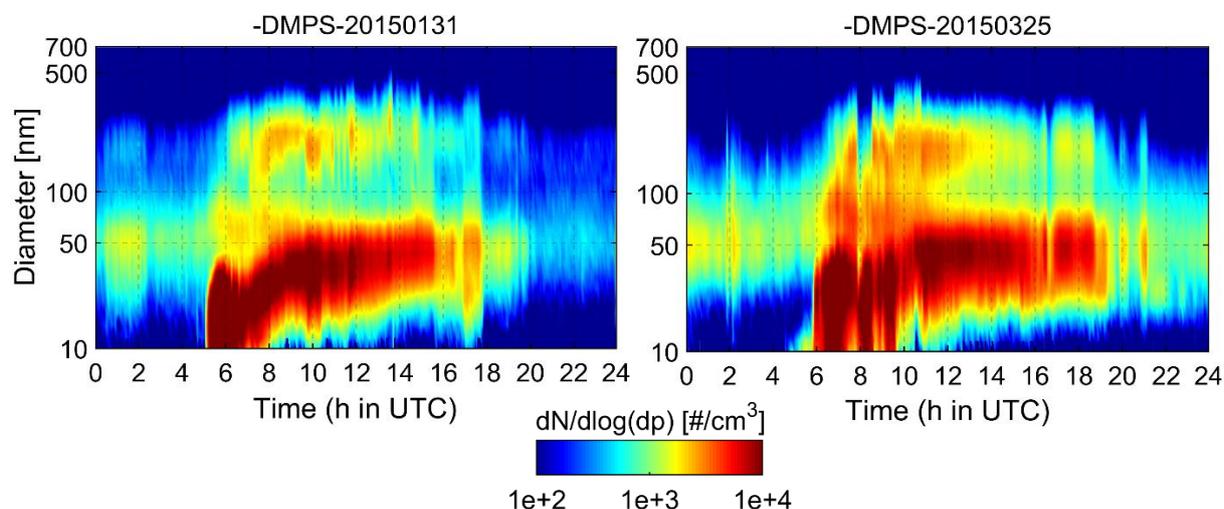
825

<i>Study</i>	Location	Environment	NPF (%)	GR (nm.h <sup>-1</sup> )	J (cm <sup>3</sup> .s <sup>-1</sup> )	CS (s <sup>-1</sup> )
<i>Dal Maso (2002)</i>	Ireland	Coastal	-	15 to 180	300 to 10000	2 × 10 <sup>-3</sup>
<i>Koponen (2003)</i>	Antarctica	Coastal	-	1 - 2	0.5	-
<i>Iida (2008)</i>	Mexico	Urban	-	18	1900 to 3000	-
<i>McMurry (2003)</i>	Atlanta, GA	Urban	-	2 – 6	20 to 70	-
<i>Venzac (2008)</i>	Nepal	Altitude	35%	2	< 0.2	2.1 × 10 <sup>-3</sup>
<i>Modini (2009)</i>	Australia	Coastal	65%	19	-	
<i>Vakkari (2011)</i>	South Africa	Remote, Altitude, Savannah	69%	2.1 to 30	0.1 to 28	4.3 × 10 <sup>-3</sup>
<i>Boulon (2011)</i>	France	Altitude	35.9%	6.20	1.382	3.7 × 10 <sup>-3</sup>
<i>Hirsikko (2012)</i>	South Africa	Remote, Altitude, Savannah	86%	8.0 ± 4.1	4.5 ± 6.1	0.8 × 10 <sup>-2</sup>
<i>Rose (2015)</i>	Bolivia	Altitude	63.9%	7.62	-	2.4 × 10 <sup>-3</sup>
<b><i>Our study</i></b>	<b>Reunion</b>	<b>Coastal, Altitude</b>	<b>65%</b>	<b>GR<sub>12-19</sub> = 19.455</b>	<b>J<sub>12</sub> = 0.931, J<sub>2</sub> = 1.531</b>	<b>2.43 × 10<sup>-3</sup></b>

This table is given to help in the comparison of NPF parameters as a function of the different types of environments. A few stations are listed here. The values of the present study are summarized in the last line and the closest ones are highlighted (yellow). Reunion Island seems to present NPF characteristics of several environment types. The GRs and Js listed in the table are in a similar range to those estimated in the present study.

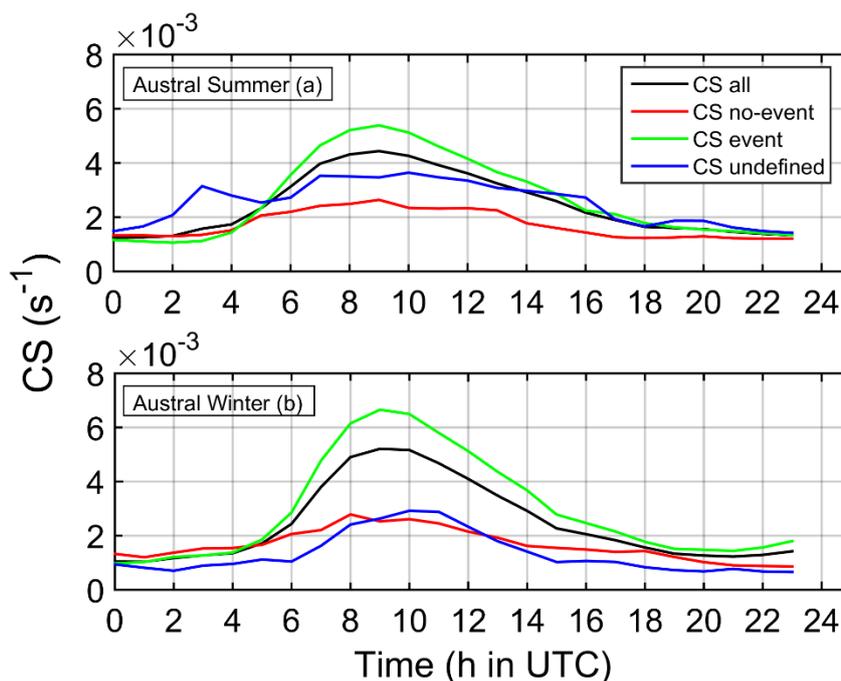
830

Figure A2: DMPS spectra for 31 January on the left and 25 March on the right. This is an evolution of the size distribution (left scale in nanometres) and of the aerosol concentration (colorimetric scale) with time (from 00 to 24 UTC).



835

Figure A3: Diurnal variation of CS



840

As the CS parameter varies throughout the day, we have also chosen to show the average daily variation of CS for summer a) and for winter b). Averages were calculated on undefined ( $CS_{un}$ ), no-event ( $CS_{noev}$ ), event ( $CS_{ev}$ ) and all days. Maximum of CS all and  $CS_{ev}$  are reached at 9:00 UTC for both seasons but are higher for winter ( $CS_{all} = 5.1 \times 10^{-3} \text{ s}^{-1}$ ;  $CS_{ev} = 6.9 \times 10^{-3} \text{ s}^{-1}$ ) than summer ( $CS_{all} = 4.5 \times 10^{-3} \text{ s}^{-1}$ ;  $CS_{ev} = 5.2 \times 10^{-3} \text{ s}^{-1}$ ).  $CS_{ev}$  really begins to increase at 4:00 UTC for summer but 5:00 UTC for winter. This corresponds to the moment when  $CS_{noev}$  becomes lower than  $CS_{ev}$ . Before these times,  $CS_{noev}$  is  $10^{-4}$  higher than  $CS_{ev}$ , implying that  $CS_{noev}$  is too large to trigger new

845 particle formation. Consequently, and thanks to this daily representation, we assume that CS affects the new  
845 particle formation trigger for both seasons in 2015. Moreover, it can be seen that  $CS_{un}$  curves follow the  $CS_{noev}$   
curves better than  $CS_{ev}$  so it is possible that no-event days were not well recognized.

850 **Figure A4: DMPS spectra for a) April and b) June months. We can clearly observe an event number difference  
between the two months according to the different FNP occurrence averages which are 93.1% for April and 46.7%  
for June.**

