## Measurement report: Insights into the chemical composition of molecular clusters present in the free troposphere over the Southern Indian Ocean: observations from the Maïdo observatory (2150 m a.s.l., Réunion Island)

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## 1. Identification of a tracer of free tropospheric conditions

Figure S1 Time series of the BLH simulated by Meso-NH for the model grid point which includes the Maïdo observatory in the innermost domain (blue) and number concentration of particles > 90 nm ( $N_{90}$ ) measured at the site (red).



5 Figure S2 Number concentration of particles > 90 nm ( $N_{90}$ ) segregated between FT and BL conditions at Maïdo. The percentage of hours for which Meso-NH indicates that the station is in the BL or in the FT for each  $N_{90}$  bin (50 cm<sup>-3</sup> width) is shown on the y-left-axis. The number of points in each bin is represented on the y-right-axis to provide further information on the distribution of the data.



Figure S3 Percentage of data for which Meso-NH indicates that the station is in the BL or in the FT for each hour of the day. For each hour, the reported statistics are based on 26 model outputs.



2. Positioning of the air masses in the FT or in the BL during the hours preceding their arrival at the site

Figure S4 Position of the air masses arriving at Maïdo in the FT along their path a. over land or ocean and b. relative to the site elevation. Twelve hours back-trajectories were computed with Meso-CAT during BIO-MAIDO and for each of the back-trajectory points along their path, air masses were considered to be over land or over the ocean (or below/above the station elevation) if 75% of the 324 trajectories in the trajectory set were over land or over the ocean (or below/above the station elevation). Otherwise, their positioning was classified as undefined.

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10 Figure S5 Positioning of the locations used in the sensitivity study performed to assess the reliability of the BL thicknesses retrieved by ECMWF ERA-5 reanalyses. The map was obtained from <u>https://www.geoportail.gouv.fr/</u> (last access: June 16, 2023).

## 3. Overview of the conditions during the OCTAVE campaign



Figure S6 72-hour back-trajectories of the air masses arriving at Maïdo when the station was in FT conditions during the nights of April a. 13 to 14, b. 15 to 16 and c. 16 to 17, 2018. The colour of each grid cell  $(0.2x0.2^{\circ})$  on the maps indicates the number of back-trajectory points falling into its area. Note that for each back-trajectory computed with the CAT model, all 125 trajectories of the corresponding set are shown in the figure.

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Figure S7 Positioning of air masses (in the FT or in the BL, as indicated by CAT) arriving in the FT at Maïdo along their 72hour back-trajectory during the nights of April a. 11 to 12, b. 12 to 13, c. 13 to 14, d. 14 to 15, e. 15 to 16 and f. 16 to 17, 2018. Note that for each back trajectory computed with the CAT model, all 125 trajectories of the corresponding set are shown in the figure.



4. Insights into the chemical composition of molecular clusters and their precursors in the marine FT

Figure S8 Time series of the fluorinated compounds normalized signal a.  $H(CF_2)_3COOH \cdot NO_3^-$ , b.  $H(CF_2)_4COOH \cdot NO_3^-$  and c.  $H(CF_2)_5COOH \cdot NO_3^-$ .

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Figure S9 Correlations of  $SO_5^-$  with a. MSA, separately in the BL and in the FT and with b. sulfuric acid during daytime (i.e. when global radiation > 10 Wm<sup>-2</sup>).



Figure S10 Similar to Fig. 10 in the main text, with in addition inter-species and inter-parameter correlations.