



**LOAC: a small
aerosol optical
counter/sizer**

J.-B. Renard et al.

LOAC: a small aerosol optical counter/sizer for ground-based and balloon measurements of the size distribution and nature of atmospheric particles – Part 2: First results from balloon and unmanned aerial vehicle flights

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Abstract

In a companion (Part 1) paper (Renard et al., 2015), we have described and evaluated a new versatile optical particle counter/sizer named LOAC (Light Optical Aerosols Counter) based on scattering measurements at angles of 12 and 60°. that allows some speciation of particles (droplets, carbonaceous, salts, and mineral dust) in addition to size segregated counting in a large diameter range from 0.2 up to possibly more than 100 µm depending on sampling conditions. Its capabilities overwhelm those of preceding optical particle counters (OPCs) allowing the characterization of all kind of aerosols from submicronic-sized absorbing carbonaceous particles in polluted air to very coarse particles (> 10–20 µm in diameter) in desert dust plumes or fog and clouds. LOAC light and compact design allows measurements under all kinds of balloons, on-board unmanned aerial vehicles (UAV) and at ground level.

We illustrate here the first LOAC airborne results obtained from an unmanned aerial vehicle (UAV) and a variety of scientific balloons. The UAV was deployed in a peri-urban environment near Bordeaux in France. Balloon operations include (i) tethered balloons deployed in urban environments in Wien (Austria) and Paris (France), (ii) pressurized balloons drifting in the lower troposphere over the western Mediterranean (during the Chemistry-Aerosol Mediterranean Experiment – ChArMEx campaigns), (iii) meteorological sounding balloons launched in the western Mediterranean region (ChArMEx) and from Aire-sur-l'Adour in south-western France (VOLTAIRE-LOAC campaign). More focus is put on measurements performed in the Mediterranean during (ChArMEx) and especially during African dust transport events to illustrate the original capability of balloon-borne LOAC to monitor in situ coarse mineral dust particles. In particular, LOAC has detected unexpected large particles in desert sand plumes.

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1 Introduction

The concentration, size and properties of atmospheric aerosol particles are highly variable in both space and time due to the large variety of aerosol sources of both natural and man-made origin, and to their relatively short residence time in the atmosphere (Holton et al., 2003). The characterization and monitoring of aerosol particles in the lower and middle Earth atmosphere is important for climate studies (e.g. Kaufman et al. 2002 and Ammann et al., 2003, respectively) and near the surface for air quality issues (e.g. Brunekreef and Holgate, 2002). When very high concentrations of ashes after volcanic eruptions are present at aircraft cruise altitude, they can severely affect air traffic (e.g. Chazette et al., 2012). In the middle atmosphere, aerosols also play a significant role in stratospheric ozone chemistry through heterogeneous reactions with nitrogen and halogen species (e.g. Hanson et al., 1994, 1996). To understand and predict aerosol impacts, it is important to develop observation and monitoring systems allowing for their characterization. In particular, small instruments adapted to balloon-borne measurements are scarce and generally devoted to stratospheric aerosols (Deshler et al., 2003; Renard et al., 2008). The aim of our study was to develop a new, relatively low-cost optical aerosol particle counter that could be launched under small balloons.

In Part 1 of this publication, a new versatile optical counter/sizer instrument named LOAC (Light Optical Aerosols Counter) was described and evaluated. It is light and compact enough to perform measurements at the surface and on-board airborne vehicles including all kinds of balloons in the troposphere and in the stratosphere and unmanned aerial vehicles (UAVs). Meteorological sounding balloons and UAVs are in particular adapted to airborne operations on alert. LOAC uses a new approach combining measurements at two scattering angles, which allows the determination of the particle size distribution and of the dominant nature of particles (mainly liquid droplets, and carbonaceous, mineral dust and salt particles) in various size classes.

In the companion paper we have presented the principle of measurements, the instrument calibration, cross-comparisons with other aerosols instruments, and field ob-

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servations validating the “speciation procedure” to estimate the main nature of the particles. In this paper, we illustrate the first airborne results obtained with LOAC on-board a UAV and under different kinds of balloons including low-altitude tethered balloons, meteorological sounding balloons, pressurized tropospheric drifting balloons, and stratospheric balloons.

2 LOAC instrument and gondola for balloon flights

The instrument description is detailed in Renard et al. (2015). In summary, particles are drawn up to the optical chamber through an isostatic tube by a small pump. The air stream crosses the centre of a laser beam of 25 mW working at the red wavelength of 650 nm. The scattered light is recorded by two photodiodes at scattering angles of $\sim 12^\circ$ and $\sim 60^\circ$. Photons travel directly to the photodiodes through pipes (without a lens), providing fields of view with a few degrees. A total of 19 size classes are defined for diameters between 0.2 and $\sim 100 \mu\text{m}$. The size classes are chosen as a good compromise between the instrument sensitivity and the expected size distribution of ambient air aerosols. LOAC can determine up to ~ 3000 particles smaller than $1 \mu\text{m cm}^{-2}$, 20 particles greater than $1 \mu\text{m cm}^{-3}$ in dry conditions and up to 200 particles cm^{-3} in fog/cloud conditions. The uncertainty (at 1σ) is of about $\pm 15\%$ for concentrations $> 10^{-1}$ particles per cm^3 and of about $\pm 30\%$ for lower concentrations.

A “speciation index” is retrieved by combining the 12° and 60° channels measurements. Speciation indices have been determined in laboratory for 4 families of particles including solid carbonaceous, mineral dust, and salt particles, and liquid droplets. The speciation indices obtained from LOAC observations in the atmosphere are compared to the charts obtained in the laboratory. The position of the data points in the various speciation zones provides the main nature of the particles. The identification of the nature of the particles works well in case of a homogenous medium, and is more difficult in case of a heterogeneous medium that generally cause the speciation index to be scattered among several speciation zones.



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To minimize the instrument weight, the optical chamber is in plastic Delrin[®]. The weight, including the pump, is of 250 g. The electric consumption is of 340 mA under 8 V, which corresponds to 3 W. Autonomy of about 3 h can be obtained with alkaline batteries. A gondola in polystyrene has been developed for flights under meteorological balloon. The data are sent in real-time by on-board telemetry. In its nominal configuration, LOAC uses the MeteoModem Company system for telemetry and GPS, and for temperature, pressure and humidity (PTU) measurements (<http://www.meteomodem.com/>). The total weight of the gondola (Fig. 1a), including the batteries and the PTU sounding, is of about 1 kg. The duration of a flight with meteorological balloons is of about 2 h, and can reach an altitude of 37 km with a latex balloon of 1200 g. One of the critical part of the instrument is the pumping system, which must work in extreme conditions in the middle atmosphere. At ground, the pump has a stability of about $\pm 5\%$. Tests have been conducted in the stratosphere during a meteorological flight up to an altitude of 34 km. The rotation speed of the pump and its stability are the same all along the flight, allowing us to conclude that the pump is insensitive to temperature and pressure variations.

A specific gondola has been developed for launch below low altitude drifting balloons developed by the French Space Agency (CNES; Fig. 1b). Such tropospheric balloons can stay in flight at a float altitude below 3500 m during several tens of hours (Ethé et al., 2002).

3 Field measurements under UAV balloons

A large number of LOAC flights under different kinds of balloons and airborne vehicles have been conducted since 2011. We present here some examples of the flight results (Table 1) and first interpretations of the measurements.

3.1 Unmanned Aerial Vehicle flights

A possible application of LOAC consists in measurements from unmanned aerial vehicles. LOAC has been mounted on a small UAV of the Fly-n-Sense (Fly-n-Sense, <http://www.fly-n-sense.com/uav-solutions/environment/>), as shown on Fig. 2. Tests have been conducted to ensure, first, that the sampled air is not affected by the motions of the propellers, and secondly that the electromagnetic radiations of the motors do not perturb the LOAC electronics. Figure 3 presents an example of a 20 min flight close to the ground performed in a field near the Bordeaux-Mérignac airport (South-West of France; 44.8° N, 0.7° W) on 18 December 2013 at 14:30 UT. The total concentration of particles larger than 0.2 μm in diameter is between 100 and 1000 particles cm^{-3} , generally decreasing with particle size as expected. Large particles up to 20 μm in diameter were observed all flight long and larger particles (up to the last channel 40–50 μm) were regularly counted. The LOAC speciation (not shown) indicates mainly carbon particles, with the presence of some mineral particles, as expected in such a location.

Because of their mobility and the possibility of stationary flights in the lower troposphere, the use of a UAV can be useful for the characterization of specific events or local (urban) pollution source.

3.2 Tethered balloons

LOAC has been operated at two different places in the cities of Vienna, Austria, and Paris, France, using a small and a large tethered balloon, respectively. Four flights under a 6 m^3 tethered balloon were performed by the Austrian Meteorological Office (Zentralanstalt für Meteorologie und Geodynamik) during the General Assembly of the European Geosciences Union between 9 and 11 April 2013, in the square of the Austria Center (conference centre) in Vienna, Austria, up to an altitude of 220 m (position in Table 1; photos in Fig. 4). Figure 5 presents the vertical concentration profile for the 19 particle size classes during the balloon ascent on 11 April 2013 at 11:00 UT. The pollution level on the ground was low with a total concentration of particles larger than

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0.2 μm of the order of few hundred of articles cm^3 . Submicronic-sized particles dominated and were observed at all levels, particles larger than 3 μm and up to more than 10 μm in diameter were often detected. The general trend is a decrease of concentrations with increasing altitude, the concentration being 4 times smaller at 220 m than at ground. A small concentration enhancement in small particles is detected between 60 and 110 m. The speciation analysis (Fig. 6) indicates a mixture of carbonaceous and mineral particles from the ground up to below 200 m. Mineral particles dominate at the altitude of the concentration enhancement (~ 80 m), probably emitted by building works going on in a tower under construction distant by ~ 250 m from the balloon (Fig. 4a). Above 200 m, only carbonaceous particles were detected, confirming the likely very local origin of the mineral particles at intermediate altitude.

Permanent measurements have been conducted at the “Observatoire Atmosphérique Generali” (OAG) in the South-West of Paris since May 2013 (position in Table 1). This observatory is a recreational 6200 m^3 tethered balloon (Fig. 7) operated in the public park André Citroën. The spring and summer 2013 measurements were contaminated by construction activities in the vicinity and are not considered here. The LOAC pump operates at 2.7 L min^{-1} and sampling is performed through a total suspended particulate (TSP) inlet having a diameter cut-off at about 100 μm . The instrument is installed in a small ventilated metallic box fixed on the side of the balloon passenger gondola with its TSP sampling inlet pointing up. The measurements can be sorted out between measurements when the balloon is at ground level and measurements during flights. From 150 to 200 days per year are favourable for flying this type of tethered balloon. The balloon measurements nominal maximum altitude is 120 m and many flights can be performed per day depending on wind conditions. Up to several flights per week can also be conducted with measurements up to an altitude of 270 m. The aim of these flights is to study the possible evolution of the nature and of the size of particles as a function of altitude, and to distinguish between local sources at ground level and the persistent urban pollution in the middle of the boundary layer.

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Most of the time, the air was well mixed and the concentrations are almost constant with increasing altitude for particles smaller than $\sim 10 \mu\text{m}$. On the opposite, some flights conducted during pollution events exhibit different trends. As an example, the 11 December 2013 (day #345) morning flight performed during anticyclonic conditions presents a temperature inversion layer at an altitude of $\sim 200 \text{ m}$, as shown on Fig. 8. A strong accumulation layer is detected between 180 and 220 m and was visually confirmed by the pilot of the balloon. The total concentration of particles larger than $0.2 \mu\text{m}$ in diameter is between more than $1000 \text{ particles cm}^{-3}$. Also, a fuzzy accumulation layer of particles is detected between 30 and 90 m. The size distribution at 3 different altitudes (Fig. 9) shows that the pollution (and thus the mass concentrations, as presented in the paper 1 detailing the instrument concept) is dominated by the smallest particles. The speciation indicates a mixture of carbon and mineral particles close to the ground in the recreation park, and only carbon particles for the highest altitudes. The analyses of flights performed later during that day and in the following days show the progressive disappearance of the accumulation layers as the wind speed increased.

The Wien and OAG examples show the interest of performing urban measurements under a tethered balloon, in order to document the size, the nature and the evolution of the particles as a function of altitude in the urban polluted boundary layer. In particular, such kind of flights can help distinguishing between local sources of pollution close to ground and accumulation/transport of aerosols in the ambient air at higher altitude in the atmospheric boundary layer.

3.3 ChArMEx tropospheric flights

LOAC was also intensively involved in the ChArMEx campaign (Chemistry Aerosol Mediterranean Experiment, <http://charmex.lsce.ipsl.fr/>). ChArMEx aims at a scientific assessment of the present and future state of the atmospheric environment over the Mediterranean basin (e.g. Menut et al., 2014; see ChArMEx Special Issue in Atmos. Chem. Phys. and Atmos. Meas. Tech.). All the LOAC balloon flights have been performed by the Centre National d'Etudes Spatiales (CNES).

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A total of 13 LOAC flights under low tropospheric pressurized drifting balloons were conducted from the Spanish Minorca Island from 15 June to 2 July 2013, and from the French Levant Island from 22 July to 4 August 2013 (station positions in Table 1), mainly during well-identified desert dust transport events. Results will be detailed in a forthcoming paper. We illustrate here one flight launched from Minorca Island during the ChArMEx/ADRI MED (Aerosol Direct Radiative Impact in the Mediterranean) campaign. Except in case of precipitation or condensation, these balloons follow a near-Lagrangian trajectory (i.e. remaining in the same air mass during their trajectory in the lower atmosphere). Their float altitude was chosen before the flight in the 400–3500 m range by adjusting the balloon density with helium, depending on the altitude of the targeted aerosol layer. Those balloons are derived from the 2 m superpressure balloon model used by Ethé et al. (2001) and Vialard et al. (2009). They are spherical with a diameter of 2.5 m (Fig. 10a). The control and transmission gondola is placed inside the envelope at the Earth pole of the balloon, (Fig. 10b). For permitting flights at 3 km altitude or more, a bit larger balloons (2.6 m in diameter) were launched unpressurized to limit internal pressure in the balloon envelope at float altitude. An aluminium foil sometimes fixed at the balloon equator (Fig. 10c) to favour evacuation of condensed or rain water from the envelope surface turned to be useful against communication problems attributed to electrostatic charged, especially encountered with balloons launched unpressurized. A hydrophobic coating may also been applied on the envelope to reduce its load by water. LOAC is fixed to the south pole of the balloon with its bevelled metallic inlet pointing on the side of its small gondola.

During such balloon flights, drifting with a speed of the balloon close to zero relatively to ambient air, the particle sampling efficiency should be unbiased, whereas sampling on-board an aircraft generally causes a cut-off diameter of a few μm at best due to high speed (e.g. Wendisch et al., 2004; Formenti et al., 2011). This should allow a good sampling by the balloon-borne LOAC of large particles, which dominate the mass flux of mineral dust transport and deposition (e.g. Dulac et al., 1987, 2002; Foret et al.,

2006) and this was one of the main reasons to deploy LOAC under drifting balloons during ChArMEx campaigns.

The integration time was chosen between 1 and 20 min. This choice was imposed by the low telemetry rate for the downlink of the LOAC measurements through the Iridium satellite communication system. Also, LOAC was sometimes temporarily shut down after a short session of measurements to save up on-board energy.

Figure 11 presents the balloon trajectory from Minorca between about 09:45 and 16:50 UT on 17 June 2013, during a Saharan dust transport event. The length of the flight was 360 km at an altitude of 2000–2050 m. The daytime average aerosol optical depth at 550 nm derived from MSG/SEVIRI shows values of about 0.3 along the balloon trajectory (Fig. 12). Figure 13 presents the evolution of the aerosol particle concentrations for the 19 size classes as a function of time along the trajectory. The mineral nature of the particles was confirmed by the speciation measurements. LOAC has detected unexpected significant concentrations of large particles inside the soil dust plume, up to 50 μm in diameter, although the plume originated from North-Africa about 3 days before (Fig. 14). Thermal anomalies from MSG/SEVIRI North Africans Sand Storm Survey (<http://nascube.univ-lille1.fr>) indicate that the dust layer sampled by the balloon was likely emitted on 14 June in the NW Algeria-N-E Morocco source region particularly active in summer (Bergametti et al., 1989). The concentrations of these largest particles remained relatively constant during the flight, suggesting no significant sedimentation of large particles during the flight or compensation by particles from above. This type of observation was found during other flights, and will need further analysis to better understand the process that can maintain such large particles in suspension during several days.

This example shows, for the first time, the time-evolution of tropospheric aerosol concentrations at constant altitude from long duration balloons.

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3.4 Upper tropospheric and stratospheric flights

Thanks to its light weight and small electric consumption, LOAC can be launched under meteorological sounding balloons, allowing a large number of flights from different places. The measurements were conducted during the ascending phase of the balloon.

The inlet was oriented toward the sky, thus towards the relative wind direction due to the ascent of the balloon at $\sim 5 \text{ ms}^{-1}$. In the May 2013–September 2014 period, LOAC has performed 41 flights under meteorological balloons from France (Aire-sur-l'Adour, South-West of France; Ury, south-west of Paris region; Levant Island, south of France; and Ile de la Réunion, tropic of Capricorn), from Spain (Minorca Island) and from Iceland (Reykjavik). The highest altitude reached by LOAC with this kind of balloon is 37 km.

During the ChArMEx campaign, a total of 19 launches have been conducted from Minorca (Spain) and Levant Island (France). In particular, flights have been conducted every 12 h between 15 to 19 June 2013, to observe the vertical distribution and the time-evolution of the desert dust plume concentrations in the troposphere. Another plume event was detected on the beginning of August 2014. Figure 15 presents the flight conducted from Levant Island (France) on 4 August 2013 between 15:30 and 17:30 UT. A sand plume was detected in the lower troposphere up to an altitude of 7 km, as shown by the speciation (Fig. 16, top). In the stratosphere, the persistent aerosol layer mainly of sulphuric droplets was detected up to an altitude of 32 km, as shown by the speciation (Fig. 16, bottom). The aerosol concentration decreased with increasing altitude, although a small enhancement was detected above 30 km. This kind of enhancement was already observed previously with another aerosol counter, STAC (Stratospheric and Tropospheric Aerosol Counter) using a totally different technique of measurements (Renard et al., 2010b). In average, STAC and LOAC measurements are in good agreement both for the background content and concentration enhancements. The nature of the particles inside the enhancements is not clearly determined by the LOAC speciation procedure and needs further analysis.

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Another example of a meteorological flight is presented in Fig. 17. The flight was conducted during the VOLTAIRE-LOAC campaign of regular measurements for establishing a stratospheric aerosol climatology at different latitudes. The flight was performed at the French Space Agency (CNES) launching base at Aire-sur-l'Adour (South-West of France) on 20 March 2014 between 11:30 and 13:30 UT. A pollution layer was detected at an altitude of 1 km, associated with a small temperature inversion layer detected by the PTU sensors on board the LOAC gondola. In the upper layer, the profile exhibits a typical situation for the vertical distribution of background stratospheric aerosols, with rather small number densities and again a concentration enhancement at around an altitude of 30 km. The speciation in the stratosphere (Fig. 18, bottom) indicates that almost all particles below 1 μm in diameter are liquid; nevertheless, some layers of solid (carbon) particles were detected around 13 and 25 km (Fig. 18, top). The presence of such a transient carbon particles layers was previously mentioned by Renard et al. (2008) from balloon measurements but were also often detected from aircraft measurements (Blake and Kato, 1995; Murphy et al., 2007). The origin of these particles could be biomass burning, aircraft traffic, but also “smoke” particles coming for meteoritic disintegration (Murphy et al., 1998; Neely et al., 2011).

Finally, for most of the vertical sounding flights, LOAC has detected few particles greater than 10 μm in the stratosphere. These detections are similar to the ones obtained by the DUSTER balloon-borne particle collector (Ciucci et al., 2011; Della Corte, 2012) and can be attributed to interplanetary dust (Brownlee, 1985).

4 Discussion

Due to its industrial production, a large number of copies of LOAC are available. They can be operated at ground, in aerial conditions, and can conduct measurements up to the middle stratosphere. LOAC ability to estimate the main nature of aerosols can be used to better distinguish between the various layers of aerosols having different origins.

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Because of its small weight, the LOAC gondola, including PTU sensors, can be launched easily with meteorological balloons. Tens of flights per year could be conducted from different locations to locally monitor the aerosols content. Also, several flights per week can be conducted to study specific events (as an example, 9 flights to be discussed in a forthcoming paper were conducted in 5 days in June 2013 from Minorca, Spain, during the ChArMEX campaign to study a sand plume over Mediterranean Sea). It is thus possible to better analyse the time and spatial variability of the aerosols content in the free atmosphere; this new measurements strategy is similar to the one already conducted with ozone soundings. Using forecast trajectories, the balloon trajectory can be estimated before the flight to optimize the probability of a safe recovery of the gondola. As an example, the recovery success was of 90 % for the flights conducted in France from Aire-sur-l'Adour and Ury in 2014. Thanks to LOAC robustness, the recovered instrument can be re-used several times.

The large set of measurements obtained in the various geophysical conditions presented above has allowed us to obtain original results on the aerosol content in the different parts of the atmosphere. Using tethered balloon, we have started to better document the urban pollution from the ground up to the middle of the boundary layer, and to determine the evolution of the size distribution and the nature of the particles with altitude. In the free troposphere, the balloon measurements inside several desert dust plumes has shown the unexpected persistence of large coarse particles of more than 15 μm and up to several tens of μm in diameter. The analysis of the LOAC and balloons housekeeping data during these flights indicate the presence of strong electrostatic fields inside the plume (but not outside) that slightly disrupted the electronics. Ulanowski et al. (2007) observed polarization effects in a dust plume over the Canary Island that they attributed to alignment of particles due to an electric field. These fields might explain the sustained levitation of these large particles, but this hypothesis needs further experimental studies. In the lower and middle stratosphere, LOAC has confirmed the presence of layers of carbon particles. Above 30 km, LOAC has also detected transient concentrations enhancements, but the nature and origins of particles

are not yet fully determined. Finally, the large size range detection of LOAC has allowed us to detect unambiguously the presence of interplanetary grains and meteoritic debris. All these first results need further flights to better document the complex content of the aerosols content in the various parts of the atmosphere.

5 Conclusions

LOAC is simultaneously involved in different projects. The LOAC ground-based and tethered balloon measurements at the Observatoire Atmosphérique Generali (Paris) will continue. The detailed analysis of the variation in concentration and the nature of the urban aerosols with altitude is still in progress, in particular during strong pollution events. Measurements at SIRTa (Palaiseau) will also continue for the detection of fog events and the time-evolution of their size distribution, and for the monitoring of sub-urban particles.

LOAC is also involved in different projects for the monitoring and the identification of tropospheric and stratospheric aerosols, using meteorological balloons and large stratospheric large balloons (zero pressure and super-pressure). In the frame of the VOLTAIRE-LOAC project, dedicated to the long-term monitoring of stratospheric aerosols, flights under meteorological balloons are conducted every 2 weeks from Aire-sur-l'Adour (South-West of France) and Ury (South-East of Paris) since January 2014. Such a strategy of recurrent balloon flights is suitable to capture events like volcanic eruptions and to derive long-term trends in the stratospheric aerosol content. Additional flights will be conducted from Reykjavik (Iceland) and Ile de la Réunion (France, Indian Ocean) to better document the latitudinal dependence of stratospheric aerosols and to identify the evolution of their nature with altitude. Some flights will be also conducted from Iceland during dedicated campaigns for the study of the vertical transport of frequently re-suspended volcanic dust (Dagsson-Waldhauserova et al., 2013), and in case of future major volcanic events. Also, the large number of flights performed each year will allow us to better estimate the mean concentrations of large particles in the

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middle atmosphere. Thus we expect to provide soon an estimate of the interplanetary dust input in the upper atmosphere.

The LOAC flights on-board UAVs have started, mainly for the measurements of urban pollution and the characterization of the aerosol sources, but other applications are under study.

LOAC is now involved in the Strateole-2 project for the study of the equatorial upper troposphere and the lower stratosphere during balloon flights lasting several months (probably in 2018).

Finally, the LOAC concept and design (in terms of weight and electric consumption) are well suited for measurements in various planetary atmospheres (like Mars, Saturn and Titan). Some electronic improvements have started recently to propose a LOAC instrument that can comply with the spatial constraints, in particular in terms of very low temperature, and robustness to vibrations and radiations.

Acknowledgements. The LOAC project was funded by the French National Research Agency's ANR ECOTECH. The instrument and the gondola are built by Environnement-SA and Meteo-Modem companies. The balloons flights of the MISTRALS/ChArMEx campaign were funded and performed by the French Space Agency CNES. The various copies of LOAC used in the campaigns were funded with the support of CNES, INSU-CNRS and the French VOLTAIRE-LOAC Labex (Laboratoire d'Excellence ANR-10-LABX-100-01).

The authors want to thank the EGU Atmospheric Sciences Division, especially Division President Oksana Tarasova, for their involvement in the LOAC measurements during 2013 EGU General Assembly, and the CNES launching team for the support and dedication that ensured the success of the LOAC flights. OMP/SEDOO in Toulouse and ICARE thematic centre in Lille are gratefully acknowledged for their support to ChArMEx browse products and database. L. Gonzales and C. Deroo from Laboratoire d'Optique Atmosphérique are acknowledged for the NAScube product based on EUMETSAT MSG/SEVIRI data.

This work is in memory of Jean-Luc Mineau.

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Table 1. LOAC balloon flights illustrated in this study.

Campaign	Launch location	Launch latitude and longitude	Launch date	Balloon type	LOAC inlet
EGU	Vienna (Austria)	48.2343° N 16.4132° E	11 April 2013	Tethered balloon	Metal, bevelled
OAG	Paris (France)	48.8414° N 02.2740° W	11 December 2013	Tethered balloon	TSP
ChArMEx	Minorca (Spain)	39.8647° N 04.2539° E	17 June 2013	Tropospheric pressurized balloon	Metal, bevelled
ChArMEx	Levant Island (France)	43.0265° N 6.4877° E	4 August 2013	Meteorological balloon	Metal, bevelled
VOLTAIRE-LOAC	Aire-sur-l'Adour (France)	43.702° N 0.262° W	20 March 2014	Meteorological balloon	Metal, bevelled

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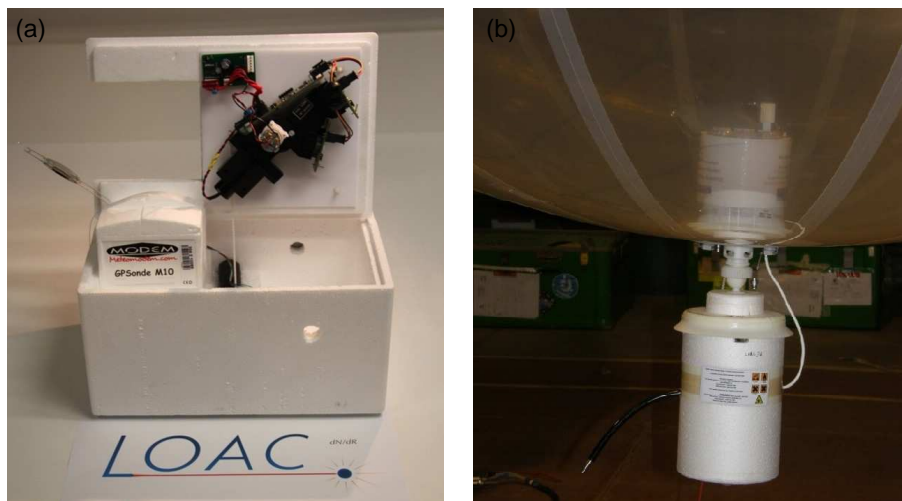


Figure 1. (a, left) The LOAC gondola with a Meteomodem Company sonde for flight under meteorological balloons; (b, right) the LOAC gondola below a low troposphere drifting balloon.

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Figure 2. LOAC on board an unmanned aerial vehicle of the Fly N Sense Company (LOAC is on the black box at the bottom of the vehicle).

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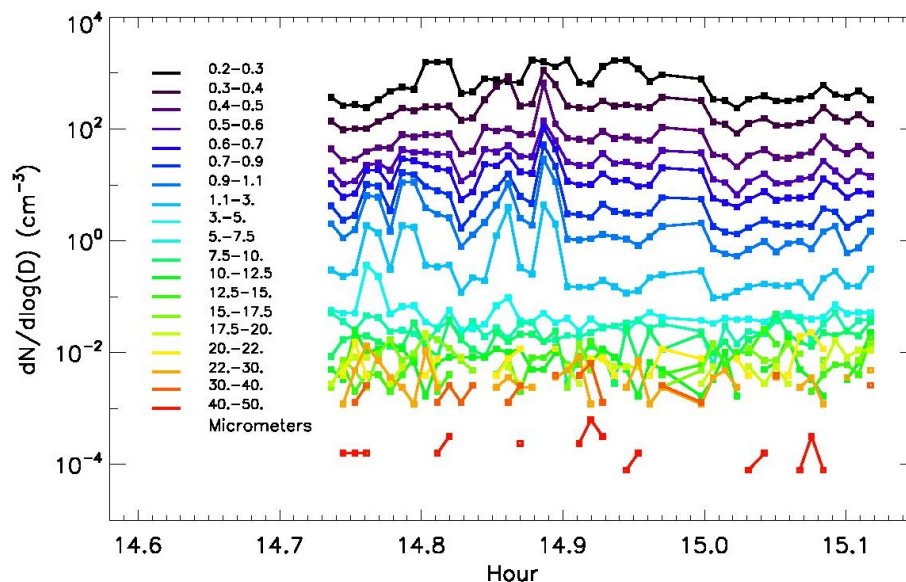


Figure 3. Aerosol particle size distribution from LOAC flight on-board an unmanned aerial vehicle flown close to the surface near Bordeaux-Mérignac (France) on 18 December 2013 at 14:30 UT.

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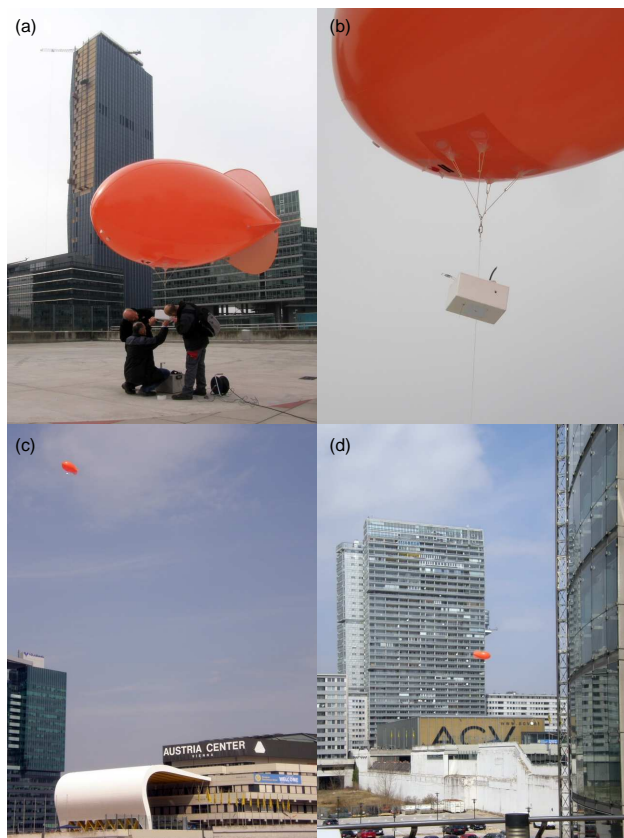



Figure 4. Pictures of the LOAC operations below a 6 m^3 tethered balloon at the Austria Center in Vienna during the 2013 European General Assembly. From left to right and top to bottom: **(a)** preparation of the launch with a view towards S on a tower under final stage of construction in the back; **(b)** view from below of LOAC in flight with its sampling inlet pointing upward; **(c)** view from the S of the balloon over the conference centre; **(d)** view from the SW of the environment of the launch site including leaving and office tower blocks and an open air car park.

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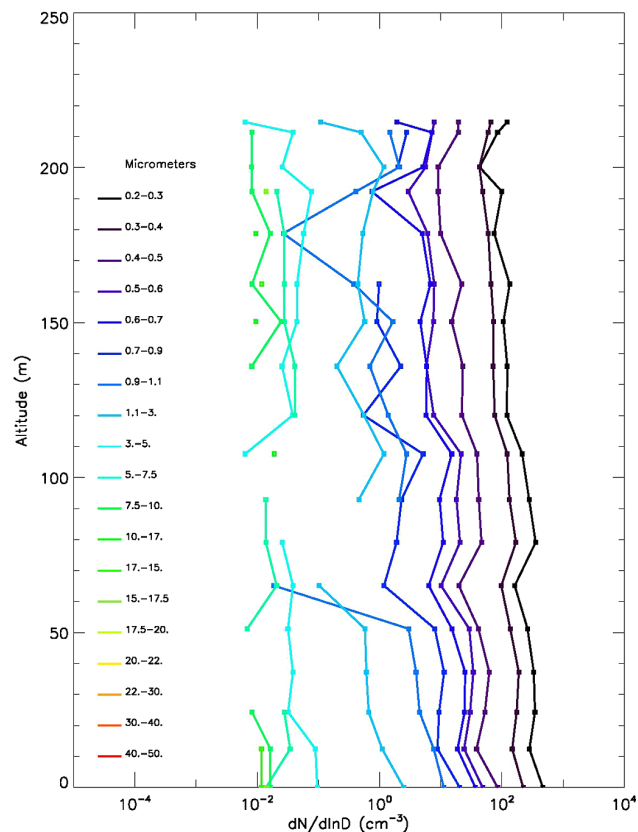


Figure 5. Evolution of the concentrations for the 19 size classes of LOAC, during a flight under a tethered balloon in Vienna (Austria) on 11 April 2013 at 11:00 UT.

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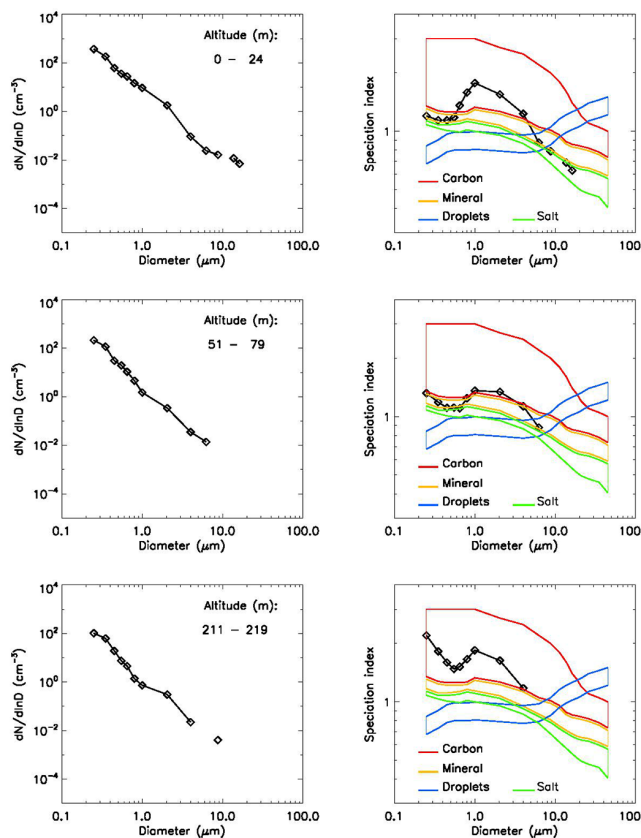


Figure 6. Size distribution and speciation at 3 altitudes during a flight under a tethered balloon in Vienna on 11 April 2013 at 11:00 UT.

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Figure 7. LOAC on the recreational OAG tethered balloon in Parc André Citroën, Paris. From left to right: **(a)** view on the balloon in flight; **(b)** view of the LOAC installed in a small box on the side of the passenger gondola with its TSP inlet above, a small WiFi antenna on the left of the box for data transmission, and a ventilation opening protection (grey) on the right.

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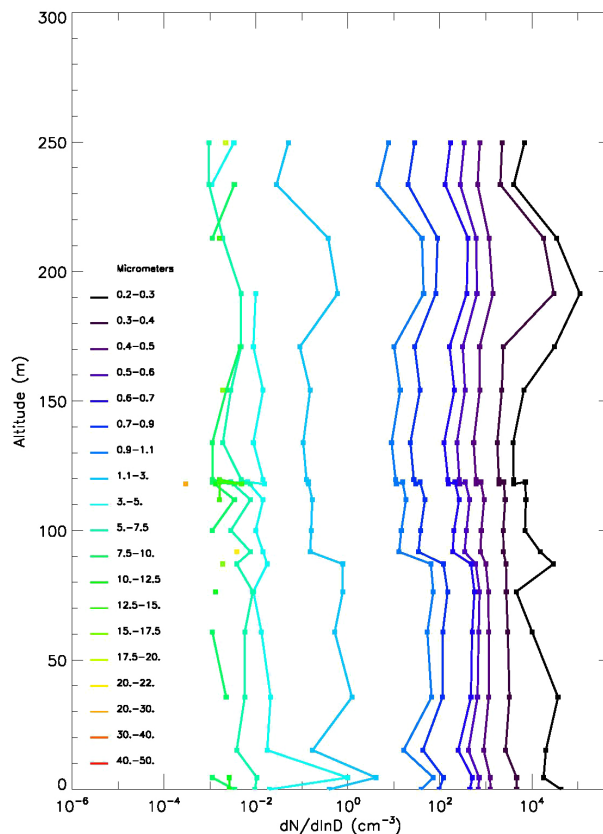


Figure 8. Evolution of the concentrations for the 19 size classes of LOAC, during a flight under the OAG tethered balloon in Paris (France) on 11 December 2013 at 10:15 UT.

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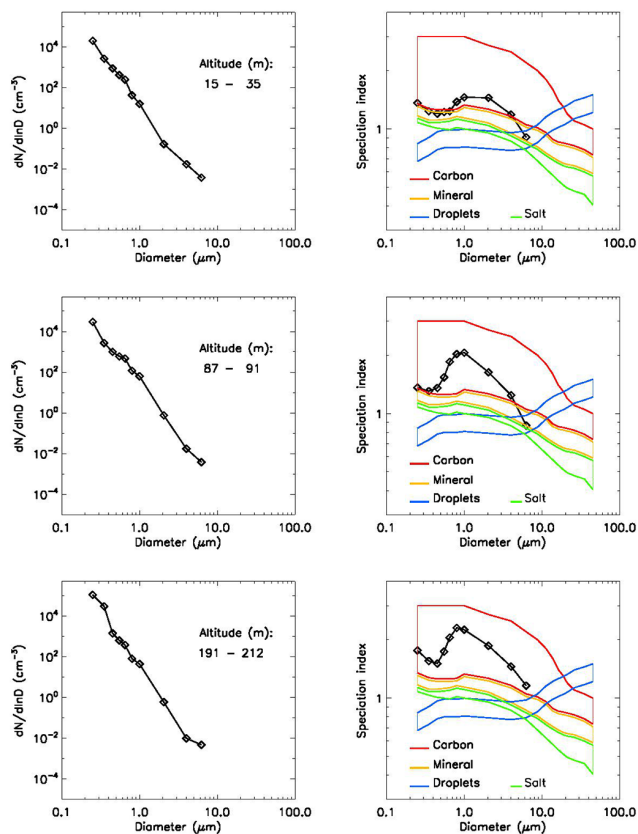


Figure 9. Size distribution and speciation at 3 altitudes during a flight under the OAG tethered balloon in Paris (France) on 11 December 2013 at 10:15 UT.

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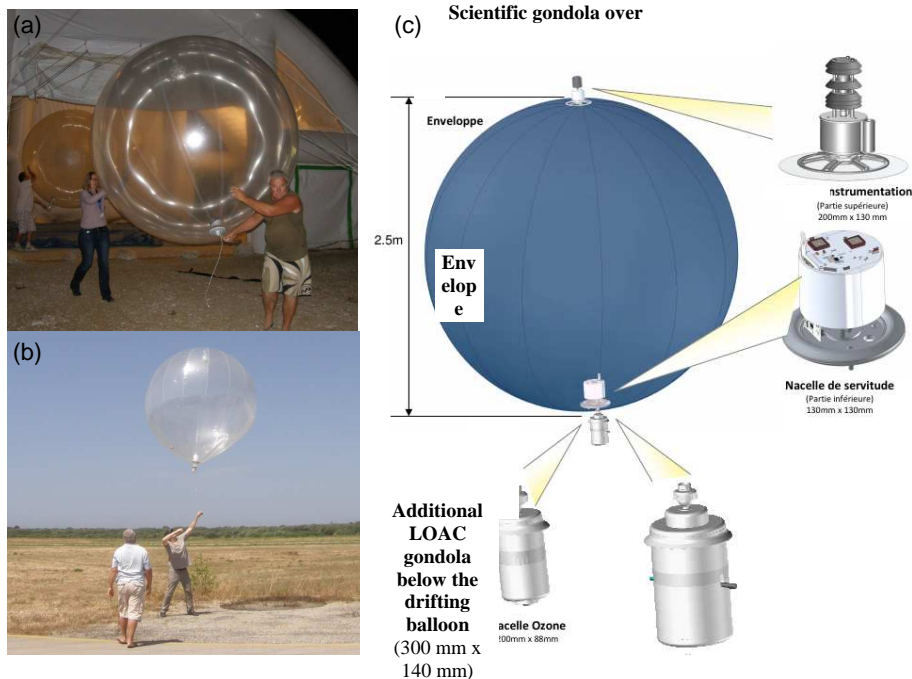



Figure 10. (a) CNES 2.5 m tropospheric pressurized balloon shortly before a night launch; (b) scheme of the pressurized balloon and gondolas; the scientific and control gondolas communicate by radio; (c) launch of balloon from Minorca on 17 June 2013, 09:45 UT.

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CHARMEX SOP1 2013 – CHARMEX-S1-B75 – (B75)
Trajectory length 364.9 km, 197.0 NM – Distance from launch site 361.7 km, 195.3 NM – QDR from launch site 352.5 degree

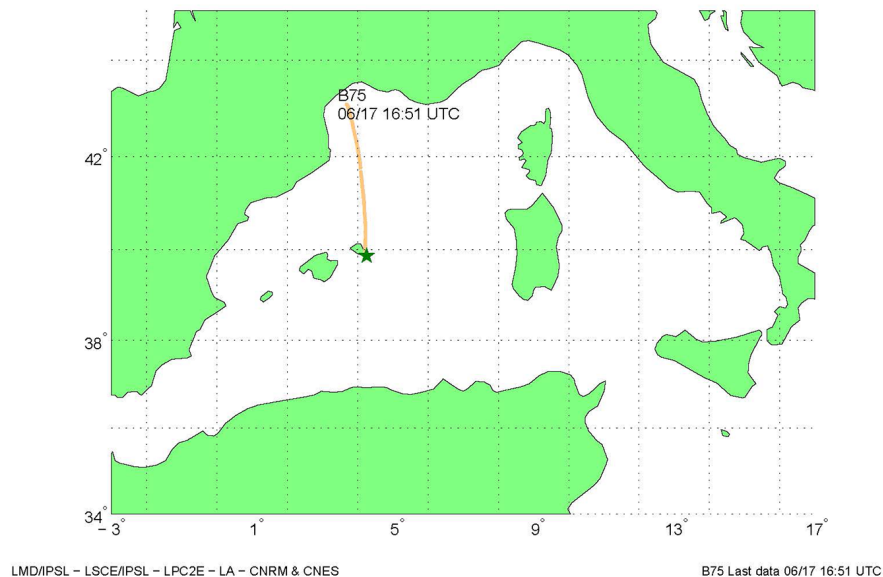


Figure 11. Trajectory of the LOAC 7 h long drifting balloon flight on 17 June 2013, at an altitude of about 2000 m.

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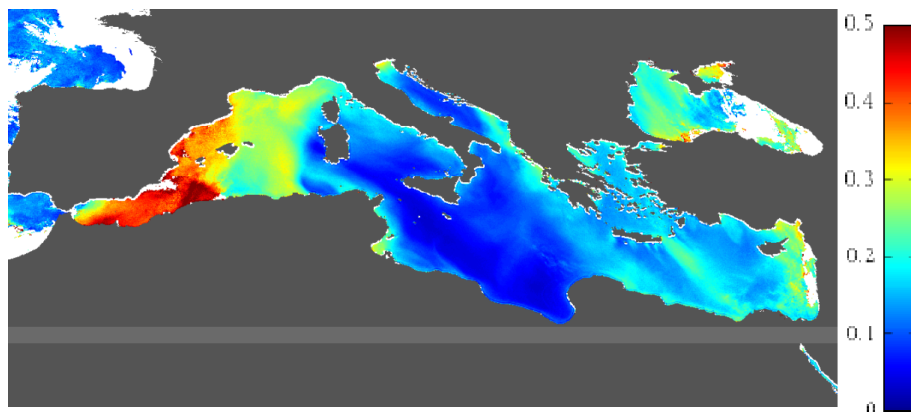


Figure 12. Daytime average aerosol optical depth at 550 nm derived from MSG/SEVIRI following Thieuleux et al. (2005; browse image courtesy ICARE/LSCE based on MSG/SEVIRI Level-1 data provided by Eumetsat/Eumetcast/LOA).

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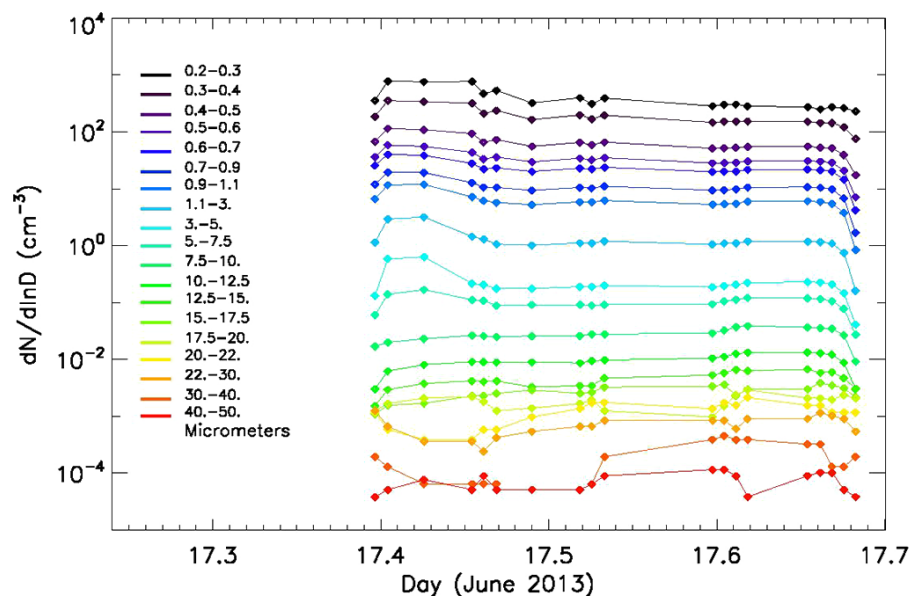


Figure 13. LOAC measurements inside a dust plume under the low tropospheric pressurized balloon during the ChArMEx campaign from Minorca, towards French coasts on 17 June 2013.

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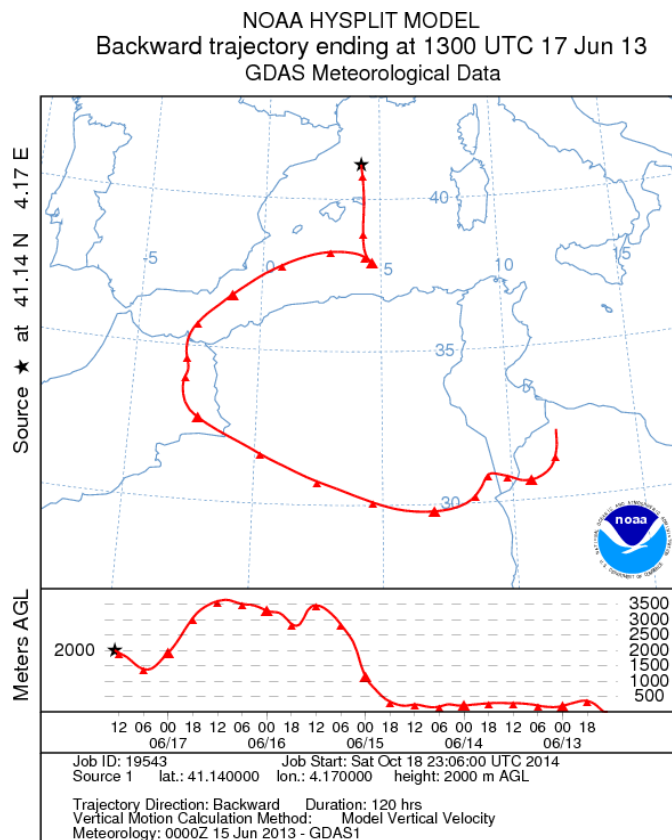


Figure 14. HYSPLIT air mass backward trajectory for LOAC balloon B75 (courtesy of NOAA Air Resources Laboratory).

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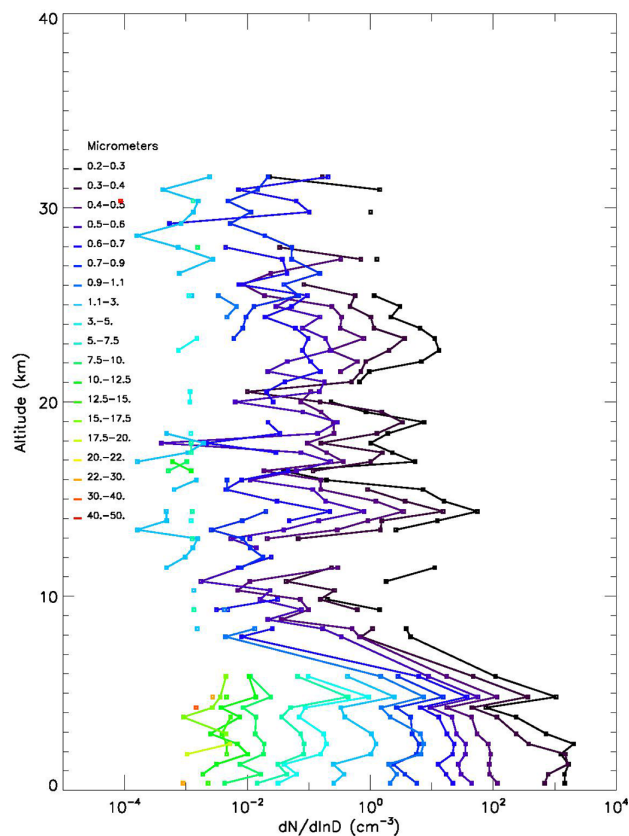


Figure 15. Particle concentrations up to 32 km in altitude from the LOAC flight under a meteorological balloon from Ile du Levant (France) during the ChArMEx campaign on 4 August 2013 between 15:30 and 17:30 UT; a sand plume is detected in the lower troposphere.

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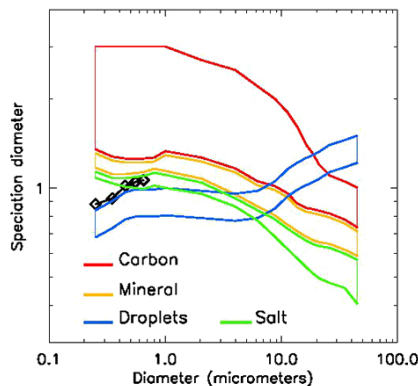
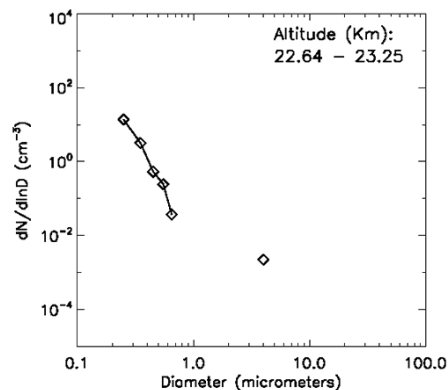
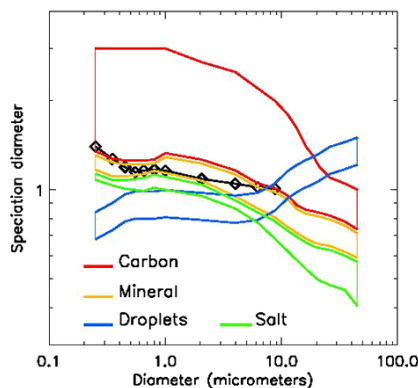
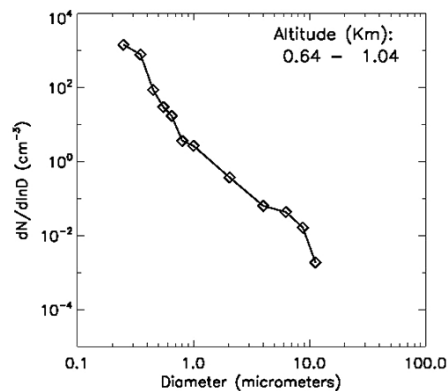


Figure 16. Examples of size distributions and speciations at two altitudes for the 4 August 2014 LOAC flight from Ile du Levant (France) during the ChArMEc campaign. At an altitude of ~ 1 km the speciation indicates mineral particles; at ~ 23 km, the speciation indicates liquid particles.

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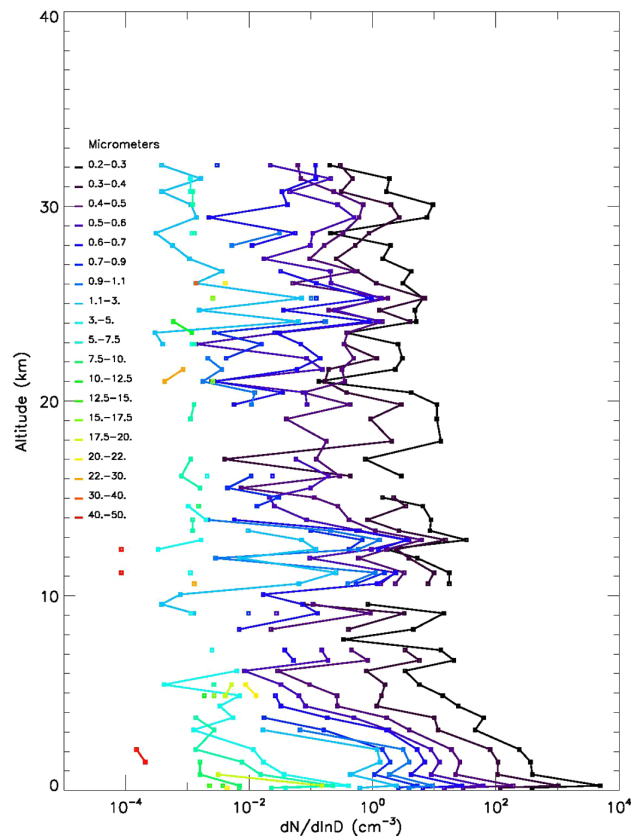


Figure 17. LOAC flight under a meteorological balloon from Aire-sur-l'Adour (France) on 20 March 2014 between 11:30 and 13:30 UT during the VOLTAIRE-LOAC campaign.

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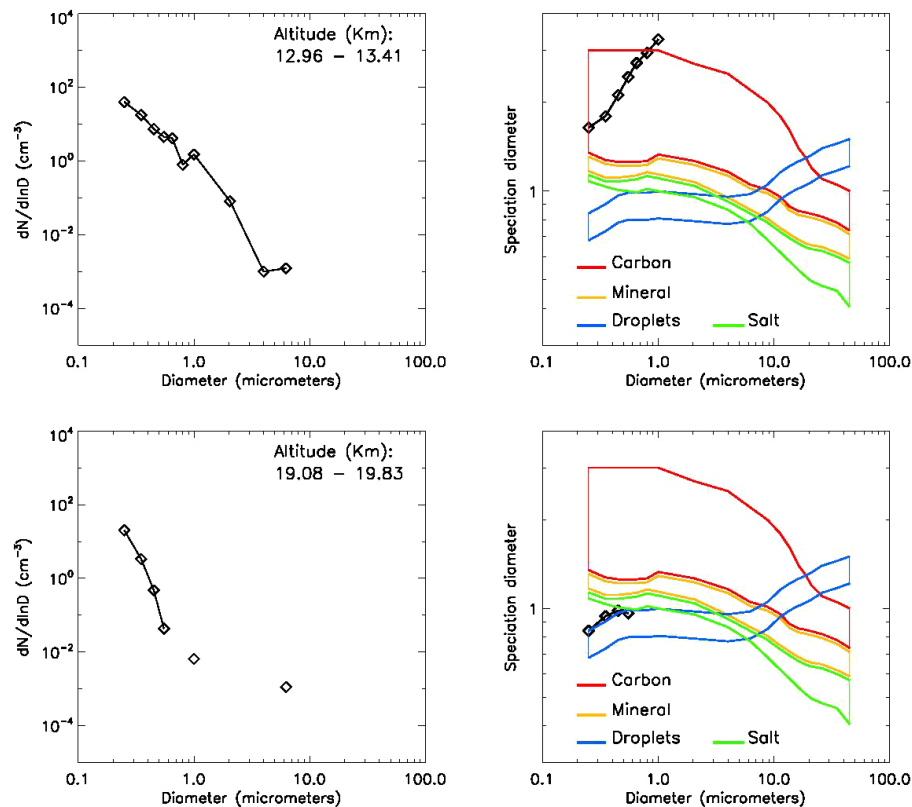


Figure 18. Examples of size distributions and speciations at two altitudes for 20 March 2014 LOAC flight from Aire-sur-l'Adour (France). At an altitude of ~ 13 km the speciation indicates carbon particles; at ~ 19.5 km, the speciation indicates liquid particles.