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Principle of  
measurements and  
instrument evaluation

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# LOAC: a small aerosol optical counter/sizer for ground-based and balloon measurements of the size distribution and nature of atmospheric particles – Part 1: Principle of measurements and instrument evaluation

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## Abstract

The study of aerosols in the troposphere and in the stratosphere is of major importance both for climate and air quality studies. Among the numerous instruments available, aerosol particles counters provide the size distribution in diameter range from few hundreds of nm to few tens of  $\mu\text{m}$ . Most of them are very sensitive to the nature of aerosols, and this can result in significant biases in the retrieved size distribution. We describe here a new versatile optical particle/sizer counter (OPC) named LOAC (Light Optical Aerosol Counter), which is light and compact enough to perform measurements not only at the surface but under all kinds of balloons in the troposphere and in the stratosphere. LOAC is an original OPC performing observations at two scattering angles. The first one is around  $12^\circ$ , and is almost insensitive to the nature of the particles; the second one is around  $60^\circ$  and is strongly sensitive to the refractive index of the particles. By combining measurement at the two angles, it is possible to retrieve accurately the size distribution and to estimate the nature of the dominant particles (droplets, carbonaceous, salts and mineral particles) in several size classes. This topology is based on calibration charts obtained in the laboratory. Several campaigns of cross-comparison of LOAC with other particle counting instruments and remote sensing photometers have been conducted to validate both the size distribution derived by LOAC and the retrieved particle number density. The topology of the aerosols has been validated in well-defined conditions including urban pollution, desert dust episodes, fog, and cloud. Comparison with reference aerosol mass monitoring instruments also shows that the LOAC measurements can be successfully converted to mass concentrations. All these tests indicate that no bias is present in the LOAC measurements and in the corresponding data processing.

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## 1 Introduction

The importance of measuring the concentration and size distribution of aerosols in the lower atmosphere has been highlighted by various studies. For instance, their presence in ambient air can have direct effects on human health (e.g. Zemp et al., 1999; Brunekreef and Holgate, 2002), and their interaction with solar radiation and clouds are affecting regional and global climate (Ramanathan et al., 2001; Diner et al., 2004; Kanakidou et al., 2005; Quaas et al., 2008). When very high concentrations of ashes after volcanic eruptions are present at cruise altitude, they can affect air traffic (e.g. Chazette et al., 2012). In the middle atmosphere, aerosols play a significant role in stratospheric chemistry through heterogeneous reactions with nitrogen and halogen species (e.g. Hanson et al., 1994, 1996), and they can affect climate through their role in the global radiative balance of the Earth (e.g. Hansen et al., 1992; Ammann et al., 2003). The concentration and size of the particles are highly variable due to the large variety of aerosol sources and properties, both of natural and man-made origin, and because of their relatively short residence time in the atmosphere. To understand and predict aerosol impacts, it is important to develop observation and monitoring systems allowing for their full characterization.

Instruments have been developed for routine measurements or for dedicated campaigns. Observations can be conducted from the ground, from unmanned aerial vehicles (UAV), from aircrafts, from balloons, and from satellites. To retrieve the physical properties of the aerosols, it is necessary to combine the information obtained with different instruments. In situ mass-spectrometers (Murphy et al., 2007) and aerosol collecting instruments (Brownlee, 1985; Blake and Kato, 1995; Allan et al., 2003; Bahreini et al., 2003; Ciucci et al., 2011) provide their composition. Optical instruments performing remote sensing measurements from the ground or from space with photometric, lidar, and extinction techniques (Shaw et al., 1973; Dubovik and King, 2000; Bitar et al., 2010; Winker et al., 2010; Salazar et al., 2013) provide indications on the size distribution and on the nature of the particles (liquid, carbon, minerals, ice, ...), generally

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assuming a priori hypotheses in the retrieval process. Complementarily, in situ optical measurements with optical particle counters can provide more accurate information on the size distributions of the particles.

The present study deals with optical aerosol particles counters (OPCs). The corresponding measurement principle relies on the properties of light scattered by particles injected in an optical chamber and crossing a light beam (e.g. Grimm and Eatough, 2009). The measurements are usually conducted at “large” scattering angles, typically around  $90^\circ$  with collecting angle of a few tens of degrees. At such angles, the light scattered is depending both on the size of the particles and on their refractive index. Conventional counters are calibrated using latex and glass beads and are post-calibrated using Mie calculations (Mie 1908) for liquid aerosols (the refractive index of latex beads and liquid aerosols is well known, assuming no imaginary part of the index i.e. non-absorbing aerosols). Some instruments can be also be post-calibrated for the observation of specific particles, as desert dust or urban pollutants, assuming a given value of their refractive index.

The refractive index dependence can be partially determined by performing measurements at different scattering angles, since the variation of the scattered intensity with scattering angles is strongly dependent on the refractive index of the particles (Volten et al., 2006; Francis et al., 2011). Thus, performing simultaneous measurements at different angles can provide an indication of the nature of the particles. Such an approach was used by Eidhammer et al. (2008) at angles of  $40^\circ$  and  $74^\circ$  mainly for the identification of mineral particles, and by Gayet et al. (1997) with a ring of detectors covering the whole scattering angle range for the identification of cloud droplets and icy particles.

Another approach was proposed by Renard et al. (2010a); in this case, measurements are conducted at small scattering angles, below  $20^\circ$ , where the light scattered is less sensitive to the refractive index of the particles. In this angular region, the scattered light is dominated by diffraction (which is not sensitive to the refractive index), at least for irregular grains as those found in the atmosphere. Such non-dependence of

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the refractive index was confirmed by measurements conducted at a scattering angle around  $15^\circ$  for different types of irregular grains (Lurton et al., 2014). In this case, the light scattered is dependent only on the size of the particles, allowing a better determination of the corresponding size distribution. However, the main problem of measurement at small angles is stray-light contamination. Thus a real-time correction of this signal offset due to the stray-light, which can vary with time, must be developed.

Aerosol particles counters are often used on the ground; some of them are used in the free atmosphere on-board aircraft or large balloons during dedicated campaigns, for example for the studies of desert dust events or volcanic aerosols (Bukowiecki et al., 2011; Jégou et al., 2013; Ryder et al., 2013) or for stratospheric studies (Rosen, 1964; Ovarlez and Ovarlez, 1995; Deshler et al., 2003; Renard et al., 2008, 2010b). We propose here a new optical particle counter concept, called LOAC (Light Optical Aerosol Counter) that is light and compact enough to perform measurements on the ground and under all kinds of balloons in the troposphere and in the stratosphere, including meteorological balloons. LOAC uses a new approach combining measurements at two scattering angles. The first one is around  $12^\circ$ , an angle for which scattering is weakly sensitive to the imaginary part of the refractive index nature of the aerosols, allowing the retrieval of the particle size distribution. The second one is around  $60^\circ$  where the light scattered is strongly sensitive to the refractive index of the particles, and thus can be used to evaluate their topology (liquid droplet are transparent, minerals are semi-transparent, and carbonaceous particles are strongly absorbing).

In this first paper, we will present the principle of measurements and calibration, and cross-comparison exercises with different instruments that detect atmospheric aerosols. In the companion paper, we illustrate first scientific results from airborne observations on-board balloons and unmanned aircraft (Renard et al., 2015).

## 2 Principle of measurements

### 2.1 Instrument concept

LOAC is a modular instrument, for which some parts can be changed depending on the measurements conditions. For measurements under balloon or on the ground in low wind conditions, the aerosols are collected by a metal profiled inlet designed to optimize the sampling conditions when oriented in the wind direction. The particles are drawn up to the optical chamber, through an isostatic tube and to the injector that focusses the flux inside the laser beam. LOAC uses a small vane-type pump (having a life-time of 3 weeks in continuous operation) working at  $\sim 2 \text{ L min}^{-1}$ . The pump is connected to the exit of the optical chamber by a flexible plastic tube. The optical chamber is open, thus the pressure is the same as outside. In-flight tests under sounding balloons have shown that the rotation speed of the pump is not affected by pressure variations.

For measurements in windy and rainy conditions, the inlet can be replaced by a total suspended particulate or TSP inlet rejecting rain droplets and particles greater than  $100 \mu\text{m}$ . For long-duration measurements, the small pump can be replaced by a robust pump; to maintain the aerosol detection efficiency, the pump flux must be in the range  $1.3\text{--}2.7 \text{ L min}^{-1}$ .

To minimize its weight, the optical chamber is in plastic Delrin<sup>®</sup>. The weight, including the pump, is of 300 g. The electric consumption is of 340 mA under 8 V (which corresponds to a power of 3 W). The optical chamber and the pump can fit in a rectangle box of about  $20 \text{ cm} \times 10 \text{ cm} \times 5 \text{ cm}$ .

LOAC is mainly designed for the detection of irregular grains, as those present in the ambient air (Fig. 1, from Institute of Physics of the University of Sao Paulo, Brazil). It uses a statistical approach for the size and concentration retrievals, as done for the laboratory PROGRA2 instruments dedicated to the study of optical properties of irregular levitating grains (Renard et al., 2002). Because of their shape, their orientation and their rotation in the air flow, the scattering properties of an individual grain vary with time at a given scattering angle (this variation could be more than a factor 2, as shown

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during laboratory tests by photodiodes and imagery measurements with PROGRA2). This must be taken into account for the calibration and data analysis. Thus, we propose here a calibration approach that can differ from the one used for other optical counters.

The sampled air crosses a laser beam of 25 mW working at the wavelength 650 nm. The homogeneity of the beam is of  $\pm 20\%$ . The scattered light is recorded by two photodiodes at scattering angles respectively in the  $12\text{--}16^\circ$  (hereafter called the  $12^\circ$  channel) and  $55\text{--}65^\circ$  (hereafter called the  $60^\circ$  channel), as shown on Fig. 2. Instead of using lenses to collect the light, the photons travel directly to the photodiodes through pipes, providing fields of view with a few degrees. The collecting area of the photodiodes is larger than the diameter of the pipes. This system prevents optical misalignment problems in case of vibrations and strong temperature variations like those encountered during atmospheric balloon flights. Such a concept of scattering measurements without collecting lenses was tested and validated by Daugeron et al. (2007).

The electronic sampling is at 40 kHz and the transit time of particles inside the laser beam is equal or lower than  $500\ \mu\text{s}$ . As said before, stray light contamination is high at small scattering angles and needs real-time correction. The stray light correction method presented in Renard et al. (2010a) was applied to the LOAC measurements. The stray light acts as a flux continuum, which can slightly vary over time due to changes in the temperature and pressure conditions and possible dust contamination in the optical chamber. The light scattered by the particles is superimposed on this continuum, which can be assumed as a continuous base-line over a short time interval. This baseline is determined before and after the intensity pulse produced by the particles that cross the laser beam.

The maximum of the intensity pulse is obtained after subtracting the stray-light contamination. Figure 3 presents an example of real ambient air measurements of the time evolution of the flux scattered by a  $5\ \mu\text{m}$  particle and by few submicronic particles. The pulse is slightly asymmetric, because the particles decelerate when crossing the optical chamber. This deceleration occurs because the diameter of the optical chamber is larger than the diameter of the inlet; thus the particles encounter pressure relaxation.

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Some secondary flux maxima may be present in the pulse and can be attributed to the rotation of irregular shaped particles in the air flow. The search for a new intensity peak is inhibited until the flux decreases to a given threshold, represented in Fig. 3 by the red line. This procedure prevents multiple counting of the same particle (of irregular shape) that exhibits secondary flux maxima.

This pulse analysis is performed by the on the on-board electronics, which provides every 10 s the concentrations detected by the 2 channels. Until now, an additional computer has been necessary to record and analyse the data.

## 2.2 Calibration

The calibration of an optical counter is not an easy task, especially for the detection of irregular particles (Whitby and Vomela, 1967; Gebhart, 1991; Hering and McMurry, 1991; Belosi et al., 2013). A first presentation of the calibration procedure for measurements at small scattering angles using a LOAC optical chamber can be found in Lurton et al. (2014).

Only the  $12^\circ$  channel, which is insensitive to the refractive index of the particles, must be calibrated. The  $60^\circ$  channel will be used as a comparison to the  $12^\circ$  channel measurements to determine the typology of the aerosol, as explained in the Sect. 2.4.

Monodisperse latex beads, which are perfect transparent spheres, have been used for diameter calibration in the  $0.2\text{--}4.8\ \mu\text{m}$  range; glass beads have been used at  $5\ \mu\text{m}$  (see Figs. 2 and 3 of the Lurton et al., 2014 paper for the LOAC response to monodisperse beads). In fact, Mie calculations show that the scattered flux encounters strong oscillations linked to small changes both in diameter and in scattering angle. Conventional aerosol counters use large field of view, typically a few tens of degrees, to average these oscillations. On the opposite, the LOAC  $12^\circ$  and  $60^\circ$  measurement channels have a field of view only of few degrees and use no lens. The detected flux at the  $12^\circ$  channel is then very sensitive to the position of the individual bead inside the laser beam, and thus to its scattering angle. Taking into account this constraint, we

considered here only the highest flux scattered by each size class of monodisperse beads.

The electronic noise is lower than 20 mV at ambient temperature and lower than 10 mV at low air temperatures. Statistically speaking, the noise is divided by the root mean square of the number of identical measurements (here the number of events detected in a given size class). To reach a 1 mV accuracy in case of 20 mV noise, which is necessary to be able to discriminate the smaller size classes and to establish accurately the size distribution, at least  $20 \times 20$  (= 400) particles must be detected for each size class.

During laboratory calibration, it is easy to reach such concentration levels using monodisperse beads. During real measurements inside the atmosphere, we must ensure that such particle concentrations are indeed present for all size classes below  $1 \mu\text{m}$ . The LOAC has an integration time of 10 s, with a pumping flow of about  $2 \text{ L min}^{-1}$ . Even in low polluted ambient air at ground (“background conditions”), all counting instruments have shown in the past that concentrations are greater than  $1 \text{ particle cm}^{-3}$  for size classes smaller than  $0.5 \mu\text{m}$  (having a  $0.1 \mu\text{m}$  width), which corresponds to  $2000/6 =$  more than 300 particles during the 10 s LOAC integration time. For particles in the  $0.5\text{--}1 \mu\text{m}$  size classes (having a width of 0.1 or  $0.2 \mu\text{m}$ ), concentrations are greater than  $0.1 \text{ particle cm}^{-3}$ , giving more than 30 particles per size class.

For all the cross-comparison exercise presented below, the measurements were integrated from 2 to 15 min. For the 2 min integration time, the number of particles given above must be multiplied by 12, giving are at least 3000 for the 3 first size classes and 300 for the other ones. For a 15 min integration time, these numbers must be multiply again by 7.5. Thus, the LOAC class identification can be conducted with the expected accuracy in the ambient air. Obviously, in case of polluted air, all these values could be also 2 to 3 orders of magnitude higher ( $1000 \text{ particles cm}^{-3}$  between  $0.2$  and  $0.3 \mu\text{m}$  is often encountered).

In case of very low particle concentrations, as those that can be encountered during flights in the stratosphere with typically less than  $1 \text{ particle cm}^{-3}$  greater than  $0.2 \mu\text{m}$ ,

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chamber due to changes in the air flow speed between the aerosols injector and the optical chamber.

Thus, the LOAC detection size range is between 0.2 and  $\sim 100 \mu\text{m}$ . LOAC, with its present calibration procedure, is operated to the detection of irregular grains and droplets, but not to perfect spherical solid grains, such as latex or metal beads for which uncertainties arise from the smoothing of Mie oscillations by the calibration curve (in this case, the total concentration is correct but the size attribution can be erroneous).

Overall, a total of 19 size classes are defined for diameters between 0.2 and  $100 \mu\text{m}$  (Table 1). The upper limit can be lower, however, depending on the sampling collection cut-off of the inlet. The size classes are chosen as a good compromise between the instrument sensitivity and the expected size distribution of ambient air aerosols.

### 2.3 Concentration measurements

Counting is conducted while the particles cross the laser beam one by one, and are classified in size classes corresponding to the scattered flux. The measurements are integrated during 10 s and are converted to number densities or particles  $\text{cm}^{-3}$ . The detectors of the two channels ( $12^\circ$  and  $60^\circ$ ) work asynchronously.

This discrete detection works well for large particles greater than  $2 \mu\text{m}$ , with uncertainty in size attribution of 10 %. For smaller particles, the size determination is within the calibration errors bars ( $\pm 0.025 \mu\text{m}$  for particles smaller than  $0.6 \mu\text{m}$ , 5 % in the  $0.7\text{--}1 \mu\text{m}$  range) if more than 400 particles are detected for each size classes.

The optical and electronic response of the system has been modelled by a numerical Monte-Carlo method, taking into account the shape of the laser beam, the speed of the particles inside the laser beam and the instrument noise. To ensure a good statistical approach,  $10^4$  particles were randomly injected for each size class. The ratio of the number of detected particles over the number of injected particles provides the detection efficiency. For the smallest particles, only the brightest part of the peak of the pulse of the scattered flux is observable and the apparent transit time in the laser beam is a few tens of  $\mu\text{s}$  (in Figs. 3 and 4 small peaks are present). Thus, some par-

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ticles cannot be detected. The detection efficiency increases as the diameter of the particles increases, and is reaching 100 % for particles larger than 1  $\mu\text{m}$ . The concentrations of submicron size particles are then corrected using these detection efficiency coefficients.

For particles larger than 1  $\mu\text{m}$ , the observed transit time in the laser beam is at its maximum ( $\sim 500 \mu\text{s}$ ) and the expected maximum concentration is of about 20 particles  $\text{cm}^{-3}$ . In fact, even higher concentrations can be determined using a statistical approach when several particles cross the laser beam almost simultaneously. The higher are the concentration, the lower is the probability that the scattered intensity peaks decreases below the threshold to start a new counting. In this extreme case, the real concentrations are inversely proportional to the detected concentrations. Another Monte-Carlo numerical modelling was conducted to establish the relationship between the number of particles  $> 1 \mu\text{m}$  detected and the number of particles injected in the laser beam (Fig. 6). In the simulations, particles were randomly injected (in time), with concentrations increasing from 0 to 500 particles  $\text{cm}^{-3}$  by step of 1 particle  $\text{cm}^{-3}$ . The response is almost linear up to 10 particles  $\text{cm}^{-3}$ , reaching a kind of saturation value at around 15 particles  $\text{cm}^{-3}$ , and decreases for larger concentrations. It is obvious that such a corrective procedure must be used only in dense aerosol media (more than 10 particles greater than 1  $\mu\text{m}$   $\text{cm}^{-3}$ ), as fog or clouds, i.e. in conditions which must be confirmed by independent measurements. At present, this procedure is applied only when large droplets are detected by LOAC using the topology procedure presented below. In this case, up to 200 large particles  $\text{cm}^{-3}$  can be detected.

For the smaller particles, the signal of the scattered flux is close or inside the noise. The photodiodes cannot detect the whole transit of the particles inside the laser beam, but just the brighter part of the peak. Thus, the effective acquisition time can be reduced down to 35  $\mu\text{s}$  instead of around 500  $\mu\text{s}$  for the largest particles. Then, a greater number of particles can be detected. Taking into account also the detection efficiency for the smaller particles, up to 3000 particles  $\text{cm}^{-3}$  can be (statistically) detected.

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For the LOAC integration time of 10 s, the counting uncertainty can be derived from the Poisson counting statistics. This uncertainty, defined as the relative standard deviation, is 60 % for aerosol concentrations of  $10^{-2} \text{ cm}^{-3}$ , 20 % for  $10^{-1} \text{ cm}^{-3}$ , and 6 % for concentrations higher than  $1 \text{ cm}^{-3}$ . Such uncertainties can be reduced by averaging the concentration measurements for each size classes or by increasing the integration time. Nevertheless, such calculation does not take into account the real instrumental uncertainties dominated by the electronic noise and the inlet sampling efficiency.

LOAC is designed to be used in various atmospheric conditions. The temperature can dramatically change, in particular during balloon flights up to the middle stratosphere. The electronics offset can change with time because of the sensitivity of the electronic components to atmospheric temperature variations. The instrument performs a check of its noise level after 10 min of measurements. If the noise differ by more than 50 % from the previous check, an electronic re-calibration is automatically performed to estimate the offset variation and to adjust the calibration. A processing software is applied after the experiment to check the offset time-evolution during the 10 min periods and to then correct the raw measurements.

## 2.4 Aerosols typology

The scattered flux recorded at  $60^\circ$  is very sensitive to the refractive index of the particles and thus to their nature (as said before this phenomenon appears at scattering angles greater than  $\sim 20^\circ$ ). The more absorbing they are, the lower the recorded fluxes. Thus, we use the “ $60^\circ$  channel” as a diagnostic for the effect of the refractive index on the scattered fluxes. This channel uses the same flux threshold (in mV) as the  $12^\circ$  channel, in order to perform a direct comparison of the counting detected by two channels. For a given size class and for a given particle concentration recorded in the  $12^\circ$  channel, the concentration detected by the  $60^\circ$  channel decreases when the imaginary part of the refractive index increases. This increase of the imaginary part leads to an underestimation of the real size of the particles, and thus produces a diameter bias in the size distribution (diameter vs. concentration) for the  $60^\circ$  channel with respect to the



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12° channel. An example of the procedure used to determine this effect is presented in Fig. 7, where the size distributions of the two channels are presented. For a given particle size of the 12° channel (noted D1), we consider the concentration value of the 60° channel. Then we search for the same concentration value on the 12° channel (a linear interpolation is used if needed). The corresponding diameter is then determined (D2). Finally, we define a so-called “speciation index” as the ratio D2/D1. The more absorbing the particles are, the higher this ratio. This procedure is conducted for each size class.

This procedure works well for irregular particles, but not for solid symmetrical particles; in this later case, the Mie oscillation will produce strong fluctuations in the evolution of the speciation index with size (we have indeed observed this effect inside some cirrus clouds). Also, this procedure must be used only for a large enough number of detected particles per size class, because of the irregular shape of the particles. Laboratory tests have shown that about 20 particles in a size class are sufficient to be able to indicate the aerosol topology. In its nominal operating mode, LOAC provides the speciation index every 1 min. For the analysis of continuous ground-based measurements presented below, we have conducted the topology detection with an integration time of 15 min (assuming that the aerosols are stationary).

Different types of particles have been tested in the laboratory to assess the amplitude of the speciation index throughout the measurement size range: organic carbon, black carbon, desert dust or sand from different origins (excluding black sand), volcanic ashes, plaster, salt (NaCl), water droplets, droplets of mixture of water and sulphuric acid. They can be classified in 4 families: carbonaceous particles, minerals, salts and liquid droplets. Figure 8 presents the curves obtained in laboratory for the various samples. Then, “speciation zones” charts (speciation index vs. real diameter) are defined by the minimum and maximum speciation index values reached by each family, taking into account the measurement uncertainties. Among solid particles, carbonaceous particles produce the higher speciation index and salt the lower, mineral particles being in between. Detailed analysis has shown that most of the carbon particles are in the

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lower part of the carbon speciation zone while some strongly absorbing particles, perhaps black carbon having fractal shape, are in the middle and upper part of the carbon speciation zone. For all solid particles, the global trend is a decrease of the speciation index with increasing size. On the contrary, the liquid droplets speciation index exhibits an increase with increasing diameter.

The case presented in Fig. 7 has  $D1 = 0.35 \mu\text{m}$  and  $D2 = 0.51 \mu\text{m}$ , leading to a ratio of 1.46, which is in the carbon speciation zone.

The speciation indices obtained from LOAC observations in the atmosphere are compared to these reference charts obtained in the laboratory. The position of the data points in the various speciation zones provides the main topology of the particles. In principle, this procedure can be conducted for each size class. In fact, due to the statistical dispersion of the results, it is better to consider several consecutive size classes to better conduct the identification. This is in particular necessary for the identification of droplets, whose speciation zone crosses all the speciation zones of the solid particles.

It is obvious that the identification of the typology of the particles works well in case of a homogenous medium, when the speciation indices are not scattered through the various speciation zones.

At present, the speciation zones are established for particles expected to be found in the troposphere and stratosphere, but it is an evolving data base. Additional laboratory measurements can be conducted to retrieve the speciation zones for specific particles in case of measurements in new specific environments.

## 2.5 Reproducibility

The instrument is industrially produced by Environnement-SA (<http://www.environnement-sa.com>); more than 100 copies were produced up to mid-2015. Tests have been conducted for the different parts of the instrument: diode, pump, photodiode and electronics, to assess their reproducibility and thus to establish the measurements uncertainty.



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specific studies where very large particles dominate, as measurements inside fog or clouds, or because of mechanical constraint if a TSP inlet cannot be mounted, the particles can be collected by a tube having a bevelled metal inlet and oriented downwards. In this case, the largest particles are generally under-sampled and a corrective coefficient must be applied, taking into account the direction and the speed of the wind.

For measurements under balloons floating at constant altitude, the relative speed between ambient air and the inlet is close to zero. The sampling efficiency assessed using the Agarwal and Liu (1980) criterion for an upward-facing inlet shows that the sampling is unbiased for particle with diameter below 20  $\mu\text{m}$ .

The sampling line used during the flights is composed of a thin wall metallic probe and antistatic tubing. The thin wall aerosol probe has an inlet diameter equal to 5.4 mm and is connected to a tube of about 20 cm long and 6.7 mm internal diameter. The sampling line is connected vertically to the LOAC. Nevertheless, due to the tube stiffness, the line can be inclined with a maximum sampling angle of 30° from vertical. The sampling efficiency of the line was assessed using modelling calculations in order to account for changes in atmospheric pressure, temperature and possible changes of the probe orientation during these flights. For that purpose, the values of pressure and temperature as a function of altitude are taken from the international standard atmosphere. Sampling efficiency calculations have been made by considering a balloon ascending velocity of 5  $\text{m s}^{-1}$ , a LOAC sampling flow rate equal to 1.7  $\text{L min}^{-1}$  and two angles of the sampling line from the vertical (0 and 30°). According to these parameters, the inlet aspiration velocity of the probe is equal to 1.24  $\text{m s}^{-1}$  (sub-isokinetic) and the flow is laminar in the tubing for all altitudes.

The mechanisms considered to calculate the sampling efficiency are the inlet efficiency of the probe in isoaxial and isokinetic sampling conditions (Belyaev and Levin, 1974; Hangal and Willeke, 1990) and particle losses in the tubing due to gravitational settling when the line is not perfectly vertical (Heyder and Gehbart, 1977). Calculations have been conducted for particles with diameter ranging from 0 to 20  $\mu\text{m}$ , and from the ground to an altitude of 30 km. Figure 9 presents the sampling efficiency for a 0°

deviation (isoaxial) and for a 30° deviation of the sampling line with respect to the vertical. Data are plotted according to the particle aerodynamic diameter which describes particle settling and inertia phenomenon.

In isoaxial conditions, results show for all altitudes an increase of sampling efficiency with the particle diameter, up to a factor > 3 for the largest particles. In this case, there is no particle deposition in the sampling line and the sampling is dominated by sub-isokinetic conditions (apparent wind velocity higher than inlet probe velocity). A sampling efficiency higher than unity is explained by the particle inertial effect resulting from the divergence of the flow field at the inlet of the probe. The increase in sampling efficiency with altitude is due to changes in air viscosity and gas mean free path with temperature and pressure.

When the tube is inclined by 30° from the vertical, the sampling efficiency is between 1 and 2. The sampling efficiency is lower than for the 0° isoaxial conditions. Firstly, the sub-isokinetic effect is reduced by the orientation of the tube, and secondly, deposition can occur in the tubing due to particle settling.

Since the tube always has a deviation of about 30° during the balloon flights, we consider only the results at 30° from the vertical. The over-sampling effect is negligible for particles smaller than 5 μm up to the lower stratosphere and for particles smaller than 2 μm in the middle stratosphere. Thus, this effect will just affect the retrieved concentrations of the largest particles.

The results of these theoretical calculations are not yet fully validated by an experimental approach with LOAC itself. Thus, all balloon measurements in the stratosphere will not be corrected from this aerodynamic effect. It could be taken into account in future work involving large particles, for example when converting concentrations to extinction by comparison with remote sensing instruments, or to estimate the real concentration of the interplanetary dust in the middle atmosphere.

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### 3 Cross-comparison with other instruments

Various cross-comparisons have been conducted in ambient air at ground and during balloon flights for concentrations and topology identification. LOAC concentrations are compared to other commercial particle counter instruments and photometer measurements, although there is no absolute reference, many of them are using different technical approaches and calibration procedures. The LOAC topologies are validated during well-identified atmospheric events of liquid and solid particles. Finally, the LOAC particle concentrations are converted to mass concentrations to be compared to commercial microbalance mass instruments used as reference instruments in air quality monitoring. Table 2 summarizes the conditions of measurements.

The LOAC was used under different conditions. When deployed underneath meteorological and tropospheric balloons, the data are transmitted in real time by telemetry. For deployments under large stratospheric balloons, the data are stored on-board using a specific module. On the OAG tethered balloon, the data are sent to the ground using a Wi-Fi link and are stored on a computer. An autonomous version for automatic ground-based applications is now also available, including an on-board computer to record the data.

#### 3.1 Concentrations and size distribution

Continuous measurements have been conducted in ambient air at the SIRTA observatory (Site Instrumental de Recherche par Télédétection Atmosphérique, <http://sirta.ipsl.fr/>) at Palaiseau, South of Paris, France (48.713° N, 2.208° E), during the ParisFog campaign, <http://parisfog.sirta.fr/>, from November 2012 to April 2013. During this period, the total concentrations of aerosols have been monitored by a WELAS aerosols counter, a Fog Monitor (counter for large droplets) and a SMPS (Scanning Mobility Particle Sizer) in common size range domains with LOAC. SMPS measurements are based on the electric mobility diameter of the particles, while the other instruments determine optical diameters. These two diameters could differ depending on the nature

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of the particles; thus the direct comparison of the measurements (even in the same air mass) could be sometimes biased.

Figure 10 presents the cross-comparison of the instruments in January 2013. Most of the measurements were conducted in background aerosol conditions, although some small fog events were detected and can be identified by concentration peaks in the Fog Monitor measurements. Roughly speaking, the order of magnitude of the concentrations is similar, although some significant discrepancies are present. To investigate their possible origin, the size distribution obtained in different conditions of aerosol content can be compared. Figure 11 (upper panel) presents an example where the agreement in total concentration during background aerosol conditions is very good between LOAC and SMPS. On the other hand, the shape of the size distribution of the WELAS instrument is unusual with a decrease of the sub- $\mu\text{m}$  aerosol concentrations with decreasing size (the opposite trend is expected for background aerosol conditions). This could be due to calibration problem of the latter instrument; Indeed, Heim et al. (2008) report a strong decrease in counting efficiency with decreasing particle diameter for submicron particles for the WELAS-2100 OPC. Finally, as expected, the Fog Monitor measurements are not useable (no fog at this time and only noise is present). Figure 11 (lower panel) presents the same cross-comparison in case of significant disagreements between all the instruments. LOAC seems to underestimate the concentrations of the smallest particles, but this can be due to the difficulty of reconciling the retrieved diameters for the different measurement techniques in case of different types of particles.

Strong fog events were observed in November 2012. LOAC, WELAS and Fog Monitor are in very good agreement during these events (Fig. 12). This result validates the correction procedure applied to the LOAC measurements in case of dense medium of liquid particles. Between fog events, LOAC and WELAS were sometimes in disagreement. This was due to the difference in the concentration values obtained by the two instruments for the particles smaller than  $\sim 0.5\ \mu\text{m}$ , which may be attributed to the WELAS undercounting. Figure 13 presents the size distribution at the beginning

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of a fog event, with the typical enhancement around a diameter of  $10\ \mu\text{m}$  (e.g. Singh et al., 2011), and at the end of the event. Both LOAC and WELAS found a bimodal size distribution although the WELAS shows a doubtful sharp decrease in concentration for the smallest sizes., but disagree for the size of the second mode at  $\sim 10\ \mu\text{m}$ . On the opposite, LOAC and Fog Monitor were in good agreement for the position of the second mode, although the population of the first size class of Fog Monitor was obviously underestimated. Finally, for the largest sizes, LOAC concentrations are in-between those of the WELAS and Fog Monitor.

A ground-based measurement session was conducted from Minorca (Spain) during the ChArMEx campaign (Chemistry Aerosol Mediterranean Experiment, <https://charmex.lsce.ipsl.fr/>) in parallel with measurements of an HHPC-6 aerosol counter in the period 12 June–2 July 2013. The orders of magnitude for the different size classes were in good agreement. In particular, both instruments captured an aerosol enhancement of large solid particles between 18 and 21 June 2013, as shown in Fig. 14 for the size distribution.

The last cross-comparison exercise was conducted during an ambient air campaign at SIRTa observatory, site 5 near Gif-sur-Yvette, South of Paris, France ( $48.709^\circ\ \text{N}$ ,  $2.149^\circ\ \text{E}$ ), in the beginning of 2014. LOAC performed measurements from 3 to 13 February 2014 in parallel with a SMPS, a Grimm aerosol counter and a HHPC-6 aerosol counter. Due to the sampling conditions that vary from one instrument to another (direct sampling, TSP inlet, dryer, direct or curved tubes), the analysis is limited to the smallest particles (diameter  $< 1\ \mu\text{m}$ ) which are almost insensitive to the sampling techniques. Figure 15 presents the temporal cross-comparison for 4 size-classes:  $0.2\text{--}0.3$ ,  $0.3\text{--}0.5$ ,  $0.5\text{--}0.7$  and  $0.7\text{--}1\ \mu\text{m}$ . In fact, the size classes of the 4 instruments are not always the same, thus the closest ones have been considered for the comparison.

Globally, all the instruments give similar concentrations for all size classes, the better agreement being for the  $0.5\text{--}0.7\ \mu\text{m}$  diameter range. Some discrepancies appear for some time periods, but several reasons can explain them. First, the SMPS instrument determines the electric mobility diameter that can depend on the nature of the aerosols,



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whereas the other instruments determine optical diameters. SMPS measurements could lead to some uncertainties in size determination, and thus in concentrations, when compared to other kinds of instruments for irregular particles (e.g. Gulijk et al., 2003). This could explain why LOAC has missed some concentration peaks detected by SMPS. Secondly, the particles size distribution of sub- $\mu\text{m}$  particles exhibits a strong decreasing when the diameter increases. Thus the uncertainty in the size calibration of a few hundredth of  $\mu\text{m}$  could induce concentration differences of at least a factor 2. This is presented in Fig. 15 for the 0.7–1  $\mu\text{m}$  comparison with the Grimm instrument for which both 0.65–1 and 0.8–1  $\mu\text{m}$  concentrations are plotted. Finally, the Grimm and HHPC-6 instruments are sensitive to the nature of the particles, and changes in the type of aerosol (for example mineral or carbon particles) could partially affect their size determination.

An indirect evaluation of the LOAC size calibration has been conducted during the ChArMEX campaign on the Balearic island of Minorca, Spain. A total of 9 flights of LOAC have been performed under a meteorological sounding balloon launched from Sant Lluís airfield (39.865° N, 4.254° E) in the 15–19 June 2013 period during a desert dust transport event. The aerosol concentration has been integrated for all size classes from the ground to the highest altitude reached by the balloon, i.e. an altitude of about 30 km, to be compared to ground-based remote sensing measurements provided by the AERONET photometer network (<http://aeronet.gsfc.nasa.gov/>) station of Cap d'En Font (39.826° N, 4.208° E), which performed measurements close the trajectory of the LOAC balloon measurements. AERONET provides the vertically integrated volume concentration of aerosols (in  $dV/d\ln(r)$ , where  $r$  is the radius of the particles) in the 0.13–30  $\mu\text{m}$  radius range (Dubovik et al., 2000).

The LOAC integrated concentrations are converted to volume concentrations by using the mean volumetric diameter  $D_v$  calculated for each size class by the formula:

$$D_v = 0.5 \times \left[ \left( D_{\min}^3 + D_{\max}^3 \right) \right]^{(1/3)},$$



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rological balloon on 16 June 2014; two LOAC flights were conducted on 19 June 2013 at the same time, the first one being under a meteorological balloon and the second being under a pressurized tropospheric balloon (see the companion paper for more information of the balloons and the gondolas, Renard et al., 2015). To be compared to lidar extinction data at 350 nm, the LOAC data were converted to extinction using Mie scattering theory, assuming spherical sand particles having an refractive index of  $n = 1.6 + 0.05i$  (such index is an average of values available in the literature). Uncertainties of the refractive index values are included in the errors bars calculations of the retrieved LOAC extinctions. Figure 17 presents the tropospheric vertical profile of LOAC and WALI lidar extinctions. Taking into account the instrumental errors bars, LOAC and WALI have captured the same main vertical structures and the extinction values are in average in good agreement in the lower troposphere.

### 3.3 Topology of the particles

The speciation zones, obtained from laboratory measurements, must be validated in real atmospheric conditions.

Urban ambient air measurements are proper for the detection of carbon particles (black and organic carbon), especially during well-identified pollution events. Permanent LOAC measurements have been conducted at “Observatoire Atmosphérique Generali” (OAG) in the South-West of Paris since May 2013 (48.841° N, 2.274° E). This observatory is a recreational tethered balloon operated in a public park; the LOAC measurements nominal maximum altitude is 120 m but some flights could be conducted up to an altitude of 270 m. The measurements can be sorted out between measurements with the balloon at ground level and measurements during flight. Figure 18 presents an example of light absorbing particles (probably carbonaceous ones) detected at the OAG on 29 December 2013 around 07:30 UT. In this example, the speciation index curve is well inside the carbon speciation zone in the whole size range up to  $\sim 10 \mu\text{m}$ .

In addition to sounding balloons mentioned above, measurements under drifting balloons launched from Sant Lluís on Minorca Island were also conducted during sev-





particle density. The mass densities were determined for each size class by identifying the typology of the particles through their speciation index. The mass densities chosen here are:

- 2.2 g cm<sup>-3</sup> for salt; a value corresponding to NaCl particles;
- 2.2 g cm<sup>-3</sup> for mineral particles; this value is a compromise for common mineral particles present in ambient air: compact sand (2.1 g cm<sup>-3</sup>), quartz (2.7 g cm<sup>-3</sup>), limestone (2.5 g cm<sup>-3</sup>) and silicon (2.3 g cm<sup>-3</sup>);
- 1.4 g cm<sup>-3</sup> for carbonaceous particles. This value was derived after detailed tests during the comparison between LOAC and microbalance measurements in the laboratory. It lies well inside the range of values proposed in the literature for such particles (e.g. Chen et al., 2010; Virtanen et al., 2006; Spencer et al., 2007). Sensitivity tests have shown that a 10% variation of this value will not induce strong changes in the results presented below.
- A value of 0.0 g cm<sup>-3</sup> was used for water droplets, for comparing LOAC measurements to those of the TEOM instrument, which tends to evaporate condensed water.

The duration of the sessions was from several hours to several days. Figure 23 presents the mass measurements for particles smaller than 20 µm, averaged on 24 h for the two instruments. The variability of the concentrations is related to the amount of particles injected into the room. The lowest values correspond to measurements without injection. In this case, LOAC indicates that only particles smaller than 2 µm were present in the air. The LOAC and TEOM measurements are in very good agreements, with a correlation of 0.97. The correlation curve has the slope of 0.98, with an offset at the origin of 2.2 µg m<sup>-3</sup>, and a mean error of 4.8 µg m<sup>-3</sup>.

Sessions of ambient air measurements were conducted in Paris and in its suburbs, to test the retrieval of PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations, with pumps working at 2.7 L min<sup>-1</sup>. The first location of measurements is at the “Observatoire Atmosphérique

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Generali (OAG)” in Paris (latitude 48.8417° N, longitude 2.2736° E). The LOAC measurements were conducted using a TSP inlet. The second location is at SIRTA observatory at Palaiseau (48.7180° N, 2.2075° E) during the ParisFog campaign. The LOAC measurements were conducted with the metal inlet directed towards the ground.

The OAG and SIRTA measurements considered here were conducted in the periods September 2013–April 2014 and September 2013–December 2013, respectively. The PM<sub>2.5</sub> and PM<sub>10</sub> LOAC mass concentrations were retrieved by combining the results for particles smaller than 3 µm and smaller than 10 µm, respectively, taking into account the sampling efficiency of the PM<sub>2.5</sub> and PM<sub>10</sub> inlets currently used by the air quality networks (cut-off at 2.5 µm for PM<sub>2.5</sub> inlet and cut-off at 10 µm for PM<sub>10</sub> inlet).

Reference mass concentrations data of urban ambient air in the Paris region are provided by the Airparif network (<http://www.airparif.asso.fr/>), operating TEOM microbalance instruments. Unfortunately, there is no Airparif station very close to the OAG site nor to the SIRTA site at the time of the measurements. Therefore, we decided to use data recorded at 3 stations that have environmental conditions close to those at OAG and SIRTA: Paris Centre (latitude 48.8528° N, longitude 2.3600° E), Vitry-Sur-Seine (latitude 48.7820° N, longitude 2.3992° E) in the south-eastern suburb area of Paris, and “Rural South” at Bois-Herpin (latitude 48.3725° N, longitude 2.2258° E) in the South of the Paris region; the last station provides background conditions measurements.

Figures 24 and 25 present the comparison of PM<sub>2.5</sub> and PM<sub>10</sub> concentrations, for the 2013 and 2014 period, respectively. The LOAC measurements are most of the time between the background and the urban conditions, the small discrepancies with the reference mass concentrations are probably due to a difference in the wind direction and to the regional-scale transport of the particles. It is worth noting that LOAC did capture well the 10–15 December 2013 and the 11–14 March 2014 pollution peaks.

These measurement sessions have been conducted with different kinds of pumps and of inlet systems. The agreement with reference mass concentration measurements is very good. This confirms that no obvious bias is present in LOAC observations for the sizes of particles considered here (~ 0.2–20 µm), and that the topology procedure is







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**Table 1.** The 19 size classes of LOAC for concentration measurements.

	Diameter range ( $\mu\text{m}$ )
1	0.2–0.3
2	0.3–0.4
3	0.4–0.5
4	0.5–0.6
5	0.6–0.7
6	0.7–0.9
7	0.9–1.1
8	1.1–3.0
9	3.0–5.0
10	5.0–7.5
11	7.5–10.0
12	10.0–12.5
13	12.5–15.0
14	15.0–17.5
15	17.5–20.0
16	20.0–22.0
17	20.0–30.0
18	30.0–40.0
19	40.0–100.0





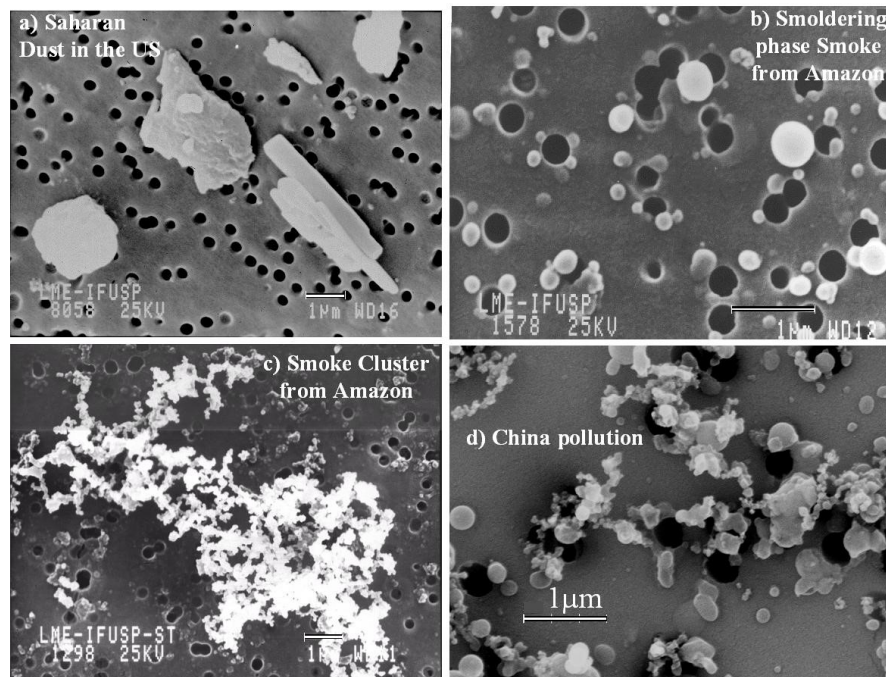
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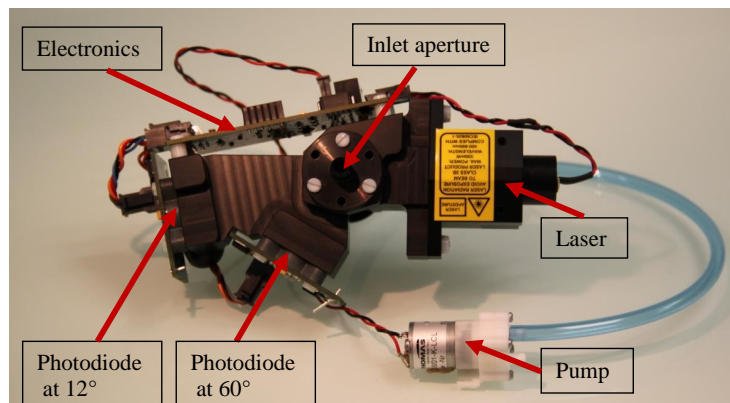
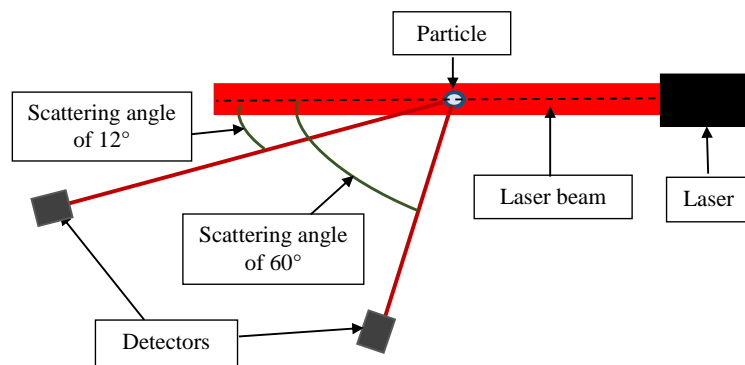


**Figure 1.** Scanning electron microscope image of ambient air aerosols (courtesy Jose Vanderlei Martins, Institute of Physics of the University of Sao Paulo, Brazil).

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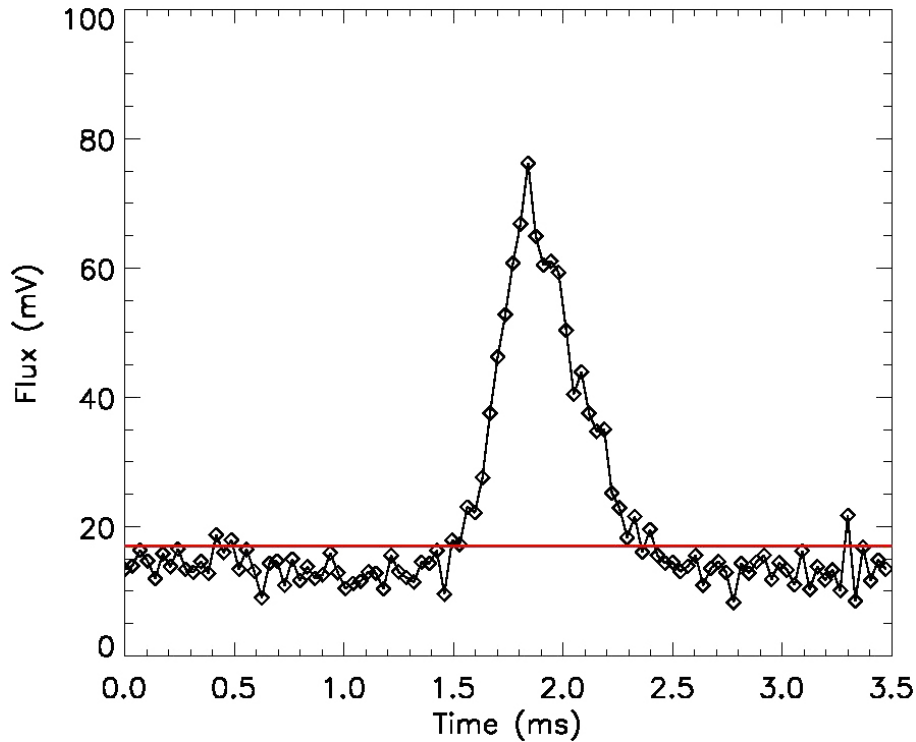
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**Figure 2.** The LOAC instrument; upper panel: principle of measurement; lower panel: picture of the instrument (the inlet tube is not presented here).

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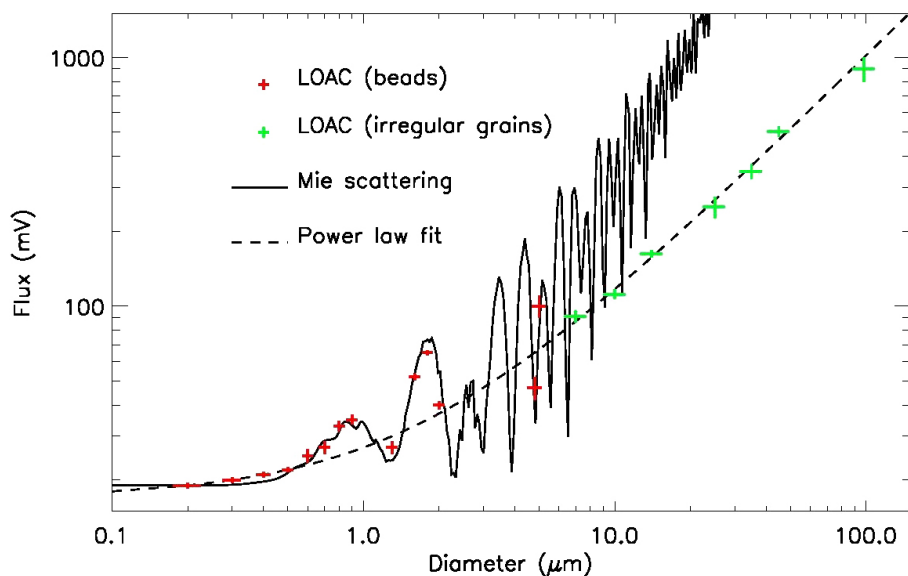
**Figure 3.** Example of the flux scattered by ambient air particles while crossing the laser beam. The red line corresponds to the threshold for the peak detection. When a particle is detected, the signal must return back below the threshold to allow the detection of the next one.

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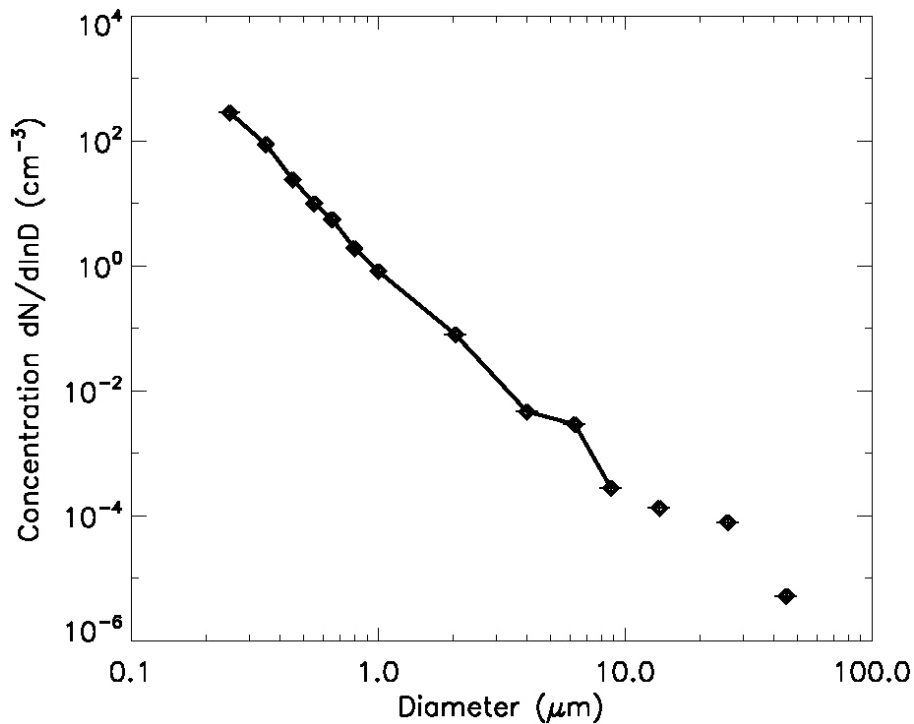
**Figure 4.** Calibration curve of the scattered flux at the 12° channel as a function of particle diameter. Beads were used in the 0.2–5.0 μm range; irregular grains selected by sifters were used for the largest size. The offset due to the electronic dark current and high frequency noise of the detector were added to the calculations; thus the curve asymptotically decreases with decreasing size to this offset value. The Mie calculations were conducted for the LOAC field of view. The difference between the Mie scattering calculations and LOAC measurements for diameters greater than 5 μm is due to the small aperture of the field of view coupled with the roughness of the particle shapes; the measurement curve is fitted by a power law (adding an offset of 17 mV).

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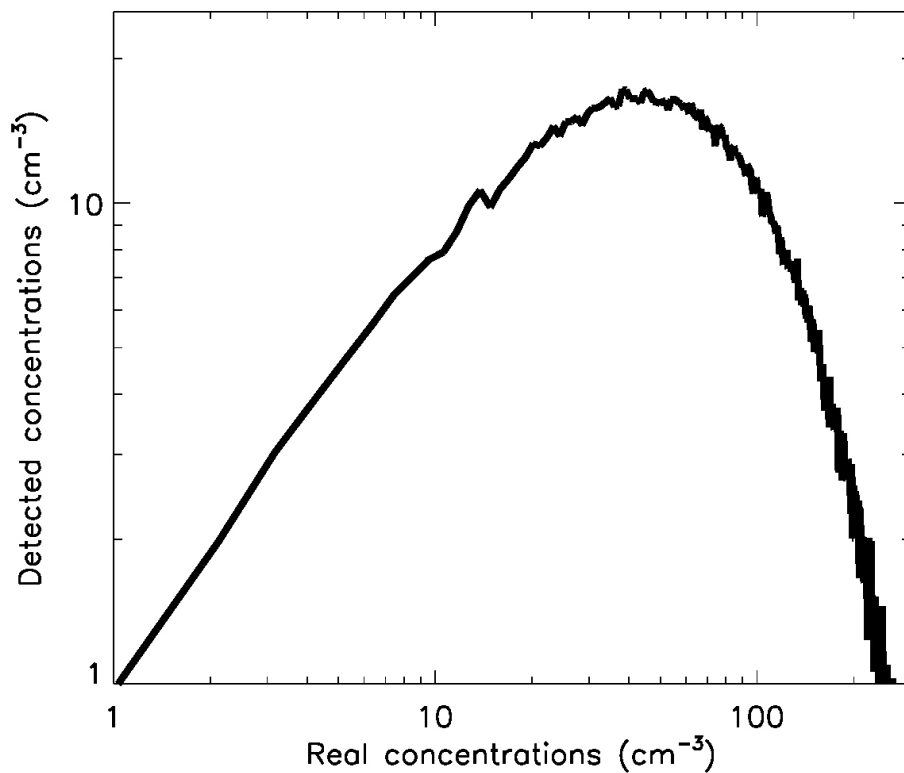
**Figure 5.** Typical size distribution in a suburban ambient air with carbon particles (Palaiseau, South of Paris) on 14 October 2013 during ParisFog campaign; the data are integrated during 15 min; the last points are not related because of zero concentration measured between them.

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**Figure 6.** Monte-Carlo modelling for the response of the counting system for particles larger than 1  $\mu\text{m}$ . The response is almost linear up to 10 particles per  $\text{cm}^3$ , and decreases for large concentrations.

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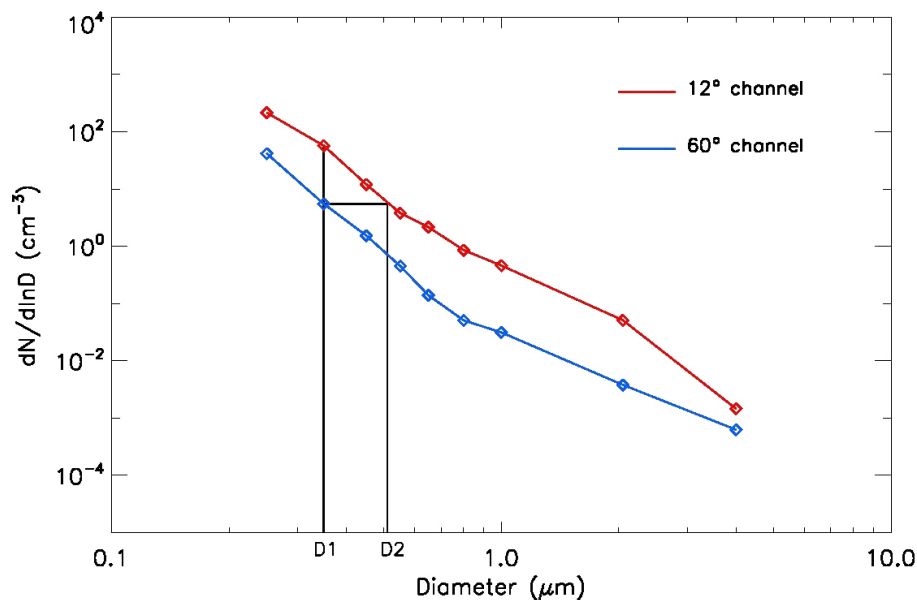
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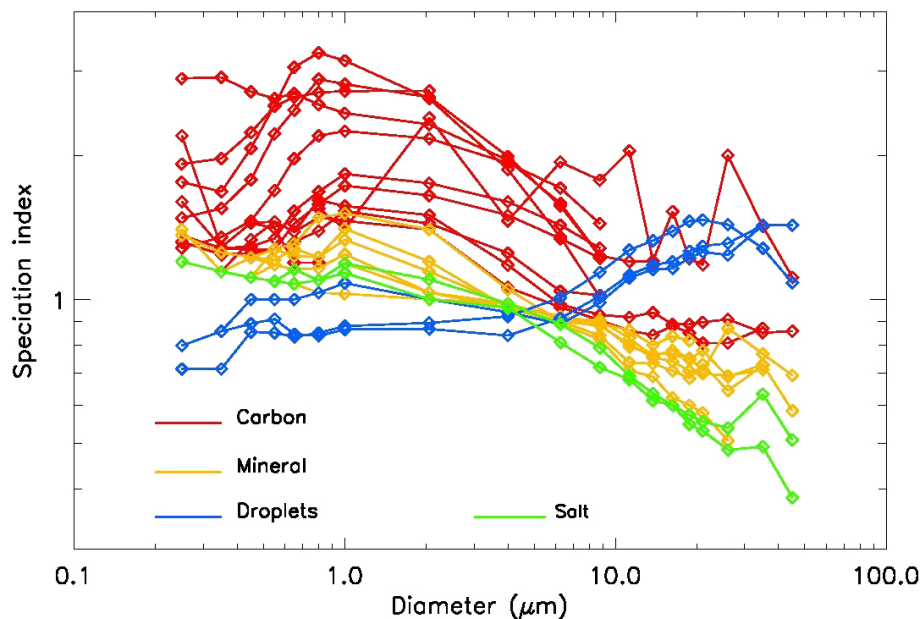


**Figure 7.** Principle of the determination of the “speciation index”  $D_2/D_1$  (the example presented here uses real measurements).



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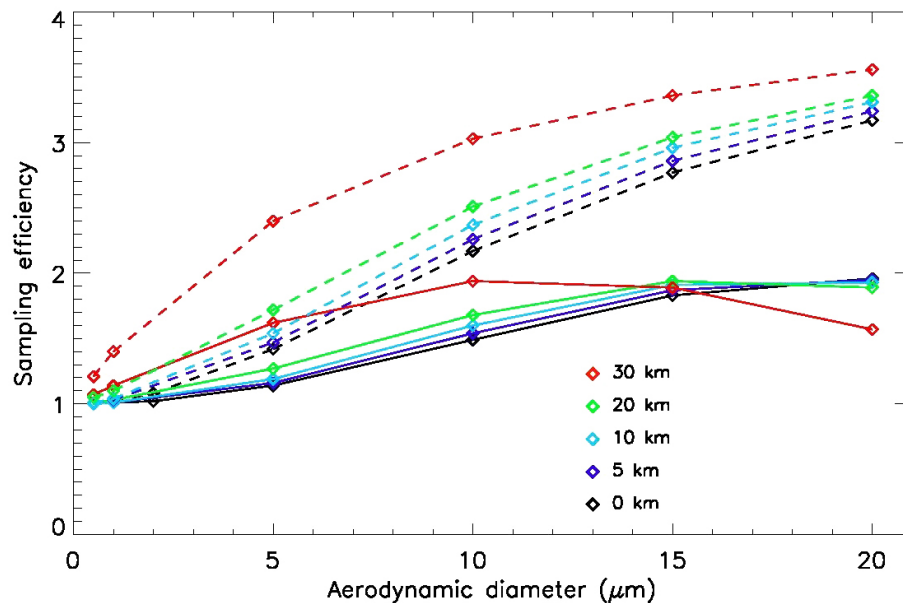


**Figure 8.** Evolution of the speciation index with diameter for various families of samples; measurements were conducted in laboratory with LOAC using pure samples.

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**Figure 9.** Efficiency of the sampling line at different altitudes from the surface up to 30 km; dashed lines: isoaxial conditions; full lines: 30° deviation from isoaxial conditions.

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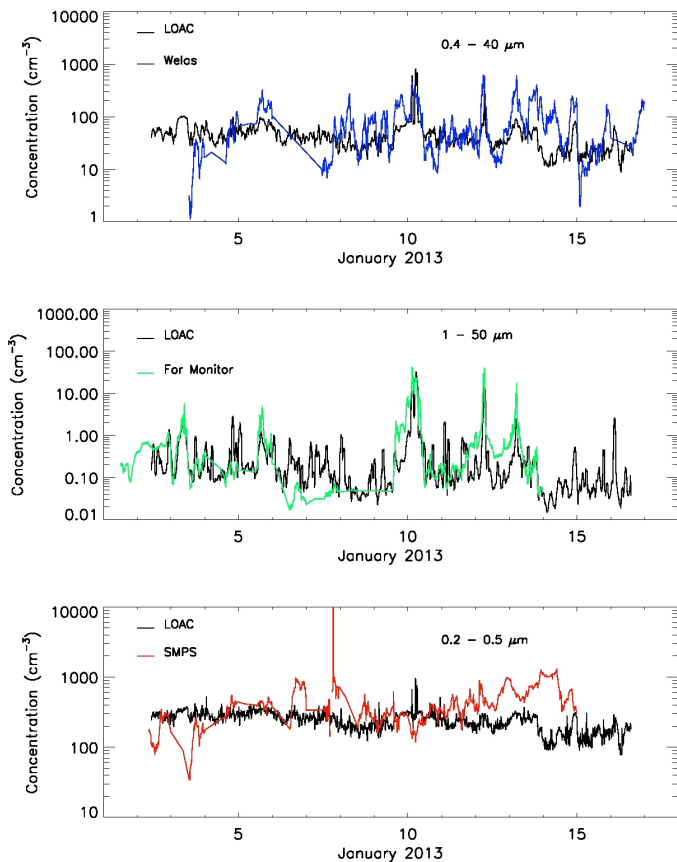
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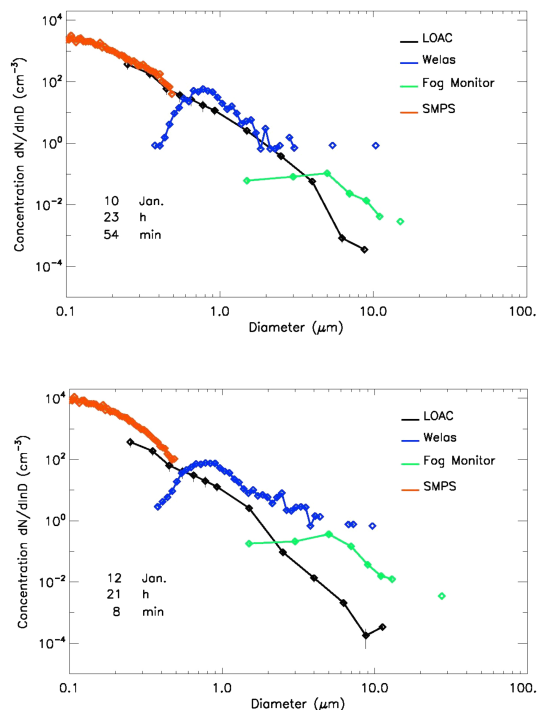
**Figure 10.** Cross-comparison of LOAC with 3 other instruments (WELAS, Fog Monitor and SMPS) for the total concentrations of aerosols in the size range domain in common, during the ParisFog campaign south of Paris. The LOAC uncertainties are of  $\pm 15\%$ .

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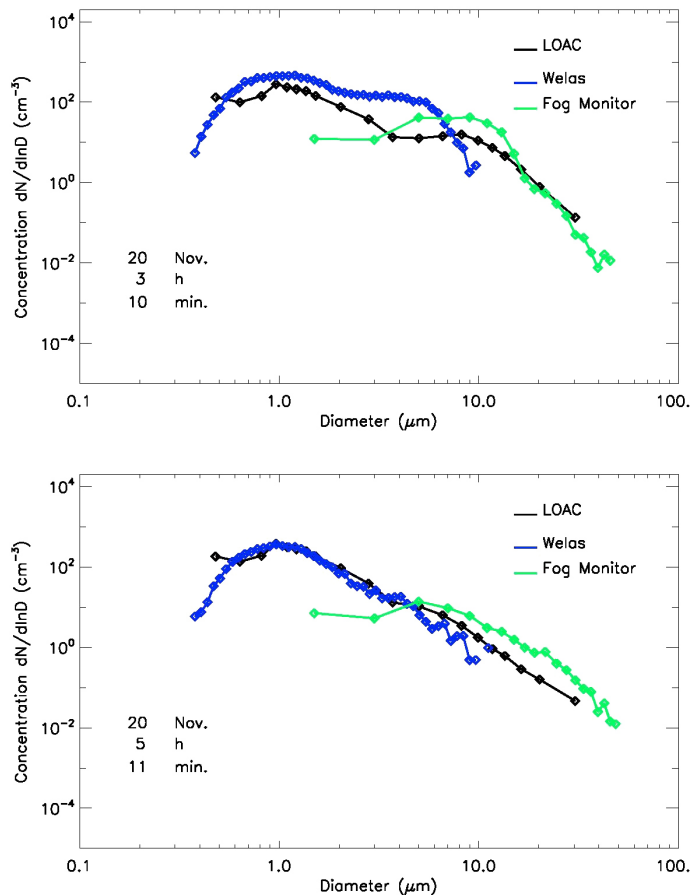


**Figure 11.** Cross-comparison of the 4 instruments during background conditions, in case of good agreement for the total concentrations measurements, during the ParisFog campaign. Upper panel: 10 January 2013, good agreement between the instruments; lower panel: 12 January 2013, poor agreement. The LOAC uncertainties are of  $\pm 15\%$ . The WELAS probably underestimates sub- $\mu\text{m}$  particles (Heim et al., 2008).



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**Figure 13.** Cross-comparison of the 3 instruments at the beginning of the fog event (top) and at the end (bottom), during the ParisFog campaign on 20 November 2012 during a fog event. The LOAC uncertainties are of  $\pm 15\%$ . The WELAS probably underestimates sub- $\mu\text{m}$  particles.

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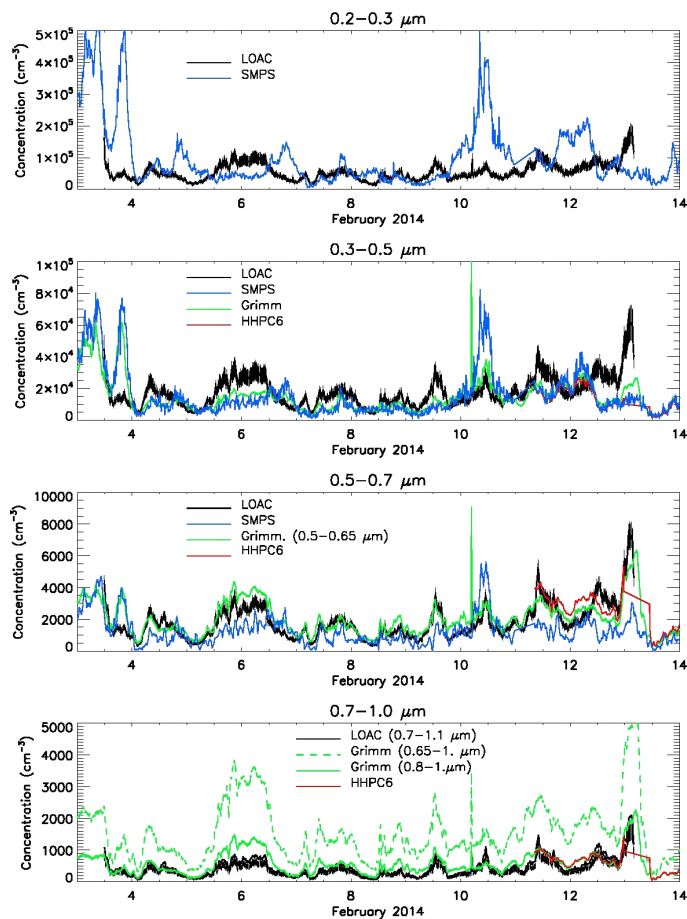






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**Figure 15.** Comparison (in linear scale) between the ambient air measurements obtained during the campaign at the SIRTA-5 station South of Paris.

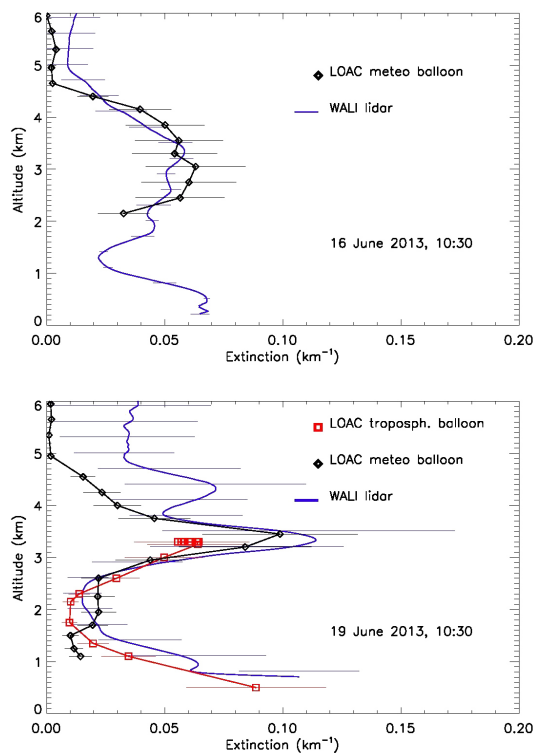
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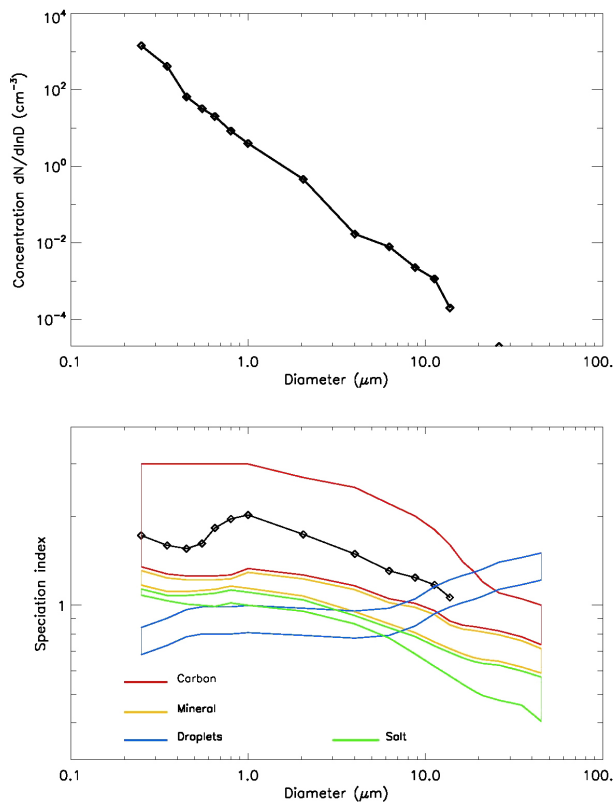



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**Figure 17.** Extinction profiles of the WALI lidar and extinction profiles calculated from LOAC measurements under meteorological and pressurized tropospheric balloons, from Minorca Island during the ChArMEx campaign.



**Figure 18.** Example of the detection of carbon particles in urban air, in South-West of Paris on 29 December 2013 around 07:30 UT, at the “Observatoire Atmosphérique Generali”; upper panel: size distribution; lower panel: topology.

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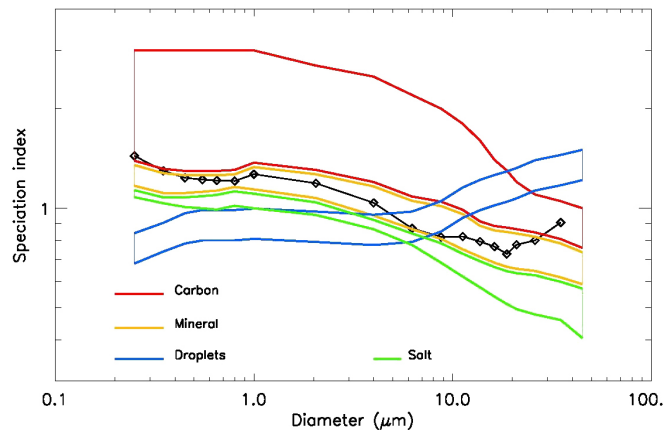
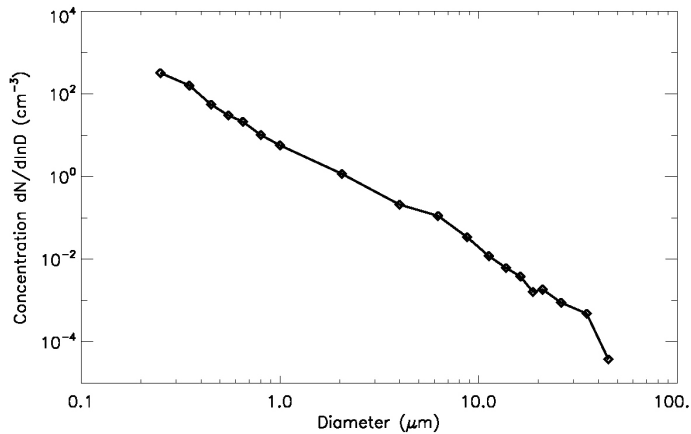
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**Figure 19.** Example of the detection of sand particles above Mediterranean Sea (close to Minorca) from a drifting pressurized tropospheric balloon on 17 June 2013 around 14:30 UT at an altitude of 2050 m, during the ChArME<sub>x</sub> campaign; upper panel: size distribution; lower panel: topology.

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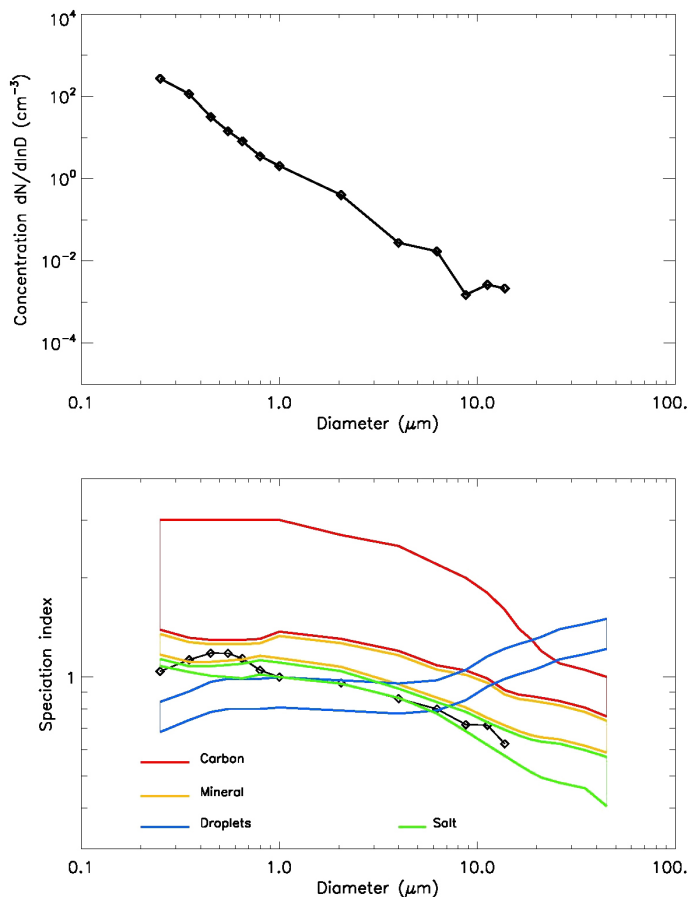
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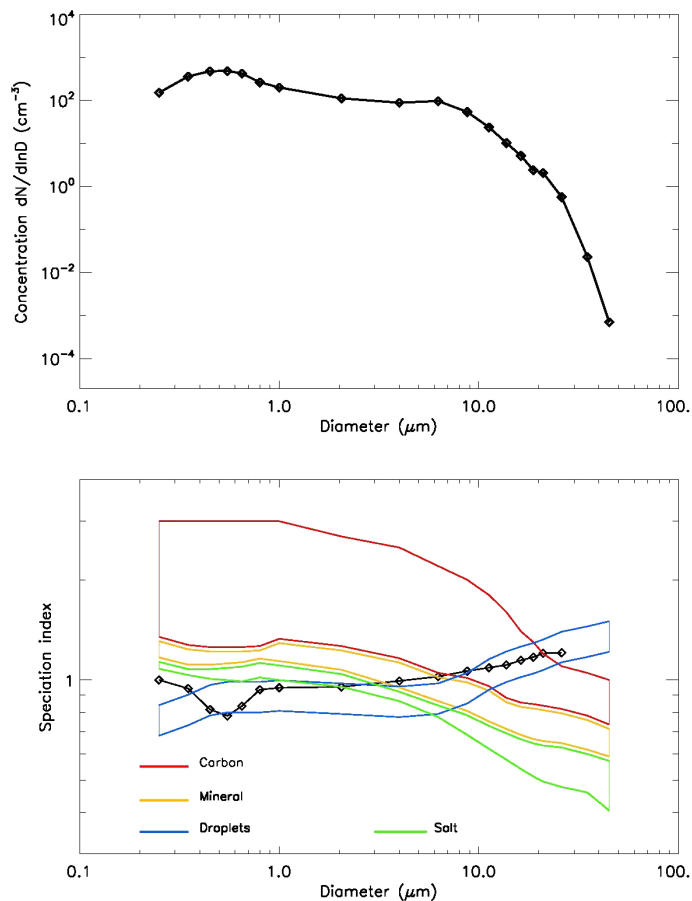
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**Figure 20.** Example of the detection of salt particles above Mediterranean Sea (close to Minorca, Spain) from balloon on 22 July 2013 at 21:25 UT at an altitude of 275 m during the ChArMEx campaign; upper panel: size distribution; lower panel: topology.

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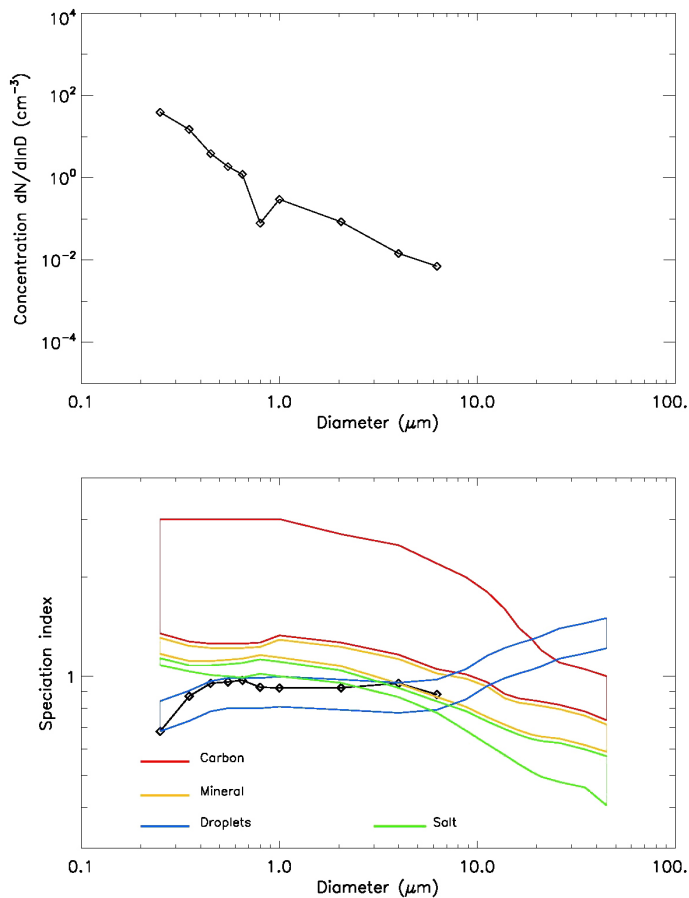


**Figure 21.** Example of measurements inside a cloud at Puy de Dôme observatory (France) on 15 May 2013 at 10:30 UT; upper panel: size distribution; lower panel: topology.



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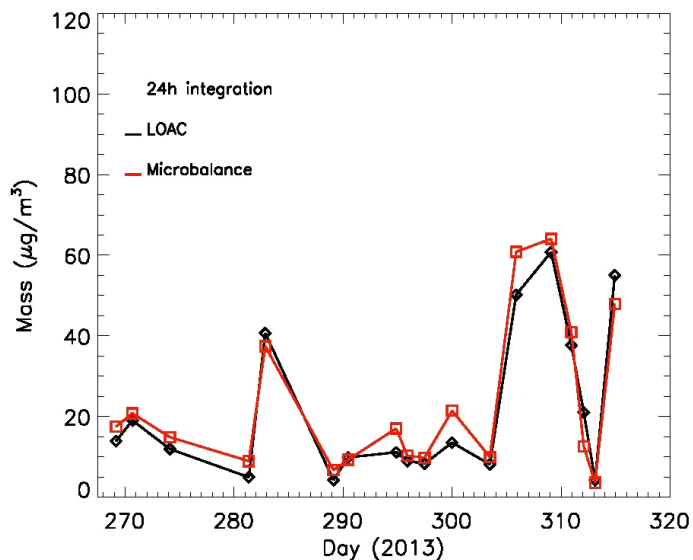
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**Figure 22.** Example of measurements inside a haze or thin cloud at an altitude of 1.2 km during a flight under meteorological balloon from Reykjavik (Iceland) on 7 November 2013 at 12:30 UT; upper panel: size distribution; lower panel: topology.

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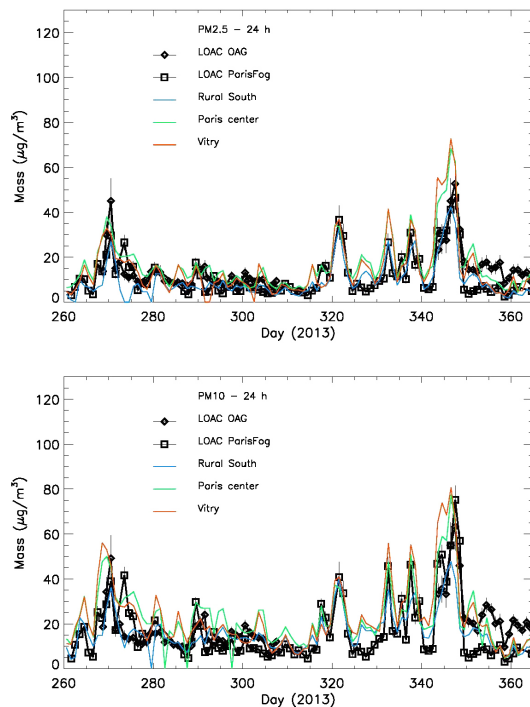
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**Figure 23.** Comparison of coincident LOAC and TEOM microbalance measurement in indoor air (averaged over 24 h); particles have been injected with various concentrations to document a large range of mass concentration.

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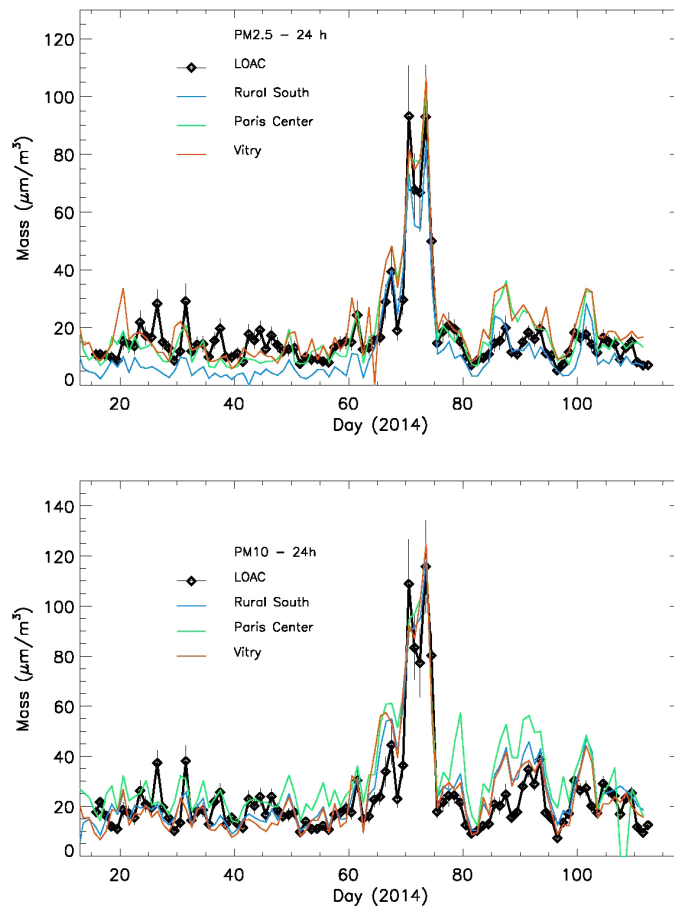


**Figure 24.** PM<sub>2.5</sub> (upper panel) and PM<sub>10</sub> (lower panel) LOAC mass concentrations measurements in 2013 during the ParisFog campaign at SIRTA Observatory in Palaiseau, South of Paris, and at the Observatoire Atmosphérique Generali(OAG) in the South-West of Paris, and comparison with reference TEOM data from the Airparif air quality monitoring network.

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**Figure 25.** PM<sub>2.5</sub> (upper panel) and PM<sub>10</sub> (lower panel) LOAC mass concentrations measurements in 2014 at the “Observatoire Atmosphérique Generali” (South-West of Paris) and comparison with reference TEOM data from the Airparif air quality monitoring network.