Evaluation of the miniature particle counter LOAC for the survey of stratospheric aerosols with meteorological balloons

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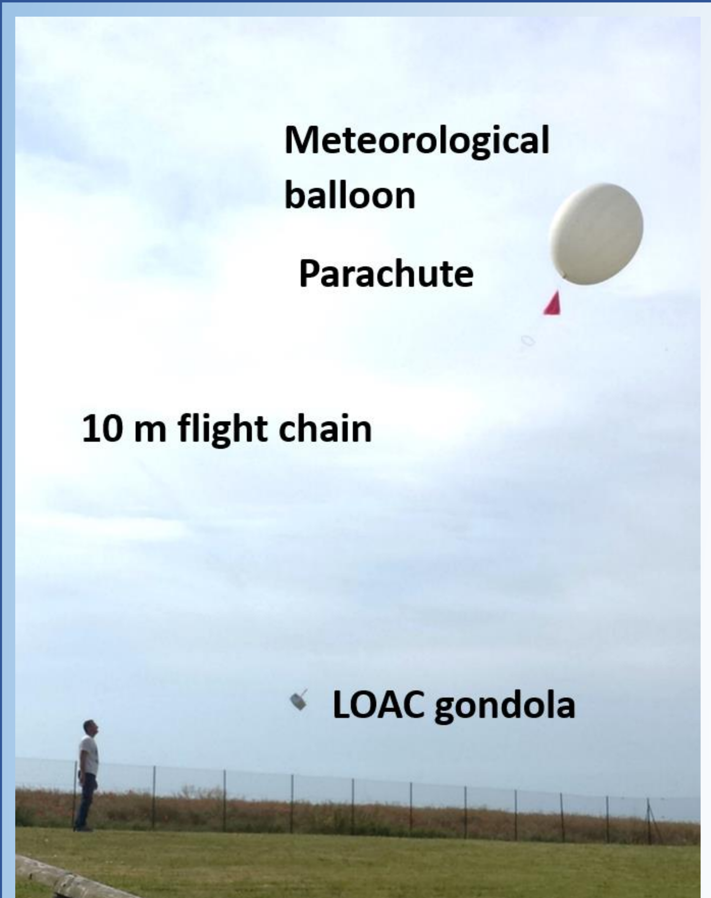
● **Summary:**

The study of the stratospheric aerosols is important to our understanding of the terrestrial radiative budget. Aerosols play also an important role on heterogeneous chemistry in stratosphere. Our current comprehension of the different types of stratospheric particles and their spatial and temporal distribution is incomplete.

In the present study, we try to show that measuring particle concentrations by the means of a new balloon-borne miniature particle counter, the LOAC, may allow us to determine the local variability in stratospheric aerosols in the size range 0.2-100 µm in diameter. In that respect, the PhD thesis sums up here consists of a first phase of a more accurate characterisation of the LOAC's performance under balloon-borne measurement. A second phase consists of comparative analysis of stratospheric aerosol content based on a LOAC dataset obtained during a continuous campaign of balloon launches in France, and along with some occasional flights abroad under particular circumstances (volcanic eruption (Iceland, Réunion Island), monsoon (India)). Thus we show that the LOAC has a detection limit that restricts the measurement of submicronic particles in volcanic Quiescent periods for concentration lower than 1 particles per cm³. Comparisons with satellites data (OSIRIS, OMPS, CALIOP), ground based lidar (LIO3s lidar OHP) and outputs from WACCM-CARMA model over the France reveal that LOAC data are more dispersed around other dataset until 25 km in altitude where the LOAC results seem converge to the detection limit.

Acknowledgements. CNES balloon launching team, ANR ("Programme Ecotech", ANR-2009-ECOT), Labex VOLTAIRE (ANR-10-LABX-100-01)

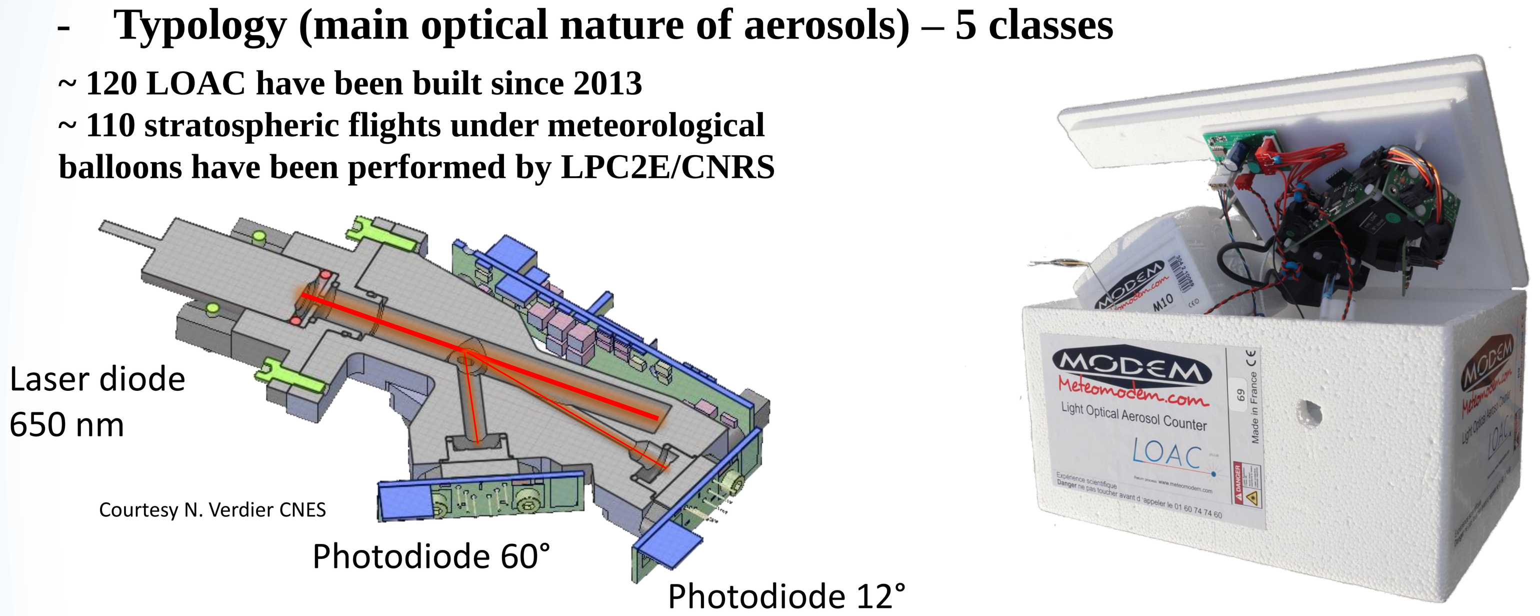
LOAC (Light Optical Aerosol Counter) – under meteorological balloon



Light miniature particle counter rose through the atmosphere clinging to meteorological balloon

- **Complete payload : 1 kg (LOAC + pump less than 300 g)**
- **3 W electrical consumption**
- **Concentrations over 19 size classes (0.2 – 100 µm Ø)**
- **Typology (main optical nature of aerosols) – 5 classes**

~ 120 LOAC have been built since 2013
~ 110 stratospheric flights under meteorological balloons have been performed by LPC2E/CNRS



Metrological performance during balloon flight

Internal temperature :

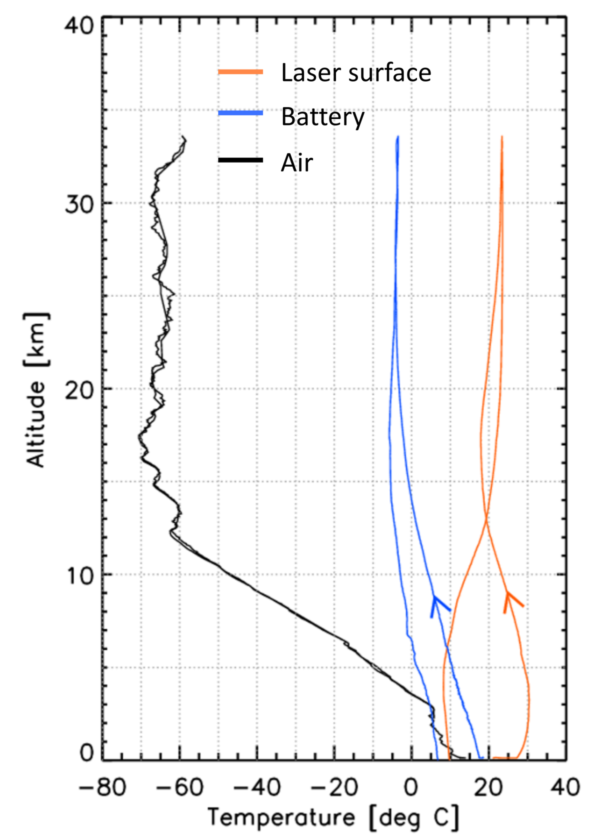


Fig 1 : Internal and external temperatures recorded in a LOAC gondola during a meteorological flight (Vignelles 2017)

In the LOAC's gondola during the ascent of the meteorological balloon, temperature is never below 0°C on batteries, and never below 15°C on the laser.

Temperature has an influence on :

- **Laser power : < 2 % in 5 → 35 °C range**
- **Volumetric flowrate : < 1 % at standard pressure and at 12 hPa**
- **LOAC's auto-calibration procedure : operating temperature between -10°C → 35°C**

Air pressure :

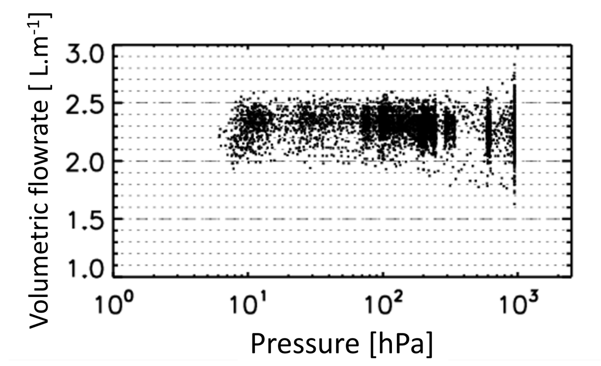


Fig 2 : LOAC pump volumetric flowrate as a function of pressure tested in a vacuum chamber at PIT Guyancourt France (Vignelles 2017)

The pressure (from 10 to 1000 hPa) has no visible impact on the volumetric flowrate of the LOAC pump (model Thomas "rotary vane pump" G01-K-LC). Tests realised at -40°C in a simulated chamber at PIT Guyancourt France.

Detection limit :

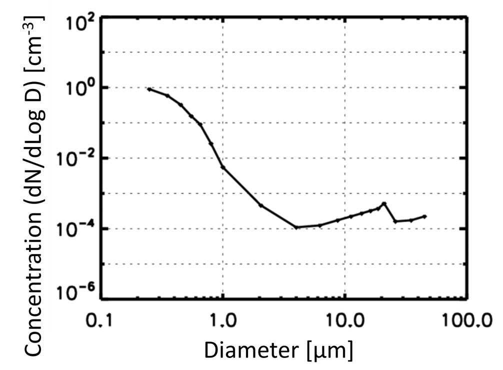


Fig 3 : Detection limit for the LOAC 19 size bins. For Ø < ~2 µm, the detection limit is due to statistical detection, for larger Ø the detection limit is due to volume sampled (Vignelles 2017)

In a particle-free atmosphere (less than 1 part.cm^{-3} of 5 nm in diameter on 10 minutes) the LOAC gives a residual concentration. From this residual concentration and its dispersion, a detection limit has been set as the mean residual concentration plus 1-σ. We consider that for a cumulative concentration on LOAC's 19 size bins of around 1 particle per cm³, the signal is only noise. Fig. 3 represents the detection limit for each size bin.

The residual extinction 532 nm is 4.10^{-5} km^{-1}, while the detection limit is 8.10^{-5} km^{-1} averaged on 10 minutes.

Inter-comparison in flight:

A first comparative test has been made with two LOAC under two balloons released 1 minute apart.
Balloons had the same trajectory and remain together within 2 kilometres horizontally.
Differences between the two air temperatures recorded on-board the gondolas are less than 4 %.

Fig 4 : Picture taken before the double flight. Each balloon is carrying a LOAC in the same configuration (Vignelles 2017)

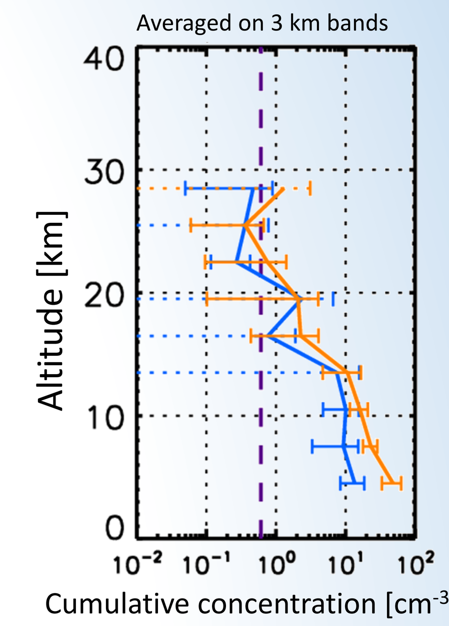


Fig 5 : Profile of the cumulative concentration over the LOAC's 19 size bins and averaged on 3 km bins for the two LOAC on two separate balloons. Error bar represents 1-σ dispersion on 3 km bins from 1 minute data. The vertical dotted line represents the detection limit (Vignelles 2017)

The compared LOAC flights indicates a reproducibility lower than the one established in the laboratory. We assumed that internal temperature variation during flight causes this lower reproducibility. Others comparative flights are under processing.

LOAC's performances appear to be limited by the low concentration limit and the low reproducibility in flight conditions. Low pressure and low temperature are without influence on flow rate and laser power.

94 meteorological balloons flight

In the period between June 2013 and August 2016, 94 profiles have been made with LOAC from several launching base in France (Fig. 6). With an averaged flight frequency of 2 flights per month, this innovative dataset represents an opportunity to study local variations of the stratospheric aerosol content.

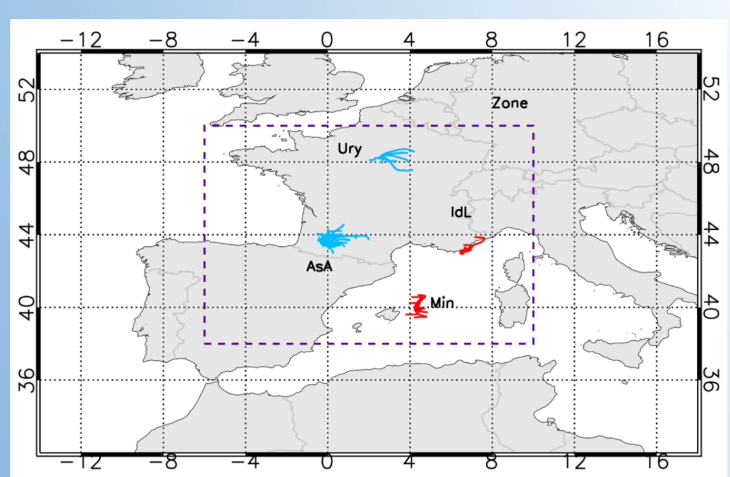


Fig 6 : Balloon launching sites for ChArMEx and Voltaire-LOAC field campaigns (Vignelles 2017)

The period between June 2013 to August 2016 over the France is a volcanic quiescent period. Mean and median per 3 km bins (Fig. 7 blue line and violet line) slowly decrease with altitude and get close to the detection limit above 30 km (Fig. 8 orange dotted line), expressed here in extinction calculated from the residual concentration (Fig. 3). A "Pinatubo Criterion" has been set in order to exclude extinctions larger than space-borne observations during the Pinatubo eruption (Fig. 7, green line). This "Pinatubo criterion" allows us to determine a new profile (Fig 7. blue dotted line).

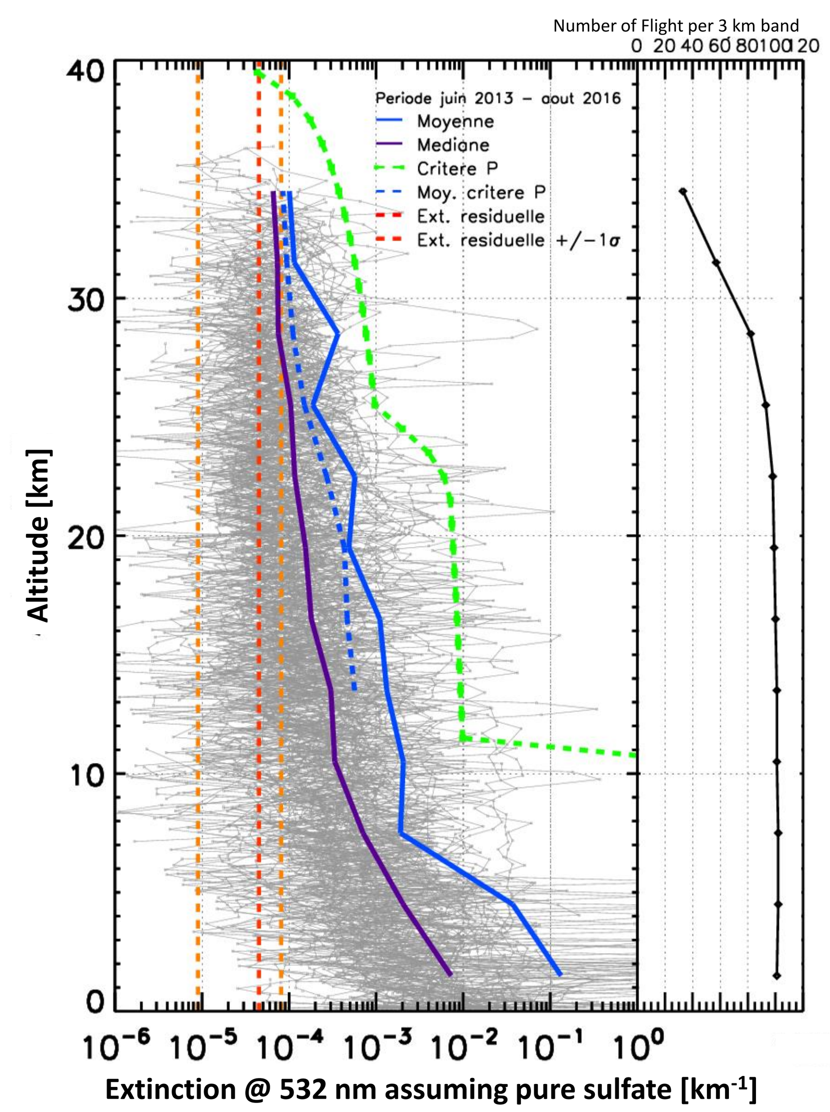


Fig. 7 : 94 LOAC profiles from France. Grey lines are individual LOAC profiles at 1 minute time resolution. Blue and violet lines are means and median on 3 km bands. Green dotted line is the Pinatubo Criterion (extinction profil during the last great volcanic eruption) (Vignelles 2017)

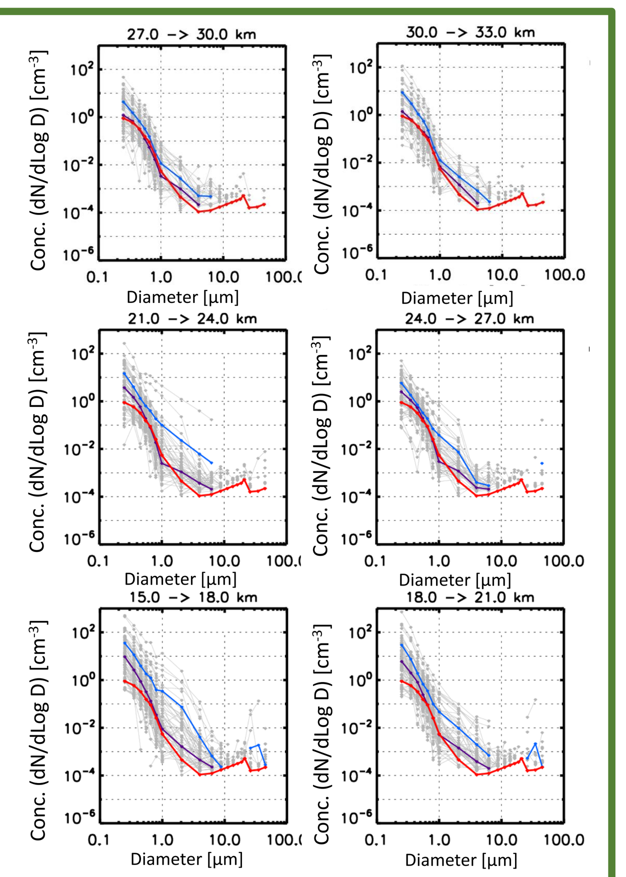


Fig 8 : Size distribution on 3 km bands. Blue and violet lines are mean and median for each size bins. Red lines are detection limit established in lab under contained conditions (Vignelles 2017)

Size distribution of concentration is given in Fig. 8 for 3 km bin. Averaged and median concentration per size bin are compared to the detection limit. Shapes of size distributions are representative of background conditions. Above 24 km, median distribution per size bin (Fig. 8 violet line) are quite close to the detection limit (Fig. 8 red line).

Comparative results with others datasets

Inter-comparisons between LOAC dataset on the period 2013-2015 have been made with measurements from OSIRIS, OMPS, CALIOP and the LIO3s ground lidar at the OHP (*Observatoire de Haute Provence*), and with outputs from the WACCM-CARMA model. The comparative results consider averaged satellite data and model outputs over France (Fig. 6 : dotted violet square / [38;50]°N & [-6;10]°E).

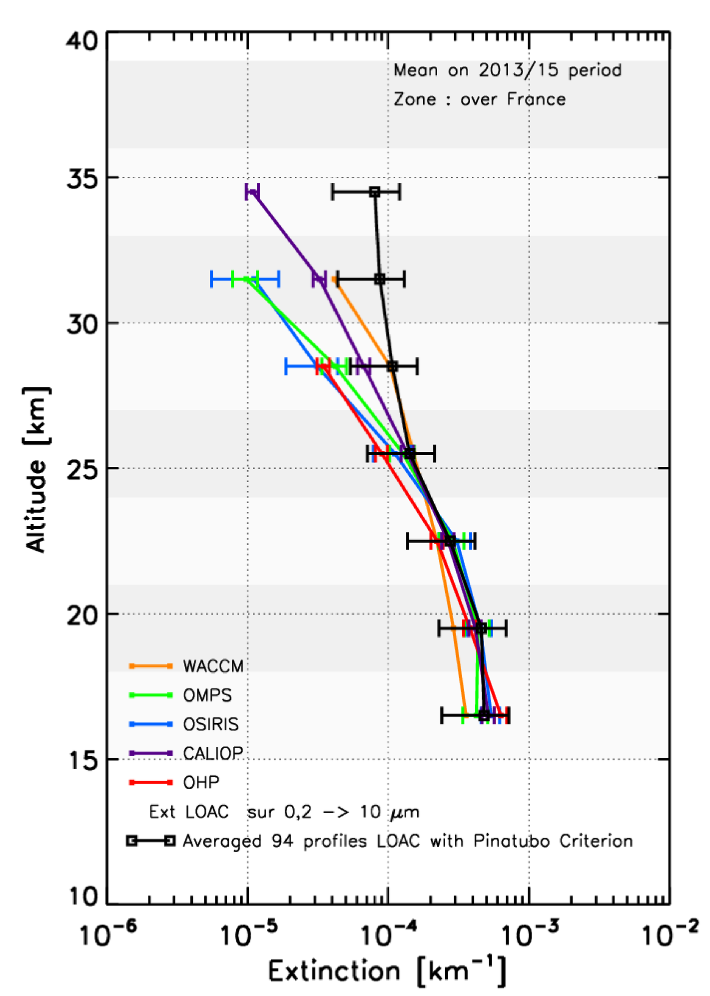


Fig. 9 : cross comparison between 3 years mean extinction 532 nm over France with LOAC, ground OHP lidar, OMPS, OSIRIS, CALIOP and outputs from WACCM-CARMA model (Vignelles 2017)

In the lower stratosphere (15-24 km) datasets are consistent with each other. Above 27 km, OMPS, OSIRIS and OHP lidar give lower extinctions than CALIOP, LOAC and WACCM. LOAC gives higher extinctions in middle stratosphere ; a combination of two reasons can be suggested in order to explain this phenomenon. Firstly the LOAC's detection limit is not removed here (Fig. 9) and closed to 8.10^{-5} km^{-1} which is approximately the mean extinction found at 35 km. Removing the residual extinction improves the comparison but it assumes that we can extrapolate residual extinction determined in lab to flight conditions. Secondly, retrievals from satellites and ground-based lidars are based on various assumptions: pure sulphate aerosols, invariable size distributions in order to derive extinction, and no extinction from aerosols above certain levels.

Higher than 25 km LOAC data averaged over a 3 year period need further investigations.

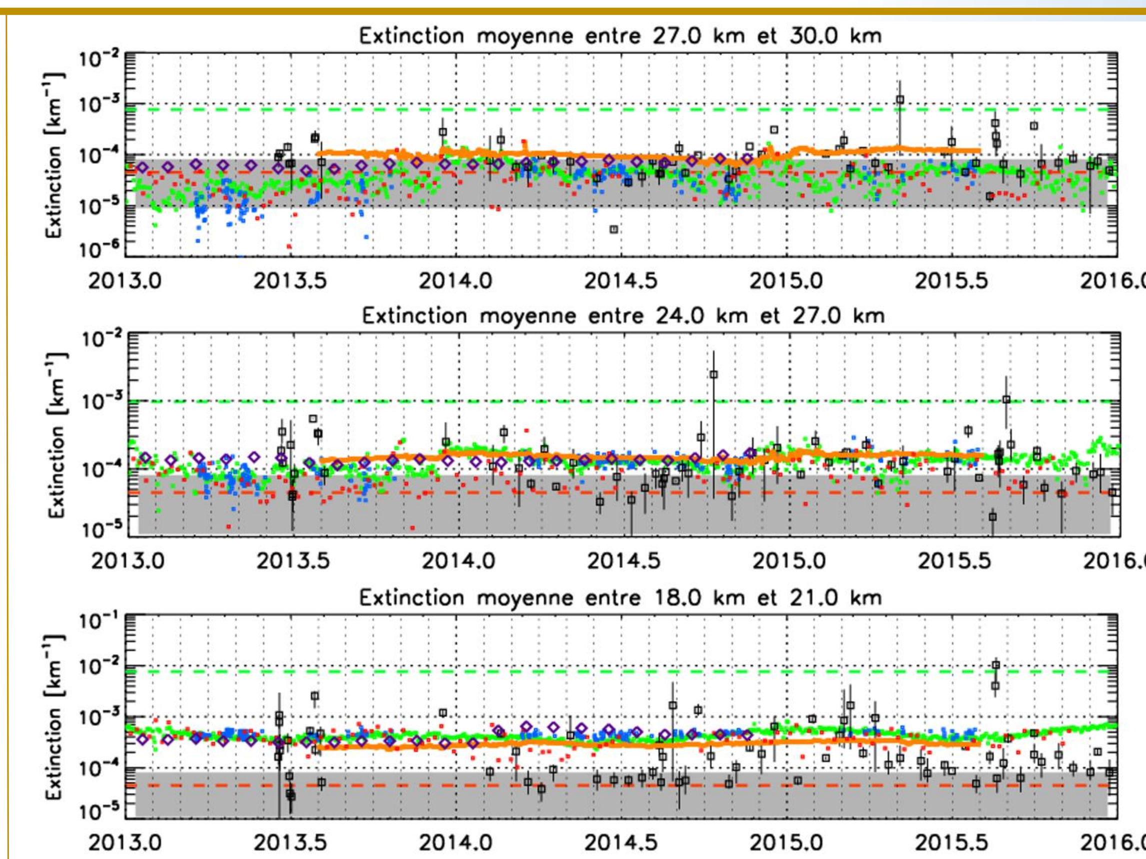


Fig 10 : Variability of extinctions over France for WACCM, OMPS, OSIRIS, CALIOP, OHP and LOAC for the period 2013-2015. Green dotted line represents Pinatubo criterion, orange dotted line represents the LOAC residual extinction and grey square represents ± 1σ around residual extinction (Vignelles 2017)

Temporal evolutions of extinction per 3 km bins (Fig. 10) show that local measurements (lidar OHP and LOAC) are more scattered. A seasonal variability related to positions of the polar vortex is clearly visible in OMPS and WACCM datasets which show the influence of stratospheric dynamics on the aerosol distribution. The OHP lidar and LOAC show different patterns of variability, keeping in mind that these specific data represent a very local view compared to satellite results which integrate large areas.

In conclusion, LOAC dataset is consistent with satellite/ground-lidar/model datasets in lower stratosphere and for a large number of flights. In middle stratosphere, instrumental limitations seem to restrict the use of the LOAC in volcanic quiescent periods. Individual LOAC flights reveal an important dispersion of extinction in time and in altitude that satellite and model data don't reveal. Representativeness of individual LOAC flight compared to the satellite one is a key problem of this kind of inter-comparisons. We point out that lidar OHP representativeness and individual LOAC flight representativeness is closer but this two local measurements do not present same time evolution with each other or with satellite and model. The comparative flight conducted with two LOAC reveals a low reproducibility in flight in the same air mass. This reproducibility lower than the one determined in laboratory must be investigated in order further discuss this results. New versions more accurate and lighter of LOAC are in development.

Renard et al. (2016) *LOAC: a small aerosol optical sounter/sizer for ground-based and balloon measurements* AMT. 9, 1721-1742. doi:10.5194/amt-9-1724-2016 / Vignelles et al. (2016) *Balloon-borne measurement of the aerosol size distribution from an Icelandic flood basalt eruption.* EPSL. 453, 252-259. doi:10.1016/j.epsl.2016308.027 / Vignelles (2017) *Caractérisation des performances du nouveau mini compteur de particule LOAC embarqué sous ballon météorologique : application à l'étude de la variabilité spatiale et temporelle des aérosols de la haute troposphère et de la stratosphère* – PhD thesis