

We thank Referee #1 for her/his comments and suggestions that helped to improve the manuscript. Our response is formatted as follows:

Reviewer's comments

Author's reply

[Changes to the manuscript](#)

All page, line, section and figure numbers in bold refer to the original manuscript, all others to the revised version.

This manuscript presents field results from airborne measurements made over the Amazon in 2014 as part of the ACRIDICON-CHUVA project. It illustrates the potential of using aerosol mass spectrometry instruments to identify ambient SOA formed from IEPOX. Airborne in-situ measurements of aerosol composition and physical properties were used to illustrate the presence of IEPOX-SOA at altitudes > 5 km. Several different approaches are used to quantify the presence of this IEPOX-SOA and subsequently for organic nitrate providing a robust analysis. These observations are original, showing the relationship between organic nitrates and IEPOX-SOA.

This manuscript is very well written, all figures and tables are clear and easily interpreted. This paper is of interest to the ACP audience and is suitable for publication. I have a small number of comments below that can be considered or discussed prior to publication.

We thank the reviewer for this positive rating of our manuscript.

- 1. Although details of the different flights are provided in other papers (Andreae et al., 2018) some flight details would be appreciated in the supplementary of this manuscript. This study includes measurements from 13 different flights, how did the meteorological conditions change during each of these flights. According to the overview paper by Andreae et al., 2018, there is some variability linked to air mass source and wind conditions. Can the vertical profiles be classified into different groups depending on meteorological sources?**

The meteorological situation was quite similar for all campaign days and dominated by convection. We added an overview table to the supplement giving information on the different flights regarding the duration, meteorological situation, and flight strategy. Air mass sources were analyzed with the model FLEXPART and addressed in the reply to question 2 of the reviewer. The following was added to **Sect. 4.1, p. 11, line 8** (p. 12, line 16):

[The meteorological situation during the ACRIDICON-CHUVA campaign was quite similar for all days. Convection was dominating the daily weather and affecting every flight. The invariance of the meteorological situation is also visible in the temperature profile \(see Fig. 2, Panel \(a\)\) that barely shows any deviation. An overview of some flight details is provided in Tab. S1.](#)

We included the following to the Supplement:

Tab. S1: Overview of all flights with date, duration, maximum altitude that was reached and the meteorological situation. Furthermore, information on the flight strategy and comments to the C-ToF-AMS measurements.

FLIGHT NO.	DATE IN 2014	DURATION	ALTITUDE [KM]	METEOROLOGICAL CHARACTERISTICS	FLIGHT STRATEGY AND COMMENTS
AC07	06.09.	7 h 35 min	13.9	Convection	Cloud profiling; no zero filter measurements
AC08	09.09.	5 h 30 min	13.8	Convection	Cloud profiling
AC09	11.09.	6 h 10 min	12.6	Convection	Cloud profiling; no zero filter measurements
AC10	12.09.	7 h 25 min	14.4	Convection, cirrus	Cirrus sampling; CVI measurements only
AC11	16.09.	7 h 25 min	12.9	Convection	In- & outflow measurements, cloud profiling
AC12	18.09.	6 h 15 min	13.8	Convection	Polluted cloud profiling
AC13	19.09.	6 h 30 min	12.9	Convection	Polluted cloud profiling
AC14	21.09.	7 h 15 min	15.2	Convection	No filter measurements
AC15	23.09.	7 h 20 min	13.8	Convection	Outflow sampling, cloud profiling
AC16	25.09.	6 h 50 min	13.2	Convection	In- & outflow measurements
AC17	27.09.	6 h 40 min	8.1	Convection	Cloud contrast measurements: clouds above forested and deforested areas
AC18	28.09.	6 h 50 min	14.4	Convection	Clean cloud profiling
AC19	30.09.	7 h 15 min	13.8	Convection, pyro-Cumulus	Marine and biomass burning influence
AC20	01.10.	7 h 05 min	14.4	Convection	Cloud profiling

2. In section 4.3, the authors state that the sources of the organic aerosol in the LT and the UT are not the same, providing air mass trajectories along the flight track would help support these conclusions.

We agree that air mass trajectories along the flight track help to support our conclusions. We analyzed FLEXPART trajectories, calculated along the flight track starting every minute and calculated backwards for 10 days providing hourly information on the location of each trajectory. Fig. S4 shows the release altitude of the trajectories against the residence time. The residence time gives the time that the trajectories spend in the boundary layer (BL, red curve) or in the upper troposphere (UT, black curve). There is only little interaction and almost no overlap between both curves. This leads to the conclusion that convection cannot be resolved with the FLEXPART model.

Fig. S5 shows maps with trajectories that are released below 4 km (lower troposphere, LT, Panel **(a)**) and above 8 km (upper troposphere, UT, Panel **(b)**). The origins of the trajectories differ overall. The trajectories released below 4 km have their origin also in the LT and show almost no interaction with higher air masses. Most of the trajectories come from the Atlantic Ocean and the southern part of South America. In contrast to this, the trajectories released above 8 km have their origin mainly above the Pacific Ocean and circulate at high altitudes above South America. Just a minor part of the trajectories origins from the eastern direction, coming from the Atlantic Ocean and/or Africa. Interactions with air masses at lower altitudes are rare, most prominent is the lifting at the Andes mountains.

We included the following to **Sect. 4.3, p. 15, line 12** (p. 16, line 12):

Air mass trajectories were calculated using the FLEXPART model. The trajectories are calculated along the flight tracks starting every minute and calculated backwards for 10 days providing hourly information on the location of each trajectory. The FLEXPART model is not able to resolve convective transport (see Fig. S4) for the ACRIDICON-CHUVA campaign. Nevertheless, the origin of the trajectories that are released in the LT (< 4 km) differs from the origin of the trajectories released in the UT (> 8 km) (see Fig. S5). The trajectories released in the LT have their origin also in the LT and show almost no interaction with higher air masses. Most of the trajectories come from the Atlantic Ocean and the southern part of South America. In contrast to this, the trajectories released above 8 km have their origin mainly above the Pacific Ocean and circulate at high altitudes above South America. Just a minor part of the trajectories origins from the eastern direction, coming from the Atlantic Ocean and/or Africa. Interactions with air masses at lower altitudes are rare, most prominent is the lifting at the Andes mountains.

We included the following to the Supplement:

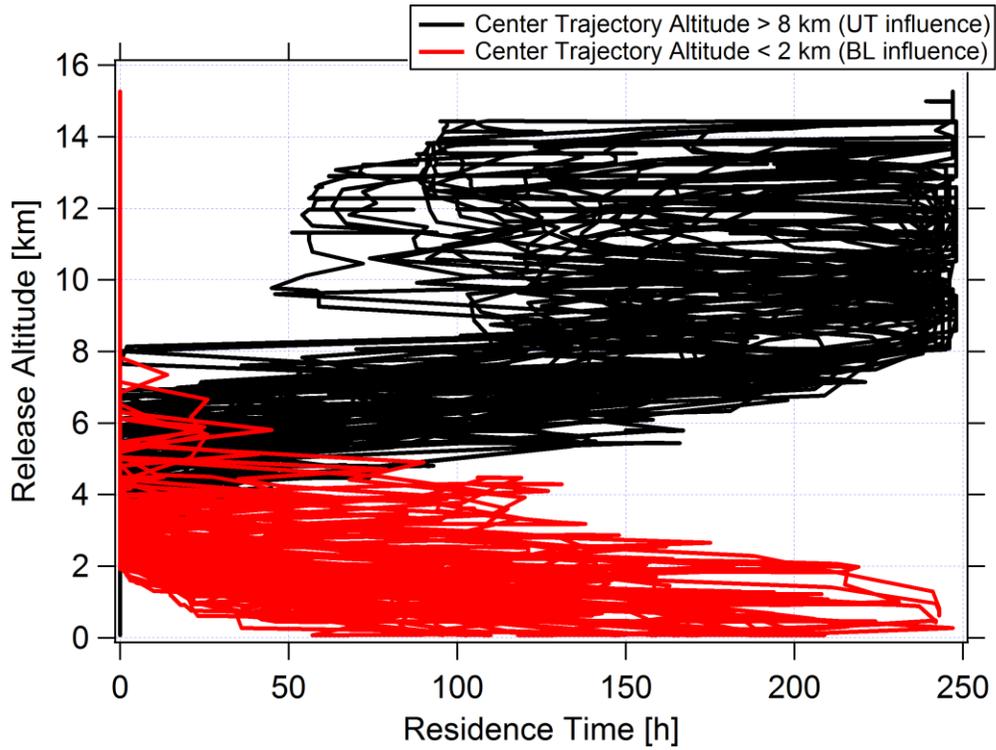


Figure S4. Release altitude of the trajectories versus the residence time, i.e. the time that the trajectories spend in the boundary layer (BL, red curve) and in the upper troposphere (UT, black curve). There is only little interaction and almost no overlap between both curves. This leads to the conclusion that convection cannot be resolved with the FLEXPART model.

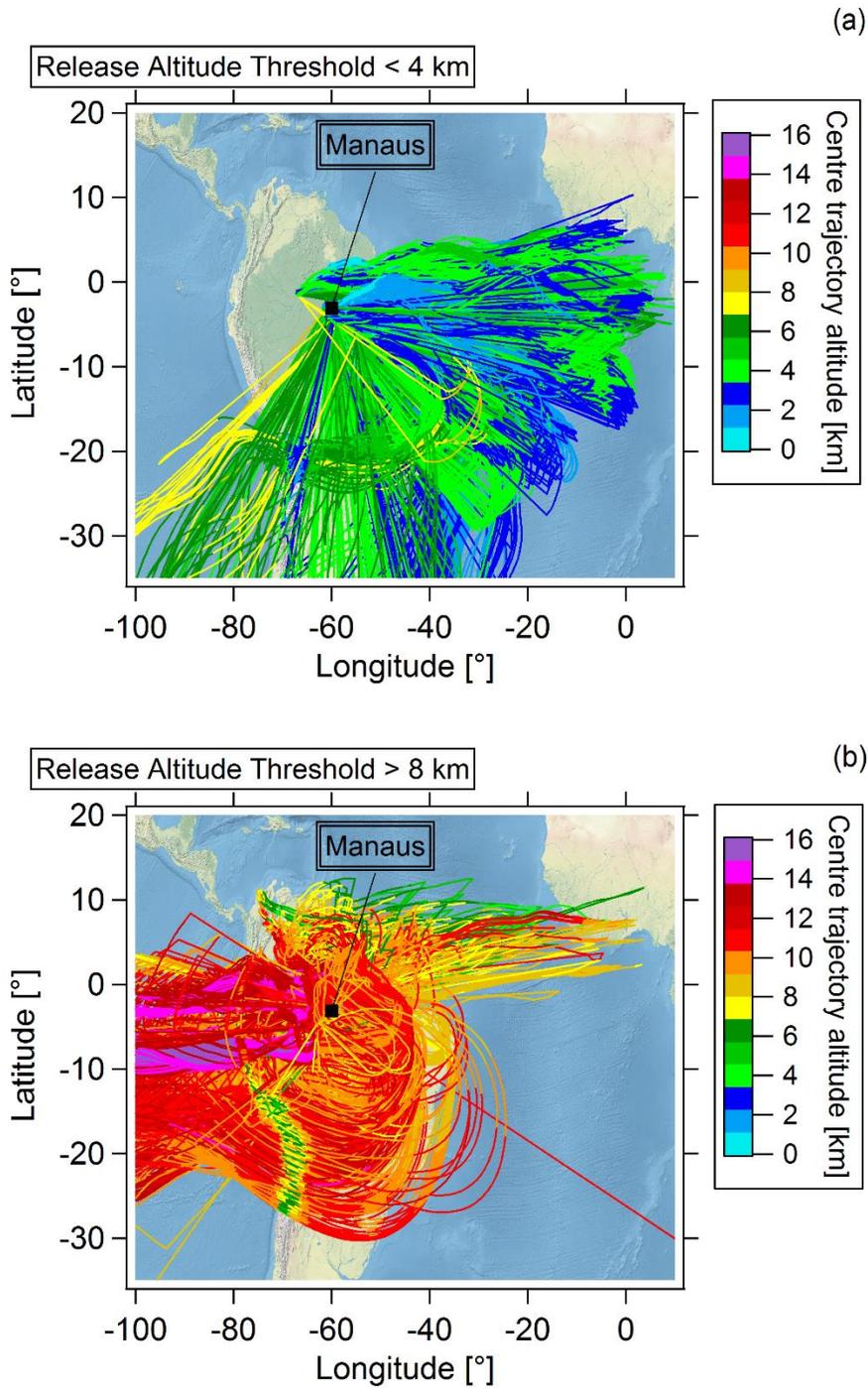


Figure S5. Maps with trajectories that are released below 4 km (lower troposphere, LT, Panel (a)) and above 8 km (upper troposphere, UT, Panel (b)). The colour code refers to the altitude of the centre trajectories.

3. The authors mention that there are 4 CPC instruments operating during these flights, with cut of diameters of 4 and 10 nm. Were two of the CPCs at 4 nm and the other two at 10 nm. In the manuscript Andreae et al. (2018) it was mentioned that the second set of CPC instruments were coupled with a DMA set up for particle size distribution measurements. Is this also the case for these flights. According to the accompanying papers, other aerosol physical parameters should be available from the UHSAS for larger diameters. These size distribution measurements may help to support the conclusions on the SOA particle growth (Page 15, Line 13).

We agree that these data would help to support our conclusions. The data were not finally processed during the manuscript preparation and therefore not used in the ACPD version of the manuscript. In the meantime, the validity of the UHSAS-A data was improved, so the UHSAS-A data are included now in the revised manuscript as follows:

Sect. 2.1.2, p. 5, line 30 (p. 5, line 31):

For particles in the size range between 90 and 600 nm data from an ultra-high sensitivity aerosol spectrometer (UHSAS-A) that was installed as an underwing probe were analyzed. The measurement system is based on the detection of scattered light from laser illuminated aerosol particles. For the ACRIDICON-CHUVA flights used here (AC07-AC10 and AC15-AC20), the mentioned size range was divided into 66 logarithmic size bins. Data for the other four flights (AC11-AC14) are recorded in a different size binning and not used here. Cloud passages and intervals with sample flow deviations were removed. The UHSAS-A was calibrated using spherical polystyrene latex particles.

Sect. 4.3, p. 15, line 15 (p. 17, line 3):

Another indication supporting this is the size information of aerosol particles with diameters between 90 and 600 nm. Figure 6 shows the vertical profile of the median and the mode of the binned size distributions measured with the UHSAS-A (Panel **(a)**). It should be noted here, that the lowest cutoff of the considered size range of the UHSAS-A is at 90 nm. Accordingly, the displayed mode diameters are confined by this lower limit. Also, the displayed size distribution medians are affected by the size range limits and should only be interpreted in this context. Whereas in the LT the median and the mode are at diameters around 150 nm (median) and 130 nm (mode), respectively, both the median and the mode are shifted towards smaller diameters with increasing altitude. The lowest value of the median is reached at altitudes above 4 km and (apparently) remains constant. The colour code in the vertical profile refers to the size distributions for the three different altitude regions in Panel **(b)** of Fig 6. Shown are the median and interquartile range of the size distributions. The size distribution in the LT shows a maximum at 130 nm. The size distributions in the MT and UT are shifted towards smaller diameters, and it is clearly seen that the highest concentrations of small particles (around 90 nm) are found in the UT.

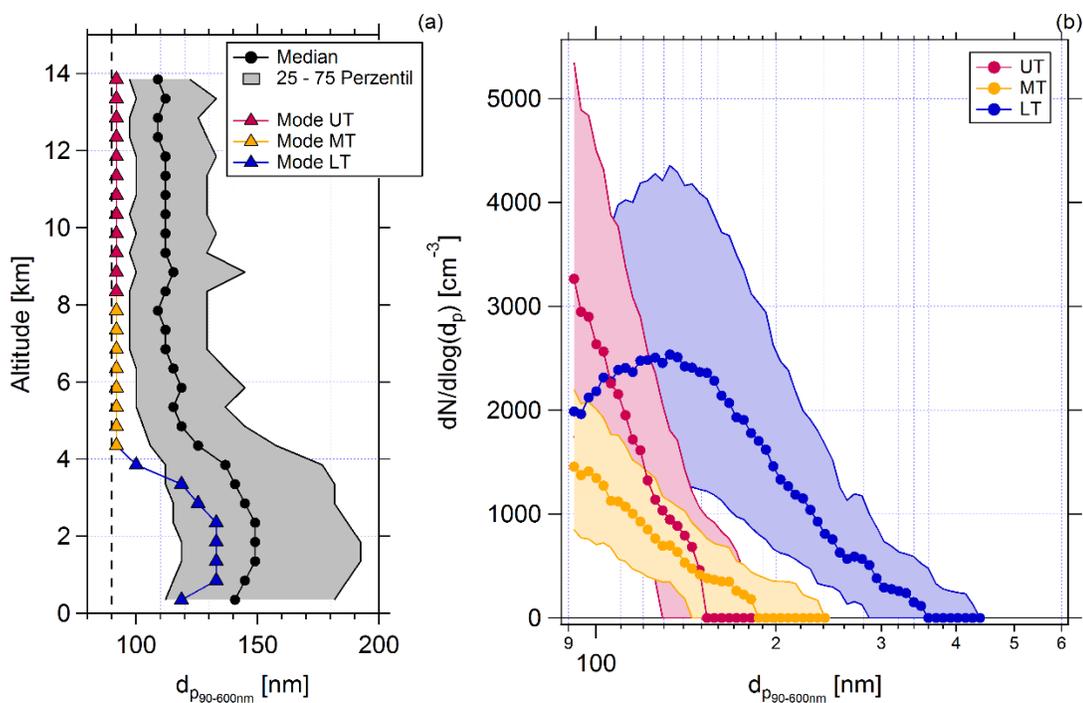


Figure 6. Vertical profile of the median (black dots) and the mode (triangles) of the binned size distributions (Panel (a)), and median and interquartile size distributions of particles between 90 and 600 nm in the UT (pink), in the MT (yellow) and in the LT (blue) (Panel (b)). The grey area in Panel (a) gives the interquartile range. The dotted line in Panel (a) indicates the lower cutoff of the considered size range of the UHSAS-A. The statistics shown in both Panels (a) and (b) are calculated from all valid UHSAS-A data from 10 flights (AC07-AC10, AC15-AC20). Data are calculated for STP conditions.

4. Page 14, Line 6: It should be mentioned here that there is significant variability of the m/z 44 (or f_{44}) among different AMS instruments and care should be taken when comparing results from different instruments (Frohlich et al., 2015, Pieber et al., 2016, Crenn et al., 2016).

We agree to that and included the following to Sect. 4.3, p. 14, line 13 (p. 15, line 25):

It should be mentioned here that there is a significant variability of m/z 44 (and f_{44}) among different AMS instruments such that no quantitative comparison can be done among the different data sets shown in Fig. 4 (Fröhlich et al., 2015, Crenn et al., 2015, Pieber et al., 2016).

5. The authors detail several different methods to provide a robust characterization of the presence of organic nitrate and IEPOX SOA during these flights. It could be stated why PMF analysis was not used to try identify the presence of these aerosols. If IEPOXSOA is contributing up to 40% of the organic mass, they should be easily extracted by a PMF analysis. In addition, would adding inorganic ions (SO_4 and NO_3) into the PMF matrix help in extracting an organic nitrate factor that could then be compared with the other methods used to identify these species?

In general, PMF can be used to deconvolute the organic matrix into several factors. However, in this study data from the entire campaign were analysed and cover a wide spatial area (horizontally

and vertically) with a fairly low temporal resolution due to high aircraft speed and low time resolution of the C-ToF-AMS. This is not the typical application of the PMF method which works best if a constant time series at constant location is analyzed. During our analysis we came to the conclusion that PMF analysis is not suited for the analysis of the data set presented here.

- 6. For the calculation of the organic nitrate concentrations. It is not clear the difference the first estimation and the third estimation. These both methods are based on the ratio of the NO⁺/NO₂⁺ ions in the instrument and how it varies from calibration values. However, using the method outlined in Kiendler-Scharr et al., is a more robust and tested method than just using the ratios alone. Can the authors comment on the added values of the first estimation compared to the third?**

The first estimation with the ratios alone can be seen as a demonstration that the potential for organic nitrates is given as it is the simplest method to check this. To give this study a more robust character the other methods were applied, compared, and discussed. According to a short comment (<https://doi.org/10.5194/acp-2018-232-SC1>) on our manuscript, a correction of the calculation was recommended and is included now.

- 7. Clarification on the contents of Figures:**

Figure 2: presents data from all flights during the ACRIDICON-CHUVA campaign.

Figure 3: presents data from 13 flights of the ACRIDICON-CHUVA campaign.

Figure 4 to Figure 10: It is not stated which flights these measurements correspond.

Can the authors provide more information on which flights were represented in figures 3 to 10 and why they were chosen over all 20 flights?

The campaign consists in total of 14 flights. Fig. 2 shows the meteorological parameters during the campaign for all flights. 13 out of 14 flights were shown in Fig. 3 - 10. During one flight (AC10, conducted on 12.09.2014) no aerosol measurements of the background air were carried out. Therefore, data from this flight are missing for the AMS and are not shown in Fig. 3 – 12.

We included the following to **Sect. 4, p. 11, line 6** (p. 12, line 13):

One flight does not provide any aerosol data (AC10, conducted on 12.09.2014). Therefore, this flight is not included in the analysis of the C-ToF-AMS data. All figures are valid for 13 flights, except where otherwise noted.

References

Crenn, V., Sciare, J., Croteau, P. L., Verlhac, S., Fröhlich, R., Belis, C. A., Aas, W., Äijälä, M., Alastuey, A., Artiñano, B., Baisnée, D., Bonnaire, N., Bressi, M., Canagaratna, M., Canonaco, F., Carbone, C., Cavalli, F., Coz, E., Cubison, M. J., Esser-Gietl, J. K., Green, D. C., Gros, V., Heikkinen, L., Herrmann, H., Lunder, C., Minguillón, M. C., Močnik, G., O'Dowd, C. D., Ovadnevaite, J., Petit, J.-E., Petralia, E., Poulain, L., Priestman, M., Riffault, V., Ripoll, A., Sarda-Estève, R., Slowik, J. G., Setyan, A., Wiedensohler, A., Baltensperger, U., Prévôt, A. S. H., Jayne, J. T., and Favez, O.: ACTRIS ACSM intercomparison – Part 1: Reproducibility of concentration and fragment results from 13 individual Quadrupole Aerosol Chemical Speciation Monitors (Q-ACSM) and consistency with co-located instruments, *Atmos. Meas. Tech.*, 8, 5063-5087, <https://doi.org/10.5194/amt-8-5063-2015>, 2015.

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