We thank Referee #2 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 paper reported an aircraft measurement in the tropical troposphere over the amazon region. The vertical profile of the main components in submicron aerosols were measured by a C-ToF-AMS. The authors focus their discussion on SOA formed through isoprene oxidation under low NO condition, i.e. isoprene epoxydiols-derived SOA (IEPOX-SOA) and organic nitrate formation. Vertical profiles of IEPOX-SOA and organic nitrate mass up to 14 km are new and very interesting. However, the analysis method of this paper has a very serious caveat which needs to be furtherly explored and addressed. I recommend a major revision due to the comments as below:**

1. **The entire analysis of this paper only depends on AMS measurement. To prove the deduction that the IEPOX-SOA can be formed in the upper troposphere. The authors should show evidences. E.g., what are the vertical profile of isoprene and NO, or even gas-phase IEPOX. The authors argued there is high organic nitrate formation due to NOx production of lighting. However, the high NO will inhibite IEPOX-SOA formation (Paulot, Crounse et al. 2009, de Sá, Palm et al. 2016, Liu, Brito et al. 2016). The addressed reason for organic nitrate formation conflicts with that for IEPOX-SOA formation. And the authors should show recalculate the PH after correcting the mass concentration of nitrate. See detailed information in the followed comment.**

   Isoprene or gas-phase IEPOX were not measured during the ACRIDICON-CHUVA campaign as there were no appropriate instruments on board. The vertical profile of NO and NO_y can be found in **Fig. 8** (Fig. 9, p. 21). We also included UHSAS-A data to provide size information of particles with a diameter between 90 and 600 nm.
   We agree to the statement that high NO will inhibit the formation of IEPOX-SOA as already mentioned in our manuscript on **p. 16, line 24**. We concluded that the oxidation of isoprene to IEPOX must occur in regions with low NO, e.g. during the vertical transport to high altitudes.
   A recalculation for the acidity of aerosol particles after nitrate correction was done and Fig. S3 in the supplement is updated.

2. **This paper reported the mass and ions measured with C-ToF-AMS, which gives an UMR spectrum. M/z 30 is mainly composed of CH2O+ and NO+ ions, and m/z 46 is NO2 and CH2O2+. The contribution of CH2O ions to m/z 30 (CH2O2 to m/z 46) is very high in areas strongly influenced by biogenic emissions. In the SE US, the aerosol composition of which is very similar to the Amazon area, UMR nitrate (with NO+, NO2, CH2O+CH2O2) is overestimated a factor of 2-3 than the high-resolution nitrate (true nitrate with NO+ and NO2) based on the default fragmentation table, due to underestimation of the organic (CH2O and CH2O2) interferences (Hu, Campuzano-Jost et al. 2017). Thus the nitrate mass concentration reported in this study are**

combination of organic and nitrates. As a result, the ratio of UMR m/z 30 and UMR 46 cannot be used to calculate organic nitrate mass concentration, unless the authors find a way to exclude the interference. A revised fragmentation table is suggested in (Allan, Morgan et al. 2014). However, its suitability for Amazon area still needs to be evaluated. Similarly, the authors cannot really report the IEPOX-SOA increases with nitrate, which is probably organics. The statement in page 17 1-7 lines are wrong due to the reason mentioned here.

As already suggested by the short comment (https://doi.org/10.5194/acp-2018-232-SC1) on our manuscript, a correction of the calculation of organic nitrate was recommended. This was done according to a recent publication by Fry et al., 2018 (https://doi.org/10.5194/acp-2018-255) and is included now. We added the following to the manuscript to **Sect. 3.4 p. 9, line 16** (p. 10, line 9):

As organic interferences on the mass spectral signals at m/z 30 (interference from CH_2O^+) and m/z 46 (interference from $CH_2O_2^+$) can occur in environments with high biogenic contribution and/or small nitrate concentrations, a correction according to Fry et al. (2018) was applied. The correction of both signals at m/z 30 and 46 is achieved by using correlated organic signals at m/z 29, 42, 43, and/or 45 derived by high resolution measurements. The organic signals at m/z 29 (CHO^+) and m/z 45 (CHO_2^+) are closest to those affected by the interference and used for the correction here. Equations 4 and 5 give the individual correction for the nitrate signal at m/z 30 and 46, respectively. The correction for NO^+ includes the total signal at m/z 30, the default fragmentation correction from the air signal (Allan et al., 2004), and a correction coefficient that depends on the m/z used for the correction (A_i). As for m/z 30 the correlated organic signal at m/z 29 is used here, the organic signal at m/z 29 (Org29) needs to be taken into account as well as the contribution of the isotopes of organic CO. For the correction of the nitrate fraction at m/z 46 a term which includes a correlation coefficient B_i and the organic signal at m/z 45 is subtracted from the signal at m/z 46. The correction coefficient A_i is in this case 0.215, B_i is 0.127 (see supplement to Fry et al., 2018). In the organic signal at m/z 28, 29, 30 and 45 the relative ionization efficiency (RIE_{Org}) is already applied and needs to be reversed for the correction of the nitrate signal.

Nitrate fraction at m/z 30:

$$NO^+ = m/z\ 30 - 0.0000136 * m/z\ 28 - A_i * (Org29 - 0.011 * Org28) * RIE_{Org} - Org30 * RIE_{Org} \quad (4)$$

Nitrate fraction at m/z 46:

$$NO_2^+ = m/z\ 46 - B_i * Org45 * RIE_{Org} \quad (5)$$

The total nitrate signal is then calculated by adding both fractions. The final nitrate mass concentrations were reduced by 0.045 µg m^{-3} (STP) on average corresponding to an averaged reduction of 39 % of the initial nitrate mass concentrations. A comparison of the initial and finalized nitrate mass concentrations can be found in the Supplement (see Fig. S2).

Furthermore, we added the following to the paragraph in **Sect. 4.5 p. 19, line 2** (p. 21, line 16):

Figure 10 shows the scatter plot of the corrected NO^+ and NO_2^+ for the LT (dark blue) and the UT (light blue). The linear fit curves for the LT and UT have an intercept of 0, proving that the applied correction is essential. Nevertheless, it has to be noted that the correction is based on correlations between different m/z signals derived from measurements at low altitudes (Fry et al., 2018), thus the application to UT data bears uncertainties, because the conditions (especially temperature) are different. However, high resolution AMS measurements at these altitudes are currently not available.

The linear fit for the LT data shows a higher slope than that derived from calibrations with ammonium nitrate. The linear fit for the UT shows also a higher slope and some data points are still significantly above the fitted ratio between NO^+ and NO_2^+. This can be seen as a first hint that organic nitrates might be observed, especially in the UT.

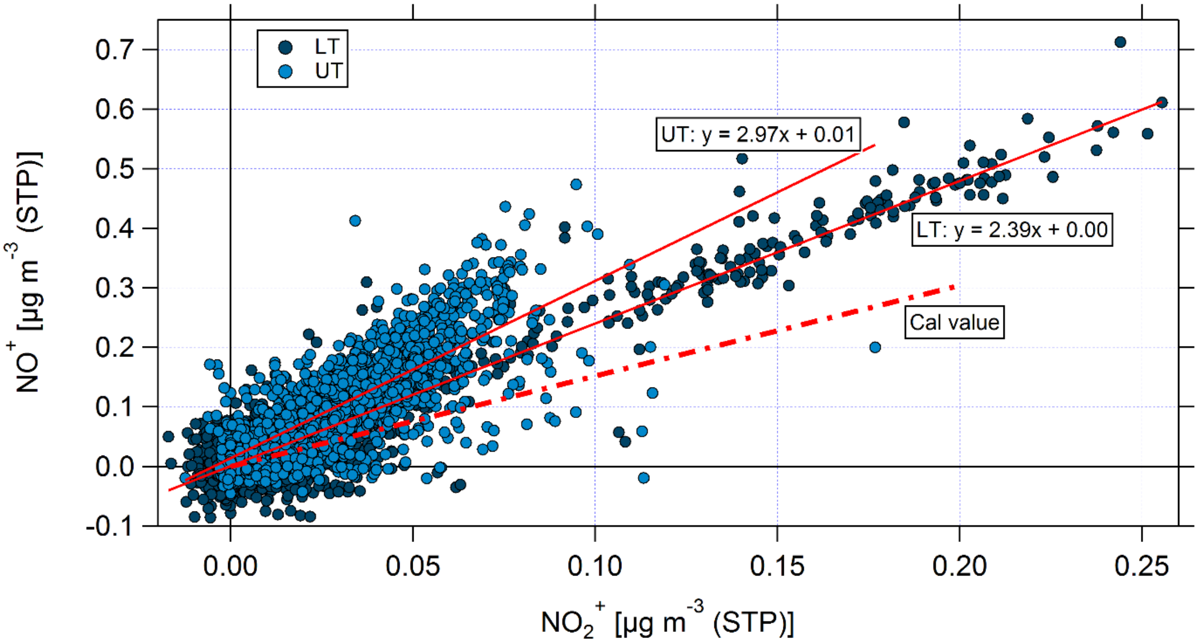

Figure 10. Scatter plot of NO^+ and NO_2^+ for the lower troposphere (LT, dark blue) and the upper troposphere (UT, light blue). The nitrate signals have been corrected for organic interference according to Fry et al., 2018. Linear fit curves are shown in red, the ratio of NO^+ and NO_2^+ derived from calibrations with ammonium nitrate is presented by the red dashed line.

We added the following to the supplement:

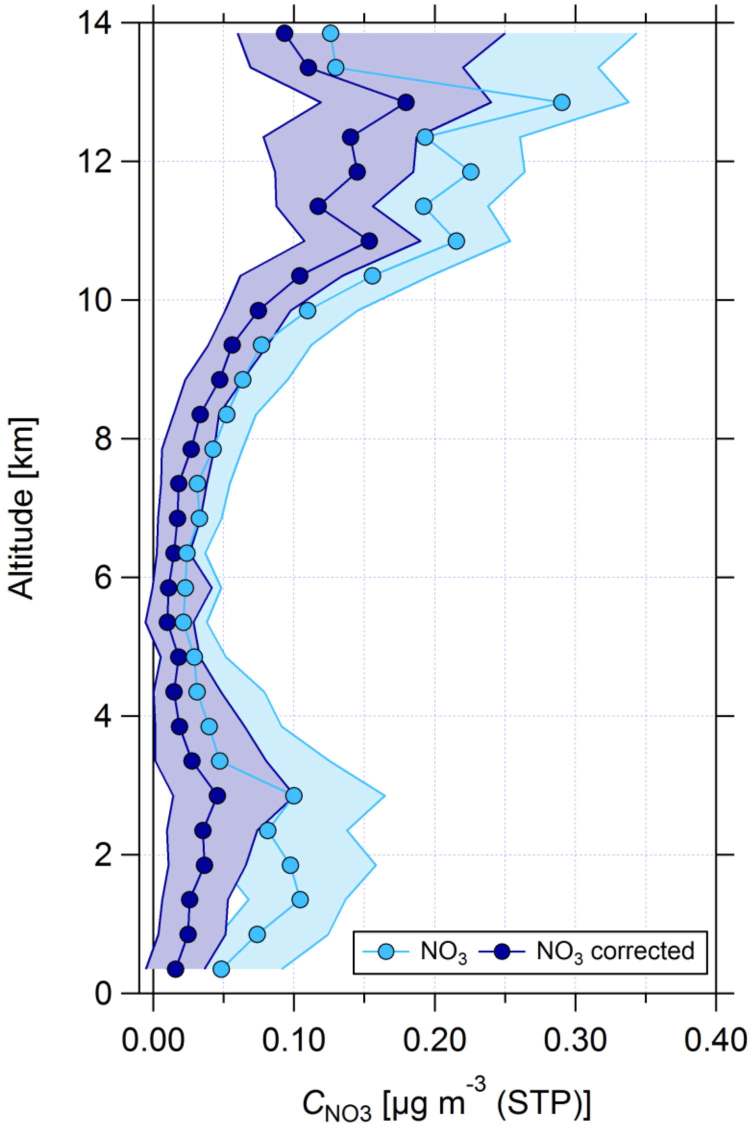

Figure S2. Vertical profile of median nitrate mass concentrations and interquartile ranges before (light blue) and after (dark blue) corrections according to Fry et al., 2018 and supplement to Fry et al., 2018.

Furthermore, we changed the paragraph in **Sect. 4.4 p. 17, line 2** (p. 20, line 8) to the following:

Although the correlation between IEPOX-SOA and nitrate is weak, it may indicate that not only sulfate, but also nitrate can provide the acidic conditions for the partitioning of IEPOX to IEPOX-SOA. For our data, taking only the inorganic species (nitrate and sulfate) into account for acidity calculations, the aerosol is mainly neutralized (see Fig. S6). Although there is a tendency in the UT above 10 km that the measured ammonium is not sufficient to neutralize the inorganic species, a quantitative statement cannot be made as the values fall below or close by the DL. The presence of organosulfates and -nitrates could also affect the acidity calculations. As for the organonitrates the data are already corrected, for organosulfates such a similar correction is not possible with data from a C-ToF-AMS as there are no different fragmentation patterns between inorganic and organic sulfates (Farmer et al., 2010).

3. **Page 20 line 15, there is no way that the calculated organic nitrate mass concentration based on Fry et al. (2013) can be higher than the total nitrate mass concentration measured from AMS. Because Fry et al. (2013) used a method base on splitting the mass of total nitrate masses with differential NO/NO2 ratios from NH4NO3 and organic nitrate. Please check furtherly.**

   After correcting the contribution of organics to m/z 30 and m/z 46 all data were recalculated. For the comparison with the method described by Fry et al. (2013) a good agreement is now achieved. The vertical profiles show similar median values (see Fig. 11). Differences in the scatter plot are in the range of uncertainty of the data (see Fig. 12).

4. **How are the vertical profiles of main components in submicron aerosols in this study compared to the amazon study reported in (Allan, Morgan et al. 2014). Also the It will be interesting if the authors can address the similarity and differences, especially for m/z 82 and f82. What is the new finding in this study compared to the previous ones.**

   The study by Allan et al., 2014 reported aircraft measurements above the Amazon rain forest at altitudes up to 5 km. Our study presents also data from higher altitudes up to 14 km, providing more information on the vertical profile of submicron aerosol chemical composition and f_{82}.
   We added a short comparison with the study by Allan et al., 2014 to the paragraph in **Sect. 4.4 p. 16, line 10** (p. 18, line 12):

   To the best of our knowledge, f_{82} data from the tropical upper troposphere have not yet been reported in the literature. Aircraft measurements above the Amazon rainforest were reported by Allan et al. (2014), but these data are restricted to altitudes below 5 km, corresponding to the definition of the LT in this study. In Allan et al. (2014), two cases are presented and show background (4 ‰) and increased values (9 ‰) of f_{82}. The highest values of f_{82} were found on top of the boundary layer, decreasing with increasing altitudes. These values are similar to the data from the LT presented here.

Minor comments:

5. **Page 7 line 12: please give details about the "a time-dependent cubic spline function was used to determine a detection limit for each data point"**

   In order to provide more details on the function that was used to determine the detection limits, we added the following to the manuscript to **Sect. 3.1 p. 7, line 11** (p. 7, line 13).

   Here, a time-dependent cubic spline function was used to determine a detection limit for each data point. This function was developed at the Max Planck Institute for Chemistry (MPIC) and is introduced and explained in more detail in a PhD thesis (Reitz, 2011). For every data point of the background signal a third order polynomial (cubic) function is calculated through the four neighbouring points (two before and two behind) while omitting the actual point. Applying this method, all trends from the background signal are excluded and just the short-term noise remains.

A quantity *R* is introduced in the algorithm and characterizes the statistical spread of the noise. *R* is defined by the squares of the deviation between the omitted centre point and the cubic function along a moving window. To relate this *R* to the standard deviation (σ) a proportionality factor of $(18/35)^{1/2}$ is needed (Reitz, 2011). The exact derivation of the proportionality factor will not be explained here, but qualitatively it accounts for the fact that not only the points are affected by noise but also the cubic function itself. Thus, *R* is larger than the standard deviation (σ). This calculation also provides a continuous σ from which the detection limit can be derived using the equation $DL = 3 \times \sqrt{2} \times \sigma$.

6. **Page 6 line 13: give full citation of Molleker et al. 2018.**

   Molleker et al., 2018 is a manuscript in preparation. We changed the citation in the manuscript to the following:

   Further details of the constant pressure inlet are subject of a separate publication (Molleker et al., manuscript in preparation).

7. **Page 8 line 15-16: Hu et al. 2015 showed an f82 value from IEPOX-SOA and background sources in the appendix.**

   We do not understand this comment. We refer to Hu et al. (2015) already.

8. **Page 8 equation 1, what is the f82 values of IEPOX-SOA used in this study to calculate the mass concentration of IEPOX-SOA.**

   The f_{82} value of IEPOX-SOA used in our study is 22 ‰ as suggested in Hu et al. 2015. We denoted this as $f_{82}^{IEPOX\text{-}SOA}$ (**p. 8, line 26-27** (p. 9, line 7-8)).

References:

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Reitz, P. (2011). Chemical composition measurements of cloud condensation nuclei and ice nuclei by aerosol mass spectrometry. Mainz: University.
url: http://nbn-resolving.org/urn:nbn:de:hebis:77-28691.