Reply to Anonymous Referee #2:

Thanks a lot to the reviewer for his/her helpful advice. Please find our point-by-point response below.

Received and published: 8 August 2018

General assessment and major comments

This study provides lidar measurements from two sites in Northern France, showing long range transported smoke in the UTLS. The absorbing nature of smoke is crucial for the stratospheric height ranges, concerning both heating rates (HR) and direct radiative forcing (DRF). The authors try to estimate the DRF and HR and their results show decrease of the radiation reaching the surface and an increased HR due to the absorption of the solar radiation at TOA. In general, I find this study very interesting and of high value. It is a study that fits well in the EARLINET special issue, since it demonstrates the value of EARLINET lidars for atmospheric research in both troposphere and stratosphere. However, before proceeding with publication in ACP, I strongly suggest that the authors would revise the following points:

1. Page 11, Lines 18-27: "The spheroid model was used to retrieved dust properties (Dubovik et al., 2006; Mishchenko et al., 1997; Veselovskii et al., 2010). But it is not clear if this model is applicable to soot particles with complicated morphology. The size of smoke particles is expected not too big so that we choose to apply regularization algorithm with sphere model." The retrieved microphysical properties seem to be associated with high uncertainties, since the shape used (spherical) does not reproduce the depolarization measurements and it should not reproduce accurately the backscattered light measurements either. The reported uncertainties in Table 2 refer to cases of spherical particles and are not representative. Please provide a better assessment of the retrieval uncertainties.

A1: It is true that spheres do not represent correctly smoke particles, neither spheroid. Our retrievals of dust particles demonstrated, that when spheres were used instead of spheroids, the algorithm was still able to provide reasonable estimates of volume and effective radius (Veselovskii et al, JGR 2010). The main errors were attributed to estimations of the refractive index. So we expect that in the case of smoke estimations of radius and volume are also possible.

2. Regarding the DRF calculations: these are based on the retrieved microphysical (point 1) properties which, as discussed above, are derived from the 3b+2a regularization inversion and are associated with (most probably) high

uncertainties. Especially for the imaginary part this uncertainty is expected to be the highest (Burton et al., 2016). Please provide a better assessment of the retrieved property uncertainties and quantify the uncertainties of the DRF calculations accordingly. If this is not possible, omit section 4.2.3 from the manuscript. This also applies to Page 14, Lines 9-13, where the derived complex refractive index is compared to other studies. Omitting 4.2.3 would not affect the quality of the paper, since the authors already provide important results on smoke optical properties and microphysical estimates.

A2: From our simulation studies we estimate errors of V (volume concentration) and R_{eff} as 30%, for m_R it is +-0.05 and m_I 50%. These are typical values and we are not able to evaluate the effect of shape of on retrievals. But basing on dust studies, we expect it to be similar.

The deficiency of using sphere model is its not being able to reproduce the depolarization effect. However, the estimation of the radiative effect is not so sensitive to the depolarizing effect of the particles. Indeed, the uncertainty of the imaginary part of the complex refractive indices is much higher than the other parameters and it is strongly dependent on the shape of the particles, but the values we present are quite reasonable for previously reported absorbing smoke. We think the estimated radiative forcing is quite representative and the heating of smoke predicted by the DRF is able to explain the ascending trend of the plume, as shown in the newly added figure. Although the values suffer from some extent of uncertainties, we would like to keep section 4.2.3 and we will mention in the manuscript that the uncertainty of the retrieved aerosol microphysical properties affects the accuracy of the DRF estimation.

3. Another issue addressed in this study is the increase in particle depolarization ratio at 532 nm which is attributed to the particle aging. The authors gathered observations of the particle linear depolarization at 532nm from previous studies and have also included the results obtained from the present study. Nevertheless, the only visible trend seen in Figure 11 results from CALIPSO measurements. From the ground-based lidars in Lille and Palaiseau, there is no obvious increase at 532nm. In conclusion, the phrase“we found an increase in depolarization versus transport time” in the manuscript abstract should be changed to “CALIPSO observations of the UTLS smoke layer suggest an increase in depolarization at 532nm versus transport time”.

A3: We agree that the main increasing trend of the depolarization is indicated by the CALIPSO measurements. However, the CALIPSO data are questionable because of the high noise level. Moreover, the RH of the smoke plumes is not known. As a result, we cannot draw really convincing conclusion about the changes of depolarization ratio

during the aging process. At current stage, we decide to remove this part from the manuscript and more efforts will be made to investigate this issue and re-assess CALIPSO data.

Minor comments

4. Page1, Line 9: “Typical particle depolarization” the meaning of the word typical should be clarified by the authors, meaning what is the definition of linear particle depolarization ratio used? (is it the cross/parallel ratio or the cross/total ratio?)

A4: After re-organizing the paper, the definition of particle linear depolarization ratio is in Section 2. The methodology is presented before the observation section, so this problem is avoided.

5. Page 1, Line 10: “The relatively high depolarization ratios and such spectral dependence are an indication of a complicated morphology of aged smoke particles” The conclusion that the spectral dependence of the depolarization ratio is characteristic of aged smoke particles can be hardly drawn by two cases, i.e. the current one and the one reported in Burton et al. (2015). Please rephrase accordingly.

A5: This conclusion is drawn in Mishchenko et al., 2016

6. Page2, Line 30: “We focus on the retrieval of the aerosol optical and microphysical properties from the Lidar measurements”. The authors should highlight that the depolarization ratio values are not reproduced in the retrieval of the microphysical properties.

A6: Yes, this message is given in section 4.2.3 as the limitation of the retrieval.

7. Page 4, Line 2: Please change the phrase “showed an increase of temperature in the stratospheric smoke layers” to “An increase of temperature due to the presence of smoke aerosols in this region” or something similar.

A7: Modification has been made in the revised manuscript.

8. Page 5, Line 3: Change the phrase “A plume with relatively high UVAI first occurred over the British Columbia on 11 August, and the intensity of the plume was moderate” to “a plume of moderate intensity and relatively high UVAI, first occurred over British Columbia on 11 August. Page 5, Line 4: Please change the phrase “and the UVAI in the center of the plume reached above 10” to “and the

UVAI in the center of the plume reached above 10, as indicated by the grey area on the plot (Fig 4)"

A8: Modification has been made in the revised manuscript.

9. Page 6, Line 1: "We have examined the temperature profiles" Did you use radiosonde measurements? Please provide more info.

A9: Yes, it is radiosonde measurements from Wyoming radiosonde stations, data can be found here: http://weather.uwyo.edu/upperair/sounding.html

10. Page 6, Line 2: "the temperature drops below -38_C, at which temperature the cloud droplets mostly turn to ice phase" Please provide relevant reference.

A10: Please refer to Kârcher et al., 2003, A parameterization of cirrus cloud formation: Heterogeneous freezing.

11. Page 6, Line 8: "The increasing trend of the depolarization ratio is probably due to aerosol aging" As discussed above, this is a hardly drawn conclusion. Please rephrase accordingly.

A11: We decide to remove this argument.

12. Page 7, Line 7: "we can calculate the optical depth of the cirrus cloud" Please change "cirrus cloud" to "UTLS aerosol layer" since this is what you refer to in this case.

A12: Corrected.

13. Page 7, Line 12: change the phrase "are considered as the major error sources of the optical depth" to "are considered as the major error sources in the estimation of the optical depth"

A13: Corrected

14. Page 7, Line 13: "based on the statistical error of photon distributions" Please provide more info on the definition of the noise of your lidar measurements. Do you take into account the systematic errors?

A14: The error of the lidar signal is estimated based on the assumption that the photon-counting detection mode of the photomultiplier follows Poisson distribution. Then signal error is given by the covariance of the Poisson distribution. Systematic error of photon-counting detection is negligible especially in nighttime measurements, so it is not taken into account in the error estimation. We estimated about 3% of error for nighttime signal. In order to account for the interference of sunlight, we roughly use 5%

for the error in daytime.

15. Page 7, Line 22: change the phrase “of the error of optical depth” to “of the error of optical depth to the estimation of the Lidar ratio”

A15: corrected

16. Page 10, Line 15: typing error, change volume depolarization ratio at 355 nm to molecular depolarization ratio at 355 nm.

A16: Corrected

17. Page 14, Line 14: “Smoke in dry conditions have higher refractive indices than that in wet condition” Provide relevant reference.

A17: After reconsideration, we think this statement is not strict. Studies have shown that fresh smoke has a broad range of hygroscopicity. The study of the hygroscopicity of aged smoke is quite limited and requires more observational and experimental efforts. Additionally, the aging process could be very complicated considering possible effects related to the photochemical process, fuel types, particle coagulation, secondary aerosol generation and so on.

We decide to remove this comment and mention in the revised manuscript that “the hydroscopicity of aged smoke is not yet well revealed.”

18. Figure 6: The x axes on CALIPSO plots should be the same in order to show the variation. Also the phrase in the caption “The profiles of backscatter coefficient and particle linear depolarization ratio (PLDR)” could be changed to “The profiles of backscatter coefficient and particle linear depolarization ratio (PLDR) at 532nm from CALIPSO” Figure 7: The points on this figure should be larger to be more visible. Also, it would be better if the colors of the points are different for the two lidar systems.

A18: Corrected.