Response to Anonymous Referee #3 :

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

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The paper provides unique measurements of an extreme event of smoke advection from Canada to Europe. Smoke could be observed within the Troposphere as well as in the Stratosphere. I consider the measurements and the resulting data as very valuable and of interest for the scientific community and the topic fits well in the scope of ACP.

However, in my opinion the paper tries to cover too much topics at once (optical properties of smoke, microphysical properties, source analysis, change of depolarization with aging time by using Calipso data, radiative transfer calculations, estimating of heating rates, temperature changes in the stratosphere etc.). Thus, the paper is partly confusing and for some of the topics the proper fundament needed for the conclusion drawn are missing.

I therefore recommend first to focus on your key expertise and present only the unique measurements, which are already shown and exploit as much as possible (optical and inversion results). Of course you should also include the satellite data for source analysis. However please focus on the lidar measurments in France. Then, in another paper(s), the other topics could be covered (i.e. radiative transfer calculations and heating rates etc.) Especially the discussion of the impact of the smoke plumes need much more detail and evidence, as for example it is not shown nor discussed that temperature increases in the stratosphere at a certain height are not caused by the complicated upper atmospheric circulation including tropopause foldings etc. I therefore recommend for the second paper to include some expertise of the upper atmospheric dynamics and probably some modelling to show and prove the impact of the smoke layer with its possible warming on the overall general condition of the UTLS region above Europe.

I also think that the use of Garrlic/Grasp radiative transfer code to obtain radiative impact must be explained much better. Currently, it is not understandable how the calculations are performed.

From the current version, I also doubt for the evidence of the increase in smoke depolarization with aging time. I think the use of Calipso data for such a study is again a topic of its own. Uncertainties in the Calipso retrieval (see comments in pdf) should be discussed. Even more important, other influencing factors as the relative humidity, the altitude of the smoke with respect to ground level but also tropopause should be investigated. Among others influencing factors (fire source, burning types).

I therefore recommend major revision with the recommendation written above. Find other comments below and as pdf-comments in the supplement.

Paper structure:

1. -The abstract is too long. Please shorten.

A1: The abstract is rewritten after all the corrections.

2. -The introduction is in my opinion a loose sequence of different paragraphs and therefore not constructive. Please revise and make it more focusing on your topic. E.g.: have such events been reported earlier? Is this the first time? . . .

A2: Modifications have been made.

3. -Several facts concluded in the observations section are again raised in the discussion. I think you should shape your paper. Either describe your observations only, and then make a discussion in a separate section or make conclusions also in the observations section but then discuss only new issues in the discussion section.

A3: The abstract, introduction, discussion as well as the conclusion have been re-shaped.

Terminology

4. -I would recommend not to use “upper troposphere/lower stratosphere UTLS” as a standard the term. This historical term covers all altitudes between 5 and 30 km and thus does not make clear that a significant portion of the smoke you detected is above the local tropopause. Please feel free and state, that there is stratospheric smoke as well as smoke in the free troposphere. In most times you anyhow refer to stratospheric smoke with your statements. . .

A4: We decided to change “upper troposphere/lower stratosphere UTLS” to lower stratosphere.

Major scientific remarks:

5. -What about the uncertainties of Calipso. For example for the PLDR, the particle backscatter coeff. is needed which is in turn calculated with a Klett-like approach, i.e. a a-priori lidar ratio. Thus the questions arises, which aerosol type was classified by Caliop and which lidar ratio was used to obtain the PLDR and is this correct for your aerosol type of investigation.

A5: This is a very useful comment. In Figure 6, we find that the observed plumes are not well classified. 1) The classification provides scattered aerosol types, such as polluted dust, elevated smoke, dust and volcanic ash. 2) The derived aerosol type sometimes oscillates profile by profile, and even adjacent profiles could be classified into different categories. Different aerosol types correspond to different lidar ratio assumptions, but Figure 6 shows the mean profiles of backscatter coefficient and PLDR over a small range (latitude ±0.01), without considering the impact of aerosol mis-classification. In this situation, the error of CALIPSO results is barely expectable.

We added in the manuscript that the plumes are not well classified in CALIPSO data processing and this decreases the accuracy of CALIPSO results. But we still keep Figure 6, in order to the transport of the smoke plume from Canada to Europe. In addition, we remove the argument about PLDR increasing with transport time, because of the unknown error level of CALIPSO PLDR product.

6. -Depolarization: The molecular depolarization ratio depends on your filters used in the lidar. Are the theoretical values you stated valid for your system? And can you neglect temperature effects? Which molecular depol ratio value did you use for the PLDR calculation? The measured one or the theoretical one?

A6: The theoretical value of molecular depolarization ratio (specific to the Cabannes line) is about 0.36%. However, due to the imperfection of the lidar optics (and ther factors), the measured depolarization ratio in the aerosol-free zone is higher than the theoretical value. Our interference filters in LILAS system well block the rotational Raman lines. Figure 1 below shows the rotational Raman lines and Cabannes lines for the laser wavelength at 532 nm (in standard atmosphere), as well as the transmission function of the interference filters. We have estimated the total molecular depolarization ratio to be 0.4%, including the Cabannes lines and rotational Raman lines and found that the impact of the included rotational Raman lines on the molecular depolarization ratio is quite minor. However, in the historical measurements, our system LILAS measured about 0.8—1.3% at 532 nm, 1.2—1.8% at 355 nm and 0.7—1.0% at 1064 nm. The depolarizing effect of the optics, the misalignment and the error in the calibration procedure are expected to be responsible for the error of measured molecular depolarization ratio. In the calculation of the aerosol particle linear depolarization ratio and its error, we use 0.4% for the molecular depolarization ratio. The error level of molecular depolarization may look a bit astonishing but, fortunately, the total error of the particle depolarization ratio is much less dependent on it when the aerosol is optically thick and depolarizing, like in the presented cases. In addition, we measured cirrus clouds below the stratospheric plumes on 24—25 August, the derived PLDRs are about 45%, without noticeable spectral dependence. The results are very consistent with previously reported PLDR of ice clouds and can be regarded as a verification of our measurements.

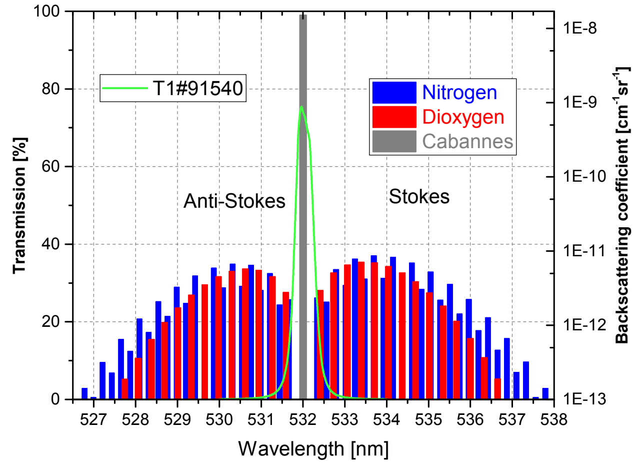

Figure 1. The molecular backscatter coefficient of rotational Raman lines and Cabannes line for the laser line at 532 nm. The calculation is made under a standard atmosphere. Only oxygen and nitrogen are considered as scatters in the atmosphere.

7. -Radiative transfer: The methodology explanation is too short. It is not reproducible how you performed the radiative transfer. Which parameters did you use as input? Which are constrained by the algorithm? and so on... thus, either you introduce a much more detailed description of this method and the paper gets longer or you shift these calculations to a second paper.

A7: We agree that the explanation about GARRLiC/GRASP is not enough for reproducing the forcing effect of the smoke plumes. More information has been added in the manuscript to describe the general strategy of GARRLiC /GRASP and the input parameters for the calculation procedure. The theories and methodology of GARRLiC/GRASP can hardly be well presented in a short section. So we suggest the readers to refer to previous publications about GARRLiC or GRASP. GRASP is an **open source algorithm**, anyone who is interested in using GRASP to reproduce the results in this paper or to invert their own measurements, is **very welcome** to download the algorithm here: https://www.grasp-open.com or contact us by email.

8. -Inversion: Is it useful to perform an inversion when having only 3 elastic signals? I mean lidar ratio is not an independent variable in your case...please discuss this!

A8: The lidar ratio is not a completely independent parameter, because we introduce an extra constraint, which is the optical depth of the smoke layer. Indeed, assuming vertically constant lidar ratio is not a favorable way in the Raman lidar community, because it looks not realistic in some cases. But do not forget that, the particle depolarization ratio is almost vertically constant in the smoke layer. It indicates that the particles are well mixed in the smoke layer. Based on this fact, we have confidence to say that the lidar ratio within the smoke layer will not show significant variations. To assure this hypothesis, we compared the backscatter coefficient calculated from Raman and Klett method. The comparison in the selected two cases is shown in Figure 2. One can see that the differences in the backscatter profile between the two methods are quite minor. It indicates that the backscatter coefficient we calculated is reliable and assuming a constant lidar ratio in the smoke layer is not far from the truth.

There are actually **5 input parameters** in regularization algorithm. As to the extinction profile, it fits the pre-calculated optical depth so the mean extinction in the smoke layer is also trustworthy.

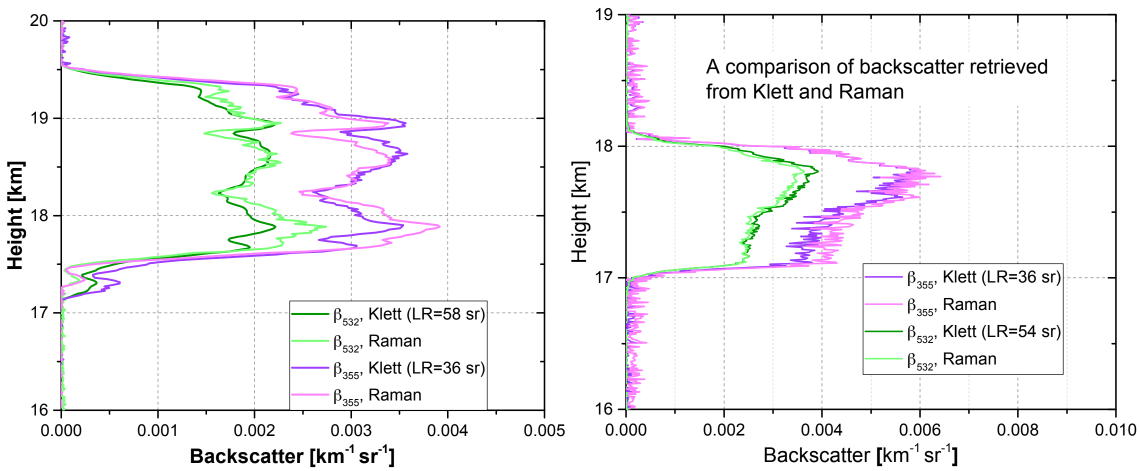

Figure 2. The comparison of backscatter coefficient, (left) 19:20—21:20 UTC, 28 August 2017, Palaiseau and (right) 20:30—00:30 UTC, 24—25 August 2017, Lille

9. What justifies using a sphere model when particle size is small.

A9: It is maybe a bit miss leading to say that the small particle size justifies the applicability of sphere model. What we really wanted to address is: the difference between spheroid scattering and sphere scattering is very minor when the particle size is small, and to not complicate the situation, we chose to use a simpler model, the sphere model.

The sensitivity of scattering of particles to the shape (spheres or spheroids) can be found in "*Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust*". Figure 3 is taken from the Figure 26(b) in Dubovik et al. 2006. It plots the lidar ratio at 532 nm as a function of the aerosol Angstrom exponent, which is an indicator of the particle size.

It can be seen that when the Angstrom exponent gets bigger, the difference of lidar ratio between spheroid and sphere model gets smaller.

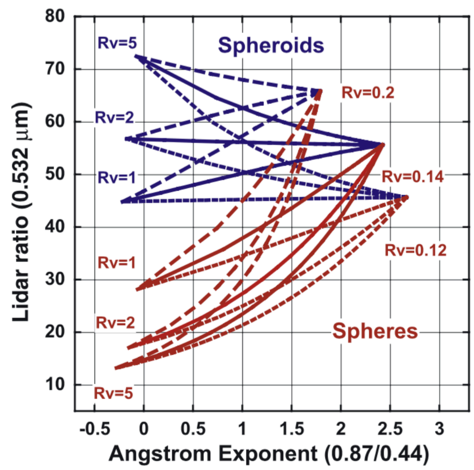

Figure 3. Lidar ratio plotted as function of Angstrom exponent. Rv is the mean radius of the size distribution. Each line shows the dependence of the lidar ratio for aerosol defined by a lognormal bimodal distribution of spherical (red labels) fine mode (with median radii 0.12, 0.14 or 0.2 mm) and coarse spherical (red labels) or spheroid mode (blue labels) (with median radii 1.0, 2.0, 3.0 or 5.0 mm).

10. -Increase in stratospheric temperature. Your explanations are not convincing concerning the temperature increase. Did you consider also all other processes in the UTLS? May this only be normal variability? How long do you observed a temperature increase? So please add more detail or shift to another paper (I think this is a topic alone).

A10: Other process, for example the variation of ozone concentration can also result in temperature changes in the stratosphere. We investigated the temperature profile measured by radiosonde at Trappes (close to Palaiseau, France) in the last two weeks of August 2017. Figure 4(a) shows that the temperature in the stratosphere has obvious variations in August 2017. The region in the magenta box is distinct with others, because it is an obvious local maximum. Moreover, the spatial-temporal occurrence of this temperature peak coincides with the occurrence of the smoke plume. The same spatial-temporal coincidence appears to Beauvechain temperature observation (close to Lille station). Based on the observations in two independent observation site, we are confident that temperature increase in the plume layers is caused by the presence of the absorbing smoke layers instead of other reasons.

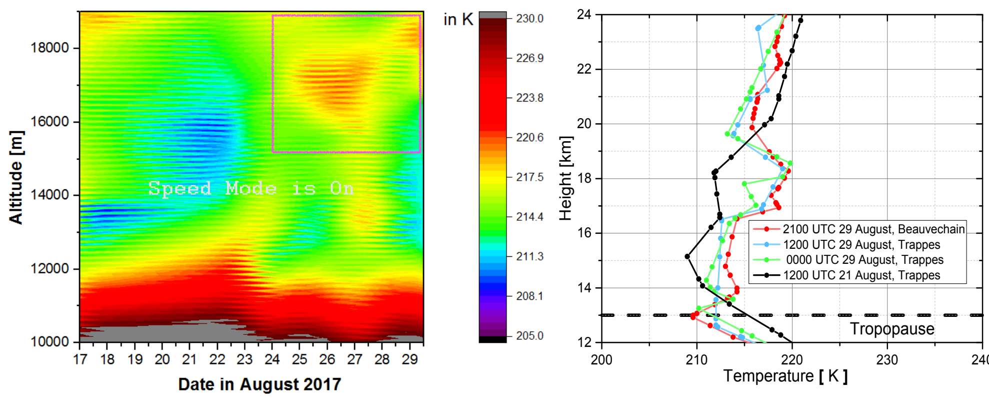

Figure 4. (a) Temperature at Trappes in August 2017. (b) Temperature profiles at Trappes and Beauvechain.

11. -Depolarization ratio vs. aging time: The provided graphic and literature review does not convince me of the given causality, please also investigate the RH, the height etc. vs depolarization ratio.

A11: We agree that CALIPSO data are very noisy from which we can hardly draw convincing conclusion about the depolarization increasing with aging time. The error level of CALIPSO measurements is also questionable regarding the stratospheric smoke layers. In addition, the RH of the smoke plume is not available in CALIPSO measurements and some ground-based lidar observations. As a result, we decide to remove the questionable argument about depolarization increasing with aging time. We need more investigations before getting more convincing results.

- Please also note the supplement to this comment: [https://www.atmos-chem-phys-discuss.net/acp-2018-655/acp-2018-655-RC3- supplement.pdf](https://www.atmos-chem-phys-discuss.net/acp-2018-655/acp-2018-655-RC3-supplement.pdf)

The modifications as below have been made following the comments in the supplement:

1. The abstract, introduction discussion and summary are reshaped.
2. Typing and grammatical errors, ambiguous statements and other minor errors pointed out by the reviewer have been corrected.
3. A table has been added to summarize the configuration of the three Lidars

4. Data will be uploaded to EARLiNET database after the final review session. More

information about data availability is added.

5. Acknowledgement is modified and author contribution is included.

6. Errors of Rayleigh scattering?

AA6: We use temperature and pressure profiles from the closest radiosonde profiles. The Rayleigh fit in the aerosol free zone is excellent so the errors resulting from molecular scattering are ignored in the error estimation.

7. Why not simply use the lidar ratio you retrieved with the other method or the ones reported by Haarig et al.? I consider using the total AOD as a constrained much more critical as you have to use one lidar ratio for all heights...you could also use the lidar ratio of the PBL obtained with LILAS or IPRAl for the MAMS Klett retrieval in the PBL.

AA7: MAMS system performed measurements between Palaiseau and Lille. There are three data points from MAMS, two of them are not collocated with IPRAL or LILAS, and only the third data point was obtained in Lille. We lack the information of the tropospheric aerosol along the road. Moreover, considering the noise in MAMS lidar daytime measurements, the AOD measurement collocated with the MAMS Lidar should be a more solid constraint. So we chose to process MAMS lidar measurements with an extra constraint of AOD.

8. Divide the paper into two papers

AA8: After re-shaping the paper and considering the advise from the other reviewers, we would like to keep the observation, inversion and radiative forcing estimation in the same paper. But, as you suggested, we added more information about GARRLiC/GRASP algorithm. We cannot present all the details about GARRLiC/GRASP because it is an integrated algorithm containing many modules. In the revised version, the basic strategies and the input parameters are introduced in more detail, so if the users want to reproduce the results, they can download this open-source algorithm and follow the instructions. The information we present in the paper is to show the general strategy of the algorithm.