

acp-2016-212, “Near-surface and columnar measurements with a Micro Pulse Lidar of atmospheric pollen in Barcelona, Spain” by Sicard et al.

General comments from the authors: First of all, we want to thank the two referees of our paper for their time and revision of our work. There is a small change in the paper that we would like to indicate to the referees. We noticed that the pollen on the last day of the event, on March 31st, after 18UT is not removed from the atmosphere (as it was said in the initial submission). A look at Figure 2b evidences it: a strong volume depolarization ratio persists in the ABL after 18UT but not below 0.5 km where it comes down to values typical of the local, background aerosol in Barcelona. This non-depolarizing plume (< 0.5 km) is at the same time associated with high values of the backscatter coefficient (Figure 2a). We checked that the detection of the pollen plume height was erroneously found at the first range bin shown (near 0.16 km), and not near the top of the ABL as it should be. We corrected our script and ran it again for the last hours of March 31st. This re-run results in changes in h_{pol} and thus on all integrated parameters (AOD_{pol} , AOD_{pol} / AOD , $\overline{\delta^v}$ and $\overline{\delta^p}$) only for March 31st after 18UT. We updated the following figures: 5, 6 and 8 and all four tables, as well as the discussion related to March 31st. We insist that these little changes only affect the data from March 31st after 18 UT, and do not change anything to the conclusions of the paper.

Answers to Referee#2's comments

Comment on “Near-surface and columnar measurements with a Micro Pulse Lidar of atmospheric pollen in Barcelona, Spain” by M. Sicard et al. More and more attentions have been paid on biological aerosols due to their significant impacts on environment and climate. The article presents an investigation of nearsurface and column characterization of atmospheric pollen in Barcelona, Spain, mainly by use of lidar measurements and sampling analysis. Moreover, impact of meteorological elements (e.g., RH, T and wind speed) and solar flux on atmospheric pollen load in the atmosphere was discussed in detail. The topic is of sufficient interest to the communities of study of atmospheric aerosol (especially bioaerosols), climate as well as human health. In general, I find this manuscript to be of interest for publication and appropriate for this journal. There are several suggestions for improvement listed below that should be considered by the authors and the editors before publication.

1. In fact, there is uncertainty during measurements of atmospheric pollen and spores. As introduced by the authors, pollen and spores was identified using a fluorescent microscope. However, the results should be affected by other fluorescent particles. It will be easier for readers to understand if the authors briefly introduce how to identify pollen and spores, obtain their concentration as well as discuss its uncertainty.

Authors'reply: Pollen and spores counts were performed by specialist technicians using light microscope, not fluorescent microscope. The explanation of pollen and spores identification has been rewritten in order to better describe the methodology used (Lines 26-34 of the initial submission):

“The drum was changed weekly and the exposed tape was cut into seven pieces, each one corresponding to one day, which were mounted on separate glass slides. Pollen and spores were counted under light microscope, at 600X magnification. Daily average pollen and spore counts were obtained following the standardized Spanish method (Galán et al., 2007), consisting in to run four longitudinal sweeps along the 24 h slide for daily data, identifying and counting each pollen and spore type found. To obtain the hourly concentrations, twenty-four continuous transversal sweeps separated every 2mm along the daily-sample slide, were analyzed, since the drum rotates at a speed of 2 mm per hour. Daily and intra-diurnal (hourly)

pollen and spore concentrations are obtained converting the pollen and spore counts into particles per cubic meter of air, taking into account the proportion of the sample surface analyzed and the air intake of the Hirst pollen trap (10 L min⁻¹)”

2. In spring, dust aerosols could be long-range transported to Barcelona. And pollen, like dust aerosols, are coarse particles and shows strong backscatter signal and large depolarization ratio from lidar measurements. Even high mass concentration of PM10 also could be seen during dust events. So the authors should explain why this is a pollination event, not a dust event. How to distinguish dust particles from pollen and spores?

Authors’reply: The synoptic situations was not favourable at all for the transport of mineral dust to Barcelona. In the Supplement 1 at the end of this document we add the forecast of mineral dust load from the BSC-DREAM8b model as well as Hysplit backtrajectories. No dust transport is forecasted by the model and the backtrajectories, identical during the five days, clearly indicate a North Atlantic (and thus clean) origin of the air masses transported to Barcelona. As Supplement 1 is not provided in the revised manuscript, the following sentence has been added at the beginning of Section 3: “To confirm that mineral dust was not transported over Barcelona during the pollination event, we used the dust transport models BSC-DREAM8b v2 (Barcelona Supercomputing Center – Dust Regional Atmospheric Model 8 bins) and NMMB/BSC-DUST (Nonhydrostatic Multiscale Meteorological Model on the B grid / Barcelona Supercomputing Center – Dust), as well as HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) backtrajectories (not shown).”.

3. Page 4 line 24: incomplete sentence, “ P_{\perp} and P_{\parallel} represent the perpendicular and parallel backscatter powers respectively”.

Authors’reply: The sentence has been completed.

4. Page 4 line 30: please delete “linear”. Strictly speaking, MPL depolarization ratio is not linear depolarization ratio.

Authors’reply: We agree with the referee that what Flynn et al. (2007) call MPL depolarization ratio, δ_{MPL} , is not the linear depolarization ratio. However the depolarization product provided by our MPL system (MPL-4B-IDS Series) is the one given in our Eq. (3), which coincides with the linear depolarization ration taking into account that $\delta_{MPL} = P_{cr} / P_{co}$ and Eq. (1.6) of Flynn et al. (2007).

5. Page 6 line 20: the authors should explain how to decide a threshold for estimating the vertical height of pollen plume. There will be better if the vertical height is estimated based on particle backscatter coefficient and depolarization ratio simultaneously.

Authors’reply: Using β_{pol} to extract the pollen top height is an estimation already based on the consideration of both the particle backscatter coefficient and the particle depolarization ratio (since both parameters are needed to retrieve β_{pol}). The value of the threshold was selected so that the integral of $\beta_{pol}(z)$ up to h_{pol} represents at least 99 % of its integral over the whole column. It has been indicated with a new sentence at the end of Section 2.3: “This empirical threshold guarantees that the integral of $\beta_{pol}(z)$ up to h_{pol} represents at least 99 % of its integral over the whole atmospheric column.”.

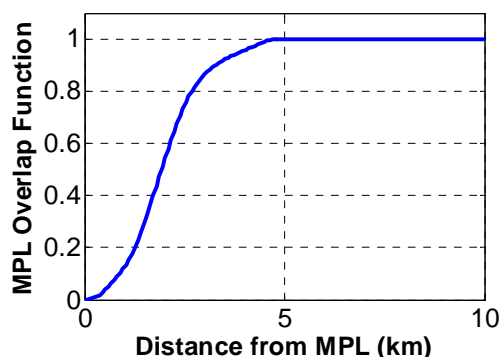
6. A peak of particle backscatter coefficient is always found at near surface (<300m), but not for depolarization ratio profiles. Overlap correction is very important before retrieval of lidar observation data, especially within boundary layer. So probably the correction is not proper, then cause this problem. Please carefully check processing of lidar data.

Authors’reply: The referee is right that overlap correction is very important. In particular for the MPL system which has a full overlap at a distance > 3 km, it is indispensable to make such

a correction if one wants to study the low troposphere. All MPL profiles shown in the paper are obviously corrected from the overlap. The overlap function used is shown in the figure thereafter. As one can appreciate, it is a very smooth function which can not produce artefacts or artificial sudden changes in the corrected lidar signals, so that we do not believe that the variation below 0.5 km are due to the overlap correction.

We had a careful look at the overall MPL data pre-processing to verify if one of the pre-processing procedures could introduce artefacts in the lidar profiles. Besides checking the overlap correction, we looked at the background subtraction, and the deadtime and afterpulse corrections. The background signal is calculated with background “bins” acquired before the laser emission. These “bins” are clearly visible before the afterpulse peak on the raw returned powers, and are guaranteed to be free of any laser-induced atmospheric signal. The deadtime correction modifies very, very little the signal in the first few hundred meters. With the naked eye, this modification is undetectable. The profile used to correct from the afterpulse effect has values different from the background signal only for heights below 100 m, and for this reason, like the overlap correction, it could not explain the differences commented by the referee.

We believe that the increase of the backscatter coefficient below 0.5 km is actually real and reflects the increase of the aerosol concentration in the surface layer in Barcelona. The fact that the depolarization ratio is not varying as much as the backscatter coefficient is not surprising: the depolarization ratio is a semi-intensive parameter (while the backscatter coefficient is an extensive parameter) and if the main contributor to the depolarization, pollen, is well mixed and its contribution to the total aerosol load remains stable, it is to be expected that the depolarization will stay approximately constant with height. Now, the fact that the backscatter coefficient is varying while at the same time the depolarization ratio is not indicates that the increase of the backscatter coefficient is due to an equal relative increase of the concentration of both pollen and non-pollen particles.



7. Page 7 line 3: add “a day” to the end of “: : pollen and fungal spore per cubic”.

Authors’reply: It has been corrected.

8. Page 8 line 15: use abbreviation at the first time, “relative humidity (RH) and temperature (T)”.

Authors’reply: Abbreviations RH and T have defined the first time they appear in the text.

9. Figure 3: why does the total pollen concentration peak precede the AOD peak of pollen? In general, high particle concentration and RH cause large AOD. However, on 31 March maximum pollen concentration and AOD were found at 3 UT and 15 UT, respectively. RH is very close but large differences between total pollen concentration (2 times). Please explain the reason.

Authors’reply: Referee#1 had a similar question about this part of the manuscript. When the pollen is from local origin, not long range transport, the simple idea behind this statement is

that the pollen has to be first released at ground level (concentration peak) before it disperses in the atmosphere (AOD peak). Our rationale is true only if pollen of local origin is considered, not long-range transport. This condition has been added in the text.

We have changed the two sentences starting by “As expected ...” at the end of Section 3 by: “As expected, every day the AOD_{pol} peak follows the total pollen concentration peak. Logically, in the case of pollen of local origin, not long-range transport, a peak of the amount of pollen in the atmosphere (parameterized by AOD_{pol}) can only happen if previously a strong release of pollen at the ground level (parametrized by the pollen concentration) has occurred”.

10. Section 4: It is very important to fix a depolarization ratio of pollen when estimate its contribution ratio and backscatter coefficient. The authors reference results reported by other researchers. However, depolarization ratios are also affected by ambient RH. So please consider the factor and discuss uncertainty of contribution ratio and backscatter coefficient caused by artificially decided depolarization ratio.

Authors’reply: We have assessed the impact of the uncertainty of δ_{pol} on CR_{pol} . A new paragraph has been added following the discussion on the selection of δ_{pol} in Section 4. The new paragraph is:

“To assess the impact of the uncertainty of the assumed pollen depolarization ratio, δ_{pol} , on the uncertainty of the contribution ratio of the pollen to the total particle depolarization, CR_{pol} , we consider an error $\Delta\delta_{pol}$ in the pollen depolarization ratio and write:

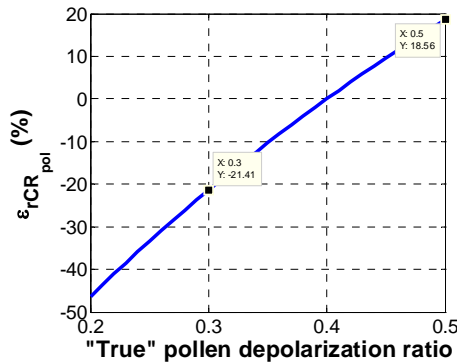
$$CR_{pol}(z) + \Delta CR_{pol}(z) = \frac{\delta^p(z) - \delta_{no-pol}}{1 + \delta^p(z)} \frac{1 + \delta_{pol} + \Delta\delta_{pol}}{\delta_{pol} + \Delta\delta_{pol} - \delta_{no-pol}}, \quad (1)$$

from which the relative error in CR_{pol} , $\varepsilon_{rCR_{pol}} = \frac{\Delta CR_{pol}}{CR_{pol}}$, can be found as:

$$\varepsilon_{rCR_{pol}} = -\frac{(1 + \delta_{no-pol})\Delta\delta_{pol}}{(1 + \delta_{pol})(\delta_{pol} - \delta_{no-pol} + \Delta\delta_{pol})}. \quad (2)$$

We have calculated $\varepsilon_{rCR_{pol}}$ for various values of the “true” pollen depolarization ratio ranging from 0.2 to 0.5, when $\delta_{no-pol} = 0.03$ and $\delta_{pol} = 0.4$ is assumed. In the range $-0.1 < \Delta\delta_{pol} < +0.1$ (i.e. $0.3 < \text{“true” pollen depolarization ratio} < 0.5$) the contribution ratio error is limited to $\pm 20\%$ approximately.”

For the referee we are attaching thereafter the figure (not shown in the paper) showing the error $\varepsilon_{rCR_{pol}}$ as a function of the “true” pollen depolarization ratio ranging from 0.2 to 0.5, when $\delta_{no-pol} = 0.03$ and $\delta_{pol} = 0.4$ is assumed. Datatips indicate that for a “true” pollen depolarization ratio ranging 0.3 – 0.5, $-20 < \varepsilon_{rCR_{pol}} < 20\%$ for an assumed $\delta_{pol} = 0.4$.



11. Page 11 line 21: The AOD is not reliable from lidar data, by integrating the profile of backscatter coefficient in the whole column and multiplying the assumed lidar ratio. Why do not use AOD from co-located AERONET sun-photometer?

Authors'reply: The referee is right: in general the AOD obtained from the integral of the backscatter coefficient profile multiplied by an assumed lidar ratio is not reliable. We can unfortunately not use co-located AERONET measurements to constrain the lidar inversion because the Barcelona AERONET sun-photometer was not in operation at the time of the pollination event. This is the reason why we had to choose a constant lidar ratio.

The selection of a proper lidar ratio (50 sr) was made given the time of year considered (March) and the type of aerosol observed (pollen). As explained at the end of Section 2.2, 50 sr falls in the range of the mean columnar lidar ratios, 46 – 69 sr, found in Barcelona during the period from February to April and calculated over a period of 3 years by Sicard et al. (2011). In that work the columnar lidar ratio was retrieved with the two-component elastic lidar inversion algorithm constrained with the aerosol optical depth from a sun-photometer (like the referee is suggesting to do in the present work). The second reason, and probably the most grounded one, is that Noh et al. (2013b) found a mean columnar lidar ratio of 50 ± 6 sr during a 6-day pollination event (mostly dominated by *Pinus* and *Quercus* pollen) in South Korea by using the two-component elastic lidar inversion algorithm constrained with the aerosol optical depth from a sun-photometer.

12. Page 17 line 31: add “are” to “: : lidar systems (with at least two channels) are able to produce continuously profiles: : :”.

Authors'reply: The verb of this sentence is a little further. The full sentence is “First, relatively simple polarization-sensitive lidar systems (with at least two channels) able to produce continuously profiles of the volume depolarization ratio are very attractive tools for modellers to validate their pollen concentration forecasting models and/or perform data assimilation”, and we believe it is correct as is.

13. Figure 2: please use particle depolarization ratio rather than volume depolarization ratio.

Authors'reply: To retrieve the particle depolarization ratio, one needs to invert the lidar signal to obtain the profile of either the backscatter or the extinction coefficient. With the Micro Pulse Lidar, which is a low energy system, such inversions typically require to average during periods of time of 1 hour. The visualization of the volume depolarization ratio with a 5-min. resolution, in opposition to 60-min. particle depolarization ratio, is an excellent way of showing qualitatively the pollen day-to-day evolution during the whole pollination event. The 60-min. profiles of the particle depolarization ratio are shown anyway in Figure 4. For these reasons, Figure 2 has been kept as a function of the volume depolarization ratio.

14. Figure 4: Too many, hard to get the points. Please 1) remove all total backscatter coefficient and volume depolarization ratio, just keep pollen backscatter coefficient and particle depolarization ratio; 2) only plot 3-4 panels per day, 9 panels are too many.

Authors'reply: To gain in readability Figure 4 has been re-formatted as suggested by the referee. We have kept 4 profiles per day, every 3 hours at 9, 12, 15 and 18 UT. The profile of the particle backscatter coefficient has been removed. The profile of the volume depolarization ratio is a key parameter in the paper and for studying atmospheric pollen with a polarization-sensitive lidar system. Our study finally shows that it is almost as sensitive as the particle depolarization ratio to pollen with the advantage that its retrieval (Eq. 3 of the paper) is much more straightforward than that of the particle depolarization ratio (Eq. 4). For this reason we have wanted to keep the profile of the volume depolarization ratio in Fig. 4.

15. Figure 5: Same problem as fig 4, please remove panels of total AOD and volume depolarization ratio, and re-arrange the figure side by side.

Authors'reply: The panel of total AOD was removed from Figure 5. For the same reason given in the answer of the previous comment, the plot of the volume depolarization ratio was maintained in Figure 5.

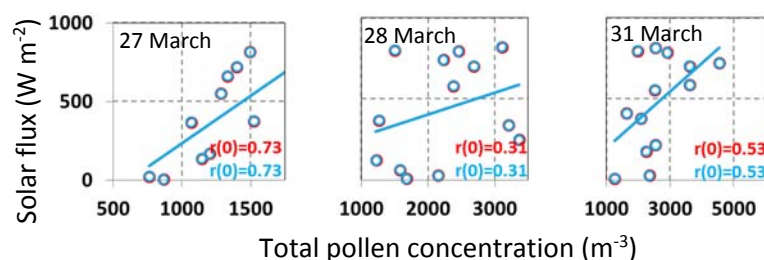
16. Figure 6: Remove the upper panels (pollen concentration vs. volume depolarization ratio).

Authors'reply: For the same reasons given in the answer of the above two comments, we have wanted to maintain the plot of the volume depolarization ratio in Figure 6.

17. Figure 8: Depolarization ratio is used in the figure, but why do not use pollen concentration or backscatter coefficients?

Authors'reply: On request of the referee we have performed the correlation study between solar radiation and total pollen concentration and pollen AOD. In terms of correlation coefficient it is equivalent to use the pollen AOD or the integral of the pollen backscatter coefficient profile since both properties are related by a multiplicative factor. The following figure (not included in the paper) shows the results for the total pollen concentration. The correlation coefficients range between 0.31 and 0.73, and a large scatter of the points is observed. No time delay could be found to maximize the correlation coefficients. These results point out a poor correlation between solar radiation and near-surface pollen concentration.

The correlation study between solar radiation and pollen backscatter coefficient has been included in the revised manuscript: Figure 8 has been updated as well as the discussion at the end of Section 5.



18. A paper about the vertical distribution of Asian dust measured by three MPL Lidars over Northwest China (Huang Z. et al., 2010) was published in JGR. Please reference this paper to increase reader understanding of lidar data retrieval and MPL performance. Furthermore, studies of fluorescent spectrum of atmospheric aerosols from a lidar spectrometer system with high spectral resolution (Sugimoto N. et al., 2012, OE) provides a new tools for investigating vertical structure of biological particles, which will be very useful for readers to understand remote sensing of bioaerosols.

Authors'reply: Both references have been added in the revised manuscript. Thank you for indicating them to us!

Supplement 1: BSC-DREAM8b mineral dust maps at 12UT and Hysplit backtrajectories arriving in Barcelona at 500, 1000 and 2000 m at 12UT

