

## **Review of manuscript with number: acp-2019-289**

Towards continuous monitoring of aerosol hygroscopicity by Raman lidar measurements at the EARLINET station of Payerne

April 30, 2019

### **Summary**

The manuscript describes the retrieval of aerosol hygroscopicity from Raman lidar measurements. Also included is a comparison of mixing ratio, temperature and relative humidity profiles from lidar with those from radiosondes. This is a comprehensive paper of, in my opinion, very high interest to the atmospheric remote sensing community. Therefore, I recommend this work to be published in ACP. However, there are some minor points that need to be addressed before publication. My comments and corrections are given below.

### **General comments**

Most importantly, I'm missing a discussion of uncertainties in the part of this study about the impact of hygroscopicity of aerosols on the radiative budget.

Another aspect to correct is the consistency within this manuscript. Generally it is recommended to use present tense describing established knowledge and previously published work, and for presentation of results (Figure 1 shows ...), and to use past tense describing methods and results, and for referencing (Author X reported ...). I have added comments in appropriate places throughout the manuscript, but it would be helpful to give the finished manuscript to a native English speaker to check the language. There are also inconsistencies in some units, especially altitude measures are given in m and km, and in date formats. In figure captions alone, there are many different date formats (8 of September 2017, 7th September 2017, 3 September 2017, 07 September 2017), please homogenise these throughout the text and captions.

### **Specific comments**

#### **Page 1**

##### **Title**

You do not highlight the automatization of the detection of hygroscopicity. Shown are case studies from automatic and continuous measurements, but the hygroscopicity retrieval was done "manually", as far as I understood. You could crop the title to "Aerosol hygroscopicity from Raman lidar measurements at the EARLINET station of Payerne".

#### **Abstract**

##### **Line 7**

"whole troposphere" is only valid for nighttime retrievals. Please rephrase.

##### **Line 11**

"rural aerosol": You only mention a mix of "local" and smoke particles once in the main text, and once in the conclusion. Elsewhere you only refer to smoke. You could update smoke references in the text to "smoky mix" or similar.

##### **Line 17**

Please see my comment on the use of "significant" in the main text (page 13, line 20).

## **Page 8**

### **Line 2, Figure 1**

Use either "r" or "mixing ratio", and either "T" or "temperature" in the text. After introducing "r" on page 5, line 12, you keep using "mixing ratio". When discussing the results you sometimes use "r", and sometimes "mixing ratio". It should be consistent throughout. Please update occurrences of "r"/"mixing ratio" in the text.

T is first used on page 6, line 8, but is not introduced as meaning temperature, nor used in other parts of the text.

### **Line 31-32**

Am I correct that you averaged the mean bias below 2.1 km asl, where it was positive? In that case, please replace "was +2.0" with "was on average +2.0", and "was -4" with "was on average -4".

## **Page 9**

### **Line 3**

You are using different altitude ranges to average mean bias (2.1 km asl) and standard deviation (2.0 km asl) of the RH nighttime comparison, and yet different when looking at temperature and RH daytime (5 km asl). The only reason I can see, is that the mean bias of RH changes from positive to negative at 2.1 km asl. Please provide justifications for your choice of altitudes in the other two cases.

### **Line 6**

You refer here to "errors". What are the errors/uncertainties of RS and lidar profiles? It is not specified. Or do you mean the difference between the profiles?

### **Line 30**

Please explain NAAPS and add reference.

### **Line 32**

I don't think that the lowest trajectory (arriving at 566 m agl, red) indicates air mass origin. It is too close to the ground throughout its journey to allow any conclusion on air mass origin.

## **Page 10**

### **Figure 5**

You show, but do not discuss PM10. Either remove this panel from the figure, or discuss what is shown.

### **Line 22**

Why are potential temperature and mixing ratio auxiliary information? Besides, you sometimes use "temperature" and sometimes "potential temperature". Please be clear and specific, when which is used.

### **Line 34-35**

Referring to "indicating a lower sensitivity of this wavelength to the aerosol hygroscopic growth": Do you mean this applies to this case? Or generally? Would it depend on the aerosol type?

## **Page 11**

### **Line 2**

Please rephrase the sentence starting with "This parameter ...". I don't understand, especially the last part "observed in the aerosol property".

**Line 6**

What do you mean with "consistent"? The trend is the same (lower at longer wavelength), but the values are different. How large is uncertainty or variability of the Hänel parameter? This information would help to judge, what falls into the "consistent" range.

**Line 21**

You write that backscatter is sensitive to wavelength and particle size, **as expected**. This contradicts somewhat the first sentence of this paragraph. Please explain, or rephrase.

**Line 22**

You write, Mie scattering regime is expected for  $x \approx 1$ . However,  $x$  is as large as 7.1 in figure 8d. Please discuss how representative or applicable your Mie simulations are in such conditions, especially for wavelength 355 nm.

**Page 12****Line 10**

Out of curiosity, did you also look at dehydration along the vertical?

**Page 13****Line 12**

Did you try this on night-time measurements using directly the Raman lidar extinction profiles? Please briefly discuss here or below, what impact the choice of LR has on AOD, ARE and FE.

**Line 15**

Is AOD in table 1 columnar AOD from the full integrated lidar extinction profiles (which height range?), or just layer AOD? As the only change (according to figure 13) occurs in one layer per case, it would also be useful to see these values for the layers only. Please discuss if it is reasonable, that there is no hygroscopicity in other altitude regions. (See my comment on figure 13)

**Line 17**

You wrote it in the table title. Please also add here a short remark that the relative values are relative to dry values.

**Line 20**

It is hard to judge significance, if no uncertainties are provided. Please specify what you mean with "significant".

**Table 1**

Please provide an estimate of uncertainties, either in the table or in the text.

**Line 31**

Please add a reason, why you chose to optimise solar zenith angle for Case 1 rather than Case 2. I assume it's because the hygroscopic effect was stronger in Case 1, but it is not mentioned.

**Page 14****Line 10**

Does this not contradict the previous sentence? It sounds to me, that it would not be necessary. As long as AOD and ARE are right, FE is not sensitive to hygroscopicity. Please elaborate, I cannot follow this statement.

## Page 23

### Figure 3 (and figure 11 a)

Please consider using a different colour map, for reasons outlined here among other sources: [https://www.mathworks.com/tagteam/81137\\_92238v00\\_RainbowColorMap\\_57312.pdf](https://www.mathworks.com/tagteam/81137_92238v00_RainbowColorMap_57312.pdf) (see page 3 for a brief overview). I find cubehelix a good replacement (or the reversed version of cubehelix, from light to dark colour).

## Page 26

### Figure 7

Did you use instantaneous lidar measurements here, or again the average from 15:00 to 15:30? Please specify.

## Page 27

### Figure 8

Is the size parameter  $x$  a result of the simulations, or was it an input?

## Page 29

### Figure 11

Show plot a) as separate figure.

Increase font size in plot b). Did you create plot 11b? If not, add source. Possibly like: "Adapted from [source]". If you plotted the model output, please keep style consistent with other figures in this manuscript.

Remove legend in plot c), or add meaningful label.

In the caption, replace "Quicklook of  $r$ , RH and backscatter coefficient at 355 nm from Ralmo measurements on" with "Same as figure 3, on". Avoid "quicklook", it's too specific to the lidar community.

## Page 30

### Figure 13

Those profiles suggest that there was no hygroscopic growth in other regions along the profile. How realistic is that? I think it would be better to, for example, plot the full wet profile as faint line, the wet profile within the studied layer as bold line, and the dry profile within the layer as dashed line, but without connecting it to the wet profile.

## Technical corrections

### Page 1

Line 14 replace "is" with "was"

Line 16 be specific about "this type of aerosol"

### Page 2

Line 4 remove "altering also in this way", and rephrase the following part of this sentence to "also altering the global radiative budget (indirect effects)(...) in this way."

Line 9 replace "has been" with "was";  
remove "in order"

Line 10 replace "kind" with "kinds"

Line 12 replace "as" with "like"

Lines 15-16 keep either "still" or "yet", remove one of them

Line 17 replace "RH" with "relative humidity (RH)";  
add comma after "water"

Line 32 rephrase sentence: “remote sensing [...] since it”, or “remote sensors [...] since they”,  
or “remote sensing techniques [...] since they”

Line 33 it is not clear what “this technique” refers to

**Page 3**

Line 3 replace “RS” with “radiosondes (RS)”

Line 10 please explain what “r” is

Line 29 replace “telescope” with “telescopes”

**Page 4**

Line 4 replace “to” with “into”

Line 11 replace “have been” with “were”

Line 13 “temperature” appears twice

**Page 5**  
no comments

**Page 6**

Line 23 replace “the particle” with “particles”

Line 28 replace “defined” with “fixed / constant”

Line 29 replace “are” with “were”

Line 30 replace “need” with “needed”

**Page 7**

Line 1 remove “the”;  
add comma after “that”

Line 3 replace “use” with “used”;  
replace “the profiles” with “profiles of”

Line 4 replace “are” with “were”

Line 6 replace “have been” with “were”

Line 11 replace “An other” with “Another”

Line 12 remove “have”

Line 25 duplication of “of”

Line 30 add comma after “critical”

**Page 8**

Line 1 replace “RSs” with “RS”;  
replace “allows” with “allowed”

Line 10 add comma after “retrievals”;  
replace “has been” with “was”

Line 12 replace “has been” with “was”;  
replace “treated” with “discussed”;  
change format of citation to “(Martucci et al., in preparation)”

Line 13 add comma after “UTC”

Line 19 add comma after “UTC”

**Page 9**

Line 1 remove “the”

Lines 7-10 I find this difficult to follow. Please rephrase. Commas are your (and your readers')  
friends!  
For example: “In any case it is important to point out the good accomplishment of  
Ralmo in retrieving RH information. This can be concluded from this intercomparison,  
in which Ralmo showed very small biases and standard deviations (below 9%RH),  
which are indicative of accuracy and precision, respectively, of our measurements.”

Line 12 add comma after “studies”

Line 15 add comma after “2017”;  
rephrase the rest of the sentence, for example: “...with smoke particles present during  
one, and mineral dust during the other.”

Line 16 replace “in” with “of”

Line 18 replace "pannel" with "panel"  
Line 19 replace "planetary" with "atmospheric"  
Line 20 replace "PBL" with "ABL", and change throughout the manuscript  
Line 21 replace "pattern" with "development";  
replace "along" with "during"  
Line 24 replace "is" with "was";  
replace "pannel" with "panel"  
Line 25 replace "for" with "during" or "in"  
Line 31 add "(blue colour map)" after "concentrations";  
replace "over" with "at" (you refer to surface concentration)  
Line 32 replace "from ground to 3 km asl" with "from 1 to 3 km asl"

#### Page 10

Line 2 replace "for" with "in"  
Line 10 remove "burning", this is already covered by "combustion"  
Line 12 Please specify "that days" (or rather "those days"). Do you mean "in the period from 6th to 8th September"?  
Line 14 replace "concentrations" with "concentration";  
replace "were" with "was";  
remove "than"  
Line 15 replace "took place" with "occurred"  
Line 16 add comma after (AAE)  
Line 20 replace "of" with "over"  
Line 24 replace "is" with "was"  
Lines 24-25 remove either "Simultaneously to this increase," or "for the same layer";  
remove "that there is also";  
replace "can observe" with "observed";  
replace "moving" with "increasing"  
Line 28 add comma after "respectively");  
replace "is" with "was"  
Line 30 replace "have been" with "were"  
Line 31 add comma after "RH";  
remove "what is"  
Line 34 replace "respect its" with "with respect to its"

#### Page 11

Line 1 replace "took a value of" with "was"  
Line 11 replace "have been" with "was"  
Line 13 remove "have"  
Line 21 replace "how" with "that";  
remove 2<sup>nd</sup> "the";  
replace 2<sup>nd</sup> "and" with comma  
Line 22 add: "where D is particle diameter" after the equation  
Line 29 remove "totally"  
Line 32 add comma after "cases";  
remove "also means that we"  
Line 33 replace "the theory of Mie" with "Mie theory";  
replace ";" with ".";  
replace "so" with "Hence," (start new sentence)

#### Page 12

Line 2 replace "For" with "In";  
replace "we can observe that r (Fig.9, top) is quite constant (...)" with "we can observe that r was quite constant (... , Fig. 9, top)"

- Line 4 remove "The"
- Line 6 remove "again"
- Line 9 remove "very"
- Line 10 replace "suffered by this aerosol" with "that occurred within this aerosol layer"
- Line 12 remove "very";  
remove "either"
- Lines 14-15 it sounds like you used a hygroscopic parameter of 0.4 for the later period (16:00 to 23:30); please rephrase;  
remove "also" in line 15
- Line 21 replace "pannel" with "panel"
- Line 22 explain acronyms "NMMB" and "BSC"
- Line 23 add comma after "Europe"
- Line 24 replace "in" with "for" or "above"
- Line 27 replace "in our station" with "at our station"
- Line 29 replace "along" with "throughout"
- Line 30 replace "along" with "during" or "throughout"
- Line 31 remove parentheses and include as full sentence
- Line 32 replace "is" with "was";  
add comma after "measurements";  
replace "analyze" with "analyzed"
- Line 33 replace "observe" with "observed"
- Page 13**
- Lines 5-6 put parenthesis and dot on same line as 0.24 (remove space?)
- Lines 6-7 rephrase, for example:  
As in case 1, we found the opposite spectral dependency compared to Lv et al. (2017).  
However, we considered a wider spectral range.
- Line 10 remove "also";  
remove "the"
- Line 11 remove "the", twice in front of "AOD"
- Line 13 remove first "the"
- Line 18 replace "16.6" with "15.6" (check if table 1 or text is correct);  
remove first "the"
- Line 23 remove "the"
- Line 24 remove first occurrence of "model"
- Line 28 add "and relative" after "absolute"
- Line 31 remove "the one of"
- Page 14**
- Line 13 replace "monitor" with "observe" or "detect"; "monitor" sounds more like an automated process
- Line 14 replace "in almost a continuous way" with "almost continuously"
- Line 16 remove "the";  
replace "the particle" with "particles"
- Line 17 add comma after "growth";  
replace "have been" with "were"
- Line 21 replace "along" with "throughout"
- Line 27 replace "in the full" with "throughout the"
- Line 33 swap order of sentence: "...in the lower troposphere (...) were very similar to the ones obtained during nighttime."
- Page 15**
- Line 14 add: "as well as the study of Haarig et al. (2017)"
- Line 20 remove "very"

- Line 21      add comma after “2017”);  
 add comma after “particles”
- Lines 26-27    remove first “the”;  
 remove “It”;  
 move “was also remarkable” to the end of the sentence
- Line 28      replace “has been” with “was”
- Line 30      remove first “the”
- Line 31      add “and relative” after “absolute”
- Line 32      please see specific comment for page 13, line 20
- Line 33      replace “was not” with “not having been”;  
 add comma after “Therefore”
- Line 34      replace “have” with “has”
- Page 16**
- Line 6      replace “AA” with “AH”?
- Page 21**
- Table 1      replace “layer width” with “layer depth”
- Page 22**
- Figure 1      start caption with: “Mixing ratio (r), temperature (T) and relative humidity (RH) [...]”
- Figure 2      add date after “UTC”
- Page 23**
- Figure 3      replace “of” with “of vertical profiles of”;  
 In the caption “r” is called “water vapour mixing ratio”, and in the figure “WVPR”.  
 Please amend figure and caption to “mixing ratio” and “r” to keep it consistent  
 throughout the manuscript and to avoid confusion.
- Page 24**
- Figure 4      replace “Total Optical Depth” with “total optical depth from sulfate (orange/red scale),  
 dust (green/yellow scale), and smoke (blue scale)”;  
 possibly replace “1500, 2500, 3500 m asl” with “1000, 2000, 3000 m asl”; Payerne  
 being roughly at 500 m asl. 3500 m asl is not consistent with the text either (page 9,  
 line 32).
- Page 26**
- Figure 6      explain in caption what the grey shaded area means;  
 add "temperature" after "potential"
- Page 27**
- Figure 8      increase font size in legends
- Page 28**
- Figure 9      add “of” after “Evolution”
- Figure 10     replace “Hannel” with “Hänel” in figure legends
- Page 29**
- Figure 12     replace “pannel” with “panel” (3 times in this caption);  
 increase size of the three panels on the left; font sizes in left and right plot should be  
 the same