We thank all the reviewers for their comments and suggestions that have helped to improve the quality of the manuscript. A point by point response to the reviewers’ comments is included below. Reviewers’ comments are noted in bold. Changes in the manuscript are noted between quotation marks. The new version of the manuscript with the changes tracked is included at the end of this file.

**Anonymous Referee #1**

Comment: This paper addresses an evaluation of the aerosol property profiles retrieved from GRASP algorithm and which uses as inputs ceilometer and sun-photometer (SPM) measurements versus in-situ measurements. The work presents different relevant aspects that show its importance and novelty. This is the first time that GRASP algorithm using as inputs ceilometer and SPM measurements (GRASPpac) has been evaluated in a long-term comparison. This new approach (GRASPpac) presents big advantages since these two instruments can be operated in a continuous and almost unattended way and its use has been expanded by networks providing much more global coverage. However, before this approach is widely used its products need to be validated as is done in this work. In addition the work have dealt with the complexity of comparing different techniques (remote and in-situ) which also cover different ranges in the Earth-atmosphere system (surface and almost full troposphere). The results presented here show a good agreement between the optical properties from techniques and larger discrepancies in the volume size distribution when fine particles are dominant. So after these comments I conclude that the paper is very interesting, well written and show the capability of GRASPpac approach to retrieve vertical information of aerosol properties based on this long-term study. I consider that this work is appropriated for Atmospheric Measurement Techniques and it should be published after some minor corrections:

About the comparison: some explanations should be given about how the in-situ measurements and the GRASP profiles are compared. How many points from the lowest part of the profiles do you take? Do you average those points? What is the altitude range that they represent? The lowest part of the remote sensing profiles are always more problematic due to incomplete overlap issues, and this might affect the comparison. However, since the overlap of the telescope and laser beam is greater than 85% at the height of the MSA station (1570 m a.s.l.), this effect is expected to be low. We have discussed these points with more detail in the revised version of the manuscript.

Answer: We agree with the reviewer that this explanation is missing in the manuscript. Since the sun/sky photometer is located at the Montsec observatory (1570 m a.s.l.) and the ceilometer is located at 800 m a.s.l., the combination of RCS from the ceilometer and sun/sky photometer measurements in the GRASP algorithm is only possible from 1570 m a.s.l. and onwards. The ceilometer RCS used as input in GRASP is normalized and averaged at 60 log-spaced points, being the first point (1/60) equal to 1570 m a.s.l. Therefore, the GRASP-derived profiles start at 1570 m a.s.l. The comparison with the in-situ measurements is therefore made at 1570 m a.s.l. and consequently the comparison is only representative of this altitude (we cannot assure that the results of the comparison are similar at higher or lower altitudes since the in-situ measurements are limited to a single point). Also, we agree with the reviewer that the lower part of the remote sensing profiles are always more problematic due to incomplete overlap issues, and this might affect the comparison. However, since the overlap of the telescope and laser beam is greater than 85% at the height of the MSA station (1570 m a.s.l.), this effect is expected to be low. We have discussed these points with more detail in the revised version of the manuscript.

Section 2.4.: It has been modified as follows: “The ceilometer is located at 800 m a.s.l., at the Center for the Observation of the Universe (COU, http://www.parcastronomic.cat/). The horizontal distance between the ceilometer and the MSA station is less than 2.5 km.”
Section 4.1.1.: We have included: “The comparison has been performed at 1570 m a.s.l., where the in-situ instrumentation is located and coinciding with the first height of the GRASPpac retrievals. Therefore, the following results and associated discussion on the comparison between GRASPpac and in-situ measurements refer exclusively to this height.”

Section 2.4: we have modified it as follows: “The RCS profiles provided by the instrument are overlap-corrected using the manufacturer’s overlap function. In addition, according to this function, the overlap of the telescope and the laser beam is greater than 85% at around 770 m a.g.l. Thus, the effect of the overlap at the height of the MSA observatory (1570 m a.s.l.) is expected to be low.”

Section 3: we have included: “As mentioned before, the RCS profiles provided by the ceilometer are overlap-corrected and, according to the manufacturer’s overlap function, the overlap of the telescope and the laser beam is greater than 85% at the MSA altitude (770 m above the ceilometer). Thus, the effect of the overlap in the GRASPpac retrievals is expected to be low, since the ceilometer RCS below 1570 m a.s.l. is not used here as input in GRASPpac.”

Comment: Looking at the histograms presented in Figure 2 I have the impression that the distributions of the relative differences are bounded to a certain positive value, how do you explain that there are no observations with discrepancies larger than +1%?

Answer: Histograms presented in Figure 2 show the relative difference between in-situ measurements and GRASP derived optical variables, taking the in-situ measurements as reference (calculated as the difference between in-situ and GRASPpac values divided by the in-situ value). We agree with the reviewer that the relative difference seems biases to lower positive relative errors. To avoid this, we have replaced relative difference by absolute difference (in-situ minus GRASPpac values) in Figure 2 and Figure 4, as well as the related discussion in the manuscript. We think that these new figures provide a better idea on the difference between GRASPpac and in-situ measurements.

Comment: Page 2, line 30: it should be also indicated that ceilometer provides continuous measurements, in contrast with most of the “more sophisticated” lidar systems.

Answer: Done.

Comment: Page 4, line 22: I wonder why you use Aeronet data level 1.5. For this long-term study level 2 data (quality assured) should be available.

Answer: We use Level 1.5 data because typically Level 2.0 data is not available after one year, when the post-calibration of the photometer is done. GRASPpac has the potential to derive aerosol properties in near real time if it is feed with Level 1.5 Aeronet data, but not if Level 2.0 data is used. Hence, we think that the study of the performance of GRASPpac feed with Level 1.5 data is more interesting than with Level 2.0 due to the near real time implications.
Comment: This paper entitled "Retrieval of aerosol properties from ceilometer and photometer measurements: long-term evaluation with in-situ data and statistical analysis at Montsec (southern Pyrenees)" provides a very detailed statistical assessment of a fairly recent algorithmic approach entitled GRASP (Generalized Retrieval of Aerosol and Surface Properties) to optimize the vertical retrieval of aerosol properties by merging vertical profile data with constraining column data from a sun/sky photometer. The novelty of the statistical comparison is that it is done using insitu data obtained from the Montsec Observatory which is _ 750 meters above the ceilometer profiling instrument which allows the authors to study the retrieval performance above the critical overlap region which even with correction generally leads to significant biases that would be enhanced by the multi-instrument retrieval. Another novelty of the paper is the long duration of the study (3 years) which allows the authors to study the effects of different meteorology and aerosol sources on the results. Based on the authors literature background, this is a significant improvement over existing algorithm retrieval validations which were limited to short duration aircraft campaigns.

Besides quantifying the retrieval characteristics and identifying conditions in which the retrieval performances is degraded (fine mode dominated), the authors are able to build a very useful profile climatology of vertical aerosol properties filtered by climatology (RH) and source locations.

In summary, the paper illustrates convincingly the usefulness of the GRASP algorithm in optimally analyzing combined Ceiometer / Sky Radiometer data. The fact that a much cheaper ceilometer is used instead of more costly and sophisticated Lidars is very useful since it opens up the possibility of developing a much larger network of such instrument sites.

Suggestions:

One suggestion that comes to mind would be to illustrate the importance of the radiometer (i.e radiance constraints) on the retrieval properties. In particular, prior algorithms that combine lidar (or ceilometers) with only total column Aerosol Optical Depth at multiple wavelengths from a sunphotometer has been used to retrieve vertical aerosol profiles. The sun photometer AOD measurements may provide an even cheaper alternative to an instrument site. Therefore, it would be very helpful to the community if some comparison between the different algorithms could be made to see how much improvement there is in the AERONET Radiometer data as compared to just sunphotometer AOD constraints on this very nice and rich data set.

Answer: We thank Prof. Gross for his positive comments on this manuscript. The GRASP retrieval using only AOD and ceilometer signal is an interesting suggestion that looks very promising since sky radiance measurements are scarcer than AOD measurements. In line with this comment, Torres et al. (2017) explored the use of multi-wavelength AOD measurements to retrieve column integrated size aerosol properties. The versatility of GRASP algorithm is promoting the use of different set of data as inputs such as sky radiances from sky cameras combined with lunar photometers (Román et al., 2017) or scattering measurements from polar nephelometers to retrieve aerosol optical and microphysical properties (Espinosa et al., 2017). In this study we focus on the evaluation of the inversion scheme proposed by Román et al. (2018) that uses sun/sky photometer and includes the RCS from ceilometer as novelty. We are currently exploring other inversion strategies and we will consider the use of only AOD as suggested by the reviewer. However, we feel that this would be out of the scope of the present manuscript in which the main objective is to evaluate the GRASPPac inversion strategy.
References:


Anonymous Referee #3

Comment: The paper is an interesting contribution to aerosol remote sensing as it highlights the potential of automated ceilometers being available in large numbers (networks operated by national weather services). The limitation in characterizing aerosols is caused by the low power and the single wavelength compared to advanced lidar systems. However, the very good spatial and temporal coverage is a big advantage (unattended continuous operation). To overcome the above mentioned limitation the joint exploitation with photometer measurements is proposed (by means of GRASPpac) – a validation of this approach is provided in this manuscript. Moreover, long-term observations of aerosol profiles (in terms of extinction and derived by the novel GRASP-approach) are presented. The paper is clearly structured, well written and relevant. There are several promising applications: the benefit of successful retrievals of aerosol profiles (backscatter or extinction) with high temporal resolution (as described in this paper) could be enormous for model validation: up to now validation is mainly confined to surface values (PM10, PM2.5) or columnar values (AOD), e.g. in the framework of AQMEII. Consequently, I recommend publication in AMT. Only minor changes are suggested. Most of them are just details in wording – nevertheless a few clarifications to avoid possible misunderstandings are strongly recommended.

In the following page and lines are given in square brackets

Comment: [2,23] "Aerosol Optical Depth". I would suggest to use lower case letters.

Answer: Done

Comment: [2,24] "range corrected signal (RCS) lidar values..." -> "range corrected lidar signal (RCS)"

Answer: Done

Comment: [2,29] "...the GRASP algorithm is a significant advance...". Maybe change "is" to "can be", as the (positive) result of this paper is not yet known when reading the introduction.

Answer: Done

Comment: [2,31] I would be cautious with the word "worldwide" (see also the abstract): There are quite different ceilometers in operation and it is not yet clear whether the conclusions of this paper (found for one CHM15k) can be transferred to other systems: Many ceilometers have a lower pulse energy (consequently a limited measurement range) than the CHM15k (e.g., CL31, CHM8k), are influenced by water vapor absorption (CL31, CL51, CHM8k) and dense networks do not exist in all parts of the world. It is not unlikely that GRASPpac can be applied to some other systems as well, but to my knowledge this has not yet been demonstrated. This topic should briefly be discussed (maybe in the conclusions). There are certainly publications on these issues that might be useful. See also comment on [5,14].

Answer: We agree with the reviewer. We have omitted the word “worldwide” in the manuscript. Also, we have included in the conclusion section a brief discussion on the applicability of GRASPpac to other ceilometers with lower capabilities than the one used here.
“Nevertheless, it is important to bear in mind that the results presented in this study are limited to day time and low cloudiness conditions due to the need of simultaneous sun/sky photometer measurements. Also, further studies investigating the performance of the application of GRASPpac to ceilometers and automatic lidars with different characteristics (i.e. wavelength of operation, pulse energy) than the one used in this study are needed to maximize its potential application. With this in mind, the implementation of GRASPpac in the frame of measurement networks will contribute to enhance the representativeness of aerosol vertical distribution providing useful information for satellite and models evaluation, and contributing to the objectives of several international initiatives (Illingworth et al., 2018) such as the EU COST Action TOPROF (Towards operational ground-based profiling with ceilometers, Doppler lidars and microwave radiometers for improving weather forecasts) or the E-PROFILE program of the European Meteorological Services Network.”

Comment: [3,1] I agree that the methodology presented in this paper can be a step forward, however, there are other options for quantitative aerosol remote sensing (including night time measurements). In the last years several publications have demonstrated that ceilometers can provide the particle backscatter coefficient (under certain atmospheric conditions, see e.g. Wiegner and Geiß, 2012). They use a different approach than Titos et al. by using calibrated ceilometer data and get an uncertainty in the order of 10% for the backscatter coefficient. How does this compare to GRASPpac (same order of magnitude?)? See also comment on [8,19].

Answer: We certainly agree with the reviewer and believe that a discussion on previous studies retrieving quantitative aerosol optical information (i.e. backscatter coefficients) from ceilometers' RCS signal is missing in the manuscript. We have included this discussion in the introduction and conclusion sections, with appropriate references.

“The use of ceilometer measurements in the GRASP algorithm can be a significant advance towards a better representation of aerosol properties with vertical resolution since ceilometers are cheaper, require less supervision, provide continuous measurements and are more extensively distributed compared to more sophisticated lidar systems (Wiegner et al., 2014; Cazorla et al., 2017; Dionisi et al., 2018). However, the main drawback of this approach is that sun/sky photometer measurements are only available during day time and under low cloudiness conditions. Other methodologies, such as the absolute calibration of the ceilometer (Wiegner and Geiß, 2012), are able to overcome this issue and provide quantitative backscatter profiles during day and night time. Quantitative ceilometer profiles could be used for evaluating dust forecast models (Tsekeri et al., 2017) such as the BSC-DREAM8b, as input to radiative transfer models (Granados-Muñoz et al., 2019) or can be assimilated in global models (Chen et al., 2018). This application represents a step-forward in the classical use of ceilometers that were originally developed for cloud base detection (e.g., Martucci et al., 2010; Wiegner et al., 2014).”

Concerning the uncertainty of GRASPpac retrieved backscatter coefficient, Román et al. (2018) estimated a mean uncertainty of 31% analyzing synthetic data under various scenarios. This estimated uncertainty is lower for the volume concentration and the extinction coefficient (21%). We have included the uncertainties in the revised version of the manuscript, and a comment on the lower uncertainty estimated by Wiegner and Geiß (2012) for their calculation.

“Román et al. (2018) estimated the GRASPpac uncertainty analyzing synthetic data under various scenarios. According to these authors, the backscatter coefficient mean uncertainty is 31%, and 21% for the extinction coefficient and the volume concentration. The uncertainty in the backscatter
profiles retrieved with GRASPpac is higher than the estimated uncertainty by Wiegner and Geis (2012) for the absolute calibration method (10%)."

Comment: [4,2] Whenever "backscattering" is used in the paper make clear whether backscattering in the "lidar sense" is meant (i.e. scattering under 180-1; in m-1 sr-1) or "hemispheric backscattering" (in m-1) as measured by the nephelometer. And explain how the comparison is made: From Section 4.1.1 I understand that it is assumed that scattering into the backward hemisphere is the same for all directions. Then, from the integral (scattering angles \(90 \leq \theta \leq 171^\circ\), extrapolated to scattering angles \(90 \leq \theta \leq 180^\circ\)) the authors calculate a mean backscatter coefficient (which – under these assumptions – can also be applied to 180\(^\circ\)) and compare this value with the GRASPpac-retrieval. As scattering under 180\(^\circ\) is typically larger than scattering under smaller angles, I would expect an overestimation by GRASP (see also discussion in [6,6ff]). That was indeed found. As this is one of the main topics of this paper, the authors should be very clear – this might induce an extension of the discussion.

Answer: That is correct, we are assuming that the scattering into the backward hemisphere is the same in all directions. In section 3 we have included the following paragraph explaining how the comparison has been done:

“Since the in-situ measurements and GRASPpac retrievals provide different information with respect to the aerosol backward scattering properties (hemispheric backscattering versus backscatter at 180\(^\circ\)) the direct comparison between both techniques is not possible. To have a sense of the performance of the GRASPpac backscatter retrieval, for the comparison we have assumed that the scattering into de backward hemisphere is the same in all directions. Therefore, we have extrapolated the backscatter at 180\(^\circ\) to the angular range 90-180\(^\circ\) in order to make it comparable with the backscattering coefficient measured with the nephelometer. This assumption constitutes an additional source of error since the actual angular scattering distribution is not known and typically backscatter at 180\(^\circ\) is larger than at smaller angles.”

Comment: [4,5] What is "lpm"? Should be "l min-1"?

Answer: Yes, it is liters per minute. We have changed the notation.

Comment: [4,32] "...using the manufacturer's overlap function." This seems to be for information only, with no consequences for the retrieval as the vertical difference between the ceilometer site and the observatory is 760 m (correct? Or what does "downslope" mean? If the vertical difference is smaller, the overlap issue should be discussed in more detail. On the other hand a horizontal distance of 2.5 km is mentioned.). Or is there another reason for mentioning this? Please avoid confusion of the reader.

Answer: The vertical distance between the ceilometer and the Montsec observatory is 770 m (there was a typo in the original version, and the height difference is 770 instead of 760 meters). The Montsec observatory where the in-situ instruments and the sun/sky photometer are located is at 1570 m a.s.l. and the ceilometer is located at 800 m a.s.l. We have clarified this point in the revised version of the manuscript, avoiding the term downslope that was misleading. On the other hand, the horizontal distance between the two sites is 2.5 km. We have included this information because a short horizontal distance between the two set of instruments will likely guarantee that both are sampling the same air mass, while for longer horizontal distances the probability of
different air masses affecting each site increases. The following information has been included / modified in the revised version of the manuscript:

Section 2.4.: It has been modified as follows: “The ceilometer is located at 800 m a.s.l., at the Center for the Observation of the Universe (COU, http://www.parcastronomic.cat/). The horizontal distance between the ceilometer and the MSA station is less than 2.5 km.”

Section 4.1.1.: We have included: “The comparison has been performed at 1570 m a.s.l., where the in-situ instrumentation is located and coinciding with the first height of the GRASPpac retrievals. Therefore, the following results and associated discussion on the comparison between GRASPpac and in-situ measurements refer exclusively to this height.”

Section 2.4: we have modified it as follows: “The RCS profiles provided by the instrument are overlap-corrected using the manufacturer’s overlap function. In addition, according to this function, the overlap of the telescope and the laser beam is greater than 85% at around 770 m a.g.l. Thus, the effect of the overlap at the height of the MSA observatory (1570 m a.s.l.) is expected to be low.”

Section 3: we have included: “As mentioned before, the RCS profiles provided by the ceilometer are overlap-corrected and, according to the manufacturer overlap function, the overlap of the telescope and the laser beam is greater than 85% at the MSA altitude (770 m above the ceilometer). Thus, the effect of the overlap in the GRASPpac retrievals is expected to be low, since the ceilometer RCS below 1570 m a.s.l. is not used here as input in GRASPpac.”

Comment: [5,12] Please explain what ”normalized” ceilometer RCS means?

Answer: The RCS is normalized at 60 log-spaced bins at different heights, as in Lopatin et al. (2013), being the minimum of these heights (zmin) the MSA altitude (1570 m a.s.l.). The maximum height (zmax) selected for the 60 log-spaced bins is 7000 m above MSA since aerosol layers are rarely detected above this height and the ceilometer signal is usually too noisy at higher altitudes due to the low power of the ceilometer’s laser (please see our next comment about the maximum height). The RCS at these 60 log-spaced bins is normalized by dividing the average of RCS in each logarithmic height interval by the integrated RCS between zmin and zmax. Therefore, the normalized RCS provides a value of 1 when it is integrated. This is done because GRASP algorithm uses normalized signals as input. Further details can be found in Román et al. (2018). For clarification we have included the following information in section 3:

“The minimum height of these 60 values corresponds to the MSA altitude. The maximum height selected for the 60 log-spaced bins is 7000 m above MSA, since aerosol layers are rarely detected above this height and the ceilometer signal is usually too noisy at higher altitudes due to the low power of the ceilometer’s laser. The RCS at these 60 log-spaced bins is averaged and then normalized by dividing each value by the integrated RCS between the minimum and maximum heights.”

Comment: [5,14] “corresponds to the MSA altitude...” So RCS between 760 m and 7760 m (above sea level, distance from the ceilometer < 7 km) are considered in the inversion? Here I would expect a comment on the measurement range of the CHM15k: data are available up to 15 km ([4,27]) but the range that can be exploited is smaller. In the framework of the CeiLinEx2015 campaign it was shown that in the free troposphere the CHM15k signals are quite noisy (the maximum range of Vaisala- and Campbell-ceilometers
is even smaller; this may influence the "worldwide"-discussion from above as well). How does this affect the GRASPpac-retrieval? Is the maximum range (7000 m) reduced, but keeping the 60 levels?

Answer: We agree with the reviewer, the quality of RCS from ceilometers decreases with height. The minimum height of the 60 log-spaced height values corresponds to the MSA observatory altitude (1570 m a.s.l.) and the maximum height has been assumed up to 7000 m above the observatory altitude, therefore up to 8570 m a.s.l. This altitude corresponds with a height of 7770 meters above the ceilometer, therefore, although RCS data from the ceilometer is available up to 15 km, only ~8 km are considered. This value is slightly larger than that considered by Román et al. (2018) in order to reach a height of around 7000 m above the observatory.

In those cases which the RCS is too noisy, the normalized RCS values at high altitude are negative and noisy, and hence, following the criteria of Román et al. (2018), an iterative process is applied to the RCS values where the maximum height (7000 m above the observatory) is iteratively reduced every 100 meters until all the RCS values are positive. This process means that the maximum altitude reached with GRASPpac is not always 7000 m above the observatory, since it could be lower. The number of height-levels is always 60 independently of the maximum height attained.

Comment: [5,20] The authors mention that the retrieved re is height-independent, but never use re in the paper. So it is recommended to mention the volume concentration V instead (or in addition).

Answer: We have clarified this point in the revised version of the manuscript. It is important to keep in mind that GRASPpac assumes that intensive properties (such as the effective radius) does not change with height while it considers changes with height in the extensive aerosol properties (like volume concentration).

“Since ceilometer measurements are limited to a single wavelength, it is not possible to vertically differentiate between aerosol modes/types and therefore vertical profiles of intensive variables such as the single scattering albedo (SSA), lidar ratio (LR) or effective radius are assumed as vertically constant by this method. As a result, for each GRASPpac retrieval we obtain aerosol profiles (at 60 points) of backscatter at 180º, scattering, extinction and absorption coefficients at 440, 675, 870, 1020 and 1064 nm, and also of aerosol size distribution (but without changes in the effective radius with height) and the aerosol volume concentration.”

Comment: [5,21] "backscattering": here "under 180_"?

Answer: Yes, we have modified this.

Comment: [5,22] Please add a short comment on the accuracy of the retrieved aerosol parameters that are used in Section 4 (see also comment [3,1]). This is an important/mandatory information.

Answer: We have included a comment on the uncertainty of the retrieved parameters. Please, see our response to comment [3,1].

Comment: [5,26] "backscattering": here "hemispheric"?

Answer: Yes, we have changed it.
Comment: [5,30] Give an equation/definition of "comparison" and "relative difference" (see also [6,2] and figures): "GRASP minus in-situ" or "in-situ minus GRASP"? Divided by the "mean of in-situ"?

Answer: We have included the definition.

Comment: [6,7] Is the angular range with respect to the backscatter configuration correct? It should be 90º instead of 10º here (cf. Müller et al., 2011a)?

Answer: Yes, the design of the instrument limits the collection of the scattered light to the angular range 10-171º. However, we agree with the reviewer that this information here is misleading since after applying the correction introduced by Müller et al. (2011a) the scattering coefficient should correspond to the angular range 0-180º. With the aim of also clarifying how the comparison of the backscatter coefficient retrieved with GRASPpac and the hemispheric backscattering coefficient measured with the nephelometer this paragraph has been completely modified. Please, see our response to comment [4,2].

Comment: [6,12] "tends to overestimate all the studied...". Is this statement trivial?

Answer: We have modified this paragraph as follows:

“The frequency distributions of the absolute errors (in-situ minus GRASPpac values) for the scattering and extinction coefficients are tailed towards negative values evidencing an overestimation of GRASPpac retrievals compared with in-situ measurements. For the extinction coefficient, Herreras et al. (2018) showed good agreement between the integrated extinction profiles derived with GRASPpac and AOD from sunphotometers located at various heights (R2>0.6). For the backscattering coefficient, Fig.2 shows that GRASPpac also overestimates the in-situ measurements, but the frequency distribution of the absolute errors is more symmetrically distributed around 0. The overestimation of GRASPpac retrieved backscattering coefficients is in agreement with the assumption made to convert the backscatter coefficient at 180º provided by GRASPpac into a hemispheric backscattering coefficient in order to perform the comparison with the in-situ measurements (see section 3). As the backscatter at 180º is typically larger than at smaller angles, this overestimation was expected. However, since overestimation of the total scattering and extinction coefficients also occurs, it is difficult to discern whether this overestimation originates in the GRASPpac retrieval or in the assumption made to compare with the in-situ data. On the other hand, this assumption might be contributing to lower the correlation between the backscattering coefficient from GRASPpac and in-situ measurements in comparison with the results obtained for the scattering and extinction coefficients comparison (Fig. 1), that shows higher correlation coefficients “

Comment: [6,22] "...there is a linear trend between scattering and extinction coefficients...": What does this mean? If scattering and extinction are the same, the single scattering albedo is = 1. This is more or less the case under clean and turbid conditions according to Fig. 3a. If scattering and extinction show a linear dependence, the single scattering albedo is constant. The fact that in general large extinction coefficients correspond to large scattering coefficients is not surprising (! is typically between say 0.8 and 1).

Answer: The idea behind this figure was not to show that there is a linear dependence between the scattering and extinction coefficients nor to show that large extinction coefficients correspond to large scattering coefficients, since as mentioned by the reviewer, this is not surprising. With
this figure, we wanted to show the different relationship between scattering and extinction for the in-situ measurements (single scattering albedo close to 1, perfect linear trend) compared to the GRASPpac retrievals. GRASPpac retrievals do not reproduce the observed behavior. The absorption coefficient is overestimated in many cases which leads to differences in the scattering-extinction pattern as it is observed from in-situ measurements. Due to the climatic relevance of the single scattering albedo, we think that the performance of GRASPpac retrieving absorption or single scattering albedo values is an important topic to discuss in the manuscript.

Comment: [8,17] "Qualitatively speaking, the volume concentration...". I don’t understand the reason for this sentence? The seasonal cycle of the volume size distribution is not shown in the paper. Is this sentence included because a similar behavior is plausible? Or because this is known from literature? Or is it an intrinsic feature of the GRASP-methodology?

Answer: We included this sentence to note that the seasonality of the volume concentration and the scattering coefficient are similar to the one shown in the manuscript for the extinction coefficient. Although it was not shown in the paper, the volume concentration and scattering coefficient vertical profiles were also retrieved and available to perform the statistical analysis. To avoid confusion we have removed this sentence from the manuscript.

Comment: [8,19] A comment on the limitation to daytime: This is caused by the combination with the sun photometer. From the ceilometer’s perspective the determination of backscatter coefficients can be provided during night time as well (likely even better) provided that a calibration is possible, see comment on [3,1]. The extinction coefficient can be estimated if the lidar ratio can be estimated (of course, subject to – maybe significant – errors). In Titos et al.’s paper the transformation from RCS to physical quantities is provided by using the photometer data as constraint; this is conceptually superior (at the expense of the two measuring systems required).

Answer: We agree with the reviewer. We have specifically noted that this limitation is caused by the combination with the sun/sky photometer instrument. See also our response to comment [3,1] on this topic. However, it is important to keep in mind that the use of the sun/sky photometer, although restricted to day-time, allows us to obtain profiles of more optical properties and not only backscatter, as well as microphysical aerosol properties.

Comment: [8,23] above -> larger than

Answer: Done

Comment: [10,1] Explain "center of mass"; e.g. by citing Mona et al. (2006) or alternatively (if you want to avoid another self-citation) Binietoglou et al. (2015).

Answer: The following sentence has been included:

"The center of mass gives in a single number an indication of the altitude of the aerosol vertical distribution in the atmosphere. Cases in which a single aerosol layer is present in the atmosphere, the center of mass gives an indication of its mean altitude; in cases of multiple layers, however, it could be located in areas without any considerable aerosol load (Binietoglou et al., 2015; Mona et al., 2006)."
Comment: [10,4] Here and in Fig. 7, the center of mass is given with respect to sea level. The numbers are correct but may lead to misunderstandings as the reader would intuitively expect much lower values (main contribution is almost always from the mixing layer). Indeed, values of 1.0–1.5 km can be found from Fig. 7 when considering heights above ground. So I recommend to add the corresponding values in brackets, at least in one or two cases.

Answer: We have included the height above the ground in this sentence and a specific statement in caption of Fig. 7 noting that heights refer to the sea level, but measurements start at 1570 m a.s.l.

Comment: [10,28] "Similar seasonal behavior...": This is not a result of this study, it is only a message from a paper by Pandolfi et al. (2014). This should be made clear.

Answer: We agree with the reviewer, we have deleted this sentence from the conclusions.


Answer: Thank you for the reference, we have included it in the revised version of the manuscript.

Comment: [Fig. 5] delete "light" in the caption; also in Fig. 6.

Answer: Done

Comment: Suggested references:


Retrieval of aerosol properties from ceilometer and photometer measurements: long-term evaluation with in-situ data and statistical analysis at Montsec (southern Pyrenees)

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Abstract. Given the need of accurate knowledge of aerosol microphysical and optical properties with height resolution, various algorithms combining vertically-resolved and column integrated aerosol information have been developed in the last years. Here we present new results of vertically-resolved extensive aerosol optical properties (backscattering, scattering and extinction) and volume concentrations retrieved with the GRASP (Generalized Retrieval of Aerosol and Surface Properties) algorithm over a 3 year-period. The range-corrected signal (RCS) at 1064 nm measured with a ceilometer and the aerosol optical depth (AOD) and sky radiances from a sun/sky photometer have been used as input for this algorithm. We perform a detailed evaluation of GRASP retrievals with simultaneous in-situ measurements performed at the same height, at the Montsec mountaintop observatory (MSA) in the Pre-Pyrenees (northeastern Spain). This is the first long-term evaluation of various outputs of this algorithm; previous evaluations focused only on the study of aerosol volume concentration for short-term periods. In general, our results show good agreement between techniques although GRASP inversions yield higher values than those measured in-situ. The statistical analysis of the extinction coefficient vertical profiles shows a clear seasonality as well as significant differences depending on the air-masses origin. The observed seasonal cycle is mainly modulated by a higher development of the atmospheric boundary layer (ABL) during warm months, which favors the transport of pollutants to MSA, and higher influence of regional and North-African episodes. On the other hand, in winter, MSA is frequently influenced by free troposphere conditions and venting periods, and therefore lower extinction coefficients that markedly decrease with height. This study shows the potentiality of implementing GRASP in ceilometers and lidars networks for obtaining aerosol optical properties and volume concentrations at multiple sites worldwide, which will definitely contribute to enhance the representativeness of aerosol vertical distribution as well as to provide useful information for satellite and global models evaluation.
1 Introduction

Atmospheric aerosol particles interact directly with the incident solar radiation by either scattering or absorbing light. These aerosol-radiation as well as the aerosol-cloud interactions influence the Earth’s radiative budget and therefore have an impact on climate. Aerosol particles are considered the atmospheric constituents with the largest uncertainty in global climate forcing estimations (IPCC, 2013). Their high spatial, vertical and temporal variability is one of the key factors contributing to their large uncertainty (IPCC, 2013).

During the last years, a great effort has been done from the Aerosols, Clouds, and Trace gases Research InfraStructure (ACTRIS, www.actris.eu) community to extend the temporal and spatial coverage of aerosol properties sampling and to harmonize measurement protocols to increase their representativeness and the comparability among sites and between measurement techniques (i.e. in-situ versus remote-sensing). In-situ observatories are widely distributed and cover a large variety of atmospheric conditions (urban, rural, background and remote sites). Moreover, in-situ instrumentation is able to provide a complete set of information in terms of chemical, optical and microphysical aerosol properties. The main drawback of in-situ observatories is that they are only representative of the atmospheric layer closest to the surface and might not be useful to infer aerosol radiative properties at elevated layers (Rosati et al., 2016). For this reason, vertically resolved aerosol observations are needed to complement surface in-situ measurements and column-integrated observations from satellites or ground-based photometers. Lidar systems are frequently used for profiling aerosol optical properties and, depending on the lidar capabilities and availability of co-located photometer measurements, vertical profiles of aerosol microphysical properties can be retrieved as well by using inversion algorithms (Chaikovsky et al., 2008, 2016; Lopatin et al., 2013).

One of the recently developed inversion algorithms is the Generalized Retrieval of Aerosol and Surface Properties (GRASP; Dubovik et al., 2014; www.grasp-open.com) code that uses the heritage of AERONET (Aerosol Robotic Network) inversion scheme (e.g. Dubovik and King, 2000; Dubovik et al., 2006). It is a versatile and open-source algorithm capable of obtaining optical and microphysical aerosol properties from different sets of measurements (Kokhanovsky et al., 2015; Espinosa et al., 2017; Torres et al., 2017; Román et al., 2017; 2018). In particular, GRASP allows the user to combine A_\text{aerosol} O_\text{optical depths} (AODs), sky radiances and range corrected lidar signal (RCS) lidar values to retrieve columnar and vertically-resolved aerosol properties. Román et al. (2018) proposed a similar approach but using the RCS values at only one wavelength measured with ceilometer instead of using multi-wavelength lidar RCS values as done before. The retrieved vertical profiles of aerosol volume concentration showed good agreement with in-situ measurements from an aircraft campaign and with in-situ measurements from a nearby mountain station during a summer campaign in southern Spain (Román et al., 2018). The use of ceilometer measurements in the GRASP algorithm is a significant advance towards a better representation of aerosol properties with vertical resolution since ceilometers are cheaper, require less supervision, provide continuous measurements and are more extensively distributed compared to more sophisticated lidar systems (Wiegner et al., 2014; Cazorla et al., 2017; Dionisi et al., 2018). However, the main drawback of this approach is that sun/sky photometer measurements are only available during day time and under low cloudiness conditions. Other methodologies, such as the absolute calibration of the ceilometer
(Wiegner and Geiß, 2012), are able to overcome this issue and provide quantitative backscatter profiles during day and night time. GRASP-Quantitative ceilometer profiles from ceilometers worldwide could be used for evaluating dust forecast models (Tsekeri et al., 2017) such as the BSC-DREAM8b, as input to radiative transfer models (Granados-Muñoz et al., 2019) or can be assimilated in global models (Chen et al., 2018). This application represents a step-forward in the classical use of ceilometers that were originally developed for cloud base detection (e.g., Martucci et al., 2010; Wiegner et al., 2014).

The potential of this new technique motivates the present study in which the GRASP code is used to retrieve long-term vertical profiles of aerosol optical and microphysical properties combining ceilometer and AERONET sun/sky photometer measurements over a 3-year period. The main objective of this paper is to evaluate the performance of the retrieved aerosol products by GRASP combining ceilometer and photometer measurements using as reference the in-situ measurements performed at the Montsec Global Atmosphere Watch (GAW) station (MSA, 1570 m a.s.l., NE Spain). Additionally, a statistical analysis of the vertical structure of aerosol properties based on the 3-years of GRASP retrievals at MSA is presented.

2 Experimental site and instrumentation

2.1 Montsec Observatory

Measurements used in this study were performed in the northeastern Iberian Peninsula, most of them at the Montsec mountain-top station (MSA; 42° 3’ N, 0° 44’ E, 1570 m a.s.l.), located in the facilities of the Montsec Astronomic Observatory (OAdM, http://oadm.ieec.cat/). The MSA continental background site is part of the Catalan Air Quality Network (Xarxa de Vigilància i Previsió de la Contaminació Atmosfèrica, http://territori.gencat.cat/) and it is integrated in the European Research Infrastructure ACTRIS and in the Global Atmosphere Watch (GAW) program. It is a remote high-altitude station situated in the southern side of the Pre-Pyrenees at the Montsec d’Ares mountain. This region is sparsely populated and isolated from large urban and industrial agglomerations (140 km from Barcelona to the northwest and 30 km from the largest city in the region). The prevailing atmospheric conditions are characteristic of Mediterranean climate, with long dry periods, sporadic but intense rains, and a prevalence of local and regional atmospheric air mass circulations and Saharan dust intrusions (Ripoll et al., 2014; Ealo et al., 2016).

2.2 In-situ measurements

Aerosol particles light scattering ($\sigma_{sp}$) and hemispheric backscattering ($\sigma_{bsp}$) coefficients were measured at three wavelengths (450, 525 and 635 nm) with a LED-based integrating nephelometer (model Aurora 3000, ECOTECH Pty, Ltd, Knoxfield, Australia) with 5-min time resolution. The aerosol flow in the nephelometer was set to 5 l min$^{-1}$. Measurements were performed at dry conditions (RH<40%) by using the internal RH-control function of the nephelometer that slightly heats the sampled air when the RH is above the threshold value. The nephelometer is periodically calibrated (four times per year) with CO$_2$ and filtered air. Zero adjustments are performed every midnight using internally filtered particle free air. The Aurora 3000 nephelometer used in this study operates by collecting light scattered within the angular range 10–171° (Müller et al., 2011a).
The main source of error is the truncation in the forward direction (0–10°) due to the inability of the nephelometer to sense near-forward scattering, which is an increasingly dominant part of the total scattering for large particles (Anderson et al., 1996). Non-idealities due to truncation errors have been corrected following the scheme described by Müller et al. (2011a). The detection limits of the nephelometer over 1 min averaging time are 0.11, 0.14 and 0.12 Mm$^{-1}$ for total scattering at 450, 525 and 635 nm, respectively, and 0.12, 0.11 and 0.13 Mm$^{-1}$ for backscattering (Müller et al., 2011a).

The aerosol light-absorption coefficient, $\sigma_{ap}$, was measured with a Multi-Angle Absorption Photometer (MAAP, model 5012, Thermo) at 637 nm (Müller et al., 2011b). A detailed description of the method is provided by Petzold and Schönlinner (2004). The MAAP draws the ambient air at constant flow rate of 16.7 l min$^{-1}$ and provides 1 min values. The detection limit of the MAAP instrument is lower than 0.6 Mm$^{-1}$ over 2 min integration. The total method uncertainty for the particle light-absorption coefficient inferred from MAAP measurements is around 12% (Petzold and Schönlinner, 2004).

An aerosol optical counter (GRIMM Spectrometer, model 1129-Sky-OPC) was used to measure particle number concentrations in 31 size bins, for particles in the diameter size range from 0.25 to 32 µm at 5 min time resolution. The working principle of this instrument is based on multi-channel light-scattering optics (Grimm and Eatough, 2009) in which the intensity of the measured scattered light is related to the size of the particles. Volume size distributions were derived from the number size distribution assuming spherical particles.

All in-situ measurements were performed at MSA station and have been referred to ambient temperature and pressure using the measurements from an automatic and collocated weather station. Measurements were performed at low relative humidity (RH<40%), as recommended by the World Meteorological Organization (WMO/GAW, 2003) and ACTRIS infrastructure.

### 2.3 Passive remote sensing measurements

Measurements of column integrated aerosol properties were determined with a CE-318 sun/sky photometer (Cimel Electronique, France) included in AERONET (Holben et al., 1998) and located at MSA observatory. This instrument performs direct sun measurements with a 1.2° full field of view at least at 440, 675, 870, and 1020 nm, which are used to derive AOD at these wavelengths. The sky radiance measurements (almucantar configuration) are also carried out at 440, 675, 870 and 1020 nm. A full description of the AERONET products obtained from this instrument can be found in Holben et al. (1998). In this work, AOD and sky radiances, both at 440, 675, 870 and 1020 nm, from version 2 of AERONET level 1.5 data are used.

### 2.4 Active remote sensing measurements

Vertical profiles of RCS at 1064 nm were performed with a Jenoptik CHM 15k Nimbus (G. Lufft Mess- und Regeltechnik GmbH, Germany) ceilometer that includes a pulsed Nd:YAG laser, emitting at 1064 nm. The energy emitted per pulse is 8 µJ and the duration of each pulse is between 1 and 5 ns with a repetition frequency of 6.5 kHz. The maximum height of the signal is 15.36 km a.g.l. equivalent to 1024 range bins. The ceilometer is located at 800 m a.s.l, 1.760 meters downslope of the MSA measurement station, at the Center for the Observation of the Universe (COU, http://www.parcastronomic.cat/). The horizontal distance between the ceilometer and the MSA station is less than 2.5 km. This instrument operates continuously with a temporal
resolution of 1 min and a spatial resolution of 15 m. The RCS profiles provided by the instrument are overlap-corrected using the manufacturer’s overlap function. In addition, according to this function, the overlap of the telescope and the laser beam is greater than 85% at around 770 m a.g.l. Thus, the effect of the overlap at the height of the MSA observatory (1570 m a.s.l.) is expected to be low. According to the manufacturer overlap function, the overlap of the telescope and the laser beam is greater than 85% at around 760 m a.g.l. Nevertheless, the RCS profiles provided by the instrument are overlap-corrected using the manufacturer’s overlap function.

3 GRASP retrievals

GRASP code is mainly based on two independent modules: 1) the forward module consisting of a radiative transfer and aerosol model which simulates the radiative measurements for a given aerosol scenario, and 2) the numerical inversion module which is not related to the physical nature of the inverted data (Dubovik et al., 2011; Dubovik et al., 2014) and is mathematically based on multi-term least square method (Dubovik and King, 2000). Detailed description of the GRASP working principle using sun/sky photometer and RCS data can be found in Lopatin et al. (2013), where the GARRLiC (Generalized Aerosol Retrieval from Radiometer and Lidar Combined data) scheme, which is part of GRASP code, is explained.

In this study, we follow the inversion strategy named as GRASP_{pac} (sub-index meaning “photometer and ceilometer”) introduced by Román et al. (2018). A GRASP_{pac} retrieval is done for each sky radiance almucantar sequence available from AERONET if sky radiances and ceilometer measurements satisfy cloud-free conditions. The following measurements are used in GRASP code for each retrieval: 1) the cloud-screened sky radiance and AOD at 440, 675, 870 and 1020 nm (AERONET version 2 level 1.5); and 2) the normalized ceilometer RCS at 1064 nm, previously cloud-screened, smoothed and averaged in a ±15 min window centered in the photometer measurement time, at 60 log-spaced heights as in Lopatin et al. (2013). The minimum height of these 60 values corresponds to the MSA altitude while the maximum height could be up to 7000 m above MSA. As mentioned before, the RCS profiles provided by the ceilometer are overlap-corrected and, according to the manufacturer’s overlap function, the overlap of the telescope and the laser beam is greater than 85% at the MSA altitude (770 m above the ceilometer). Thus, the effect of the overlap in the GRASP_{pac} retrievals is expected to be low, since the ceilometer RCS below 1570 m a.s.l. is not used here as input in GRASP_{pac}. The maximum height selected for the 60 log-spaced bins is 7000 m above MSA, since aerosol layers are rarely detected above this height and the ceilometer signal is usually too noisy at higher altitudes due to the low power of the ceilometer’s laser. The RCS at these 60 log-spaced bins is averaged and then normalized by dividing each value by the integrated RCS between the minimum and maximum heights. In addition, Bidirectional Reflectance Distribution Function (BRDF) is needed to make the GRASP_{pac} retrievals and, to this end, an 8-days climatology (2000-2014) of the MCD43C1 product (V005 MODIS Terra+Aqua BRDF/Albedo 16-Day L3 0.05Deg CMG) of MODIS (MODe rate-resolution Imaging Spectroradiometer) is used (Schaaf et al., 2011).

Since ceilometer measurements are limited to a single wavelength, it is not possible to vertically differentiate between aerosol modes/types and therefore vertical profiles of intensive variables such as the single scattering albedo (SSA), lidar ratio (LR)
or effective radius are assumed as vertically constant by this method. As a result, for each GRASP\textsubscript{pac} retrieval we obtain aerosol profiles (at 60 points) of backscatter at 180°, scattering, extinction and absorption coefficients at 440, 675, 870, 1020 and 1064 nm, and also of aerosol size distribution (but without changes in the effective radius with height) and the aerosol volume concentration and also of aerosol volume concentration and size distribution. The estimated uncertainty for the backscatter coefficient retrieved with GRASP\textsubscript{pac} is 31%, and 21% for the extinction coefficient and the volume concentration (Román et al., 2017). The uncertainty in the backscatter profiles retrieved with GRASP\textsubscript{pac} is higher than the estimated uncertainty by Wiegner and Geiß (2012) for the absolute calibration method (10%). Since the in-situ measurements and GRASP\textsubscript{pac} retrievals provide different information with respect to the aerosol backward scattering properties (hemispheric backscattering versus backscatter at 180°) the direct comparison between both techniques is not possible. To have a sense of the performance of the GRASP\textsubscript{pac} backscatter retrieval, for the comparison we have assumed that the scattering into de backward hemisphere is the same in all directions. Therefore, we have extrapolated the backscatter at 180° to the angular range 90-180° in order to make it comparable with the backscattering coefficient measured with the nephelometer. This assumption constitutes an additional source of error since the actual angular scattering distribution is not known and typically backscatter at 180° is larger than at smaller angles. For the backscattering coefficient comparison, it is important to bear in mind the intrinsic differences among the variables compared. The nephelometer design limits the collection of the scattered light to the angular range 10-171°, while the ceilometer measures the backscatter signal at 180°. Additionally, for the backscattering coefficient at 180° retrieved with GRASP\textsubscript{pac} we assume for the comparison with in-situ data that the hemispheric backscattered radiation is symmetric along all hemispheric angles, which might not be true for all cases and might contribute to lower the correlation coefficient.

### 4 Results and discussion

#### 4.1 GRASP\textsubscript{pac} – in-situ comparison

**4.1.1 Optical properties comparison**

In-situ extensive aerosol optical properties (i.e. hemispheric-backscattering, scattering and extinction coefficients) measured at MSA over a 3 years period (April 2014 - March 2017) were used for evaluating the retrieval of aerosol optical properties from a ceilometer and a sun/sky photometer using the GRASP\textsubscript{pac} method in a long-term frame. Previous evaluations of this algorithm with in-situ data focused on aircraft campaigns (2-3 study cases) (e.g. Benavent-Oltra et al., 2018; Tsekeri et al., 2017) or short-term periods (Román et al., 2018). Figure 1 shows the comparison between the GRASP\textsubscript{pac} retrievals and in-situ measured coefficients at low ambient RH (RH\textsubscript{ambient} < 50%). This restriction has been imposed to avoid cases affected by hygroscopic growth and consequent enhancement of the optical coefficients detected by the remote sensing instrumentation. To merge both datasets (GRASP\textsubscript{pac} and in-situ), the data have been averaged in 1-hour intervals. The comparison has been performed at 1570 m a.s.l., where the in-situ instrumentation is located and coinciding with the first height of the GRASP\textsubscript{pac} retrievals. Therefore, the following results and associated discussion on the comparison between GRASP\textsubscript{pac} and in-situ
measurements refer exclusively to this height. Figure 2 shows the relative differences between optical parameters measured in-situ and retrieved by GRASP$_{pac}$ optical parameters. In general, the GRASP$_{pac}$ retrievals are in agreement with the in-situ measurements. The coefficients of determination span from 0.49 for the backscattering coefficient to 0.77 for the scattering coefficient and 0.73 for the extinction coefficient (see details in Figure 1). For the backscattering coefficient comparison, it is important to bear in mind the intrinsic differences among the variables compared. The nephelometer design limits the collection of the scattered light to the angular range 10°-171°, while the ceilometer measures the backscatter signal at 180°. Additionally, for the backscattering coefficient at 180° retrieved with GRASP$_{pac}$ we assume for the comparison with in-situ data that the hemispheric backscattered radiation is symmetric along all hemispheric angles, which might not be true for all cases and might contribute to lower the correlation coefficient. For both the aerosol light scattering and the extinction coefficients the slope and intercept of the regression are >1, while for the backscattering coefficient the slope is <1. Figure 2 shows the absolute differences between optical parameters measured in-situ and retrieved by GRASP$_{pac}$ optical parameters. The frequency distributions of the absolute errors (in-situ minus GRASP$_{pac}$ values) for the scattering and extinction coefficients are tailed towards negative values evidencing an overestimation of GRASP$_{pac}$ retrievals compared with in-situ measurements. As it can be seen in Fig. 2, GRASP$_{pac}$ tends to overestimate all the studied coefficients with higher occurrence of negative relative errors. In this sense, all frequency distributions are tailed towards negative values. The agreement of GRASP$_{pac}$ with in-situ data shows relative differences of ±1.25% for 84%, 75% and 68% of the backscattering, scattering and extinction coefficients, respectively. For the extinction coefficient, Herreras et al. (2018) showed good agreement between the integrated extinction profiles derived with GRASP$_{pac}$ and AOD from sunphotometers located at various heights ($R^2$ > 0.6). For the backscattering coefficient, Fig. 2 shows that GRASP$_{pac}$ also overestimates the in-situ measurements, but the frequency distribution of the absolute errors is more symmetrically distributed around 0. The overestimation of GRASP$_{pac}$ retrieved backscattering coefficients is in agreement with the assumption made to convert the backscatter coefficient at 180° provided by GRASP$_{pac}$ into a hemispheric backscattering coefficient in order to perform the comparison with the in-situ measurements (see section 3). As the backscatter at 180° is typically larger than at smaller angles, this overestimation was expected. However, since overestimation of the total scattering and extinction coefficients also occurs, it is difficult to discern whether this overestimation originates in the GRASP$_{pac}$ retrieval or in the assumption made to compare with the in-situ data. On the other hand, this assumption might be contributing to lower the correlation between the backscattering coefficient from GRASP$_{pac}$ and in-situ measurements in comparison with the results obtained for the scattering and extinction coefficients comparison (Fig. 1), that shows higher correlation coefficients.

[Figure 1]

[Figure 2]

Figure 3 shows the relationship between the scattering and extinction coefficients measured in-situ and retrieved by GRASP$_{pac}$. The color scale represents the difference in the single scattering albedo measured in-situ and retrieved with GRASP$_{pac}$. For the in-situ data, there is a linear trend between scattering and extinction coefficients ($R^2 = 1$), denoting that the aerosol light-extinction is dominated by the scattering process, which is in accordance with previous in-situ studies performed at MSA.
(Pandolfi et al., 2014). On the contrary, for the GRASP\textsubscript{pac} retrievals the correlation is also good but the data points deviate from the 1:1 line as the difference in the SSA between in-situ and GRASP\textsubscript{pac} increases (yellowish colors). In general, GRASP\textsubscript{pac} retrievals yield lower SSA values (average SSA of 0.88 ± 0.14) compared with in-situ SSA (0.93 ± 0.04). These discrepancies in the absorption could be related to the differences in the SSA at ground level (as measured in-situ) and the SSA associated with the total atmospheric column (GRASP\textsubscript{pac}) due to absorbing aloft layers. However, the largest disagreement (yellowish colors in Fig. 3b) coincide with Atlantic air masses influence, that as it will be shown in Section 4.2, are characterized by low aerosol load and low impact of decoupled aerosol layers. On the other hand, Andrews et al. (2017) showed a systematic difference in the SSA from AERONET retrievals compared with integrated in-situ profiles, revealing that AERONET retrievals yield higher aerosol absorption than in-situ measurements, especially at low aerosol load. MSA is a remote site with predominantly low aerosol load and low contribution of absorbing particles. Furthermore, Román et al. (2018) found with synthetic data that SSA values retrieved by GRASP\textsubscript{pac} reproduce better the real SSA values for moderate-high aerosol loads. In a similar way, AERONET, in version 2, only provides SSA values with quality assurance if the AOD at 440 nm is higher than 0.4 (Dubovik et al., 2000; Dubovik et al., 2002; Holben et al., 2006). Then, most of the obtained SSA differences could be associated with the low aerosol load conditions, where the SSA uncertainty is high in GRASP\textsubscript{pac} values.

**4.1.2 Volume size distribution comparison**

Figure 4 shows the comparison of the total aerosol volume concentration (V) determined with GRASP\textsubscript{pac} and measured in-situ at MSA height over the study period. The color scale represents the ratio $V_{\text{fine}}/V$ that quantifies the contribution of fine particles (diameter below 1 µm) to the total volume concentration, as determined from the in-situ measurements. As we can see in Fig. 4a, there is a lack of correlation, showing different relationship depending on the ratio $V_{\text{fine}}/V$. When fine particles predominate (i.e. $V_{\text{fine}}/V > 0.75$, yellowish colors) the volume concentration measured in-situ is significantly larger than the volume concentration retrieved from the ceilometer and photometer data using GRASP\textsubscript{pac}. On the contrary, when coarse particles predominate the volume concentration provided by GRASP\textsubscript{pac} is larger than the one determined in-situ. Limiting the comparison to those cases with $V_{\text{fine}}/V < 0.75$ (Fig. 4b) the correlation improves significantly (R² = 0.65), and shows absolute differences within ±25 µm³/cm³% for 98.5% of the data (Fig. 4c). Similar to the extinction and scattering coefficients comparison, GRASP\textsubscript{pac} retrievals yield higher aerosol volume concentrations compared with the in-situ measurements. Similar overestimations comparing GRASP\textsubscript{pac} and in-situ data have been reported before. In particular, Román et al. (2018) compared the GRASP\textsubscript{pac} retrievals using also ceilometer and photometer data as input with in-situ measurements performed in a mountain station located ~25 km apart from the ceilometer and at around 2000 m above it during an intensive field campaign. Their results show that GRASP\textsubscript{pac} overestimates the volume concentration with a slope of the comparison around 1.5. We found similar results, revealing that in general, GRASP\textsubscript{pac} overestimates the aerosol volume concentration (slope of the comparison of 1.34). However, the comparison between GRASP\textsubscript{pac} and in-situ measurements shows significant discrepancies when fine particles predominate ($V_{\text{fine}}/V > 0.75$). The reduced number of cases with $V_{\text{fine}}/V > 0.75$ (~15% of the
total number of data points) makes it difficult to draw conclusive results concerning the total volume concentration in atmospheric conditions dominated by fine particles. Previous evaluations of GRASP algorithm were mainly conducted during Saharan dust events with predominance of coarse mode particles. Benavent-Oltra et al. (2018) found similar coarse volume concentrations between GRASP retrievals and in-situ profiles during two flights performed under dust-dominated conditions, with slight underestimation of GRASP in the aloft dust plumes, while significant overestimation was reported for the fine volume concentration. Overestimation of fine volume concentrations obtained with GARRLiC (Generalized Aerosol Retrieval from Radiometer and Lidar Combined data algorithm) algorithm compared with in-situ data were also observed under a dust-dominated and a marine polluted cases (Tsekeri et al., 2017). Using synthetic data, Román et al. (2018) showed higher discrepancies in the retrieval of fine volume concentrations than in coarse ones for GRASP\textsubscript{pac}. The reason behind these differences was partly attributed to the use of a long wavelength (1064 nm) as RCS in the retrieval which is less sensitive to fine particles than shorter wavelengths. Nevertheless, despite the differences among studies, all of them evidence that the retrieval of fine volume concentrations is particularly challenging while good results can be obtained for the coarse volume concentration or total concentration if the size distribution is dominated by coarse particles.

Finally, several environmental and topographic factors can be brought forward to partly explain the differences observed among techniques, namely the measurement atmospheric conditions (temperature, pressure and RH) and orographic effects affecting wind patterns and atmospheric boundary layer structure and causing spatial inhomogeneities. Concerning the atmospheric conditions at which the aerosol properties are measured in terms of temperature, pressure and relative humidity, we expect low effect on the comparison since the in-situ data has been converted to ambient T and P and the comparison was restricted to cases with ambient RH < 50%. Although hygroscopic growth can occur even at low RH (Zieger et al., 2017), we limit the study to ambient RH < 50% in order to minimize the RH effect in the comparison (Titos et al., 2016). As can be seen in Figure S1 of the supplementary material, the comparison shows no dependency on RH for RH\textsubscript{ambient} < 50%. Another possible factor that could affect the comparison is the fact that the in-situ and photometer measurements are not performed exactly over the ceilometer vertical. However, due to the short horizontal distance (< 2.5 km), this fact is expected to have little impact in our results.

### 4.2 Statistical analysis of aerosol profiles

In the following section, we focus on the extinction coefficient since it is the most relevant climate variable from the ones retrieved with GRASP\textsubscript{pac}. Qualitatively speaking, the volume concentration and the scattering coefficient show similar trends in the vertical distribution. Figure 5 shows the seasonality of particle extinction profiles retrieved with GRASP\textsubscript{pac} using ceilometer and photometer data as inputs. It is important to recall that GRASP\textsubscript{pac} retrievals are performed only during daytime and clear sky conditions caused by the combination of the ceilometer with the sun/sky photometer data (see Section 3 for further details), which might bias the statistical analysis presented in this section compared to continuous measurements. Figure S2 of the supplementary material shows the frequency distribution of the number of profiles retrieved by month and hour of
the day. As it can be seen, the GRASP_{pulsed} retrievals are restricted to daytime conditions and solar zenith angles larger than 40° (mainly from 6 to 9 h in the morning and from 14 to 16 h). Accordingly, there are also less GRASP_{pulsed} retrievals during autumn and winter.

In average terms, the largest extinction coefficients are observed at the lowest altitudes sounded. A nearly exponential decrease with height of the median extinction coefficients is observed during all seasons up to 4000-5000 m a.s.l. Exponential decreasing trend of the extinction coefficient has been observed in several statistical lidar studies in Europe (Mattis et al., 2004; Amiridis et al., 2005; Navas-Guzmán et al., 2013). There is a clear seasonal behavior in the vertical distribution of aerosol particles, evidencing that during winter most particles are confined to the first few kilometers above the surface while the median profile in summer shows the presence of particles at higher altitudes. Also in summer, the extinction profiles display a larger interquartile range compared with the other seasons denoting high variability in the vertical distribution of aerosol particles.

Concerning the extinction coefficients in the lowermost part of the profiles, Pandolfi et al. (2014) reported a similar seasonality for continuous in-situ measurements at MSA, with the highest extinction coefficients observed in summer and the lowest ones in winter.

Air masses arriving at MSA have been classified in four sectors following the procedure of Ripoll et al. (2014): Atlantic (ATL), North-African (NAF), Regional (REG) and European and Mediterranean (EU+MED). Figure 6 shows a statistical overview of the extinction profiles from GRASP_{pulsed} classified according with their air mass origin. There are significant differences in the extinction vertical distribution depending on the origin of the air masses affecting the Montsec area. The lowest median extinction coefficient occurs under Atlantic air masses. This result is in agreement with the low extinction coefficients found in winter, given that during colder months the site is frequently affected by Atlantic air masses and is located within the free troposphere (Ripoll et al., 2014). These profiles also show low variability (smaller interquartile range). A similar behavior is obtained for the MED+EU sector, although the extinction coefficient displays higher variability; especially pronounced close to the surface (high 90th percentile). For air masses with origin in North Africa the extinction coefficient vertical profiles show the highest variability; denoting the strong variation in intensity and aerosol-layers stratification among events. The average extinction coefficient for the lowest atmospheric layer is slightly lower than the average extinction coefficient found during dust events at surface level in MSA using in-situ techniques (Pandolfi et al., 2014). This discrepancy can be attributed to the different study period and therefore different NAF episodes included in the calculation with varying intensity and frequency. The air-masses grouped in the REG sector include transport from the Iberian Peninsula as well as re-circulation processes associated with the land-sea breezes regime (Millán et al., 1997). In this case, the extinction coefficient profiles show high variability up to 6000 m a.s.l., indicating layering and accumulation of pollutants under regional re-circulation conditions. During these episodes, pollutants are raised up to upper levels resulting in the stratification of aerosol layers along the vertical atmosphere (Pérez et al., 2004). On the other hand, MED+EU and ATL sectors show a low 90th percentile and interquartile range above 3000 m a.s.l., suggesting that the likelihood of aloft aerosol layers under these atmospheric scenarios is significantly reduced compared with REG sector and, more remarkable, with NAF sector.
The air mass classification and the seasonality of the extinction vertical profiles are clearly linked. NAF and REG episodes are more frequent during spring and summer while ATL episodes are more frequent in autumn and winter (Ripoll et al., 2014). The seasonal cycle observed is mainly modulated by a higher development of the ABL during warm months, and higher influence of REG and NAF episodes (e.g. Ealo et al., 2018). This combination leads to high extinction coefficients at higher altitudes and strong variability (large difference in the 10th and 90th percentiles and interquartile range) during warmer months. However, in winter, MSA is frequently influenced by free troposphere conditions and venting periods (Ripoll et al., 2014), and therefore lower extinction coefficients. NAF episodes also affect MSA during winter (i.e. Titos et al., 2017), but their frequency of occurrence is low and their impact in the extinction vertical profile is not observed in the median and 90th percentile profiles (Fig. 6).

5

[Figure 6]
Figure 7 shows the center of mass calculated for the median extinction profile, and the 25th and 75th percentile extinction profiles following the procedure described by Cazorla et al. (2017), as a function of the air mass origin sector. The center of mass gives in a single number an indication of the altitude of the aerosol vertical distribution in the atmosphere. Cases in which a single aerosol layer is present in the atmosphere, the center of mass gives an indication of its mean altitude; in cases of multiple layers, however, it could be located in areas without any considerable aerosol load (Binietoglou et al., 2015; Mona et al., 2006). The highest center of mass is achieved under NAF air masses, evidencing the influence of aloft dust layers. During an intense dust outbreak in February 2016, Cazorla et al. (2017) calculated a center of mass of 3000 m a.s.l. (1430 m a.g.l.) at MSA in the most intense day. An interesting feature of Fig. 7 is the difference in the centers of mass retrieved from the percentiles and median profiles for REG and NAF sectors, while for ATL and MED+EU the difference in the 25th and 75th percentiles is small. This fact evidences the high variability in vertical distribution of aerosol particles occurring during NAF and REG episodes.

[Figure 7]

5 Conclusions

In this study, we present a systematic application of the GRASP algorithm using ceilometer RCS and sun/sky photometer measurements (GRASP pac) over an extended period of time (3 years). Our unique experimental set-up allows us to perform a long-term evaluation of the GRASP pac retrievals versus in-situ measurements under different atmospheric conditions. The output variables studied here are the aerosol backscattering, scattering and extinction coefficients and the volume concentration. The results show an overall good agreement between GRASP retrievals and in-situ measurements, especially good for scattering and extinction coefficients (R^2 > 0.7). The volume concentration comparison shows differences depending on the predominance of fine or coarse particles, with poor agreement when the contribution of fine particles to the total volume concentration is > 75%, and good agreement otherwise. Restricting the comparison to cases with V_{fine}/V < 0.75, GRASP pac and in-situ measurements show good correlation although GRASP pac yield higher volume concentrations. Similar
overestimation of GRASP_{pac} is found for the scattering and extinction coefficients. We found slight discrepancies in the scattering-extinction relationship obtained with GRASP_{pac} compared to in-situ data. In general, GRASP_{pac} retrievals yield lower SSA values (average SSA of 0.88 ± 0.14) compared with in-situ SSA (0.93 ± 0.04). This result can be linked with previous evaluations of AERONET retrievals that were shown to yield higher aerosol absorption than in-situ measurements, especially at low aerosol load. Evaluation of GRASP_{pac} algorithm at different environments with variable aerosol load and SSA characteristics will contribute to better understand and constrain the validity and limitations of GRASP_{pac}.

The statistical analysis of the extinction coefficient vertical profiles retrieved with GRASP_{pac} shows a clear seasonality as well as significant differences depending on the air-masses origin. The observed seasonal cycle is characterized by higher extinction coefficients during summer with strong day to day variability while during winter the extinction coefficient is lower in the whole atmospheric column and shows lower variability. Similar seasonal behavior is obtained from the in-situ measurements at ground level. This seasonality is associated with a higher development of the atmospheric boundary layer during warm months, favoring the transport of pollutants to MSA. Additionally, the higher influence of regional and North-African episodes in summer contributes to the observed seasonality. On the other hand, in winter, MSA is frequently influenced by free troposphere conditions and venting periods, and therefore lower extinction coefficients that markedly decrease with height.

The use of automated lidars and ceilometers systems for the determination of vertically-resolved aerosol properties has increased in recent years thanks to their low operation requirements and costs, and their capability of providing continuous unattended measurements. Together with this increase use of ceilometer systems, there is a growing need of being able to convert the ceilometer signals into usable aerosol properties. In this context, the overall good results obtained in our validation are encouraging and emphasize the potentiality of implementing GRASP in ceilometers and lidars networks for obtaining aerosol optical properties and volume concentrations with height resolution and wide spatial coverage. Compared with previous studies, the present evaluation of GRASP_{pac} retrievals with in-situ data has been performed over a 3 year-period, being therefore representative of varying atmospheric conditions. Nevertheless, it is important to bear in mind that the results presented in this study are limited to day time and low cloudiness conditions due to the need of simultaneous sun/sky photometer measurements. Also, further studies investigating the performance of the application of GRASP_{pac} to ceilometers and automatic lidars with different characteristics (i.e. wavelength of operation, pulse energy) than the one used in this study are needed to maximize its potential application. With this in mind, the implementation of GRASP_{pac} in the frame of measurement networks will enhance the representativeness of aerosol vertical distribution providing useful information for satellite and models evaluation, and contributing to the objectives of several international initiatives (Illingworth et al., 2018) such as the EU COST Action TOPROF (Towards operational ground-based profiling with ceilometers, Doppler lidars and microwave radiometers for improving weather forecasts) or the E-PROFILE program of the European Meteorological Services Network.
Authors contribution

GT analyzed the data and wrote the manuscript, ME operated the MSA in-situ station, RR performed the GRASP retrievals, AC processed the ceilometer data in the frame of ICENET, YS operated the sun/sky photometer at MSA, OD provided feedback on the GRASP algorithm, AA designed the experiment, MP operated the ceilometer, designed the experiment. All authors provided comments on the manuscript.

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References

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Figure 1: Scatter plots of the hourly averaged aerosol light backscattering, scattering and extinction coefficients determined with GRASP pac from the ceilometer and photometer data at MSA height versus the measured in-situ coefficients. This comparison is restricted to situations with low ambient RH ($R_{\text{H ambient}} < 50\%$). The linear regression and the 1:1 line are also shown.
Figure 2: Histograms of the relative absolute difference between in-situ measured and retrieved with GRASP\textsubscript{pac} optical parameters (aerosol light backscattering, scattering and extinction coefficients) at low ambient RH (RH\textsubscript{ambient} < 50%).
Figure 3: Scatter plots of the hourly averaged aerosol light scattering and extinction coefficients measured in-situ (left panel) and retrieved with GRASP$_{pac}$ algorithm (right panel). The color scale represents the relative difference in the single scattering albedo, SSA, between in-situ and GRASP$_{pac}$ data.
Figure 4: (a) Scatter plot of the hourly averaged aerosol volume concentration determined with GRASP_{pac} from the ceilometer and photometer data at MSA height versus the in-situ concentrations at low ambient RH (RH_{ambient} < 50%), with the color scale representing the contribution of fine particles to the total volume concentration. (b) Same than (a) but restricted to situations with contribution of fine particles to the total aerosol volume concentration < 75% (V_{fine} / V < 0.75). (c) Frequency of occurrence of the relative absolute difference between the volume concentrations measured in-situ and determined with GRASP_{pac} for situations with V_{fine} / V < 0.75.
Figure 5: Seasonal variability of vertical profiles of aerosol light-extinction coefficients at 675 nm. The line represents the median and the shadowed area is the interquartile range. The dashed-lines represent the 10th and 90th percentiles. Seasonal statistics are based on daily averaged profiles. Spring corresponds with March, April and May; Summer with June, July and August; Autumn with September, October and November; Winter with December, January and February.
Figure 6: Particle light-extinction coefficient profiles at 675 nm classified by air mass origin (ATL = Atlantic, REG = Regional, MED+EU = Mediterranean and European, NAF = North African). The line represents the median and the shadowed area is the interquartile range. The dashed-lines represent the 10th and 90th percentiles. Statistics are based on daily average profiles.
Figure 7: Bar-plot of the center of mass in m a.s.l. (note that MSA observatory is at 1570 m a.s.l.) of the 25th percentile (P25), median and 75th percentile (P75) profiles, separated as a function of air mass (ATL = Atlantic, REG = Regional, MED+EU = Mediterranean and European, NAF = North African).