Anonymous Referee #2

Received and published: 11 August 2019

Authors would like to thank the reviewer for his/her interest in the work and the constructive suggestions. Corrections allow us to clarify the method used for clustering and highlight the main goals of the paper.

The atmospheric stability is used as a tracer to differentiate the atmospheric layers. As recommended, Stable Layer (SC) and Turbulent Layer (TL) are have been replaced by Stable Conditions (SC) and Turbulent Conditions (TC). This was also requested by other 2 reviewers.

In the following, we provide answers to the reviewer's comments and list the modifications made in the manuscript.

This work reviews the optical properties of aerosol sampled in a mainly free tropospheric site in Bolivia for a period that spans 4 years. The authors have performed a very comprehensive and thorough analysis of their results categorizing the optical properties of aerosol in the area based on the layer sampled (FT or PBL), based on source region and on seasons. The manuscript provides a rather complete picture of the aerosol optical properties of Chacaltaya with the only information missing is the composition of the measured particles. Even though, it is understood that such information (on composition) cannot be included in this work, a short summary would be more than welcome. I recommend that this work is published with only some minor additions which I list below.

Please add a table and summarize in a small paragraph what type of particles (dust,urban, ..etc) are expected to be sampled in each season, layer and source region based on the types of categorization performed in this work. This information is available, but scattered throughout the manuscript and if you compile into one small paragraph the reader will be greatly assisted in understanding your work.

Answer: Three tables have been added to the text which summarize information from the text. The Table 1 summarizes ranges of Angström exponent for the different aerosol types. Table 2 details the median values of the Angström exponent for each cluster, season and atmospheric stability. Table 3, on the conclusion, suggests a new Angtsröm exponent definition for the different aerosol types.

Modifications:

Aerosol type	SAE	AAE	SSAAE
Dust	Close to 1	Close to 1	Below 0
Urban pollution	Close to 2	Close to 1	Higher than 0
Biomass burning		Close to 2,1	

Table 1: Expected aerosol type and their optical properties for each cluster according season and atmospheric stability.

Cluster	season	SAE	AAE	SSAAE	Aerosol types
NA	WET	2,04 (1,42)	0,58 (0,56)	0,18 (0,15)	urban (dust/urban)
	DRY	1,91 (1,80)	1,00 (1,01)	0,01 (0,004)	urban (dust)
	BB	1,92 (1,87)	1,10 (1,26)	0,03 (0,02)	dust/BB (dust/BB)
SA	WET	1,2 (1,40)	0,74 (0,68)	0,11 (0,11)	urban (urban)
	DRY	1,69 (1,70)	1,04 (0,96)	0,02 (0,03)	dust (dust)
	BB	2,16 (2,02)	1,23 (1,20)	0,005 (0,01)	BB (BB)
LP	WET	1,71 (2,09)	0,86 (0,82)	0,08 (0,10)	urban (urban)
	DRY	1,64 (1,74)	1,05 (1,07)	0,02 (-0,01)	urban (dust/urban)
	BB	1,49 (1,93)	1,09 (1,29)	-0,02 (-0,02)	dust (dust/BB)
ATL	WET	1,93 (2,11)	0,75 (0,65)	0,11 (0,15)	urban (urban)
	DRY	1,77 (1,94)	1,00 (1,05)	-0,001 (0,006)	dust (dust/urban)
	BB	1,80 (1,81)	1,23 (1,08)	0,008 (0,01)	dust/BB (urban)
APO	WET	2,15 (2,04)	0,84 (0,82)	0,11 (0,10)	urban (urban)
	DRY	1,39 (1,38)	1,06 (1,10)	0,006 (-0,02)	dust (dust)
	BB	1,56 (1,61)	1,14 (1,20)	-0,008 (-0,01)	dust/BB (dust/BB)
NES	WET	2,05 (1,67)	0,72 (0,66)	0,13 (0,12)	urban (urban)
	DRY	1,74 (1,83)	1,06 (1,09)	-0,008 (0,003)	dust/urban (dust)
	BB	1,89 (1,80)	0,95 (1,07)	0,002 (0,02)	dust/urban (urban)

Table 2: Median aerosol Angström exponents of turbulent condition (stable condition) for each cluster and seasons measured at the CHC station and resulting aerosol types.

Aerosol type	SAE	AAE	SSAAE
Dust	-	> 0,9	[-0,05 ; 0,05]
Urban pollution	> 1,4	< 0,9	> 0,05
Biomass burning	-	> 1,1	[-0,05 ; 0,05]

Table 3: Updated Angström exponent values expected for aerosol types at the CHC station.

l.127: "As a summary, Table 1 shows expected Angström exponent for dust, urban pollution and Biomass Burning particules according the different referenced works (Dubovik et al., 2002 ; Collaud Coen et al., 2004 ; Clarke et al., 2007 ; Russel et al., 2010). This information has to be taken with caution since source influences are expected homogeneous and have been reported from several regions."

l.482: "Table 2 summarizes the median Angström exponents measured at the CHC station for turbulent conditions (stable conditions in parenthesis). According to these values and as discussed above, aerosol types for the turbulent conditions (and stable conditions in parenthesis) are given."

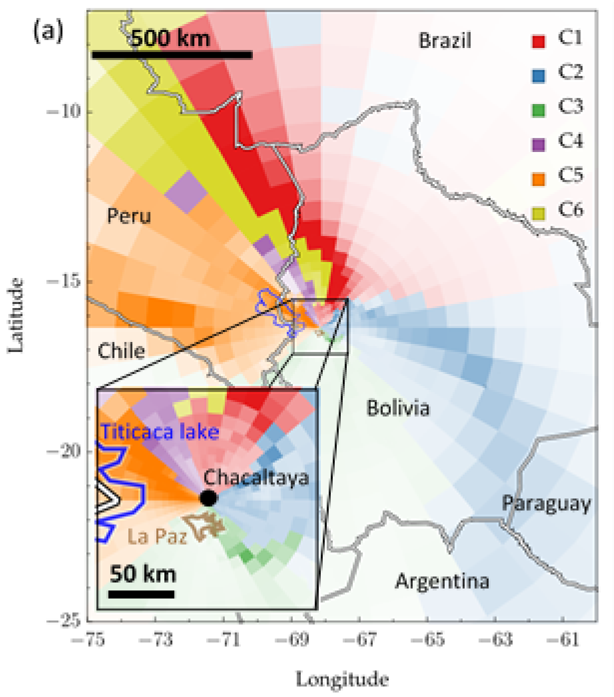
l.525: "A new Angsrtöm exponent classification can then be defined for measurement at the CHC station and is reported Table 3. Thresholds are close to the ones proposed by previous works (Dubovik et al., 2002 ; Collaud Coen et al., 2004 ; Clarke et al., 2007 ; Russel et al., 2010) but adapted to CHC's instruments and particular atmospheric conditions."

The source region analysis performed in this work is puzzling. I am not sure how source regions have been distinguished. As an example C6 and C4 seem to overlap on Fig 4b. The same holds for source regions C1 and C2. There is a second graph in the lower left corner of Fig 4b for which I could not find any explanation. What is this graph about and how it is different than the main one of Fig4b? Please improve the caption of Fig 4 to include all information so that the reader can decipher the plots easily. Some of the info required to do so are found in the text but definitely the info provided is not enough. Since you have performed this analysis for 4 years, the individual trajectories for each source region should be shown in a separate (for each source region) graph in the Appendix. Please also add another plot showing the average trajectories for each source region on a map. Hysplit has this ability to produce average trajectories and so other software that are free to use. Personally I would recommend that the average trajectories graph for each cluster to be included in Fig 4. However it is not mandatory.

Answer: Cluster definition is based on the statistical method described by Borge et al. (2007) now better described in the text. Hence, the six clusters correspond to the main directions of the BTs as shown by the map figure 4. C1 and C2 show clear difference between their main origins (less transparent cells in Figure 4a). About C4 and C6, the difference is much on the distance range from CHC station which can be seen on the zoom in on the lower left corner. C4 corresponds to shorter distance (within 100 km) and C6 corresponds to longer distance. Maps of the first 10% BTs of each cluster and selected for the study are added in the appendix and clearly helps to visualize these features.

Modifications:

Figure 4 has been improved according RC1 recommendations. Scales have been added and geographical references improved.



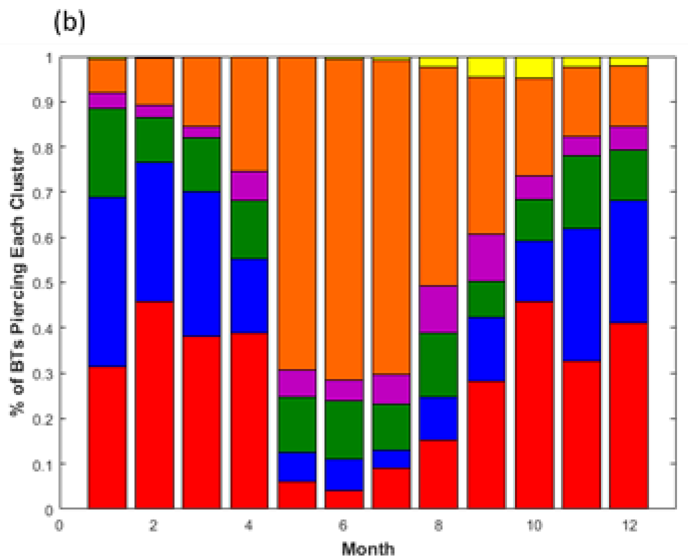


Figure 4: a. Trajectory frequency plot in La Paz region in Bolivia, with a scale of 500 km and 50 km in the lower left corner centered on CHC station. b. Monthly variation of the percentage of back-trajectories (BTs) for each cluster.

Appendix A1:

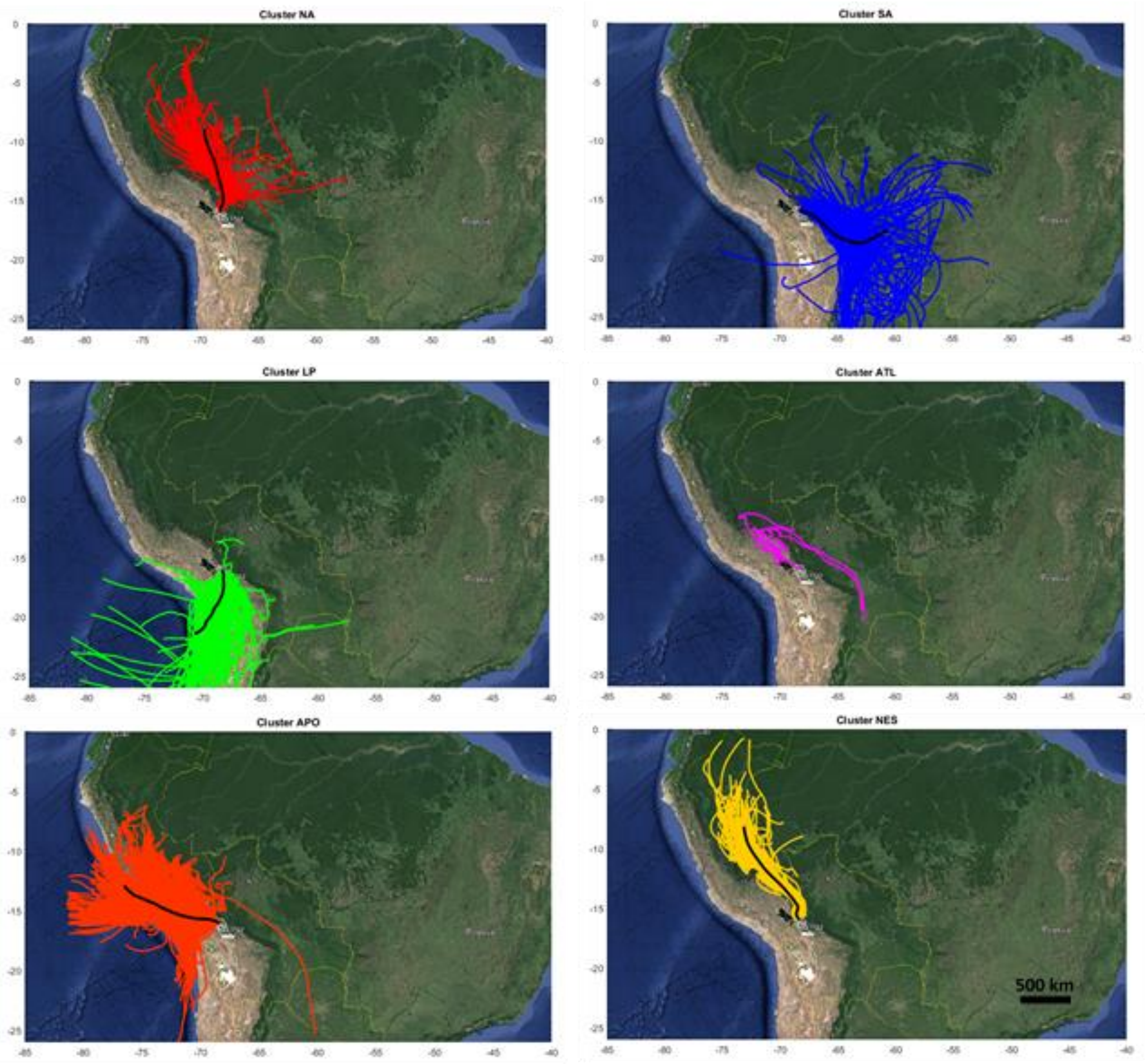


Figure A1: Selected 96-hours back-trajectories for the six clusters obtained from the Borge et al. (2007) method. The black line corresponds to the main back-trajectory.

l.554: "For each hour of the period of the study, nine back-trajectories have been used to describe the mean influence at Chacaltaya station. The nine BTs start within a square of 2 km by 2 km around the station. The mean BT has been calculated from these nine BTs and generated every hour from January 2012 to December 2015. Clusters are defined according the Borge et al. (2017) method using a two-stage technique (based on the non-hierarchical K-means algorithm). The Borge et al. (2007) method allows to attribute to each mean BT a fraction of each cluster according to their time residence into the cluster and their distance from the CHC station. Hence, BTs are sorted according to their representativeness in each cluster. The first 10% of them are used in the present study and are reported in Fig. (A1)."

l.301: "Finally, cluster 6 (NES) has properties close to cluster 1 but with less influence from the Amazonian Basin and close to cluster 4 but with aerosol sources further from CHC station (> 100 km)."

There is a problem with the term ε in Eq 11. Is sa and ca are the sine and cosine of the same angle then by definition $\varepsilon=0$ regardless. This is due to the well known formula of $\cos2\theta+\sin2\theta=1$. I suspect a typo.

Answer: s_a and c_a are the average value of sinθ and cosθ in the 15 minutes time interval. As described in Yamartino et al. (1984), $s_a \neq \sin\theta_a$, $c_a \neq \cos\theta_a$ and $s_a^2 + c_a^2 \leq 1$.

Modifications:

l.231: Definition of ε has been corrected : " $\varepsilon = \sqrt{1-(s_a^2+c_a^2)}$ "

l.232: definition of sa and ca have also been corrected : "with the averages $s_a = \frac{1}{N}\sum_{i=1}^{N} sin\theta_i$ and $s_a = \frac{1}{N}\sum_{i=1}^{N} cos\theta_i$ of N the number of horitontal wind direction (θi) recorded in 15 minutes."

In addition if σθ corresponds to the 15 minute average wind direction as stated in Line 179 what is the θ(15), θ(30), θ(45), θ(60) of Eq. 10. I thought they denoted different time intervals. Please spend some effort to explain further how the classification shown in Fig.2 is performed. In other words explain further what is discussed in Lines 181 and 182.

Answer: $\sigma_{\theta(15)}^2$ corresponds to the squared standard deviation of the horizontal wind direction calculated every 15 minutes. These quantities are later hourly averaged.

Indeed, atmospheric layer definitions are largely discussed on aerosol and dynamic studies, and according to the method used, the definitions of the layers are slightly different. In the present study, because we use atmospheric stability as tracer for atmospheric layers, it is more appropriate to use the two different regime "stable conditions" and "turbulent conditions".

Modifications:

In the full document, "layer" has been replaced by "condition" when needed, and SL – TL has been replaced by SC – TC (Stable and Turbulent Conditions).

l.220: "In addition, a residual layer can also be present at CHC station during nightime, resulting from low dispersion of the daytime convection. Because no clear distinctions between the mixing, the free tropospheric, and the residual layers can be strictly obtained from in-situ measurements only, the present dataset recorded at Chacaltaya station is separated in terms of stability conditions (turbulent and stable)."

l.227: "This method is based on the hourly averaged value of the standard deviation of the horizontal wind direction (σ_θ in Eq. 10) calculated every 15 minutes"

l.229: "with $\sigma_{\theta(15)}$ the standard deviation of the horizontal wind direction calculated on the first 15 minutes of every hour, and $\sigma_{\theta(60)}$ the last 15 minutes of every hour."

l.237: "As described in Rose et al. (2017), the classification depends also on the σ_θ value in the 4-hour time interval across the time of interest. Interface cases correspond to unclassified data which mainly show a high variability of the standard deviation between the two categories of dynamic. For clarity, the interface cases are excluded from the dataset in the rest of the paper."

Despite that most of this work relates to phenomenology, there are two important findings. These are the very low AAE reported during the wet season and the linear relationship between SSAAE and AAE observed during the wet and dry seasons. I am wondering if such low AAE have been reported elsewhere in literature. Please discuss. Can the authors provide an explanation on the linear relationship observed in Fig9a?

Answer: The present study shows AAE reaching 0.5. These values are also observed by Russel et al. (2010) and corresponds to "urban industrial" impacts. AAE values from 1,5 to 3 correspond to dust particles.

The linear relationship between SSAAE and AAE can mainly be explained by a similarity on the sensibility to the two properties to two aerosol type. When air masses are mainly influenced by urban particles, especially during the wet season for every clusters, AAE values are close to 1 (or lower than BB influences) and SSAAE values are higher than 0. In the other hand, when air masses are mainly influenced by dust and BB particles (dry season for every clusters), AAE values are close to 2 (or much higher than during urban influences) and SSAAE values are close or below 0. A mixture between these two aerosol types will lead to intermediate values located in line with them.

Modifications:

l.445: "As shown in Fig. 5, low AAE values, especially during the wet season, can be explained by important reduction of dust and less biomass burning particles due to more efficient removal."

l.450: "Thus, the wet season presents positive SSAAE and AAE close or lower than 0.9, while dry season and BB period present SSAAE close to 0 and AAE higher than 0.9. A linear relationship between AAE and SSAAE values is observed and illustrates that mainly urban emissions drive aerosol particle properties during the wet period, and that mainly dust emissions drive aerosol particle properties during the dry season and the BB period."

There is a typo in the caption of Appendix Fig A1. Aerosol should probably be absorption and for the entire dataset instead of the all dataset

Answer: True

Modifications:

"Figure A2: Weekly variation of the Absorption Angström Exponent (AAE) for the whole dataset from 2012 to 2015. The medians and their 25th and 75th percentiles from Sundays to Saturdays are represented. "

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-510, 2019.

References:

Borge, R., Lumbreras, J., Vardoulakis, S., Kassomenos, P. and Rodríguez, E.: Analysis of long-range transport influences on urban PM 10 using two-stage atmospheric trajectory clusters, Atmos. Environ., 41(21), 4434–4450, 2007.

Clarke, A., McNaughton, C., Kapustin, V., Shinozuka, Y., Howell, S., Dibb, J., Zhou, J., Anderson, B., Brekhovskikh, V., Turner, H. and Pinkerton, M.: Biomass burning and pollution aerosol over North America: Organic components and their influence on spectral optical properties and humidification response, J. Geophys. Res. Atmospheres, 112(D12), D12S18, doi:10.1029/2006JD007777, 2007.

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O. and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmospheric Measurement Techniques, 3(2), 457–474, doi:10.5194/amt-3-457-2010, 2010.

Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanre, D. and Slutsker, I.: Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atmospheric Sci., 59, 590–608, doi:Review, 2002.

Rose, C., Sellegri, K., Moreno, I., Velarde, F., Ramonet, M., Weinhold, K., Krejci, R., Andrade, M., Wiedensohler, A. and Ginot, P.: CCN production by new particle formation in the free troposphere, Atmospheric Chemistry and Physics, 17(2), 1529–1541, 2017.

Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O. and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, Atmospheric Chem. Phys., 10, 1155–1169, doi:10.5194/acp-10-1155-2010, 2010.

Yamartino, R. J.: A comparison of several “single-pass” estimators of the standard deviation of wind direction, J. Clim. Appl. Meteorol., 23(9), 1362–1366, 1984.