

Final response

Our responses are marked in *italic and blue* and were directly inserted below each comment of the referees. Changes in the manuscript are described below each comment and are highlighted in the attached version of the paper.

We thank both referees for their helpful comments and the time they spent with the review.

5 Comments from Referee #1

1. Introduction: The authors list studies done for many different high-altitude sites. However, one observatory which is not mentioned at all is Pic du Midi in the French Pyrenees. There was a recent study by Hulin et al. (2019) on atmospheric composition and the detection of thermally driven circulations with different methods, which should be cited as well.

Thanks for pointing out that study. In the new manuscript version, the study is cited in the introduction.

10 2. p. 4, l. 22: "available at 1 min intervals". Are the values averaged over 1 min intervals or instantaneous?

The values are averaged over 1 min intervals. In the new manuscript version, we use the phrase "available as 1 min averages".

3. p. 5, l. 2: What is the temporal resolution of the meteorological data?

15 *The standard meteorological data was available as 10 min averages except at the Schneesfernerhaus where 1 min averages were available. This is now mentioned in the manuscript.*

4. p. 5, l. 3-5: How are the aerosol layer heights detected? With the manufacturer software or with a algorithm developed by the authors?

The manufacturer software was used, which is now mentioned in the manuscript.

5. p. 5, l. 4: ">1": What is the unit? dB, B,...?

20 *This statement was taken from (Heese et al., 2010) who expressed the signal-to-noise ratio in a dimensionless number.*

6. p. 5, l. 7: Is daylight saving time taken into account?

We checked whether the time stamp referred to daylight saving time during the summer half year but this was not the case. However, some measurements were recorded with UTC time, which was converted to local standard time.

7. p. 5, l. 8: Why did the authors use a threshold of 66 % and not something else for the availability?

25 *We agree that the minimum data availability of 66 % per interval is somewhat arbitrary. However, it guarantees that the averaged values are representative for most of the time interval and not only for a small part of the interval in the case of data gaps.*

8. p. 6, l. 1ff: Why is there a time offset between the sites? Where the sites not synced to a time server? If the sites are not synced was there a shift of the time offset with time? E.g. what is the difference between Sept. 2013 and March 2014? How high is the correlation coefficient?

30 *After contacting the manufacturer of the weather stations, we realized that the time stamps are synced to a time server and should not differ much between the sites. The initially supposed time offsets based on crosscorrelation may be due to an inexact horizontal alignment of the pyranometers. Therefore, we repeated the analyses without shifting the time and updated the text and the figures. This had a negligible effect on $\Delta\theta_v$ and the statistical air mass classification because initially, a time offset had only be supposed for the sites ZPLT, Schachen, Kreuzalm, and Felsenkanzel and not for GAP, Brandwiese, and ZSG. Omitting the time shift, had a small effect on the mechanistic classification and slightly improved the agreement between the mechanistic and the statistical classifications.*

9. p. 6, l. 19: "exp" should not be italic.
Thanks, this is now corrected.
10. p. 7, l. 16: Why only 89 % at the beginning? What happened to the remaining 11 %.
During 11 % of the time, the ceilometer could not determine any aerosol layer height, most likely due to fog or precipitation.
- 5 11. p. 8, l. 2: Why four classes? In the following lines only three air mass classes are defined?
"Four" was a mistake, it should be three. We have corrected this now.
12. p. 8, l. 24: What are "most suitable variables"? How are they defined? What are the criteria?
To be more precise, we changed the phrase to "variables with an expected unambiguous link to vertical transport processes and a high data availability". These variables include CO, CH₄, CO₂, O₃, specific humidity, air pressure at Garmisch-Partenkirchen, and $\Delta\theta_v$ and are now mentioned directly afterwards in the manuscript.
- 10 13. p. 9, l. 17: The figures should be referred to in the correct order, i.e. 8 not before 4.
Figure 8b is now Fig. 3b so that the figures are referred to in the correct order.
14. p. 9, l. 19: What are the remaining measurements?
The remaining measurements refer to the gases NO_y, NO_x, ²²²Rn, ⁷Be, HCHO, ²²²Rn, the aerosol quantities N₉₀, eBC, PM₁₀, and the standard meteorological variables precipitation, relative humidity, temperature, global radiation. Because these measurements are already mentioned in Sect. 2.2, the new manuscript version summarizes them as "remaining chemical (e.g. NO_y, NO_x, ²²²Rn) and standard meteorological measurements (e.g. precipitation, relative humidity)".
- 15 15. p. 9, l. 25: I was surprised to read about a marine boundary layer considering the location of Zugspitze. Where does the marine air mass come from? The authors speculate about that later in the manuscript, but I think a hint about its origin should already be given here.
We adopt this suggestion and now mention the Atlantic Ocean as a potential origin of air masses in that part of the manuscript.
16. p. 10, l. 8ff: What is the typical stability distribution? This could e.g. be checked with a histogram.
We address this question by adding a short section in the supplement, including histograms of $\Delta\theta_v$ for the periods June–July and December–January (Fig. S1a). Note that we have additionally changed the sign convention for $\Delta\theta_v$ (see comment 13 of Referee #2). $\Delta\theta_v$ was almost always positive, indicating stable conditions. In June and July, $\Delta\theta_v$ ranged between –5 K and +21 K whereas in December and January, it was generally more positive with values between 4 K and 31 K. Additionally, we included the histogram, from which the threshold of $\Delta\theta_v = 8$ K between anabatic and katabatic winds was determined (Fig. S1b).
- 25 17. p. 10, l. 12: GAP is a valley floor station. Why is this station used to detect strong synoptic forcing? Wouldn't it make more sense to use high-elevation sites?
We restricted the wind velocities at ZPLT and GAP to $< 3 \text{ m s}^{-1}$ when detecting katabatic winds. This criterion was based on the idea that katabatic winds are favored by a weak synoptic forcing and are expected to have low wind velocities. After applying the wind direction and stability criteria for katabatic winds, the wind velocity still ranged up to approximately 10 m s^{-1} at ZPLT while it was almost always $< 3 \text{ m s}^{-1}$ at GAP (Fig. R1). Cases with high wind velocities ($\geq 3 \text{ m s}^{-1}$) were suspected to be synoptically driven and were discarded. For almost all of these cases, the wind velocity threshold was exceeded at the high-elevation site ZPLT, not at GAP. For the reason of consistency, the wind velocity threshold was also applied to GAP, even if only very few cases were affected.
- 35

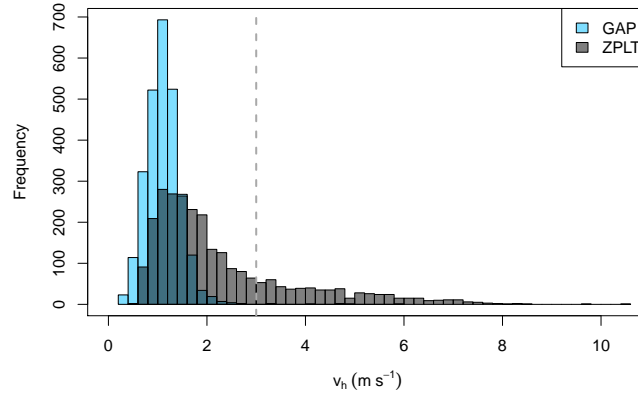


Figure R1. Histograms of the horizontal wind velocities (v_h) at GAP and ZPLT for potential katabatic winds, i.e. for downvalley wind direction sectors at both stations and strongly stable conditions ($\Delta\theta_v \geq 8$ K). The dashed line shows the threshold of 3 m s^{-1} for katabatic winds.

18. p. 10, l. 16: Why does condition a) requires anabatic wind OR UFS below MLH_{GAP} ? Why not AND?

The mixing layer height can be heterogeneous, especially in mountainous terrain. MLH_{GAP} is measured in the center of the valley atmosphere. Due to thermally driven upslope winds, boundary layer air can reach higher altitudes at the mountain slopes compared to the valley center (Henne et al., 2004; Gohm et al., 2009). Thus, the UFS could be influenced by the boundary layer while MLH_{GAP} is below the UFS level. Additionally, a residual layer could result in boundary layer air masses at the UFS during night while anabatic winds are absent.

19. p.10, l. 30-31: The authors state that PC1 was always a meaningful indicator while PC2 did not always allow for an unambiguous interpretation. On what is that assumption based?

This statement is explained in the following paragraph in which the PC loadings are discussed. To make that clear, we inserted the phrase ", which will be explained in the following".

20. Sect. 3.1: I found this part of the result section hard to follow and it might be difficult for readers not familiar with PCA to understand the interpretation of the results. It might be helpful to give a detailed example at the beginning on how to read and understand the loading diagrams in Fig. 6. What does a low score mean? Low absolute values or large negative values? In the text (p. 11, l. 1-6), the authors talk about scores while in Fig. 6 loadings are shown. How does this relate?

With low score, we mean large negative linear combinations according to Eq. 5. In the new manuscript, we explain that variables with a high absolute loading largely determine the PC scores and give an example on how loadings affect PC scores. If an original variable is much higher than its 2-month mean value and its loading on PC1 is strongly negative then the variable will strongly contribute to a large negative score of PC1.

21. p. 11, l. 32: "... air masses (Fig. 7a-g): compared ..." To make clear that the explanation why it was consistent is following.

We adopted the suggestion of inserting a colon.

22. p. 14, l. 15: This additional criteria of no clouds below 4 km, should be moved to Sect 2.5.

As suggested, we moved this criterion to Sect. 2.5, in which the mechanistic approach is described.

23. p. 14, l. 21: " .. this winds were not thermally induced BUT the MLH_{GAP} suggested non-ML air masses." BUT does not make sense. It should be AND.

We agree and replace BUT by AND. The same applies to two similar sentences in the same section and in Sect. 2.5.

24. p. 14, l. 28ff: What about LRMD and MBL/UFT air mass classes?

- 5 *LRMD and MBL/UFT are subclasses of HYBRID. To make that clear, we write "HYBRID including LRMD and MBL/UFT" in the new manuscript version.*

25. p. 15, l. 27: What differed between the six 2-month periods?

- 10 *The first two principal components, their interpretation, and thus, the mapping of air mass regimes to air mass classes differed between the 2-month periods. We changed the sentence into the following one: "but the principal components and their interpretation differed between the six 2-month periods".*

26. Fig. 7: Why not stick to the numbers I-IX for the regimes instead of introducing the long names for the regimes. This would make it more comparable with Figs. 3 and 8. The colours for the air mass classes should be brighter like in Fig. 8 and 9. Adjust the scales of the subplots to make them clearer to read (e.g. Figs. 7o, 7p, 7q, 7r).

- 15 *To make Fig. 7 more comparable with Fig. 3 and 8, we replaced the long regime names by the numbers I to IX. The same was done for the figures in the supplement. Also, the colours and scales in Fig. 7 were adjusted as suggested.*

27. Fig. 8: I probably understand it wrong, but how can regime VI belong to ML? In Fig. 3 there is no connector between ML and regime IV. Also, how can I and II belong to HYBRID? It would be good to enhance the boxes of the label to make the hatched areas better visible.

- 20 *In Fig. 3, the mapping between of air mass regimes to air mass classes is only shown for February and March. In other 2-month periods, the mapping was different, which is now explained in the figure caption. In Fig. 8, the legend is now bigger and the hatched areas are better visible.*

28. Fig. 9: Maybe add "UFS below MLH_{GAP} ..." or "UFS above..." to the label and maybe refer to the text (p. 10, l. 14ff) where the 3 conditions are explained.

We adopted these suggestions.

25 **Comments from Referee #2**

(1) Pre-processing of the data before use in the statistical classification method is limited to standardization (that is, adjustment of the sample mean to 0 and of the sample variance to 1). I am wondering whether any slightly more sophisticated pre-processing could be beneficial.

For instance:

- 30 (a) Some of the variables in the data matrix have well-defined seasonal and diurnal cycles. Would it be possible to determine average annual and daily cycles, and to remove them from the data set? Performing the analysis on deviations from the average cycles might improve classification results.

- 35 *In our analysis, the seasonal course was roughly removed by subtracting the 2-month mean in each of the 2-month periods when standardizing the data. We added a sentence in Sect. 2.4.2 to make that clear. In principle, it would be beneficial to remove the seasonal course more accurately so that the leading principal components would only depend on the shorter-term variability of the time series. In the beginning of the data analysis, we considered the determination and removal of the seasonal course with a spectral approach such as a wavelet filter. However, this idea was abandoned because it requires knowledge of the time series within a window centered at the time of interest and is thus not applicable to real-time operational mode. It would be possible to remove an average seasonal cycle from each variable based on a larger, multi-year data set. Due to interannual variability, however, this approach would only partly remove the seasonal course. For example, Scheel et al. (1999)*
- 40

showed that the monthly mean O_3 mixing ratios at Mt. Zugspitze can strongly differ between individual years and the 10-year ensemble. Therefore and because a large fraction of the seasonal variability is already removed by subtracting the 2-month mean values, we doubt that the removal of an average seasonal cycle would substantially improve the classification results.

The mean diurnal cycles of the analyzed variables reflect shifts between air masses that we aim to identify. Afternoon maxima of atmospheric constituents such as water vapor, CO , ^{222}Rn , and NO_y at high-alpine sites have been explained by thermally induced uplift processes including anabatic winds (Forrer et al., 2000; Zellweger et al., 2003; Griffiths et al., 2014). These conditions are typically accompanied by afternoon minima of atmospheric stability and air pressure. The latter is associated with plain to mountain winds in the Northern Alps (Lugauer and Winkler, 2005). Removing the average diurnal cycle would complicate the classification of air masses in the case of thermally induced vertical transport.

(b) PCA does not require the data to follow multivariate normal distributions, but its results can often be interpreted more easily if they do. It strikes me that most variables are concentrations, therefore their PDF will certainly be markedly non-Gaussian. Would a cleverly designed variable transformation allow bringing more variance into the leading principal components?

Among the PCA input variables, especially CO , CH_4 , and in the winter half year also CO_2 tend to have right-skewed PDFs. A logarithmic transformation, for example, would reduce the influence of the "long tails" of the PDFs on the principal component loadings. However, a variable transformation would not completely remove the "long tails" because the PDFs rarely follow an idealized distribution such as a log-normal distribution and they vary with the 2-month periods. Additionally, a variable transformation has the following side effect. If a logarithmic transformation is used for example and the original variable increases by a certain amount of units, then the transformed variable will not increase by a certain amount of units but by a certain factor. Although a variable transformation might somewhat increase the fraction of variance explained by the leading principal components in some 2-month periods, we think that it would not significantly improve the classification of the entire data set.

(2) The matching between air-mass regimes (I-IX) and air-mass classes (ML, UFT/SIN, HYBRID) is different in each two-month period (see Figure 8). The manuscript text contains little or no information about the overarching logic. Why was this necessary? What criteria were used to attribute regimes to classes, how did these criteria change with the season?

In my opinion, the ad-hoc tuning of the method is a serious shortcoming. It is clearly a subjective component of the classification, and as such it cannot be exported to other sites. The authors do not explain this point in a satisfactory manner, and they probably should. Why wasn't it possible to design a fully objective classification rule? Formal methods to identify classification rules exist and could be used (see for instance chapter 14 in Wilks, 2011, Statistical Methods in the Atmospheric Sciences. DOI: 10.1016/B978-0-12-385022-5.00014-2).

We realize that the mapping of air mass regimes to air mass classes was only shortly described in the manuscript. Now, more details are included in Sect. 2.4.3. The loadings of the leading principal components changed with the 2-month periods (Fig. 6), which can be explained by seasonal changes in chemical processes (e.g. CO_2 emissions and uptake, photochemical O_3 production) and atmospheric dynamics (e.g. thermally induced uplift). Therefore, the characteristics of the air mass regimes (I-IX) changed with the 2-month periods and required a separate interpretation in each 2-month period. The air mass regimes were assigned to the air mass classes by visually comparing boxplots of the original input variables between the air mass regimes (Fig. 7). In the winter half year, the class ML (now renamed as BL, see comment 4 of Referee #2) was assigned if CO , CH_4 , CO_2 , and q were relatively high and $\Delta\theta_v$, p_{GAP} and O_3 were relatively low compared to the other regimes; note that the sign of $\Delta\theta_v$ was changed so that low values indicate a low static stability (see comment 13 of Referee #2). The class UFT/SIN was assigned in the opposite case and the remaining regimes were assigned to the class HYBRID. Apart from CO_2 and O_3 , the same criteria were used in the summer half year; CO_2 was required to be relatively low for the class BL and O_3 was not considered.

We agree that this subjective mapping of regimes to classes is a shortcoming. Nevertheless, it is based on typical qualitative differences between lifted and subsided air masses. The present study has the character of a pilot study to develop a novel multivariate approach for air mass classification. In future studies, a more objective and robust mapping of regimes to classes could be achieved by using a metric such as the difference between the median of a regime and the overall median to define a threshold for "relatively high/low" characteristics and by using data from multiple years or more observatories. The supervised

classification methods, which are described in Chapter 14 in Wilks (2011), could only be applied to our problem if a reliable test data set with known air mass classes was available and included a variety of meteorological conditions.

(3) Note: the line numbering in pages 2-end is wrong, i.e., the 6th line from the top is labelled as "5" and so on, as if the first line were 0. In what follows, I'm using this unusual "zero-based" system.

5 *This issue has been corrected.*

(4) Nomenclature. The first air-mass class is labelled ML, for "mixing layer". I'm wondering if this is appropriate. A mixed layer, by definition, has nearly adiabatic lapse rate. The boundary layer (BL) is not always well-mixed, especially at night. On page 1, line 12, it is stated that "the terms ML and BL can be defined synonymously ...". In my mind, the two concepts are quite distinct. I'd rather say that the BL might sometimes include a ML. I don't really have a strong opinion on this matter, but anyway I suggest renaming the first air-mass class to BL, for "boundary layer". A similar comment applies to "mixing layer height" (MLH). This should probably become "boundary layer height" (BLH), in particular because, according to the description of the wavelet detection algorithm, MLH/BLH potentially includes multiple aerosol layers. These typically develop in connection with inversion layers, i.e., non-mixed parts of the atmosphere.

15 *We agree that the definitions of the boundary and mixing layers are based on different concepts. The boundary layer (BL) is affected by surface forcings such as the turbulent transfer of momentum, heat, and matter while the mixing layer specifically refers to the dispersion of surface-emitted atmospheric constituents (Stull, 1988; Seibert et al., 2000). In the new manuscript version, we cite definitions of the boundary and mixing layers. We point out that we consider a residual layer and elevated aerosol layers, which were influenced by the surface within a time scale of one diurnal cycle, as parts of the boundary and mixing layers, as suggested by Reuten et al. (2007). Because the term mixing layer is usually restricted to the well-mixed layer adjacent to the surface, we renamed the air mass class ML as BL. However, we keep the term mixing layer height (MLH) because the ceilometer measures the aerosol backscatter profile, which reflects the dispersion of surface-emitted particles.*

(5) Page 1, title. "Discrimination" or "classification"? The two terms have slightly different meanings. See again chapter 14 in Wilks 2011.

25 *We now use the word "classification" instead of "discrimination" because discrimination would be based on training data, for which the groups (air mass classes) are already known, while classification refers to the attribution of test data to groups (Wilks, 2011).*

(6) Page 1, lines 8 and 12. Use of the word "classifiable". I believe these statements should be formulated more clearly. As they are now, they seem to allude to the intrinsic "ability" of the methods to separate the events, and seem to suggest that the statistical method permits to obtain a meaningful classification much more often than the mechanistic one (78 % of the time as opposed to 25 %). Instead, these two percentage only represent the availability of the input data for the two methods. Please use something like: "Due to data gaps, only x % of the investigated year could be classified".

To avoid misunderstandings, we avoid the expression "classifiable cases" and now use the phrases "... input data was available in 78 % ..." and "Due to data gaps, only 25 % of the cases could be classified ...". In the rest of the paper, we use "classified cases" instead of "classifiable cases".

35 (7) Page 2, lines 10-11. Foehn flows are listed among processes that cause air mass lifting. This is inexact and quite confusing. Foehn is a fall wind: its dynamics are inextricably tied to air-mass descent (not lifting!) on the lee side of a mountain range. That said, intense foehn certainly causes mechanical mixing of the lower atmosphere, which may result in transport of chemical species from the PBL to the free troposphere. Zellweger et al (2003, cited in the manuscript) list foehn among the meteorological conditions in which free-troposphere air masses are mixed with PBL air masses (page 781, top of second column).
40 Correctly, Zellweger et al (2003) do not mention "lifting" in relation to foehn. Please revise. The comment also applies to page 2, line 23 and to page 12, line 15.

In the case of south foehn, Mt. Zugspitze is located on the lee side of the Alps where the air flow descends. Nevertheless, foehn events can be associated with lifting on the windward side of the mountain range and thus transport air masses from the BL to

- high-alpine sites. If the southern Alps experience north foehn, Mt. Zugspitze is located on the windward side of the Alps where the air flow typically ascends and lifts BL air masses. In the new manuscript version, we list foehn among transport processes that cause air mass lifting or mixing. We also mention that foehn winds descend on the lee side of a mountain range and can be associated with air mass lifting on the windward side. On page 12, we now speak of an "influence of BL air masses" with respect to foehn instead of an "uplift of BL air masses".
- (8) Page 2, line 30. "...because the MLH is a meteorological quantity". Wording could be more careful here. MLH/BLH is not a directly measured quantity, but rather an estimate obtained from measurements of other quantities. The determination of MLH/BLH from a vertical profile can be quite tricky, too. I'd rather say something like: "...because determination of the MLH from vertical profiles of measured quantities requires a-priori meteorological knowledge".
- We adopted this suggestion.
- (9) Page 5, line 17. "...set zero" → "...set to zero".
Done.
- (10) Page 5, line 21. Please delete the blank space before the full stop.
Done.
- (11) Page 6, line 17. "...using the Clausius Clapeyron equation". Or rather a numerical approximation? There are many such formulas: Goff-Gratch, Magnus-Tetens, Bolton ... which one?
We used the following form of the Clausius Clapeyron equation (e.g. Wallace and Hobbs, 2006),
- $$\frac{1}{e_s} \frac{de_s}{dT} = \frac{L_v}{R_v T^2}, \quad (\text{R1})$$
- where e_s (hPa) is saturation vapor pressure, T (K) is temperature, L_v (J kg⁻¹) is latent heat of evaporation, and R_v (J kg⁻¹ K⁻¹) is specific gas constant for water vapor. Integration of Eq. R1 yields
- $$e_s(T) = e_s(T_0) \exp \left\{ \frac{L_v}{R_v} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right\}, \quad (\text{R2})$$
- where T_0 (K) is a reference temperature with known e_s .
- (12) Page 6, line 19. I think the standard notation should be either e^x or $\exp\{x\}$. Also, please replace T_v by \bar{T}_v , to indicate the vertical averaging.
- Done.
- (13) Page 7, line 2. I personally find the sign convention counterintuitive. Although static stability conventionally corresponds to $d\theta/dz > 0$, here $\Delta\theta$ is greater than zero when θ decreases with height. Why not computing $d\theta/dz$?
Noting that the sign convention for $\Delta\theta_v$ was counterintuitive, we changed this sign convention. Now, positive values indicate stable conditions. This change corresponds to multiplying $\Delta\theta_v$ by -1 . Consequently, the sign of the PC loadings of $\Delta\theta_v$ changed as well (Fig. 6). Dividing $\Delta\theta_v$ by Δz would be an alternative but would not significantly change the results because Δz does not vary much.
- (14) Page 7, lines 20-21. "The MLH attribution was based on the idea that the MLH varies only gradually". I am not sure this is always appropriate over mountains. Horizontal advection of aerosol layers due to mountain venting can cause spatial and temporal discontinuities in MLH.
- We notice this shortcoming and now mention it in the manuscript. For most of the time, however, we expect a gradual variation of the MLH.

(15) Page 10, lines 9-10. Why using -8 K to discriminate "weak" and "strong" static stability?

The threshold of -8 K (now $+8$ K because of the changed sign convention, see comment 13 of Referee #2) was determined by comparing the histograms of $\Delta\theta_v$ for winds coming from the upvalley or downvalley wind direction sectors at GAP and ZPLT. The histograms are now included in the supplement as Fig. S1b. Using the intersect of the histograms corresponds to
5 *minimizing the number of data points that are excluded from potential anabatic and katabatic winds.*

(16) Page 11, line 29. Are CH_4 and CO_2 pollutants?

Originally, we considered CH_4 and CO_2 as pollutants. However, the paragraph under consideration was removed because the long names of the air mass regimes were replaced by roman numbers. See comment 26 of Referee #1.

Please note that marked-up manuscript versions of the main article and its supplement are included in the supplement to
10 *this response.*

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