Manuscript ACP-2019-211

Multivariate statistical air mass discrimination for the high-alpine observatory at the Zugspitze mountain, Germany

Introduction

The objective of this study is to devise a method for real-time air mass classification based on routine chemical and meteorological measurements at high-altitude sites. Compared to existing classification approaches, which typically consider the concentration of single chemical constituents or at most the ratio of two concentrations, this method adopts a multivariate approach based on principal component analysis (PCA). The study considers one year of nearly continuous measurements. Based on PCA scores, nine different "air mass regimes" are identified. In turn, these regimes are mapped to three "air mass classes", respectively mixed-layer air, free tropospheric/stratospheric air, and air with hybrid characteristics. The mapping from air mass regimes to air mass classes is not rigidly prescribed, it varies seasonally, and it introduces a subjective element into the classification. The results of the statistical classification scheme are compared to those of a so-called "mechanistic approach", where individual cases are classified based on meteorological measurements and a-priori knowledge of local wind circulation patterns. The comparison highlights some limitations of the proposed statistical classification scheme.

Recommendation

The manuscript is well organized, well written and appropriately concise. Figures are of good quality. The subject matter is well within the thematic scope of ACP. I have the feeling that some characteristics and implications of the proposed classification method have not been fully considered (see in particular comments 1 and 2 below). However, given the originality and novelty of the core idea (classifying air masses using a multi-variate method based on chemical and meteorological measurements) the manuscript is probably a good candidate for publication. A request for revisions is recommended.

General comments

(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:
(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.
(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?

(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).

Specific comments

(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 the first line were 0. In what follows, I'm using this unusual "zero-based" system.

(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.

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

(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). 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.

(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".

(9) Page 5, line 17. "...set zero" → "...set to zero".

(10) Page 5, line 21. Please delete the blank space before the full stop.

(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?

(12) Page 6, line 19. I think the standard notation should be either e^x or exp{x}. Also, please replace T_v by $\overline{T}_v$, to indicate the vertical averaging.

(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 $\Delta\theta/\Delta z$?

(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.

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

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