

Answers to referee's comments on "Planetary boundary layer influence at the Jungfraujoch analyzed by aerosol cycles and synoptic weather types" by M. Collaud Coen et al.

The authors thank both referees for their constructive comments that help us to improve the clarity and the quality of the manuscript. In the following, the comments of all referees are answered and the modifications introduced in the revised manuscript are described.

In the following answers to the referee's comments, the figure numbers correspond to the revised version of the paper. Since a new figure with the measuring stations was added, all further figure numbers are incremented by 1.

Anonymous Referee #1

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General comments

This manuscript describes relationship between synoptic meteorological pattern and concentrations of pollutants (aerosols, CO etc) at JFJ. This manuscript contains potentially useful data of seasonal frequency variations of weather types at JFJ, but the current manuscript is lacking new findings after previous publications such as Lugauer et al. (1998; 2000), Zellweger et al. (2003), and Henne et al. (2004; 2005).

This manuscript also fails to show clear aim for further analyzing these data at JFJ.

Several findings presented in this paper are new and were not published elsewhere until now. Lugauer et al. (1998, 2000) published a climatology of the aerosol surface area and Nyeki et al. (1998) a one-year data set of the measurements of the GAW monitoring program. However, the climatology of the optical aerosol parameters has not yet been published. The contribution of each synoptic weather types to the aerosol seasonal cycle has not yet been analyzed either. None of the previously published papers on the PBL influence at the JFJ took into account the new particle formation (NPF) and growth at the measurement site (JFJ) as well as over the Swiss plateau. The consideration of NPF allows explaining some aerosol behavior, particularly the diurnal cycles of N and the scattering coefficient in February-April during synoptic lifting. Finally a climatologic mean of the PBL air export to the JFJ is quantified by the relative increase of CO concentration and the specific humidity.

The referee comment is however justified in the sense that all these new findings and/or unpublished results were not well described in the last paragraph of the introduction. The paragraph (p. 988, lines 13-28) was consequently completely rewritten to clarify the aim of this study.

Methods of data analysis (last paragraph of section 2.2) and weather statistics were not clear to me.

Since this point was mentioned by both referees, the paper was substantially revised to clarify both the methods of analysis and the description of the synoptic weather types.

Discussion on meteorological conditions of new particle formation (NPF) during convective lifting might be interesting to deduce favorable meteorological condition of NPF at this site, but the authors limited to show qualitative discussion and no further analysis was given.

A further analysis of the dependence of the NPF is not within the scope of this paper, since this paper really focuses on long-time series analyses. Measurements by an aerosol mass spectrometer or ion spectrometer were only performed during campaigns and particle size distribution measurements were only recently added to the operational instrumentation. Moreover the NPF evidenced during convective lifting occurred on the Swiss plateau and not directly at the site. Operational measurements of the aerosol parameters related to NPF are

only performed in highly polluted environments on the Swiss plateau (motorway and city) that cannot be used for our study.

Combined above, I recommend major revision of the current manuscript for publication in ACP.

[Title] I feel that the word “aerosol cycles” does not fit with the context of the manuscript.

The title has been modified to **“Aerosol climatology and planetary boundary influence at the Jungfraujoch analyzed by synoptic weather types”**.

[1 Introduction] The aim of this study is not clearly stated. This section needs the reason “to refine” previous reports.

The focus of this study has been more carefully described in the introduction (see answer to the first general comment of the referee). The text has been modified as follows:

“Lugauer et al. (1998 and 2000) analyzed long time series (9 years and 7 years) of the aerosol surface area concentration, aerosol-attached radon decay products and the mass concentration of Total Suspended Particles (TSP) as a function of synoptic weather type (SYNALP) over the whole Swiss region defined by Schüepp (1979). Nyeki et al. (1998) and Weingartner et al. (1999) presented one-year time series of aerosol parameters measured operationally in the GAW program. In this paper, climatologies (14-year data set, 1995-2008) of the optical aerosol parameters (b_{abs} , b_{scat}) and particle number N is presented. The contribution of the SYNALP synoptic weather types to the median aerosol seasonal cycle is determined. Correlations between the aerosol seasonal cycle and the local meteorological conditions are also evaluated. Since aerosols are one of the most sensitive parameters to the influence of the PBL (due to a strong vertical gradient), this paper also presents a study of the transport of air masses from the PBL to the JFJ analyzed by the diurnal cycles of N , b_{abs} , b_{scat} and $\dot{a}_{scat}$ as a function of the year periods and synoptic weather types. The absorption coefficient is mostly determined by the black carbon concentration arising from combustion processes, and is hence more characteristic of polluted air masses. N is the most sensitive parameter to the formation and subsequent growth of new particles, because the number concentration is governed by Aitken mode particles whereas b_{abs} is proportional to an equivalent mass of black carbon and b_{scat} is sensitive to accumulation and coarse mode particles. N is however also sensitive to primary pollution sources. The scattering coefficient is sensitive to both primary pollution and condensational growth of the particles and thus combines characteristics of both b_{abs} and N . In combination with SO_2 and NO_x concentrations (which are precursors for aerosol number and mass concentration, respectively) and with meteorological parameters favorable for new particle formation, N allows to determine new particle formation events at the JFJ and on the Swiss plateau. In addition, as CO is not washed out by precipitation in contrast to aerosols, it can also be used as a tracer for polluted air masses during weather situations with precipitation. However, CO has a less pronounced vertical profile than other chemical species due to a longer atmospheric lifetime, resulting in higher background concentrations and less pronounced diurnal cycles (Zellweger et al., 2000). Finally, the mean export of PBL air to the JFJ will be quantified by the relative increase of CO and the specific humidity (remains constant during vertical motion in the absence of precipitation). ”

Unnecessarily sentences must be removed, such as p987 L3-4 and p987 L27-p988 L3.

These sentences have been deleted.

[2.2 & 2.3 data analysis] More description is required for data analysis (the last sentence of the last paragraph in section 2.2). How did you calculate contributions of weather types to the annual average?

The contribution of each weather types to the annual average has been better described by adding the equation:

"For the annual cycles of aerosol variables, precipitation and gaseous compounds, the contributions of each synoptic weather type to the averaged annual cycles were determined by weighting the measured parameters during the defined weather type by its frequency of occurrence:

$$Y = \sum_{i=1}^{nb_SYNALP} Y_{SYNALP(i)} = \sum_{i=1}^{nb_SYNALP} \bar{Y}_{SYNALP(i)} \cdot \frac{m_{SYNALP(i)}}{m_{total}} \quad (1)$$

where Y is the averaged annual value, with the summation performed over all weather types. $\bar{Y}_{SYNALP(i)}$ is the averaged value during the SYNALP(i) weather type, $m_{SYNALP(i)}$ is the number of cases with SYNALP(i) weather type and m_{total} is the total number of cases (= number of years). $Y_{SYNALP(i)}$ is the contribution of the i^{th} SYNALP weather type and is represented with a different color on the figures 3, 5 and 8."

To separate weather types, what kind of meteorological data (at what time? 00UT?) did you use? Were weather types clearly separated always?

The Schüepp classification system uses the 12 UTC measurements. Ambiguous cases are separated by taking another pressure level and/or another time of the day. These specifications have been incorporated into the paper. The following sentences have been added to the manuscript:

"If the parameters cannot be unambiguously determined from the 12 UTC surface map, the corresponding 850 hPa heights or the 15 UTC or the 9 UTC surface map are additionally taken in account (SMI, 1985)."

We refer the readers to the cited papers for a more detailed description of the Alpine Weather Statistic: Stefanicki et al. (1998) and Wanner et al. (1998) gave a complete summary of how the SYNALP are determined, whereas SMI (1985) described the complete procedure.

[3.1 title] The title of section 3.1 needs to refine, because AWS is Alpine weather statistics.

The AWS is indeed a complete set of 34 daily meteorological parameters determined by MeteoSwiss since 1945. The synoptic weather type classification system determined by Schüepp is one of the 34 AWS parameters. According to Lugauer et al. (2000), we change AWS into SYNALP (for synoptic weather types of the Alps) in the manuscript.

[3.4] p999 L5 For what purpose?

The amplitude of the diurnal cycle is a useful criterion to estimate the PBL influence during the afternoon at the JFJ. This was now stated in the manuscript.

[4] p999-p1000 This part needs revision. I cannot follow this discussion. Is this part just memorandum for the next sections?

Yes, this part was a memorandum. Following this comment the information given in this first discussion part was moved either to the introduction, the experiment or the discussion part.

[4.1] To show location of Payerne, adding a map is helpful to readers.

A map has been added with the 4 stations from which data were used.

p1002 L15-16 In February – March, N for CC during am in Fig. 6 is not so low. More discussion (with data) of aerosol surface area may be needed but should be separated for N and surface area. As mentioned in p1005 L15, precipitation would frequently be associated with the CC type. Cloud particles might scavenge precursor gases from the air. This leads less chance to nucleate new particles.

The sentence was misleading. N during CC in February and March is really high. These high values are more visible due to the very low N concentration during all other weather types. The sentence was therefore changed:

“The contribution of new particle formation during CC is particularly visible in February-March due to the very low aerosol number concentration during all other weather types.”

This study does not present any aerosol surface area data, because the Epiphaniometer measurements were discontinued at the JFJ since about 10 years ago.

The referee is right in saying the cloud particles might scavenge precursor gases. However the global radiation W is relatively high during CC in February and March, leading to the fact that, if precipitation does occur, part of the day is out of the cloud. New particle formation and subsequent growth are probably enhanced after the precipitation. This is not described in the paper since it is partly speculative.

p1003 L3 “FT conditions prevail during the night”. This statement needs evidence and proper reasoning.

The paragraph has been changed to emphasize the proofs for FT conditions prevailing during the night:

“In April and May, the CA and CI weather types become dominant, presenting the major contribution to all aerosol variables (Fig. 5). For subsidence (CA), large diurnal cycles are found for all aerosol variables and their amplitudes are a maximum in April for b_{abs} and b_{scat} , and in May for N (Fig. 7). The CO concentration also shows a clear diurnal cycle during subsidence (Fig. 9). April is the only month presenting a clear diurnal cycle of the scattering Ångström exponent indicating a larger aerosol size (low scattering Ångström exponent) from 2:00 to 12:00 and a smaller aerosol size (high scattering Ångström exponent) from 14:00 to 24:00 (see insert in Fig. 7). The large diurnal cycles of aerosol variables, of CO concentration and of the Ångström exponent and the higher concentration of aerosols from more polluted air masses during the afternoon show that the influence of the PBL due to thermal convection occurs from April onward during the CA weather type. The minima of aerosol variables, CO concentration and of the Ångström exponent remain however similar to the values during winter and during April-May in case of all other synoptic weather types. These low aerosol and CO concentrations during the night associated with the presence of larger particles (Ångström exponent) prove that FT conditions prevail during the night for all weather types. This alternation of FT and PBL air masses leads to a maximum amplitude in diurnal cycles during the CA weather type. All other weather types exhibit a small diurnal cycle reflecting a very small and unsystematic influence of the PBL during the afternoon, while the AS weather type exhibits no diurnal cycle.”

p1003 L10-12 This part sounds speculative.

The sentence was changed: “Aerosol variables are greatly enhanced in summer (June-August) during subsidence (CA) even if the frequency of occurrence of the CA weather type is not really higher than during the rest of the year; the higher aerosol concentration is due to mountain venting induced by higher temperatures and allowing the export of PBL air into the FT, as was also observed by Henne et al. (2005b).”

[4.2] Again, to show locations of Payerne and Stabio, adding a map is helpful to readers.

A map was added.

p1004 L22 Henne et al., 2005?

Right, it was changed.

Anonymous Referee #2

Received and published: 22 March 2011

General comments:

This study analyze 14 years of aerosol parameters (bscat, babs and N) in relation to weather types, trace gas concentration and annual and diurnal variability. Long term studies are indeed beneficial, and thus merit publication. However, the current manuscript suffers from major drawbacks:

First, the discussions and arguments presented are hard to follow and the analysis of the data is sometimes very patchy, which makes it difficult for a reader to follow.

The manuscript, principally the results and discussion parts, was carefully revised to increase the readability of the arguments. Most of these modifications are described below.

Secondly, the article contains some unsupported statements, especially those relating to new particle formation. Since this study does not employ any size segregated information on the particle population, but only integral number, not much can in principle be said about new particle formation. One may discuss the possibility, but additional data and analysis would be needed in order to support the statements given. Thus, the discussion and conclusions must be somewhat revised.

Page 1000, line 8-12: Is this shown? Would N as the other parameters be sensitive to primary emissions as well? I do not follow the reasoning.

Page 998, line 11: What is meant by “: its greater sensitivity to the Aitken mode aerosol: :” and how is it related to the discussion?

Particle size distributions were only measured during short-term field campaigns and not at an operational basis at the JFJ. However N is more sensitive to Aitken mode particles since they constitute the greatest number of particles but do not contribute much to the absorption coefficient for example. This sensitivity was clearly proved by Weingartner et al (1999) for example on Fig. 6, which shows that N measures both the new particle formation and the PBL influence the first one having the largest contribution in winter and the second one in summer. Chamaillard et al. (2006) showed that Nephelometers measure particles larger than 200 nm. Differences between the N and b_{abs} behavior can therefore be related to the number of particles in the Aitken mode.

A better description of these relations is given in the revised manuscript, principally in the introduction. In the discussion, a reference to size distribution measurements differences between CA and CC weather type was added (Weingartner et al., 1999). These results are based on one year of measurement and show that higher number concentration of Aitken mode and nucleation mode particles was present during CC.

Recommendation:

Below, I outline a number of points that caught my attention. These are a number of suggestions that need to be addressed prior publication. I do however recommend changes beyond these detailed comments in order to meet the standards of ACP. My recommendation is that the paper undergoes a major revision prior publication in order to remove overly quantitative statements as well as improving the readability of the

paper. More attention should be given to increase the flow of the text, and the analysis need to be better structured. This also applies to method description.

Detailed comments:

P 988, line 22: ..is the most sensitive: : :” remove “the” or add “parameter”

This correction was done.

p.996, line12-17: What is the meaning of the quantity provided in figure 4b? As I understand it, it somehow represents the percent of “accumulated” observations at the site. Is this correct? How is it relevant? Please clarify how and why this is calculated.

The experimental part was revised. The procedure to calculate the weather type contribution of Fig. 5a is now described with a clear equation. Moreover, the quantity provided in figure 5b is now extensively described:

“The 1995-2008 median annual cycles of dry b_{abs} , b_{scat} and N are shown in Fig. 5a, the contributions of the different synoptic weather types are shown in colors; they are calculated accordingly to Equation (1). Similar graphs with the relative contribution ($Y_{SYNALP(i)}/Y$) of each SYNALP weather type to the total value Y of the aerosol variables are provided in Fig. 5b. Note that these numbers are influenced by both the absolute values of the aerosol variables and the frequency of occurrence of the weather type, as stated by Equation (1).”

P996, line 25: What is meant by cause here? In relation to what? Integrated observations? Again, what is the purpose of this calculation?

These numbers correspond to the sum of the relative contribution of CA and CI weather type presented in Fig. 4b. The word “cause” was however not well chosen and was replaced by “have a relative contribution of”. The Fig. 4b is also now explicitly mentioned in the text. The purpose of this calculation is to evidence the very high contribution of CA and CI to the aerosol load during summer.

“The summer and particularly the end of July is dominated by the influence of CA (subsidence) and CI situations, which together have a relative contribution of 75%, 72% and 66% of b_{scat} , b_{abs} , and N , respectively (see Fig. 4b).”

P997, first paragraph: I do not follow. Please expand on the reasoning.

The unclear points that were not important for the main conclusions of the paper have been suppressed and the sentence has been rephrased:

“The summer is dominated by the influence of CA (subsidence) and CI situations, which together have a relative contribution of 75%, 72% and 66% of b_{scat} , b_{abs} , and N , respectively (see Fig. 5b). The CC type (lifting) has its greatest impact in the February-April period particularly on N . All three aerosol variables have their minimum in winter, when the greater influence of advective types on the scattering and absorption coefficients is related to a greater frequency of occurrence of these weather types (Fig. 2).”

Page 997, second paragraph: Is it shown that new particle formation is the cause of the difference in annual cycle?

Yes, this was shown in Weingartner et al. (1999) as it is referenced in the manuscript.

Page 997, line 25: It is stated that there is no prevalence of any particular weather type. Is this now based on figure 4b? Is then AW+N+E treated as a separate weather type? In that case I do not agree. Please clarify what is meant here.

This sentence was not well formulated. We wanted to emphasize that no specific weather type lead to high aerosol concentration. But AW, N and E have a higher frequency of occurrence than the other weather types that leads to higher relative concentration particularly in the absorption and scattering coefficient. This has been corrected in the revised version of the manuscript:

“First, the November-January period is characterized by low values of the aerosol variables and by a high frequency of occurrence of AW+N+E weather type that is particularly contributes to b_{abs} and b_{scat} .”

Section 3.3 overall: I think this section need to be rewritten. The discussion is hard to follow.

Large parts of section 3.3 have been modified and we hope that this will clarify this result section.

Page 998, line 12: Comparing results shown in figure 5, I think it is not evident that babs show least distinct diurnal cycles. Speculate on the reasons why babs would show less pronounced diurnal behavior.

As explained in manuscript, b_{abs} , contrarily to N , is not really sensitive to the growth of newly formed particles, because it measures an equivalent BC concentration. b_{abs} is therefore not sensitive to small secondary aerosols.

Page 998, line 13-15: I do not agree that babs show no diurnal behavior in feb-march. At least not from a visual inspection. Is the discussion based on other methods? I would say there is a quite pronounced diurnal cycle for at least M and AS weather types.

You are right, b_{abs} has a diurnal cycle for M and AS in February-March. The manuscript was accordingly changed: “The November-January and February-March periods display the weakest diurnal cycles of the year, with b_{abs} exhibiting no diurnal cycles for most of the weather types.”

Page 998, section 3.4.: it must be better clarified what variables of bscat, babs and N that are discussed in terms of annual cycles. It is very hard to follow the discussion and the statements by the author seem to contradict the results shown in the figure. I suggest to treat each variable separately and consecutively. This would allow the reader to better follow the discussion.

Similarly to section 3.3, the manuscript was largely modified in section 3.4. The various aerosol variables are however not discussed separately, since this would lengthen the manuscript without clarifying the different results found often by correlation between the different aerosol measurements. The answer to the next comment will already clarify a large part of this comment.

Page 999, line 3-4 + figure 5: What is the meaning of continuous diurnal decrease? What is clear from AS weather types during is that the diurnal cycle is not continuous. The decrease is argued to be due to wet removal. I think this statement not is supported by the data. Why would precipitation rapidly stop 24:00, and be followed by a sudden increase in the parameter value. I think that the calculations here should be checked again, it seems kind of wrong.

Page 999, line 8: Please verify these calculations. Seems odd with continues decrease.

The diurnal cycles that are shown in this paper are calculated by averaging the measurements of the variables as a function of the hours of the day. It does not correspond to a kind of “running mean seasonal cycle”, so that there is no reason that the values at the end of the day have a continuity with the value at the beginning the day. The day after may correspond to another weather type.

In this sense a continuous diurnal decrease corresponds to the fact that the aerosol parameters are higher at the beginning of the day than at the end and that this change is continuous. During AS, there is precipitation south of the Alps that decreases the aerosol concentration in the air masses that reach the JFJ some hours later.

Figure 6: I think the discussion would benefit by adding eg percentile ranges showing the range of amplitude encountered. As a reader it is hard to draw any conclusions from the figure.

Non-parametric confidence levels were added to the figure 6.

Page 999, line 11-16: Please revise the discussion to agree with what is actually shown in the figures.

These lines were revised: the word “significant” was added to take into account the information given by the confidence limits:” The annual cycles of the diurnal amplitude are almost similar for the three described aerosol variables. For the CA type (subsidence), the diurnal cycles are significantly more pronounced in the spring (April-May) and in the autumn (September-October) and lower in summer. For the CC type (lifting), the diurnal cycles of b_{scat} and N are significantly more pronounced in January-March and in July. During advective situations from W, N and E, the diurnal cycles are most pronounced in summer for b_{abs} and b_{scat} , which is not observed for N .”

Page 999, line 16-21: Please comment on the lack of diurnal variation for Å for all month except April.

The manuscript was modified: “The diurnal cycle of the aerosol size between the FT aged aerosol in the morning and the PBL aerosol in the afternoon is therefore only observed in April for the subsidence (CA) case. This means that for all other weather types and months, the scattering Ångström exponent cannot detect a diurnal evolution of the aerosol size distribution.”

Page 1000, line 2: how can a coefficient be characteristic of polluted airmasses. Should be “:elevated values of absorption coefficient...” I guess.

The word “characteristic of” has been replaced by “sensitive to”

Page 1000 line 23 and p. 1002,. Line 23: “a maximum”! “at a maximum”

The change was done.

Page 1001, line 4-5: how can b_{abs} and b_{scat} be dominated by weather types? Please clarify.

The sentence is the following: “During this period, b_{abs} and b_{scat} are effectively dominated by the contribution of advective weather types”. The absorption and scattering coefficient are effectively not dominated by the weather types, but by their contributions to the mentioned coefficients.

Page 1002, line 1-2: This is not shown here. Replace “lead to” with “favor”.

The replacement has been done.

Figure 9: Add ranges to the values shown in figure.

Confidence limits for the N diurnal max-min were added as well as uncertainties on the x-axis.

P1002, last paragraph: hard to follow

The paragraph has been rewritten to clarify the reasoning and the conclusions.

Page 1003, line 8-12: I do not follow

Page 1003, line 8 and onwards: To what figure does this discussion relate to? 9 or 10?
Please clarify.

The lines 8-12 have been rewritten to clarify the purpose. All this paragraph relates to figure 10. This is now mentioned in the text.

Figure 10: To what does the errorbars correspond to? Further more: wouldn't the discussion benefit from adding maximum and minimum values as a range around median in one plot.

The error bars correspond to upper and lower confidence limits that were calculated by a nonparametric method defined by an order statistic and using the cumulative binomial distribution, as it is explained at the end of section 2.2.

Adding the maximum and minimum values as a range around median in one plot renders the figure completely unreadable due to number of curves. We also try to do one such plot for each weather type, but we really found that the present figures give the largest information and correspond well with to the discussion. This also allows minimizing the number of figures.

Page 1004, first paragraph: Revise this section for clarity.

The section was revised to clarify the text and to shorten the sentences.

Page 1005, line 8-9: what is meant by a complete replacement of JFJ by PBL.

A complete replacement of JFJ by PBL air means that there is no concentration difference between the JFJ and the Swiss plateau: The sentence has been changed in this sense:

"When the vertical motion is zero then $r_i = 0\%$, whereas similar concentrations measured at the JFJ and on the Swiss plateau would yield $r_i = 100\%$."

Figure 11: Check x-labels

Avr was changed in Apr.

Page 1005, line 11: write " r_i reaching 35% and 45%"

The change was done.

Page 1006, line 11: Is thiit shown that this is due to growth of newly formed particles?

Yes, this is shown in Fig. 9 and explained in the text.

Page 1006, line 20-21: "with enhanced new particle formation". One may speculate that this is the case, but this statement is not supported by the analysis given in this paper.

This statement is supported first by the large diurnal cycle of the scattering coefficient and N in September and October in case of CA and CI, second by the enhanced N maxima in September-October connected to the absence of high diurnal maxima of the absorption coefficient during CA. Finally a similar statement was also published by Boulon et al. (2010).