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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Large-scale European source and flow patterns retrieved from back-trajectory interpretations of CO_2 at the high alpine research station Jungfraujoch" *by* C. Uglietti et al.

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General comments:

The paper report on a backtrajectory-based analysis of >4 years of continuous measurements of CO2 and O2 at the well-established Jungfraujoch (JFJ) mountain observatory. The authors introduce here two case studies of elevated CO2 concentrations and a backtrajectory analysis of the CO2 and O2 "high frequency" anomalies. These

continuous measurements extend by a few more years the dataset presented by the authors in an earlier paper (Uglietti et al., 2008). This paper may be considered as a useful characterization of the JFJ station in terms of O2 and CO2 observations, with a valuable discussion of airmasses origins and processes for different CO2-APO signatures. Overall, the essential novelty of this paper should be more clearly identified and stated. Discussing high frequency variability of CO2 and O2 at a remote, mountain site is challenging and the authors deserve credit for attempting to tackle this issue seriously. However, the technique used (relatively low resolution back trajectory) has several limitation for this particular situation, including poor representation of convection and boundary layer processes, which are indeed noticed by the authors but not sufficiently addressed in the paper. Backtrajectory analyses of transport of atmospheric compounds are numerous and the technique is well established, with now more elaborated tools available, such as Lagrangian particle disperson models (LPDM, such as FLEXPART) less sensitive to transport errors and more suitable for source region identification (see Han et al., 2005). The authors acknowledge that situation in their conclusions (p. 835, l. 14). Given the free availability and relative computational efficiency of these models, it is surprising that the authors did not use directly a LPDM. The authors further state (p. 835, l. 10) that "this study demonstrate the potential of trajectory analyses for classifying airmasses..."; I would argue that in fact the authors do not perform their classification based on backtrajectories but, instead, based on their measurements by selecting arbitrary regions in O2 and CO2 concentration space, which represent a significant difference. Automated, multivariate chemical composition based clustering has been shown as in, e.g., Lewis et al. (2007) for pollutants at Mace Head. Furthermore, it should be noted that the first demonstration of backtrajectory-based cluster analysis for atmospheric compound has been performed in the 80's (Moody and Galloway, 1988), and that improved LPDM-based cluster analysis now exists (e.g. Paris et al., 2010; Hirdmann et al., 2010). Choosing between the two approaches (observation-based vs. trajectory based) depends on the objectives of the study. In the light of some inconsistencies in the text such as the one noted above, this choice should clearly be better documented in the paper (see e.g. Cape et al., 2000 for such a discussion). The paper is rather well written and clear, with good quality figures. However the flow of the paper is impeded by two case studies of high CO_2 concentrations that do not bring new elements to our current knowledge. Instead, case studies could be better used to illustrate and validate some of the "cluster" analyses and discussions.

Response to general comments:

The referee noticed a few inconsistencies in our paper and we agree with that. It is true that we did not perform the classification of source areas based on the properties of the back trajectories but instead basing the selection on the CO_2 and O_2 scatterplot. Indeed the selection has been done subjectively (but not arbitrarily) while a more objective method such as a clustering approach would be more desirable. Many thanks for pointing us to the interesting study by Lewis et al. (2007) applying an air mass clustering based on trace gas observations over the North Atlantic. It is not feasible, however, to perform a similar cluster analysis based on our observations of CO_2 and O_2 only. This is quite obvious when looking at figure 6 which doesn't reveal easily identifiable clusters of points. We fully agree that LPDMs describe transport more realistically and are aware of the limitations introduced by using the simpler trajectories. In response to the concerns of reviewer 2 regarding the limitations of trajectories, partially also raised by reviewer 1, we decided to replace all trajectory results with simulations with the Lagrangian Particle Dispersion Model (LPDM) FLEXPART. Despite this important change in terms of transport model used, the necessary changes to the manuscript are rather small and our conclusions need not to be revised. Figures 10-15 presenting the air mass origins associated with the different air mass clusters as well as Figures 7 and 8a presenting the maps for the two case studies can be readily replaced by the results from the FLEXPART simulations. The FLEXPART results are very, and maybe surprisingly, similar indicating that the reported patterns are dominated by advective transport which is captured both by trajectories and the LPDM. Switching to the FLEXPART sim-

ulation results has several advantages: (i) The simulations were performed at a much higher resolution of the ECMWF meteorological input data at 0.2°x0.2° as compared to the previous trajectory simulations at 1°x1° resolution. Note that simulations at this high resolution were only possible after February 2006 when ECMWF switched to a higher resolution model (from T511->T799 and from 60->91 levels). The FLEXPART simulations for the period Jan 2005 to Feb 2006 are based on the lower resolution. (ii) FLEXPART accounts for the dispersion of a plume by turbulent and convective motions. (iii) Transport is described much more comprehensively: 50'000 trajectories are computed per time step instead of only 5 as before. (iv) Several concerns and questions raised by the referees addressing the limitations of trajectories are no longer relevant.

Specific comments:

Referee: The title of the paper mentions "flow patterns: : :retrieved", this is slightly misleading because the paper does not really retrieves the flow patterns as such, it rather establish source receptor relationship (based on reanalysis flow patterns); a better title could be coined in this respect. I would also suggest mentioning O_2 in the title for better visibility of the work performed.

Authors: The title will be changed to: "European source and sink areas of CO_2 retrieved from Lagrangian transport model interpretation of combined O_2 and CO_2 measurements at the high alpine research station Jungfraujoch."

Referee: p.814 l.5-6: actually the CO_2 and O_2 signatures are used to classify air masses, contrarily to what is suggested in this sentence

Authors: The corresponding sentence will be changed to: This study investigates the transport of CO_2 and O_2 towards Jungfraujoch using backward Lagrangian Particle Dispersion Model (LPDM) simulations and utilizes CO_2 and O_2 signatures to classify air masses.

Referee: p.815 l.7 for the logic of the flow I would suggest to replace "greenhouse gases” by “CO_2” after “For that reason: : :”

Authors: yes, done.

Referee: l.8 remove “constantly”

Authors: corrected.

Referee: l.10 remove “of the changes in time”, it is redundant with “evolution”

Authors: corrected.

Referee: p.816 l.7”partly from oceans”: a word may be missing here.

Authors: Indeed, there were a few words missing: gases become less soluble and are transferred from the ocean to the atmosphere.

Referee: l.11 “consummation” should probably be “consumption”? otherwise the sentence could be somewhat clarified.

Authors: Right, “consumption” is the correct term.

Referee: l.12 replace “like” by “i.e.” after “different fuels”

Authors: corrected.

Referee: p.817 l.2 replace “it’s” by “its”

Authors: corrected.

Referee: l.6 “One of the most important sites in Europe: ” I would suggest avoiding superlatives and ranking here and rephrase to say that JFJ is an important site to monitor atmospheric composition

Authors: Yes, done.

Referee: l.10 maybe the result of the papers cited here relevant to this study could be briefly mentioned.

Authors: The last paragraph of the introduction section will be changed to the following: In this paper, we present an analysis of 4.5 years (January 2005 - June 2009) of continuous measurements of CO_2 and O_2 at the High Altitude Research Station Jungfraujoch in the Swiss Alps, which is an important measurement site in Europe to monitor atmospheric composition particularly in the lower free troposphere. Our main focus is on investigating, by means of Lagrangian Particle Dispersion Model (LPDM) simulations the origin of short-term deviations of CO_2 and O_2 from background concentrations at Jungfraujoch and on understanding their seasonally changing characteristics. Many studies have been carried out on local and long range transport and on identifying source and sink regions of polluted air reaching Jungfraujoch (e.g. Seibert et al., 1994, 1998; Forrer et al., 2000; Henne et al., 2005a; Cozic et al., 2008; Balzani Lööv et al., 2008, Tuszon et al., 2011). However, only few studies have done this in a systematic way using a statistical approach in which multiyear trace gas observations are interpreted by means of a large number of trajectories or LPDM results, which has the significant advantage that errors in individual transport simulations tend to cancel out. Examples for this approach are the studies by Kaiser et al. (2007) studying the origin of NOx, O_3 and CO at several sites in the Alps and those by Balzani-Lööv et al. (2008) and Cui et al. (2009) investigating background concentrations and the role of stratospheric intrusions and intercontinental transport, respectively. Here, we follow such a statistical approach to classify air masses with distinct O_2 and CO_2 signatures with respect to their origin. Rather than clustering air masses according to transport histories, i.e. trajectory clustering (Moody and Galloway, 1988; Henne et al. 2008) or LPDM clustering (Paris et al., 2010; Hirdmann et al., 2010), the classification is based on identifying specific regions with the CO_2 - O_2 correlation and then analyzing the air mass origin for each class. Similar to the study by Lewis et al. (2007), this approach puts more weight on the trace gas observations as compared to the transport modeling, which is likely better suited to identify different source and sink processes. However, while Lewis et al. (2007) performed an objective clustering of air masses based on simultaneous observations of multiple species, our approach is to focus on two closely interrelated species only which precluded such a cluster analysis

Referee: l.13-14: here again trajectories are not used to classify, but CO_2 and O_2 anomalies are used to classify. Please rephrase correctly.

Authors: See above changes to the last paragraph of the introduction section.

Referee: Section 2.1. Please include longitude and latitude of JFJ

Authors: Yes done: (7° 59' 2" E, 46° 32' 53" N).

Referee: l.23: remove "also" after "North is: : :" (at the previous sentence it was northwest!)

Authors: corrected.

Referee: p.818 l.14-15: I would suggest to remain factual and simply write that JFJ is a GAW station.

Authors: Jungfraujoch is one of the Global Atmosphere Watch stations (GAW) within the framework of WMO activities.

Referee: Section 2.2. Two O_2 analysers are described, what measurements exactly are used in this study. This section should include a quantitative report on the uncertainties in O_2 and CO_2 measurements.

Authors: Yes, true the O_2 measurements are performed by using both fuel cells and a paramagnetic cell. In this study only the results from the paramagnetic cell are presented and discussed. The long-term reproducibility of the measurements was calculated from a standard gas which is normally measured twice a day in the same way as the sample air (every gas is measured for 6 minutes). Data allocation is on one second basis but when processing the data, the first 242 seconds over the measured 6 minutes were skipped and the average of the following 115 seconds was computed in order to avoid the abrupt transient changes occurring during the switching between different gases. The standard error of the measurements was computed during a period of one month and the resulting reproducibility for CO_2 was determined to be less than ± 0.1 ppm (around 0.06 – 0.08 ppm) and for O_2 to be in the range of ± 8 per meg. The CO_2 value matches the target precision of 0.1 ppm. For O_2, on the other hand, the value is still higher than the target precision of 5 per meg for O_2 (Uglietti, C.: Understanding the carbon cycle through atmospheric carbon dioxide and oxygen observations, PhD thesis, University of Bern, 2009). This information will be provided in the revised manuscript.

Referee: p.819 l.19-22: please provide the values of the concentrations used in the calibrations

Authors: As will be stated in the revised manuscript, the calibration is twice a day. We use a high CO_2 calibration gas (430 ppm), which correspond to the low O_2 calibration gas (-1300 per meg), a low CO_2 calibration gas (356 ppm) corresponding to the high O_2 calibration gas (35 per meg) and finally the working gas which should have CO_2 and O_2 concentrations in the range of the air sample values.

Referee: l.24-26 In apparent contradiction with the sentence ("Data are archived through: : :") I could not find the JFJ CO_2 data neither from the GAW's WDCGG, nor from the IMECC website. Non-CO_2 data are nevertheless available from WDCGG. At least regarding GAW the sentence is misleading and needs to be changed. If the data are indeed available on one of these databases, web links would be useful. Note that GAW in itself is not an archiving service, but WDCGG is (on behalf of GAW).

Authors: The online data are archived through IMECC (preliminary dataset, https://ramces.lsce.ipsl.fr/index.php?option=com_content&view=article&id=36). We checked the IMECC database and found that the JFJ site is no longer on the Web. Therefore we sent a request to the IMECC office to resolve this issue because usually the data are daily updated and sent to the database (http://www.icos-infrastructure.eu/index.php?p=nrt_av). It is correct that the CO_2 data are not yet available through WDCGG. The CO_2 data record presented in this study will be submitted to the WDCGG database by

May 2011. The flask data are only available through the CarboEurope database.

Referee: p.820, last paragraph: here it is explained that 5 backtrajectories are run for each observation at different receptor points in the horizontal plane near JFJ, in a fair attempt to reduce transport error. However, why is the same not done in the vertical (i.e. 7 backtrajectories, with 2 above and below JFJ) ? Mountain stations' altitude is notoriously difficult to represent in models, partly due to coarse topography (as noted later by the authors), and the uncertainty due to transport in the vertical may be more critical than in the horizontal especially for the identification of possibly thin plumes related to short term variability. I would also suggest reporting the number of vertical levels in the reanalysis used and the approximate vertical resolution near JFJ's altitude. These remarks are complementary with respect to the following comment.

Authors: Since the transport analysis is performed with FLEXPART in the revised version, this question is no longer relevant.

Referee: p.821 L.1-5 this CO-based transport model validation exercise is very important and showing results (some text mentioning compound agreement values, e.g. offset and std dev, and/or a figure) would lend the whole study an improved credibility.

Authors: Please see the response to general comments.

Referee: l.23 backtrajectories are notoriously not so good at representing convection and other BL processes. By selecting trajectories in the BL, the introduction of a bias in sourcereceptor relationship patterns is therefore probable. Could the authors explain how they tackle the problem of poor representation of convection?

Authors: Since the new FLEXPART simulations account for convection this question is no longer relevant.

Referee: Sections 3.1 and 3.2: At the beginning of section 3.2, the distinction between long term, seasonal and short term variability is exposed, but in Section 3.1 the authors have already discussed the seasonal variations, whereas they go on with long term and

short term after the distinction is made, in Section 3.2. I suggest reorganizing the text more logically: 1) define distinction long/seasonal/short, 2) long term, 3) seasonal, 4) short term.

Authors: Referee 1 had raised the same point. We therefore rearranged the whole section as described in our answer to Referee 1.

Referee: p.823 l.7-18: this paragraph is not a result and may therefore not pertain to this section 3 entitled "Results". I suggest that the text be reorganised and the section's title changed

Authors: This is a good point. We will move this statement to the introduction.

Referee: l.19-22: These two sentences should be in the "Methods" section, as are some parts of section 3.2. Could the authors detail in a short sentence or two how they de-trend the data beyond citing this reference, as this is highly relevant to the paper?

Authors: The de-trending method has already been explained in some detail in the text: the mean seasonal cycle was derived from the background data by subtracting first the long-term trend (for every background value the corresponding trend value was subtracted calculated according to the method of Nakazawa et al. (1997) using a smoothing cubic spline as stated also in Valentino et al. 2008 and Uglietti et al 2008. The resulting values were then averaged as monthly mean values for the different years. Before calculating monthly means the data were linearly interpolated to daily values.

Referee: l.20: please change "Morimoto, S., et al." to "Morimoto, et al."

Authors: corrected

Referee: l.25 The same group observed (in Valentino et al., 2008, further citing Sturm, 2005) "a strong oceanic component contributing to the oxygen seasonal cycle even at the continental sites JFJ and PUY" based on other JFJ data. On the opposite, here, the statement that the seasonal APO amplitude is smaller than at other CarboEurope stations is in apparent contradiction. Does the high resolution dataset bring new information that is challenging previous flask data analysis? Maybe this is worth discussing here.

Authors: Neither is there a contradiction between the flask and online measurements, nor between the statements given by Valentino and Sturm. There is a significant oceanic component present at JFJ but this signal is smaller than at other stations.

Referee: p.824 l.9: could the authors explain in the paper how they estimate the mean seasonal cycle? (simple monthly means? Harmonics?)

Authors: A fit curve with two harmonics was computed as seasonal cycle by means of the method described by Nakazawa et al. (1997)

Referee: l.13 "as it would be linear": what is meant here? The whole sentence (starting with: Nevertheless we calculated: : :") is not very clear, please rephrase. Authors: It means that we calculated the mean annual growth rate by computing the linear trends year by year and not using the smoothing cubic spline as for the entire trend line.

Referee: l.19: in the iterative procedure, what percentage of data points is rejected (and hence do not intervene in the calculation of the background)?

Authors: 26

Referee: l.25 I would suggest to add at the end of the sentence "for high frequency anomalies" as they removed the large scale flux effect by removing the seasonal pattern.

Authors: Corrected.

Referee: p.825 l.11 ">10 ppm above average": isn't 'background' intended here instead of 'average'? Please clarify.

Authors: Yes, background, not average

Referee: l.20 Flask sampling: this is not described in the "Methods" section. Is the sampling automatic? At regular intervals? Only upon specific events?

Authors: Right. The flasks measurements began in October 2000 and the sampling is performed by the custodians of the research station. Duplicate air samples are collected into 1L glass flasks. All flasks are equipped with two Viton O-ring high-vacuum valves at both ends (Louwers Hapert, Netherlands). Flasks were sampled biweekly until April 2007 and after that the sampling frequency increased to weekly sampling in order to ensure a high time coverage. For a detailed explanation of the sampling procedure, please see Sturm et al 2005. This information will be added to the revised manuscript.

Referee: l.22 please report the estimated uncertainty in the flask measurement.

Authors: The inter-pair flasks reproducibility for CO2 flasks measurements is $\pm$ 0.30 ppm and for oxygen is $\pm$ 8.3 per meg.

Referee: p.828 l.15 I would argue that these are not really "clusters", and the wording needs to be changed (here and at other places in the paper) to e.g. "subsets", as in section 4.2's title. Here, the cluster is too strong (Fig. 9) and no group of points is obviously separated from the rest of the data.

Authors: Yes, corrected

Referee: Section 4.2: it would be useful to have the number or percentage of points in each subset relative to the total number of observations.

Authors: The fraction of points per cluster is: Cluster 1: 62These percentage will be mentioned in the figure captions of the corresponding subsets.

Referee: Section 4.2.3 4.2.4, titles: rephrase "negative CO2" into e.g. "negative CO2 excursion" or "depleted CO2", or any other reference to a delta-CO2 relative to the defined background. Same remark for "positive APO", etc: : : Please check the text for other occurrences.

Authors: Yes corrected.

Referee: Conclusion: see remark in the General comments above.

Referee: p.846 Fig 4: why are the error bars the measurement uncertainty and not e.g. the std dev of the data? If it is the measurement uncertainty, why does the measurement uncertainty vary with time (e.g. 30

Authors: Right, there was an error. The error bars are the standard deviation of the data.

Referee: Figs. 7, 8, 10-15: - I recommend adding a sign for the JFJ position in these figures. - I also suggest to change the colour code to a light tone (toward white) instead of dark blue for low values (towards zero), in order to better visualize the relative weights of the residence time in the BL across the map and between figures. - Please mention in the captions, which subset number it has (in reference to section titles 4.2.1 and following) - Here, also please avoid the expression "negative CO2" and similar ones.

Authors: Yes, added a symbol for the position of JFJ. Added the subsets in the captions. Concerning the colour tone we think the current variation from dark blue (low values) to dark red (high values) with bright colours in between around a neutral value of 1 is clear enough.

Referee: p.856 fig. 14: please move the sentence starting with "The main provenance of air masses: : :" to the main text instead of figure caption.

Authors: Yes done

Referee: References Cape, J. N., Methven, J., and Hudson, L. E.: The use of trajectory cluster analysis to interpret trace gas measurements at Mace Head, Ireland, Atmos. Environ., 34, 3651– 3663, 2000. Han, Y. J., Holsen, T. A., Hopke, P. K., and Yi, S. M.: Comparison between backtrajectory based modeling and Lagrangian backward dispersion modeling for locating sources of reactive gaseous mercury, En-

viron. Sci. Technol., 39(10), 3887–3887, 2005 Henne, S., Klausen, J., Junkermann, W., Kariuki, J. M., Aseyo, J. O., and Buchmann, B.: Representativeness and climatology of carbon monoxide and ozone at the global GAW station Mt. Kenya in equatorial Africa, Atmos. Chem. Phys., 8, 3119-3139, doi:10.5194/acp-8-3119-2008, 2008. Hirdman, D., Burkhart, J. F., Sodemann, H., Eckhardt, S., Jefferson, A., Quinn, P. K., Sharma, S., Ström, J., and Stohl, A.: Long-term trends of black carbon and sulphate aerosol in the Arctic: changes in atmospheric transport and source region emissions, Atmos. Chem. Phys., 10, 9351-9368, doi:10.5194/acp-10-9351-2010, 2010. Lewis, A. C., et al.: Chemical composition observed over the mid-Atlantic and the detection of pollution signatures far from source regions, J. Geophys. Res., 112, D10S39, doi:10.1029/2006JD007584, 2007. Moody, J. L. and Galloway, J. N.: Quantifying the relationship between atmospheric transport and the chemical composition of precipitation on Bermuda, Tellus B, 40(5), 463–479, 1988. Paris, J.-D., Stohl, A., Ciais, P., Nédélec, P., Belan, B. D., Arshinov, M. Yu., and Ramonet, M.: Source-receptor relationships for airborne measurements of CO2, CO and O3 above Siberia: a cluster-based approach, Atmos. Chem. Phys., 10, 1671-1687, doi:10.5194/acp-10-1671-2010, 2010.

Authors: Added all the suggested references.