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Interactive comment on “Variability and budget of CO₂ in Europe: analysis of the CAATER airborne campaigns – Part 1: Observed variability” by I. Xueref-Remy et al.

Anonymous Referee #1

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The study reports very interesting results about the CO₂ concentration distribution in Western and Eastern Europe through two surveys carried out on May 2001 and October 2002 from ~300 m up to 4000 m. As far as this referee knows, not such wide campaigns studying the distribution of CO₂ have been previously made in Europe. The study characterizes the distribution of measurements and links the observed horizontal gradient with the air origin through a Backtrajectories Analysis. Furthermore, the second campaign reported in the manuscript sampled carbon monoxide, providing an alternative way to national inventories to characterize the CO₂ signal related to combustion processes through the CO:CO₂ ratio. Moreover, the authors study the variability in the boundary layer and in the free troposphere as a key parameter to test

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the proper representation of the vertical gradients in atmospheric inversion models. However, even if most of the data needed to exploit the measurements are included, the text fails at drawing conclusions from them. Some key questions raised by the authors are not dealt properly (for example Section 6). Furthermore, some pieces of the manuscript appear to be out of context, such as Section 2.3.4.; which reports the instrument used to analyze the Radon-222 concentration but any of the results are commented latter in the paper; or Figure 9 that only the way how it has been calculated is explained in the text and not the conclusions extracted from it.

Other aspect that I am concerned about relates to the back-trajectories analysis dealt in Section 3.2 and 3.3. The back-trajectory analysis is used to explain the differences of the measured CO₂ mixing ratios along the campaigns in different regions/countries. However, any mention to the change of the altitude flight is done when comparing the data (when a change of altitude took place).

I would strongly recommend the authors to revise the aforementioned before its final publication. Furthermore, it would need a throughout reading to correct improper English sentences.

ABSTRACT Page 5666, line 11. Add “(± 1 standard deviation)” after “the mean CO₂ concentration” Page 5666, line 13 and further occurrences: air masses (as two words). Page 5666, lines 13-14. The authors talk about pollution when measured high CO₂ concentrations. As CO₂ is not properly an atmospheric pollutant, I would suggest rewrite the sentence as “air masses get the signal of anthropogenic fluxes... from Benelux and Western Germany” Page 5666, line 20. I would remove “the so-called dirty thirty from WWF” as this given name is irrelevant to the description of the study in the Abstract section. Page 5666, line 28. Page 5667, lines 3, 8. I would remove the acronyms of the stations. The acronyms are not defined in the abstract and since they are mentioned further in the text, I would just keep the type of sites (ground stations; mountain sites...). Page 5666, line 28. Change “stations located near the ground” by “stations sampling within the PBL” Page 5667, line 1. Add “regional” after “local”

Page 5667, lines 3-4. Rewrite “Stations located several 100 km away...in the free troposphere” for “Stations separated by hundreds of km differ few ppm in their measurements indicating the existence of a gradient in the FT.” Page 5667, line 9. Is the variability expressed in terms of standard deviation? Please, stated how is calculated the reported variability. Page 5667, line 19-21. I would remove the last sentence as it is not relevant for the present study (maybe the authors want to move the sentence at the end of the Introduction or Conclusion section where they mentioned the model study done using the measurement reported in Part 1).

1. INTRODUCTION Page 5668, line 13. One of the main reasons why the inverse models are still uncertain is because most of the studies are based on measurements done on remote and marine sites, far from where terrestrial fluxes are taking place. Page 5668, line 26. I would remove “In a recent paper” as it is already 3 years old. Page 5669, line 2. Define what FT stands for and not in line 13.

2. DESCRIPTION OF THE CAMPAIGNS AND OF INSTRUMENTATION Page 5670, line 16. List which meteorological parameters were measured. Page 5670, line 17. Is there any reason why the flight paths are different for both surveys? Which were the criteria to choose these flight paths in both campaigns? Page 5671, line 27-28. I would move them at the end of page 5670. A sentence why the authors are also using other in-situ stations should be mentioned here. Page 5672, line 5. Add the mean altitude for 850 hPa. Page 5673, line 8-13. It is confusing how the calibration gases are measured. I guess that the authors would say that the last minute of acquisition is kept to build the calibration curve. When the outside air is analyzed, it is also the last minute kept to calculate the actual concentration? Page 5672, line 25. LSCE is not defined before in the text (either in the author’s affiliation addresses). Page 5672, line 25. I am not convinced that a figure of the CO₂ instrument is needed. It seems, however, that the analyzer has been described previously in Filipi (2002). Page 5673, line 10. Add the point in the uncertainty concentration of the High calibration gas. Page 5673, line 14. How often calibrations are done? Page 5673, lines 19-26. These lines appear out

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off-context. I would suggest moving them at the last paragraph of the Introduction section or at the beginning of the results. Line 23. It is confusing the expression “in the following at two places”. Please, rewrite it and be clearer. Page 5674, line 5. “The CO₂ concentration was measured by gas chromatography in LSCE”. Page 5674, line 20. Remove “due to rotation of the filter wheel”. Page 5674, line 23. Write the name of the IR detector rather to just state the chemical abbreviation (PbSe). Page 5675-6. I would remove Section 2.3.4 as the Radon-222 measurements are not reported in the present study.

3. ORIGIN OF SAMPLED AIR MASSES. In this section is lacking a description where / at which altitude backtrajectories are computed. Which is the longitude/latitude range where the backtrajectories presented in Figures 4, 5 and 6 are representative for? Are the release points defined as a single point? Or it is defined as a range of longitude/latitude? Which type of backtrajectories are used? Please, describe better how backtrajectories were computed. It seems that the altitude of the flight changed along the campaigns. Changes in the altitude are important as there is a change of the measured concentration in altitude linked to the vertical mixing and then, the vertical spread of surface fluxes. Linked to this point, it would be useful to have an estimation of the boundary layer height along transects where the results are reported (mainly Section 3.2 and 3.3) as it would help the reader to know whether the measurements were taken within the BL or in the FT. Linked to that point, for the flight done on the 23th May (Figure 4a), there is a clear decrease of the CO₂ concentration in the region from -2° to 2°E (~360ppmv) compared to the region from -4 to -2°E (~373 ppmv), that is coincidental with a change of the altitude of the flight (the altitude of the flight was between 500 and 1000 masl in the -2° to 2°E region whereas it was between 0 and 500 m in the -4 to -2°E one). Was the vertical structure of the atmosphere influencing on the CO₂ horizontal distribution? Another example of the change of the CO₂ concentration in the altitude was presented on the 26th May 2001 (Figure 4d). For that day, in the first paragraph of page 5678 is stated that “air masses coming from the west and north-west, which were more exposed to more urban areas are associated with CO₂

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higher by 25 ppm above this minimum". I am not really convinced that this difference is only related to the advection of polluted air masses as the backtrajectories show that air masses were always above 2000 m, then, uncoupled with the surface fluxes. In these cases it is difficult to make a clear statement that the CO₂ variability is only related to changes of the air masses origin rather than CO₂ concentration sampled at higher altitudes have more concentration compared to air masses sampled below, close to vegetation which is uptaking CO₂ by photosynthesis during the growing season/daytime when the campaign was undertaken. Page 5677, line 9. Remove "(37%)" as the percentage is stated before. 4. RELATIONSHIP BETWEEN CO AND CO₂ IN POLLUTED AIR MASSES Again, as the authors are mainly reported the CO₂ measurements with the aim to understand the underlying flux, I think that it is inappropriate to talk about polluted air masses. I would suggest titling the Section "RELATIONSHIP BETWEEN CO AND CO₂ IN ANTHROPOGENIC INFLUENCED AIR MASSES". Other aspect that I am concern is that the authors are comparing the CO/CO₂ ratios obtained from measurements with the annual national inventories. In one hand, anthropogenic fluxes show a strong diurnal cycle that is smoothed with the annual inventories. In the other hand, authors are comparing the national inventory even only a small region of one particular country is sampled. If the authors have no access to inventories with more temporal resolution, they can make a weighted mean of the countries that a sampled air mass is influenced for and maybe they can match better the observations with the inventories. To all R2, an estimation of its significance is lacking (add p-values to all R2). Page 5681, lines 5-10. Remove sentences from "In pollution loaded. . . (Palmer et al., 2006), after (e.g. Levin and Karstens, 2007) in page 5680, line 26. In lines 26 (page 5680) and 11(5681) the adjective complicated appears twice. Remove this adjective and explain better the constraints of the application of this method. Page 5682, lines 5-11. Why referring to the NDVI maps and not the terrestrial fluxes shown in Figure 1 when talking about the photosynthetic activity? Page 5682, line 28. What do you mean "which can be >0 in the North and <0 in the south"? 5. COMPARISON OF AIRCRAFT WITH SURFACE STATIONS MEASUREMENTS Page 5683, lines 11-13. How do you

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compute the mean concentration for the surface stations? Is the mean concentration calculated for the entire time of the surveys, that is, for CAATER1, from the period 23-26 May taking only the midday values? Or just taking into account the time when the aircraft was flown close to the station? Maybe a better approach would be calculating the mean CO₂ concentration for the surface sites only during the period when the aircraft observations could match in time and space. Following this approach, the early morning and night values should not be discharged whenever the aircraft observations were carried out simultaneous. Like this, a better picture of the 3D CO₂ distribution in Western Europe would be attained and the aircraft and ground stations more comparable. Page 5683, line 25 and latter occurrences. Why now talking about ABL? It would be better to keep consistence through all the manuscript and kept the defined PBL. Page 5683, line 24. As the Figures are showing the CO₂ measurements, I would suggest changing “show a strong variability” by “shows a large range”. I would also add the range of CO₂ measured mixing ratios in the PBL and in the FT for both campaigns in the text. Page 5684, line 15. Change “profil” by “profile”. Page 5684, line 17. Change “has encountering airmasses...West” by “was sampling air masses coming from the South, East and West as well.”. Page 5684, line 22. Add “s” to “observation” Page 5684, lines 25-26. Remove the first sentence and start the paragraph “Both PUY and SCH stations are located on the top of mid-elevation mountains...” Page 5685, line 3. Change “polluted” by “influenced by the valley processes” 6. ANALYSIS OF THE VERTICAL VARAIBILITY Page 5685, line 21. Change “comparing” by “compared” Page 5685, line 24. Reported the mean and standard deviation values for CAATER 2 with the same significant numbers as CAATER 1 (two decimals are given in the first campaign, only one in the second). Same in page 5686, line 2, give the CO₂ jump with two decimals. Page 5686, lines 6-11. Why the air masses are here separated depending on its origin from the LMDZ model and not from the HYSPLIT analysis presented in the current study? Page 5686, line 11. Add “(Figure 12)” after CAATER 1. Page 5686, line 11. I would suggest the “CO₂ range” rather than “the CO₂ variability” as the authors are afterwards stating the range of the measured values and not any estimation of the vari-

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ability. Page 5686, line 11. Change “high” by “large”. Page 5686, line 23. Add “(Figure 13)” after CAATER 2. Page 5686, line 23. I would suggest the “CO₂ range” rather than “the CO₂ variability” as the authors are afterwards stating the range of the measured values and not any estimation of the variability Page 5686, line 29. When talking about an advanced fall in the west part in Europe compared to the East, why not contrast this hypothesis with the surface fluxes show in Figure 1? From the NEE map on Figure 1e, it is observed that South/West Europe fluxes are $\sim 0\text{gCm}^{-2}\text{d}^{-1}$, whereas positive fluxes are shown in Eastern Europe. Page 5687, lines 2-5. The same about when talking about CO₂ emissions. Why only comment the national inventories from UNFCC? Figures 1 c and 1f show the anthropogenic fluxes (with the NEE). Why not discuss this fact using the provided information? Page 5687, lines 10-15. I don’t see the point that to understand a measured gradient in October 2002 more regular flights are needed and then, the CARBOEUROPE network is essential to understand this gradient. Do the authors think that this gradient is persistent through all seasons? I think that in the current study the authors present a huge amount of information (meteorological parameters, NEE, anthropogenic and oceanic fluxes; backtrajectories analysis, etc.) and they should be able to point the observed gradient. Which are the mean fluxes in that area? How is the vertical stability? CONCLUSIONS Page 5687, line 25. I don’t know if averages of CO₂ measured in both campaigns are comparable since different flight paths were followed. Maybe a sentence of the mean surface fluxes of the underlying paths should be mentioned. Page 5688, lines 9. A “c” is lacking in bac-trajectories. TABLES Table 4. It lacks “°C” in the Temperature row for the CO analyzer Table 5. It would be nice to have an additional column with the “mean” inventories slopes for each flight. FIGURES Figure 1. “(for the days of the campaigns) Figure 2. Last sentence in figure caption “Longitude (horizontal scale) is given in °E and latitude (vertical scale) in °N. Figure 11. CO₂ in subscript. Figure 12. A way to express the y-axis units for the left panel would be “z/z_i”. Right panel is missing axis labels. Figure 12. CO₂ in subscript in the left panel x-label. Right panel is missing axis labels.

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