

Interactive comment on “Atmospheric oxygen and carbon dioxide observations from two European coastal stations 2000–2005: continental influence, trend changes and APO climatology” by C. Sirignano et al.

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Dear Editor, dear referees, dear commentator, dear editorial office, dear readers, we must admit that the final response phase for our ACP-manuscript should have been finalized by the beginning of march 2009. Unfortunately, through personal circumstances of the first author who is no longer affiliated to University of Groningen even since before the first submission of the manuscript, it took us extraordinarily long time to complete the revised version. However, now we finally assimilated all suggestions given by the three referees and the received comment. The manuscript has been greatly improved by doing so, and we are very grateful for the work of the referees

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and the writer of the comment, especially to Dr. Andrew Manning for his careful and extensive work!

So for all of you it is a long time ago that you have been reading through our manuscript. Still now, we hope that the editors can honor the work accomplished by the referees (and also ourselves) and accept the revised version of our manuscript for final publication in ACP. Below please find our reply to all referees' reports and Dr. Tych's comment.

One editorial remark we would like to add at this point: Deviating from the original registration and all communication with Cosis (be it automatic or personal), the author list has been reshuffled in the final ACPD publication. Unfortunately it seems that we thought we could take the right order for granted and overlooked the fault in the galley proofs. It is our fault as well. So for the final publication in ACP please return to the original order, putting back H.A.J. Meijer's name to the last-author position.

Title: Atmospheric oxygen and carbon dioxide observations from two European coastal stations 2000-2005: continental influence, trend changes and APO climatology

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Looking forward to a hopefully positive response, and ready to finalize the publication with truly short response times, with kind regards,

Rolf Neubert and Carmina Sirignano for all authors

Groningen, january 2010

Reply on the referee report by Dr. Andrew Manning

First of all, we authors would like to express our special gratitude for your very extensive, careful and detailed, and thus very helpful, but also timeconsuming review! It really helped us to improve the manuscript in many aspects, surely including the english language.

Reply on the general remarks: 1. We welcome very much any help by native En-

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glish speakers and gratefully took the chance to improve the applied language at many points.

2. The analysis method section got much more attention and has been considerably extended for the O₂/N₂ measurements. For the gas chromatographic measurements more detailed references have been included, a reference to work of Worthy et al. as well as of our own group.

3. Indeed the wording for trace gas concentrations and O₂/N₂ ratios had not been consistent, and unfortunately even wrong in some cases. We now always use 'concentrations', given in ppm or ppb, for trace gas mixing ratios, and always use 'O₂/N₂ ratios' as a measured proxy for oxygen concentrations. In the theoretical description of the carbon and oxygen cycles we also use the wording 'O₂ concentration'.

4. 'ppm' are used consistently now throughout the whole manuscript.

5. We double-checked and updated the reference list.

6. The use of 'anthropogenic' and 'fossil fuel-derived' has been rectified.

7. Oxygen and carbon dioxide are now consistently written as 'O₂' and 'CO₂', resp.

Reply on the page-specific remarks and suggestions.

In principle all of the remarks were assimilated into the text as suggested by Dr. Manning, removing ambiguous expressions and incorrect wording. Given the large number of small (but important) improvements, we will here mention only those that were for some reason not introduced as suggested, need more explication, e.g. for a certain choice we made, or lead to more substantial changes.

p. 20121/4. and other places: CH₄ and CO were not included in the computation of APO. This is now consistent in the text and the equations.

p. 20121/5. Our GC is still a real Hewlett Packard one, acquired before Hewlett Packard changed names to Agilent.

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p. 20124/3(2): At this point the time lag between CO₂ production and O₂ usage is important. While soil respiration processes lag behind CO₂ uptake (and O₂ release) by the plants by hours (root respiration) to seasons and years, no long time lag would be expected between the O₂ uptake into and CO₂ release from the soil.

p. 20125/3. 113 per meg is the right value, now consistent in text and table.

p. 20126/1.2. Here we want to point more on the temporal evolution of the trends than on the spatial differences, therefore we have chosen a time period that has no overlap with the records reported in our study. Furthermore here the focus is on airborne O₂ and CO₂ trends more than on the source partitioning from APO based calculations, therefore we think that here a general reference reporting a consensus average for these two species, such as the Third IPCC report is an adequate reference to cite. Further on in the text we will refer extensively to Manning and Keeling (2006) and Tohijma et al. (2008), for comparison with our findings.

p. 20128/2. These are marginal effects (artefacts) of our (quite short) record and do not reflect the physical truth. This is stated now in the text as well.

p. 20129-31: Section 4.3 has been largely changed according to the suggestions of Dr. Manning, especially regarding the calculation methodology (p.20130, 1.).

p. 20132/1. APO discussion has been rewritten according to the suggestions.

p. 20134/3. We did not mention a land biosphere model as no land biosphere model has been included. As we only compare model output APO with APO calculated from measurements, and not O₂ concentrations or O₂/N₂ ratios, the land biosphere part would be removed from the model output anyhow.

p. 20134/4. As mentioned above, we did not use CH₄ and CO for the calculation of APO, so consistency between measurements and modelling is given. This is stated clearly now.

p. 20135-37/3. APO calculated from measurements includes the real atmospheric

fossil fuel oxidative ratio, i.e. the difference between the biospheric oxidative ratio of 1.1 and the real fossil fuel oxidative ratio of generally 1.4 (or in our case even higher), for the portion of fossil fuel derived CO₂. The APO definition does not take a fossil fuel CO₂ fraction into account. By changing the oxidative ratio of the fossil fuel fraction and including its seasonal cycle in the model output we can try to make the modelled APO meet the measured APO even better. We tried to clarify this in the text.

p. 20135-37/4. We included the meanwhile (dec. 2008) published paper by Kozlova et al.

p. 20135-37/6. We introduced ϕ^* , in analogy to the already earlier defined APO*.

p. 20135-37/7. The significance is not too high, looking at the uncertainties, but still the one value at Mace Head is a bi-monthly mean. The same structure can be detected at Lutjewad as well, by the way. We added a few words about the significance.

The figures have been improved, they had been printed smaller than expected.

Reply on the referee report by Anonymous Referee 2

The authors thank you very much for the review of our manuscript! We addressed and rectified all the specific issues mentioned by you and - also with the help of Dr. Manning - used a more concise language. The references are tidied up, something that should have been finalized before submission but obviously went wrong in a few cases. The long-term stability of the internal CIO oxygen scale is certainly of crucial importance to the significance of the observed trends, it will be addressed by another manuscript in preparation, for the whole of the CIO oxygen measurements until 2009. Also for this aspect, a longer time series helps to judge on the long term behaviour and eventual trends. Furthermore, in principle we do agree with you in leaving out the factor (1000 for permil or 1000000 for per meg) in the definition of the delta values. However, nowadays it seems to be common use to include the factor into the formula, so we keep the factor (adjusted to the correct 1000000) in as well.

Reply on the referee report by Anonymous Referee 3

The authors thank you very much for the review of our manuscript! In the revised version of our manuscript we include the resp. numbers for the pair agreement of the flask measurements (i.e. including sampling, storage and analysis), which indeed is valuable information. We hope that the text is now clearer than in the first version, e.g. regarding the influence of the Gulf Stream on the north-western european continent and its terrestrial biosphere.

Reply on the comment by Dr. W. Tych

The authors thank you very much for your comments on our paper, which prompted us to improve on the manuscript by emphasizing the (low) significance of the trend analysis! You are perfectly right in saying that we enter 'slippery terrain' when looking for trends in this short time series of O₂/N₂ and CO₂ concentration measurements. We are certainly aware of the fact that our short time series is not sufficient to determine trends with satisfying precision. However, as the field is still in development, even the short series are worth having a look at, e.g. to compare with other published data sets (especially the measurements in Europe by Valentino et al., 2008). As we are investigating system Earth, we also expect the trends and seasonal changes not to be constant over time, but subject to interannual and longterm changes. For CO₂ this has been thoroughly investigated by Bacastow et al. (1985) and Keeling et al. (1995) on their time series of 23 years and 17 years respectively. Only with much longer time series from Mace Head and Lutjewad we will be able to support or reject this analysis of the first five years. With more samples in the minima and maxima we will hopefully be able to better define the seasonal cycle and thus also the trend. In our case the relative importance and impact of marginal effects is much higher, making the fits more susceptible for e.g. the season of the beginning of the series. Unfortunately our analyzed samples are also not evenly spread over time, giving rise to still more uncertainty in the determination of both trends and seasonal variations. When dealing with atmospheric data, it is hardly possible to devide air parcels into

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regionally and locally influenced ones. That is why we used somewhat arbitrary CH₄ and CO-concentrations in order to judge on this.

In the revised text we much more emphasize that even the linear trend can only be weakly determined, that the shown non-linear trend is the result of the Loess-procedure, only used to detect outliers but not meant to derive significant trend numbers. We also emphasize that there are not enough data points to allow for a meaningful analysis of changing seasonal amplitudes. The same applies to the selection of the nodes for piecewise linear trends, which have not been chosen fully arbitrary but according to the "visibly" linear partition of the Loess curve, and thus indeed still not fully objectively. We tried to describe more clearly our data processing and outlier determination procedure in the revised text.

Bacastow, R. B., Keeling, C. D., Whorf, T. P.: Seasonal amplitude increase in atmospheric CO₂ concentration at Mauna Loa, Hawaii, 1959-1982, *J. Geophys. Res.*, 90, 10,529-10,540, 1985.

Keeling, C. D., Whorf, T. P. Wahlen, M., and van der Plicht, J.: Interannual extremes in the rate of rise of atmospheric carbon dioxide since 1980, *Nature*, 375, 666-670, 1995.

Manning, A. C., and Keeling, R. F.: Global oceanic and land biotic carbon sinks from the Scripps atmospheric oxygen flask sampling network, *Tellus B*, 58, 95-116, 2006.

Tohjima, Y., Mukai, H., Nojiri, Y., Yamagishi, H., and Machida, T.: Atmospheric O₂/N₂ measurements at two Japanese sites: estimation of global oceanic and land biotic carbon sinks and analysis of the variations in atmospheric potential oxygen (APO), *Tellus*, 60B, 2138211;225, 2008.

Valentino, F. L., Leuenberger, M., Uglietti, C., and Sturm, P.: Measurements and trend analysis of O₂, CO₂ and ¹³C of CO₂ from the high altitude research station Junfgraujoch, Switzerland - A comparison with the observations from the remote site Puy de Dôme, France, *Sci. Total Environ.*, 391, 203-210, 2008.

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