

Continuous records of the atmospheric greenhouse gases CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O and their radiative forcing since the penultimate glacial maximum

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This paper presents continuous histories of greenhouse gas variability and radiative forcing over the past ~150,000 years based on a carefully selected compilation of previously published ice core records and simple, generally well-described time-series smoothing techniques. The authors are experts in the field of ice core gas measurements and well qualified to undertake this data compilation study (albeit they are not always the originators of all the data used here). The atmospheric histories presented here will be of interest to a number of modelling studies (particularly transient simulations) currently underway and to a wide-range of paleoclimate researchers seeking the best ice core gas data.

I have a number of comments on the paper that are individually minor and can probably be addressed with further clarifications in the text. However, in aggregate they probably amount to a major revision. I will outline a few broader points, then digress with a suggestion, and end with some line-by-line comments.

### **Edge effects and stitching the splines together**

The use of splines to smooth atmospheric gas records (particularly CO<sub>2</sub>) is relatively wide-spread and justified here. However, there are a number of pitfalls that may be encountered and need to be addressed with further clarification in the text. My main concern is that splines can sometimes “overfit” the data if they are weighted too heavily to minimizing the data fit while allowing the curvature to vary greatly. In practice this can cause the spline to produce “overshoots” in between data points. The authors state that “ideally, a smoothing spline removes all high frequencies (sic) sine functions”. I would suggest that it’s also possible for splines to produce erroneous high-frequency variability if the cutoff is too small. Though I believe this is generally not the case (but see comments on N<sub>2</sub>O and CH<sub>4</sub> in the Holocene), how the analysis has tested for these possible errors needs to be discussed.

It was unclear to me about how the final, continuous spline is constructed from a number of different, discrete time intervals. Are the splines calculated with overlapping data and then truncated at both ends? Or are the splines simply spliced end-to-end? The tables provided show tstart and tstop intervals that are almost always separated by intervals that are much greater than dt (in other words there appear to be gaps between the splines). I suspect some crucial information on the method is missing here. My worry is that there can be significant edge effects with splines and therefore the truncation method could induce some artificial features in the final spline. By the eye this doesn’t appear to be a problem but a little more information is needed.

### **A thorough description of the smoothing induced by the splines, including modification of the data files.**

At the moment, the description of the spline fitting routine is fairly broad, relies a little too heavily on other references, is not exactly precise, and contains some grammatical errors. See line-by-line comments for more details, but broadly I would like to more emphasis on how the Pcutoff value relates to the smoothing of the data.

The authors state the code used to construct the splines is “based” on Enting, 1987. As there are also a number of software packages capable of spline fitting (Matlab, IGOR, even Excel, etc.) which other researchers may apply to the data for comparison it would be useful to have little information about the actual code used. Is there a specific source for this code (I believe the Enting used to be available online) or will the new code for constructing the splines be made available? I didn’t see anything in the ESSD requirements regarding code availability, so it doesn’t appear necessary for publication, but perhaps it should be considered by the authors in the spirit of open-access/open-source. Also, there are a wide variety of smoothing splines techniques, so more information is needed on what specific type of spline technique has been used.

The authors have done an excellent job at producing well referenced and easily understood data files. I am particularly glad to see a complete list of references for the original data with tags to individual data points. However, one important aspect of the study has not been transferred to the data files is the smoothing induced by the splines. In the spline files, I would suggest including the Pcutoff value so a user of the data can readily assess the degree to which the data has been smoothed.

I was somewhat surprised to not see any uncertainty analysis carried out in this study. Varying the Pcutoff values and the data within their measurement uncertainty in a Monte Carlo analysis is relatively standard. Perhaps because the error in the final data product is the radiative forcing dominated by other uncertainties, this was deemed unnecessary? However, I don't see any error analysis carried over into the radiative forcing histories either (see comments below on radiative forcing).

### **Calculating radiative forcing**

The radiative impact of the greenhouse gases are calculated based on Myhre et al., 1998. First, in all three equations presented in the text (5,6, and 7) all the terms need to be defined. Second, how does this approach differ from previous calculations of past radiative forcing (Joos and Spahni, 2008; Schilt et al., 2010)? I was under the impression that it is standard to include the interacting effects. A naïve question: is there a set protocol for calculating radiative forcing from a body like the IPCC, CMIP or the PALMOD project that this study is focused on?

### **Comparison with Schilt et al., 2010**

In a similar vein, I would strongly suggest adding a brief comparison with the results of Schilt et al., 2010, which produced a very similar data product of radiative forcing. What new data has been added? What improvements had been made in chronology? Has the method for calculating radiative forcing changed?

### **One possible suggestion**

As argued in this paper, data compilations like this are very useful for getting the best data into the hands of non-specialists. However, I've also found that a compilation can act as a stand-in for a substantial body of work and obscure the original research publications within the scientific literature. Whether because of convenience or unrealistic limitations on references in some journals, a compilation is often cited in place of the original work. It also seems to me that even when a new ice record supplants older data as the new "reference record", that new work owes a great deal to everything that came before. I wonder if the paper would benefit from briefly mentioning a wider body of ice core gas research, perhaps as a brief introduction to each GHG section.

Within the relatively narrow scope of the paper (and journal) this doesn't seem completely necessary. But perhaps it is necessary in the sense that the reader cannot evaluate if the data being presented is truly the "best" by reading deeper into the literature. Since the reference list is nearly complete already, I think this would only involve mentioning a few of the very early studies. Again, just a suggestion to discuss.

### **"ESSD Living Data"**

Are there plans to update the compilation as new data emerge?

### **Line-by-line comments**

P.1 L.22 "within sight" sounds a little too informal

P.2. L. 13 "extend" = extent

P.3. L.10 last "or" = and

P.3. L.20 semi-colon is out of place, probably best to use "and"

P.3 L.26 Why isn't the cost function shown? Also, plural problem with "cost functions". The phrase "spline/second derivative" is very confusing, is it the curvature of the spline or is it the curvature of second derivative that is being minimized?

P.3 L.27. Starting off this paragraph with a general description of Fourier analysis leads to quite a bit of confusion, as the spline fitting and Fourier analysis are very different techniques. I believe the authors were attempting an analogy with Fourier techniques but I found the results quite muddled. Also, stating

“ideally, a smoothing spline removes all high frequencies (sic) sine functions” sounds strange given that the spline fit is not specifically designed to remove sine functions (as is Fourier analysis) but rather reduce the curvature of spline and the data-spline fit (as stated in the text). I would suggest refocusing the paragraph on the Pcutoff values and it’s relation to attenuating the amplitude of variability (at various frequencies) and then turn to an analogy with other techniques. I also would like to see a clear demonstration that the spline does indeed reduce amplitude by 50% at the Pcutoff frequency or have a stronger reference attached to this statement so the reader knows where to look. Visually, it would be useful to have a figure where the effect of the spline fitting is displayed on artificial time-series (for instance a set of sine curves possibly including some with red noise), but this is just a suggestion.

#### **Atmospheric CO<sub>2</sub>**

P.5. L.15 “goes back in time until” = extends to

P.6 L.12 Simply quantifying the offset between WDC and EDC would be sufficient here. The sentence is a bit convoluted at the moment and needs to be rewritten.

P.7 L.5 Need to explain all the terms in this equation

P.7 L.22 Need a better reference to the uncertainty than “the NOAA website”. At least a URL.

P.7 L.29 “the knowledge of abrupt changes” why not something like “to preserve the rapid variability described in Marcott et al.,”

P.7 L. 30 use of “could be” makes it sound like this is simply a possibility and not what was actually done.

#### **Atmospheric CH<sub>4</sub>**

P.9 L.14. “The interhemispheric gradient of the NH...” Something is missing in this and the following sentence. Also, please state exactly how the 0.1 Wm<sup>-2</sup> calculated even if it is straightforward. Are models sensitive to the spatial pattern of radiative forcing from GHGs? The earth must be sensitive to the spatial pattern of (the much larger) changes in insolation otherwise we wouldn’t have glacial-interglacial cycles so why not GHGs?

P.9 L.22 It seems to me there are some offsets between WDC and Law Dome from 0-500CE but this could be due to age model problems.

P.9 L.24 I have already suggested to the authors in a correspondence that they should consider using the WDC continuous CH<sub>4</sub> data (Rhodes et al., 2015) as their primary CH<sub>4</sub> record from the early Holocene to the last glacial period. Not only is the highest-resolution data available, but it has already been processed by the original authors to be near continuous (2 year resolution). The authors may be interested in this recently published paper on the reproducibility and nature of centennial-scale features in the continuous dataset (Rhodes et al., 2017).

The other thing to note regarding the WDC CH<sub>4</sub> is that the complete record is actually the combination of two different labs: Oregon State (late-Holocene, deglaciation) and Penn State (early-Holocene and last glacial period). I would suggest the authors correspond with these groups to make sure the datasets provided online have been harmonized for any small differences in procedural corrections.

Finally, it is not clear whether the spline fit to the CH<sub>4</sub> data in the early-Holocene have been overfit from the figure provided. In fact it looks like there is a decrease in the magnitude of variability around 4ka, which is right when the Oregon State data (Mitchell et al., 2013) starts. A clearer figure spanning the entire Holocene, a comparison with the continuous data, or references that described this high-frequency variability in the early Holocene are needed to convince the reader that the spline has faithfully captured real variability.

P. 10 L. 21 Regarding the reference to Ivanovic et al.. 2016, my understanding is that those experiments are transient and thus need continuous data. I think a brief but closer comparison with the data in Ivanovic is warranted given that they basically used WDC CO<sub>2</sub> with EDC CH<sub>4</sub> (despite the obviously higher-quality of the WDC CH<sub>4</sub>).

#### **Atmospheric N<sub>2</sub>O**

P.11 L.1 Please reword some of the informal terms (e.g. “get...right” = obtain accurate....)

P11. L.3 Remove “again”

I was surprised to see the spline fits capture high-frequency variability in N<sub>2</sub>O during the Holocene. The Pcutoff in this study is set to 1000 years, yet the original data publication (Flueckiger et al., 2002) use a

cutoff of 1500 years (only very tentatively) and 3000 (deemed the robust features). It seems this paper have overfit the data here. This and possibly some of the early-Holocene CH<sub>4</sub> data are the only places I could see the potential for improper use of the spline fitting procedure.

Overall, I was less convinced by the (difficult) choices the authors made regarding the N<sub>2</sub>O reconstruction. For all intents and purposes, the authors have averaged a number of different ice cores that appear to each their own unique biases. Given that Schilt et al., 2010 focused on reconstructing N<sub>2</sub>O and subsequently produced a very similar set of radiative forcing histories, I would like to see a brief comparison with further justification for the inclusions of the NGRIP data. It seems clear that quite a bit of work on reconstructing N<sub>2</sub>O is still needed.

When discussing the differences between NGRIP and the Antarctic cores, it should be noted that such a large interhemispheric gradient is impossible to maintain given the residence time of N<sub>2</sub>O in the atmosphere (~100 years).

P. 12 L.24 remove “also”

## References

P.15 L.10 Baggenstos, D., reference can updated to a paper in discussion in *Climate of the Past*  
P.15 L. 23 “CO<sub>2</sub>” subscript  
P.15 L. 32 “&ndash” is an incorrect  
P.17 L.10 Subscript problem  
P. 17 L.19 Is Meinhausen now accepted?  
P. 18 L.19: “N<sub>2</sub>O” subscript

## Tables

Table 2: In the reference to Monnin et al, 2004 for Holocene CO<sub>2</sub> data, it says only EDC data was used. This paper also presented EDML and Taylor Dome CO<sub>2</sub>, so I just wanted to check that these data were excluded from the Holocene CO<sub>2</sub> record.

Table 4/6/8.

Pcutoff (the realised cutoff period) should be defined in the text.

## Figures

Figures mix “yr BP” and “kyr BP”. They should be made consistent, ideally with all the conventions in the manuscript. Given the use of both CE and BP (and the more confusing B2K used in some ice core publications), a definition of BP is needed in the main text. This can sometimes be confusing for researchers outside the paleoclimate field, who will likely be interested in this paper.

Data files on Pangaea:

Methane is misspelled in the data labels as “Methan”

Cordially,  
Thomas Bauska

References in this review:

Enting IG (1987) On the use of smoothing splines to filter CO<sub>2</sub> data. *J Geophys Res Atmospheres* 92:10.

Baggenstos D, et al. (2017) Atmospheric gas records from Taylor Glacier, Antarctica, reveal ancient ice with ages spanning the entire last glacial cycle. *Clim Past Discuss* 2017:1–26.

Rhodes RH, et al. (2015) Enhanced tropical methane production in response to iceberg discharge in the North Atlantic. *Science* 348(6238):1016–1019.

Flückiger J, Stocker, TF, Chapellaz, J, Reynaud, D, Barnola, JM, Monnin, E, Stauffer, B, Schwander, J (2002) High-resolution Holocene N<sub>2</sub>O ice core record and its relationship with CH<sub>4</sub> and CO<sub>2</sub>. *Glob Biogeochem Cycles* 16(1):10-1-10-8.

Rhodes RH, et al. (2017) Atmospheric methane variability: Centennial-scale signals in the Last Glacial Period. *Glob Biogeochem Cycles*:n/a–n/a.

Schilt A, et al. Atmospheric nitrous oxide during the last 140,000 years. *Earth Planet Sci Lett* 300(1–2):33–43.

Monnin E, et al. (2004) Evidence for substantial accumulation rate variability in Antarctica during the Holocene, through synchronization of  $\text{CO}_2$  in the Taylor Dome, Dome C and DML ice cores. *Earth Planet Sci Lett* 224(1–2):45–54.