

Interactive comment on “Carbon isotopic signature of coal-derived methane emissions to atmosphere: from coalification to alteration” by G. Zazzeri et al.

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Received and published: 16 June 2016

The authors provide a survey of stable carbon isotopic signatures of methane, emitted from coal deposits of different rank, collected around opencast mining areas and in the vicinity of vents from underground mines. They analyzed atmospheric grab samples with different elevated CH₄ concentrations and used the Keeling plot method to derive mean isotopic signatures of the CH₄ emissions. Their results obtained from different mining areas in England and Wales as well as in Upper Silesia, Poland, and in Australia fit well into the range of earlier studies. However, they are considerably different, i.e. more depleted, compared to the values generally used in global atmospheric methane budgeting studies. A dependency of the isotopic signature on the coal type (rank), and

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also on the situation of the sampled coal deposit is found.

The study provides very valuable new data that deserve publication. However, I feel that the manuscript would largely benefit from restructuring/streamlining and focusing, as far as possible, also on a generalization of the findings. This would facilitate future use of the new data by non-specialists and make them much more valuable as input into global and regional CH₄ budgeting studies.

I have a number of general points, which should be addressed by the authors in order to meet the required scientific quality for publication in ACP:

1. The results are currently presented in a very descriptive way, giving a lot of detail that makes part of the manuscript hard to read. I feel that a lot of this text, e.g. about locations, setting, would better fit into Table 1.

2. For each campaign, the authors report on the maximum CH₄ signal and present detailed figures about the measured concentration distributions. However, this information is not evaluated in any way in the manuscript (e.g. by applying a dispersion model to estimate emission rates). The concentration signals largely depend on the actual meteorological situation at the time of sampling; therefore, a value of e.g. 10 ppm, which certainly confirms that there are indeed releases from the mining area, does not tell anything quantitative about the strength of the emission. I therefore feel that most of the figures are not required. Instead, in order to give the reader a feeling about the homogeneity of the Keeling-plot derived isotopic signature at each individual location and campaign, it would be more appropriate to present, perhaps in one multi-panel figure, individual Keeling plots from ALL campaigns, therewith showing the individual data points and regression lines in a standardized way (e.g. similar as Figure 5c, may be indicating also the maximum concentration in addition to 1/CH₄).

3. As also indicated by referee #1, I would strongly recommend to prepare an additional figure that summarizes all results, may be including the already published values from Table 2, in a diagram showing the relation between isotopic signature and coal type.

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This would be of tremendous help for people who want to use these results in their (modelling) studies. Currently, the last sentence in the Abstract leaves the reader/user somehow alone with the problem that rather no representative or at least improved input value for $\delta^{13}\text{C}$ of CH_4 from coal is available. It looks to me that with the background knowledge of the authors, they could largely help to improve the current unsatisfactory situation of models using a value, which is definitely not correct/representative, no matter what the scale of the model is.

4. The authors very often claim that secondary processes, e.g. bacterial methane production (or molecular diffusion) have occurred. However, this seems to be a (pure) speculation, because no measures of these processes are presented in the manuscript. These statements need more justification!

Specific comments:

Abstract:

Line 16: Here you promise "... this study provides representative $\delta^{13}\text{C}$ - CH_4 signatures to be used in regional and global models ..." is this really the case? See my general comment #3.

Line 19: "Progression in coal rank and secondary ..." See my general comment #4

Introduction:

Line 38: "calculated" should perhaps better read "reported"

Line 48: Here the reference "Hein et al., 1997" should be added

Sec. 1.1. (there should be a section 1.2 if there is 1.1)

In general, in particular for a non-specialist, it would be more clear if the processes during coalification (many million years ago) and what the situation is today, when these coal deposits are sampled, would be discussed here separately.

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First paragraph: I am a bit confused here – is it already called "coalification" when fatty acids are converted to CH_4 ?

Line 87: should be " $\delta^{13}\text{C}$ " not " ^{13}C "

Line 108 – 112: Please clarify this sentence, do you mean ranges of 30/25 permil?

Line 117-118: See general comment #4: Has the theory (interaction with water) really been tested in the current study?

Section 2:

Line 143: Should read "km²" (not km)

Line 164: the word "origin" is missing

Line 183: Is it important to mention here the precision of $\delta^{13}\text{C}$ - CO_2 measurements?

Line 185: Please add " CH_4 "

Line 197: Please give reference to the ArcGIS software (if these maps are not removed, as suggested above).

Line 204: Please be precise: It is not possible to measure mole fractions of emissions

Section 3: A lot of this detailed information may better go into a table.

Line 248: Do you really mean +33.3 permil?

Line 255: Please be precise: an emission cannot be "above background"

Lines 292 ff and line 310: please see general comment #4

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-235, 2016.

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