

Interactive comment on “Measurements of atmospheric He/N₂ as an indicator of fossil fuel extraction and stratospheric circulation” by Benjamin Birner et al.

Anonymous Referee #1

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This is a very well-written manuscript describing a new technique for an improved quantification of He/N₂ ratios in air samples; a development that could contribute to answering many important questions in tropospheric and stratospheric science. My main criticism is that there is a lack of detail in places, but I would recommend publication after the comments below have been dealt with appropriately.

L24 Why presumably? Are there other possible causes e.g. discussed in the 7 papers cited here?

L34 14C is neither a trace nor a greenhouse gas.

L42-44 This is correct for the tracers listed here, but not for more recently introduced

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ones in e.g. Leedham Elvidge et al., ACP, 2018. Also, as stratospheric air can be several years old, these gases are governed by their respective tropospheric growth rates and its variability over time (and so is the He/N₂ ratio).

L50 Isn't this limit dependent on the amount of air used?

L53 This should probably be just Boucher et al., 2018c.

L82 Please cite Fuller et al. properly.

L96 Does "the level of roughly ± 30 per meg per year" describe the analytical precisions or the actual trend? Also, consider splitting this very long sentence into two.

L110 This should be "on the order of" or perhaps "below" (considering Table 1, where estimates in the 10⁸ range are listed). Do these estimates include the impact of large volcanic eruptions?

L124 "Fig. 2C" creates the impression that the figure consists of several separate sub-figures which is not the case. Consider rephrasing e.g. to "C in Fig. 2".

L124-126 It's not clear to me from this description a) how the pistons move the tubing, b) how stress to the tubing moving outside the chamber is dealt with and c) how often this part of the system develops leaks (or whether this has been checked at all). See also my comment on the caption of Fig. 2.

L130 Please specify which vacuum grease is used.

L131 Why does the outlet capillary have to be thermally insulated? What is the required temperature stability? Also, to which diameter has the capillary been crimped and why?

L134 Please quantify "high purity".

L145-146 Which criteria are used to judge whether the Ti needs replacing? What are the volume and dimensions of the getter oven and how is it heated?

L152-154 How is a "complete flush-out" ensured? What is the internal volume of the

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flushed-out parts? Shouldn't the getter oven be flushed for longer since the flow in there breaks down by a factor of ~ 100 ?

L171 How does one adjust the crimping of the capillaries, especially without risking to break them?

L165-166 At which temperature?

L166-167 That sounds dangerous. How is compliance with Health and Safety regulations ensured?

L171-173 What is the nature of this cold trap and how is its temperature stability ensured? What is its internal volume and how often does it need cleaning/exchanging? What mechanisms are in place to prevent ice building up in the vicinity?

L186 Have any other cycle times been tested?

L203-205 If the uncertainty of the correction is 6 per meg, why does it increase the analytical uncertainty by only 2 per meg? Also, what about the uncertainty of 1.5 h measurements?

L211-213 What are the uncertainties of the corrections?

L216 I'm not sure that the derivation of this relationship should be part of a Discussion section as it's rather a methodological part of the manuscript. Also, how much larger than relative changes in N_2 are relative changes in CO_2 ?

L217-265 This section focuses on the multitude of possibilities that the new technique might enable, but fails to draw much attention to its limitations (e.g. the large amount of air required, which is not easily acquired from the stratosphere, or the long analysis times, which will limit constraints on spatial gradients in volcanic plumes or near oil or gas facilities).

L235 Do the authors perhaps mean "possible complications that affect $3He$ measurements" here?

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L273 2 * "avoids the need"

L278-282 I'm not sure that it's necessary to list all these references again. It certainly doesn't help the readability.

L451-453 Very little detail is provided (in both the figure and the caption) on how the piston system looks and operates. Since this is a crucial part of the inlet system I urge the authors to expand their explanation and perhaps include a cross-section of this chamber. Also, how does the pneumatic control work?

L477 How many sigmas?

L490 Table 1 has a lot of empty space and at the same time an enormous amount of footnotes, which make it rather difficult to follow. Consider reorganising, e.g. through naming the references in the table or moving some of the explanations into the caption. Also, what uncertainty range is connected to the assumption of $3\text{He}/4\text{He} = 3\text{e-}8$?

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