Atmospheric Measurement Techniques Discussions

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Comment on amt-2022-242

Anonymous Referee #2

Referee comment on "Short-term variability of atmospheric helium revealed through a cryo-enrichment method" by Benjamin Birner et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2022-242-RC2, 2022

Evaluation of « Short Term variability of atmospheric Helium…"by Birner et al.

Variations in the abundance of atmospheric helium have the potential to trace the anthropogenic cycle of CO2. Radiogenic helium-4, generated as an alpha particle by the decays of U and Th in the crust, is readily released to the atmosphere together with carbon during exploitation of natural gases. The flux rate of C is well constrained from mining statistics but the potential sinks of CO2 are less known, and using an independent tracer of carbon release le 4He can help to better understand the carbon cycle over the last decades. The Scripps group has developed a novel analytical method to measure variations in the amount of atmospheric helium by measuring at high precision the 4He/N2 ratio in air sampled at different periods during the Anthropocene. The rationale is that, assuming that N2 is constant, secular increase of the ratio should be the result of release of hydrocarbon-related 4He from the crust. The group has validated the method by reporting an evolution curve of 4He/N2 since 1974 that fits well the expected flux of 4He released by hydrocarbon exploitation. This remarkable achievement, which constitutes a world premiere, required processing large quantities of air and long duration measurements. It was not possible to investigate short-term variations over short periods of time like weeks or months that could have a potential interest for tracing local sources ort air mass movements.

To circumvent this problem, the group has improved the analytical method by concentrating the amount of 4He with a cryogenic trap and by automatizing the whole process including regeneration of the trapping mechanism. This contribution describes in detail the new system and reports validation tests in terms of analytical precision, reproducibility, and short term variation of 4He concentration in air daily sampled on the Pacific coast.

This is a well written technological paper that opens new areas of research in atmospheric chemistry and climate change monitoring. The paper is well suited for publication in Atmospheric Measurement Techniques and I recommend its acceptance after minor

corrections.

The overall rationale of the method is somewhat difficult to follow in the first part as it is based on a diagram (Figure 1) where the reader has to guess the significance of the acronyms, and the essence of the method becomes clear only in the second part of the paper. I suggest the authors to add a paragraph explaining clearly the method and the use of the different devices displayed in Fig. 1. The conversion of He/M to He to N2 needs to be better explained. It is not clear to me why the direct measurement of the 4He/N2 ratio (4/28 peak ratio) cannot be done. I suspect that there are some mass spectrometric issues (e.g., magnet hysteresis) that should be explained. The use of two mass spectrometers, if I understand well, is also unclear. Maybe the authors could add a flow chart diagram of the method. What are the use of the different calibration tanks and what are their composition?

- There are a few typos, e.g. "reference" in Fig. 1
- Explain meg in abstract
- Why not using the mu notation for 1E-6 like "solid" isotopists ?
- Is it possible to have C3+ and 04+ contributing to mass 4 ?
- What are the compositions of reference gas, and what is the composition of the standard ?