Atmospheric Measurement Techniques Discussions

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Comment on amt-2022-242

Anonymous Referee #1

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-RC1, 2022

The group of Birner has developed new techniques to analyse with high precision helium in atmospheric samples in oredr to resolve a long-standing question: the potentail rise of crustal 4He in the atmosphere since the beginning of large-scale hydrocarbon exploitation, i.e the 20th century. Results have been published elesewhere but here authors present an improved system to measure atmospheric variability of helium and associated gases (CO2) in urban area air samples to check possibly short and long-term variability and its association to natural (i.e., gas transport and degassing from oceanic breeze) and anthropogenic sources (urban traffic and other releated sources). The paper is well organized and first results interesting, I suggest some improvement to clarify a few technical points and scientific ones.

Now to explain the improvements done on their system to analyse with precision variations in He, the previous system should be explained a little bit better (lines 35-40). Sentences as "The observed changes in He/M are then converted to equivalent changes in helium to nitrogen ratio (He/N2) using separate observations of CO2, O2, and Ar" are not really clear for people unaware of teh previously developed system and this papsr should be read as a "stand-alone". The previous sentence means that you measure variations in CO2, O2, Ar and then use constant atmospheric ratios with N2 to extrapolate N2 data? It is unclear to me, likely worst for a non-specialist.

Everywhere the analytical system is indicated as MS (Mass spectrometer). At lines 79 is indicated as a GV Isoprime 100 magnetic sector mass spectrometer, which is unusual for measuring He, while in previous papers of Birner a MAT253 was used, also unusual for measuring helium. Can clearly state from beginning which MS was used in the older system and which one is now used in the improved system (possibly adding it into Figure 1' caption)? Helium is historically measured by quadrupole MS or noble-gas-MS (NGMS). Can you briefly explain the advantages and disadvantages of using an Isoprime compare other magnetic sector gas-source MS usually used by noble gas geochemists?

At lines 207-210 the diurnal variations in He and CO2 are presented. During day marine breeze would bring natural He and CO2 from the ocean, while during night, reverse air circulation would bring anthropogenically enriched He+CO2 mixtures. I suppose that the mechanism is diffusive degassing of dissolved He and CO2 at ASSW conditions. Can the CO2/He ratios sufficiently precise to support this mechanism? Or eventually can you slightly develop this point air-seawater noble gas exchange has been the focus of several papers (Seltzer et al., 2019; Hood et al., 1997) but not all community is aware about.

Lines 284-296. This last part of the future applications is less clear. I don't understand the interest of measuring neon isotopes or concentrations in the atmosphere except the fact that atmospheric Ne should be constant and thus help, if I well understand, separated atmospheric N2 variability by some geological source. Why not Kr, is mostly constant in the Universe and has no problems of double charges which could hamper your measurements in dynamic mode. About H2 would be more interesting, but authors do not relay expand this part. I would suggest stopping at line 284 their future application considerations.