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Harmonisation of atmospheric methane isotope ratio measurements from different laboratories: Procedures and Protocols

BIBHASVATA DASGUPTA[1] AND THOMAS RÖCKMANN[2]

[1]Utrecht University

[2]Institute for Marine and Atmospheric Research Utrecht, Utrecht University

Presenting Author: b.dasgupta@uu.nl

As atmospheric trace gas isotope measurements get more common and analytical techniques become more diverse, combining, and merging observational records is challenging because of significant offsets between measurements from different laboratories. To address calibration challenges in atmospheric science, we need to standardise calibration procedures, conduct regular inter-laboratory comparisons, provide adequate training and education, use accurately characterised reference materials, continuously improve calibration procedures, and encourage collaboration and communication between laboratories and research institutions. The most recent worldwide comparison among laboratories, concentrating specifically on the IRMS measurement method and encompassing analyses carried out from 2003 to 2017, revealed disparities of up to 0.5 ‰ for δ13C and 13 ‰ for δ2H isotope ratio measurements in ambient air samples. These differences are equivalent to 25 and 13 times the compatibility targets set by the WMO-GAW network, respectively. The variations were attributed to differing calibration methods and approaches to calibration propagation (Umezawa et al., 2018).

Here, we present a synthesis of data from seven laboratories experienced in high-precision isotope measurements of methane to consolidate and harmonise the available data. Anchoring the MPI-BGC laboratory as the reference frame, the data from the other six laboratories has been compared for overlapping periods employing three independent methods: (a) Average of one-to-one differences, (b) Difference between averaged data, and (c) y-intercept of correlation plot, to evaluate offsets between for each laboratory. Next, the offset corrected data has been merged based on the hypothesis and observational evidence that methane and its isotope composition are relatively homogeneous at these high-latitude sites. The data harmonisation protocol was applied to raw (event specific measurements) and smoothed (polynomial and harmonic function fitted interpolation) data to further evaluate the quality of the methods. We report near-identical results for the three methods, raw and smooth treatments, and compare them to the previously established offsets from an evaluation of numerous laboratory inter-calibration studies by Umezawa et al. (2018). The harmonisation exercise was performed on an extended dataset (1988-2023) and reported a maximum discrepancy up to 0.12 ‰ for $\delta^{13}C$ and 2.12 ‰ for $\delta^{2}H$.