Supplementary Materials:

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Error Propagation:

The equations used to calculate errors shown in Table 1 are provided below, along with brief example calculations.

i) Error related to path length (E_{PL}) was calculated as:

$$E_{PL} = \frac{0.02\ m}{PL} \quad (1)$$

For example, with a 1.1 m path length, E_{PL} = 0.018, or 1.8%.

ii) Error related to the standard concentration (E_{CH4}) was calculated as: (2)

$$E_{CH4} = 0.02\ [CH_4]$$

Where the concentration was certified to ±1% accuracy, and we used 2% (i.e. 0.02) for this analysis.

iii) Error from a gap of air between the device and calibrating chamber (E_G) was estimated assuming the gap was at an ambient concentration of 1.8 ppm, and from 4 to 5 cm wide: For example, with a gap of 5 cm, PL = 2.0 m, and standard concentration of 30.6 ppm, we calculated E_G = -4.2% using this equation:

$$E_G = \frac{[(PL - 0.05m)[CH_4] + (0.05m \cdot 1.80\ ppm)] - PL[CH_4]}{PL[CH_4]} \times 100\% \quad (3)$$

iv) Error from temperature correction was calculated as the difference in temperature correction coefficient at two temperatures differing by 2°C, i.e.

$$E_{CT}=(C_{To}-C_{T2})/C_{To}, \quad (4)$$

The manufacturer-supplied equation for temperature correction is as follows, where C_T is the correction coefficient, T is the temperature in °C:

$$C_T = 1 \times 10^{-11}\mathrm{T}^4 - 3 \times 10^{-9}\mathrm{T}^3 + 7 \times 10^{-6}\mathrm{T}^2 + 0.0032\mathrm{T} + 0.9328, \quad (5)$$

For example, at 20°C, C_T = 1.000, while at 22°C, C_T=1.007. Thus, E_{CT}=0.007, or 0.7%.

v) Error from pressure correction was calculated as the difference in pressure correction coefficient at two pressures differing by 2kPa, i.e.,

$$E_{CP}=(C_{Po}-C_{P2})/C_{Po}\ , \quad (6)$$

The manufacturer supplied equation for the pressure correction coefficient (C_P) is as follows, where P is the pressure (kPa), a and b are constants (a = -0.28862, b = 16041.81):

$$C_P = a\left[\frac{4P^2}{b} - \frac{\sqrt{2}P^2}{b}\left(\frac{\left(\frac{b}{P^2} + 2\right)\left[2 + \frac{2b}{P^2}\right]^{1/2}}{\left(1 + \frac{b}{P^2}\right)}\right)\right]^{-1} \quad (7)$$

For example, at 100 kPa, C_P=0.9917, while at 102 kPa, C_P=1.004. Thus, E_{CP}=0.012, or 1.2%.

Allan Variance:

The Allan variance was calculated for a period when the GF3 was sampling stable methane concentration. The log-log plot decreases with a -1 slope, and does not reach a minimum in this time period, suggesting that longer sampling duration would further reduce noise.

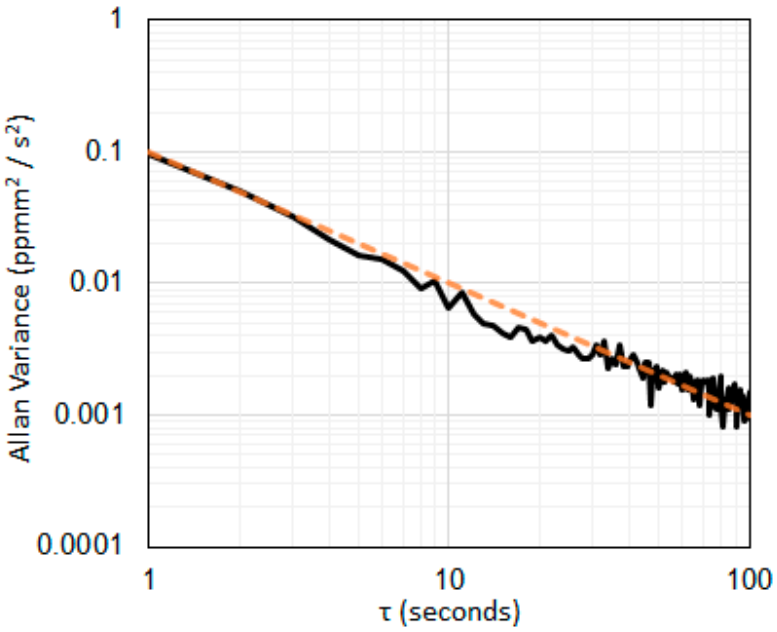

Figure S1. Allan variance (black line) during sampling of stable methane concentration. The dashed orange line shows the slope of -1.

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