Supplementary

Atmospheric CH_4 at regional stations of the Korea Meteorological Administration/ Global Atmosphere Watch Programme: measurement, characteristics, and long-term changes of its drivers

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APPEDIX A. Additional Figures

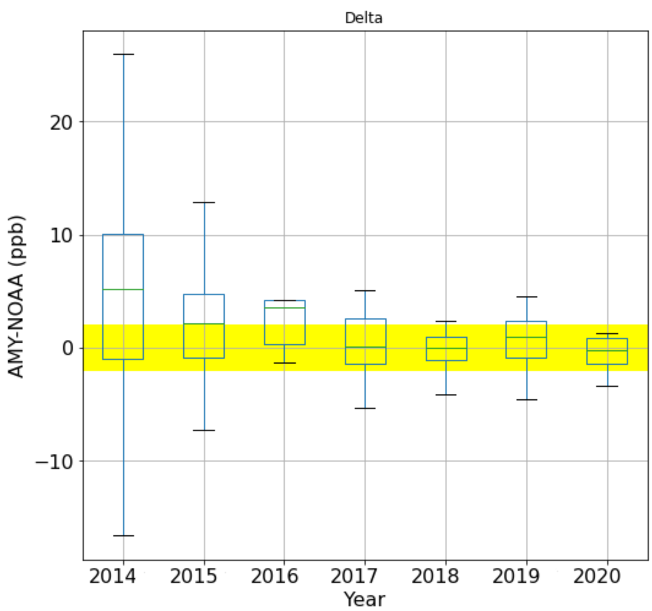
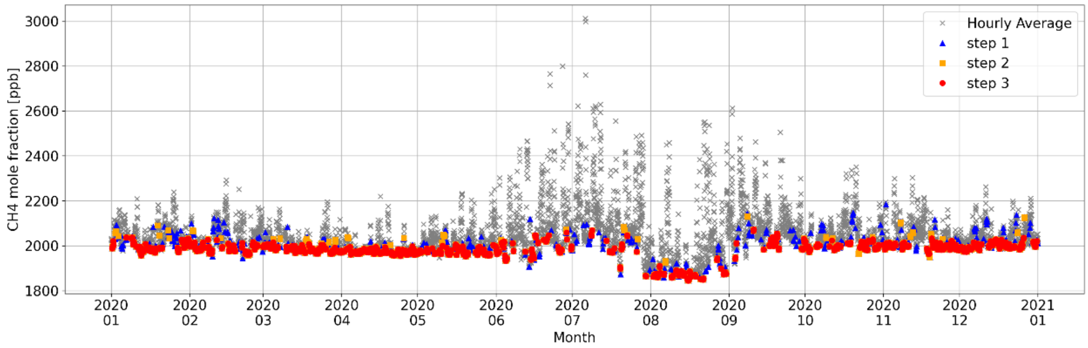
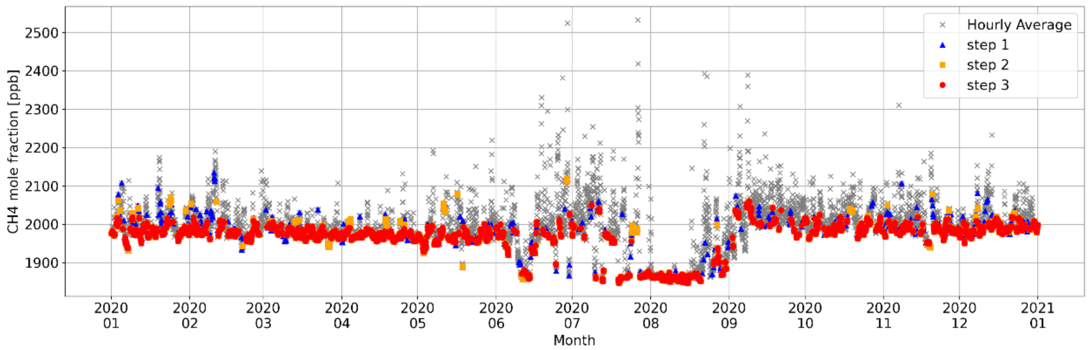


Figure S1. The box plot of CH_4 difference between AMY quasi-continuous hourly data and NOAA weekly flask data. The mean of difference is 2.2 ± 11.8 ppb.

(a)



(b)



(c)

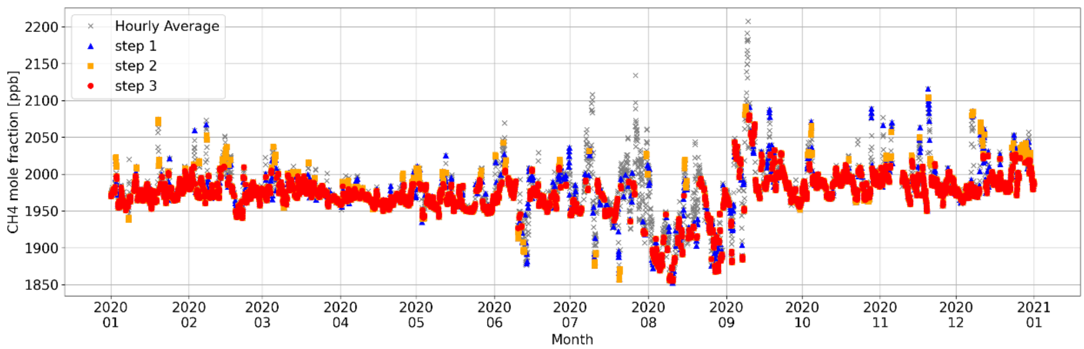


Figure S2. The time series of hourly CH_4 data through our selection method from step 1 to 3 at (a) AMY (b) JGS and (c)ULD in 2020.

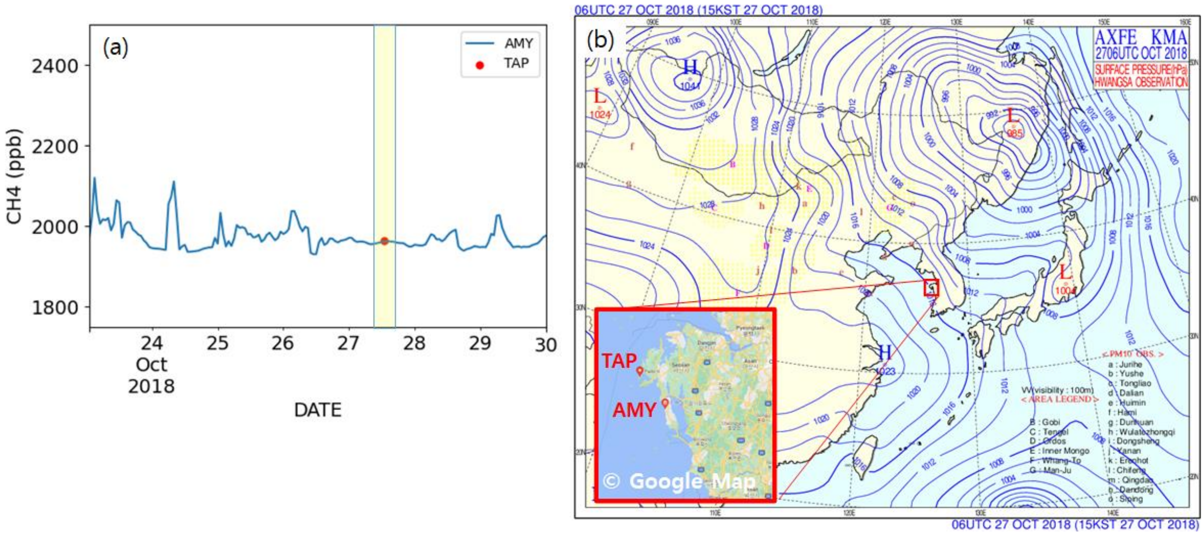


Figure S3. (a) CH_4 mole fraction observed from continuous data CRDS at AMY and from discrete flask samples at TAP (28 km away from AMY) at 15 KST, 27 Oct. 2018. (b) Surface weather chart on the same date. AMY and TAP locations were pinned on the map (derived from google map) with almost 25 times higher resolutions compared to the weather chart. Due to their close distance, two stations were affected by similar synoptic conditions. In the sampling time, the difference of CH_4 mole fraction between AMY (hourly mean) and TAP (flask) was 2.4 ppb which are close to the GAW compatibility goal(+/- 2 ppb).

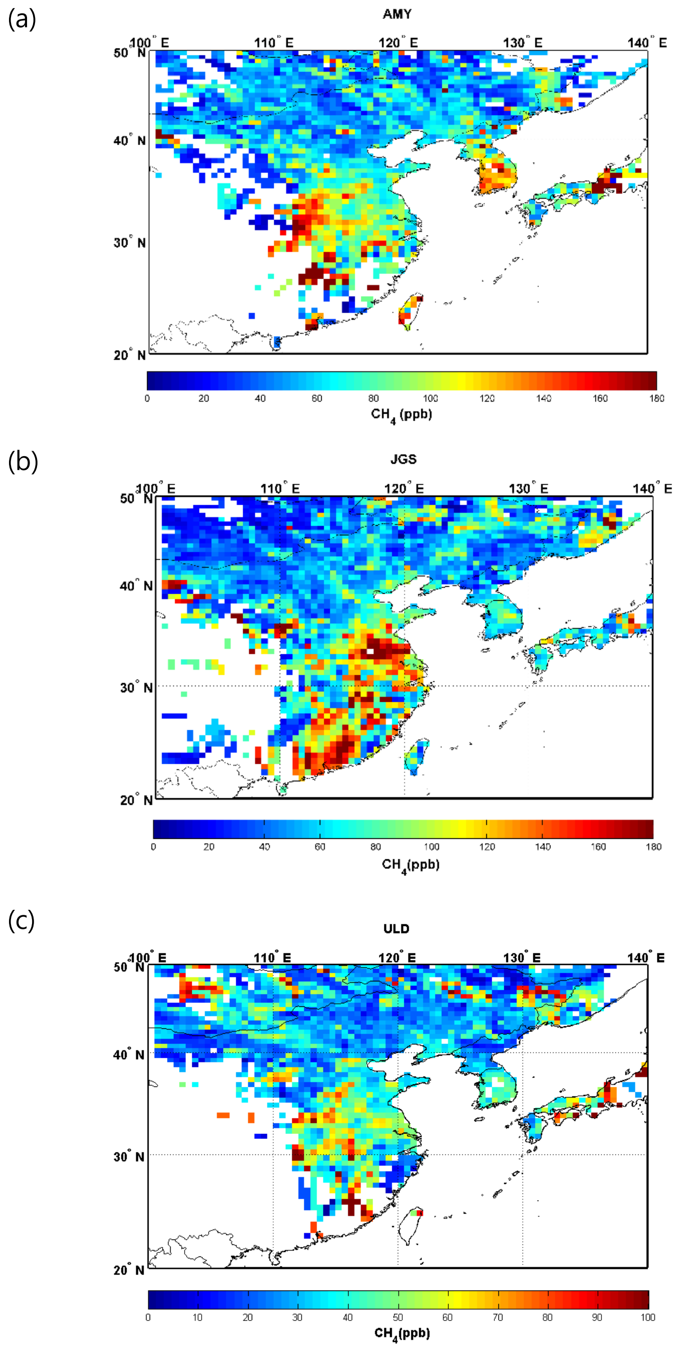


Figure S4. Distributions of Potential Source Strength (PSS) calculated with trajectory statistics for CH_4xs observed at (a) AMY, (b) JGS and (c) ULD from 2016 to 2020. The colour bars in ppb denotes the residence-time-weighted CH_4xs mean mole fractions for each grid cell computed by superimposing the back-trajectory domain on the grid matrix as described detailed in section 2.7, main manuscript. The colour bar scales are different because the synoptic wind patterns are different at each station. While the main potential emissions sources that calculated with CH_4 observation from three stations AMY, JGS, ULD are well matched, that are located widely over southern China and South Korea

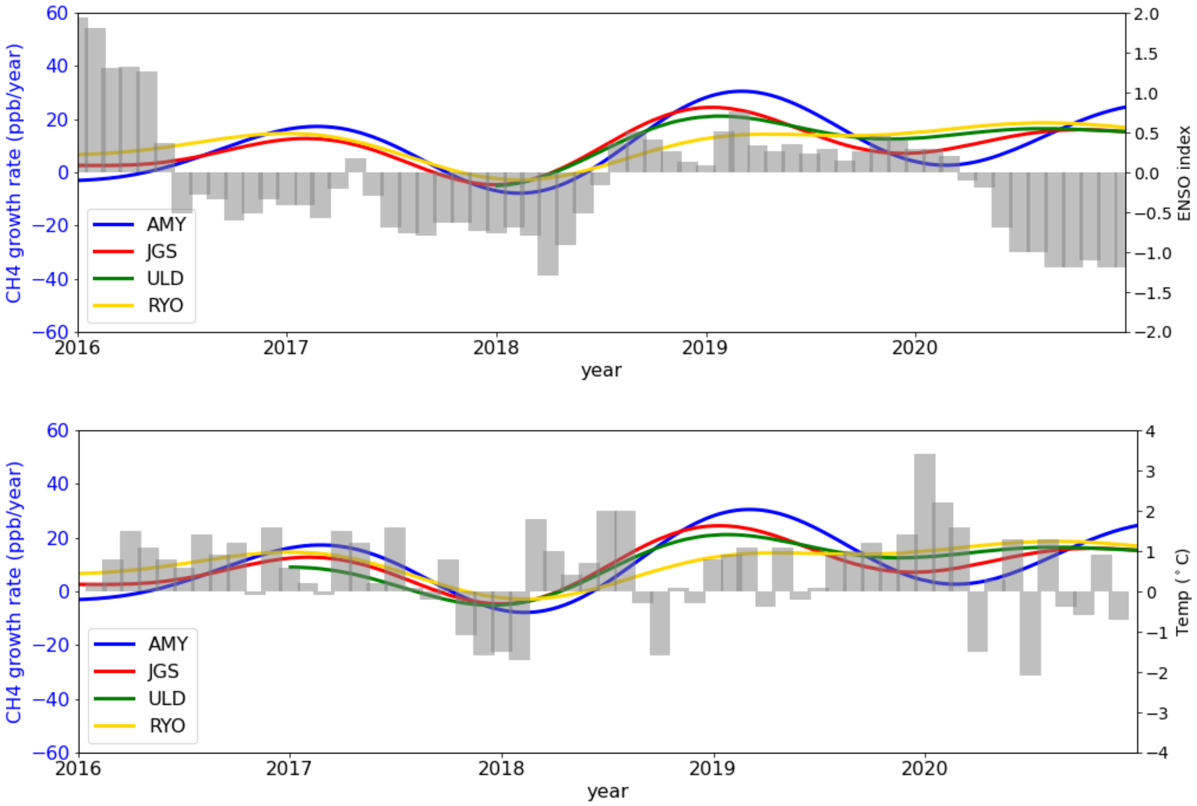


Figure S5. The trend of growth rate and ENSO (upper panel) and Temp (℃) (bottom) at four regional stations (AMY, JGS, ULD and RYO). The Temperature data were downloaded from https://data.kma.go.kr/stcs/grnd (last access: 23 Aug. 2022). These values are calculated the difference from the mean temperature anomaly.

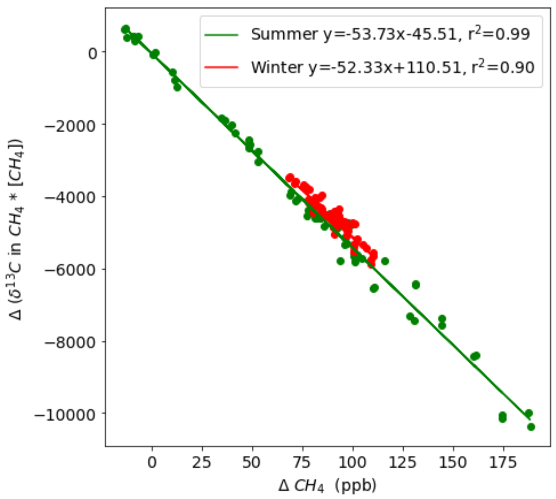


Figure S6. Miller-Tans plots showing the source signature of methane increments (Anmyeondo, AMY) into background air (Mauna-Loa). Collected from AMY station during summer (Jun, July and August, green scatters) and Winter (December, January, and February, red scatters).

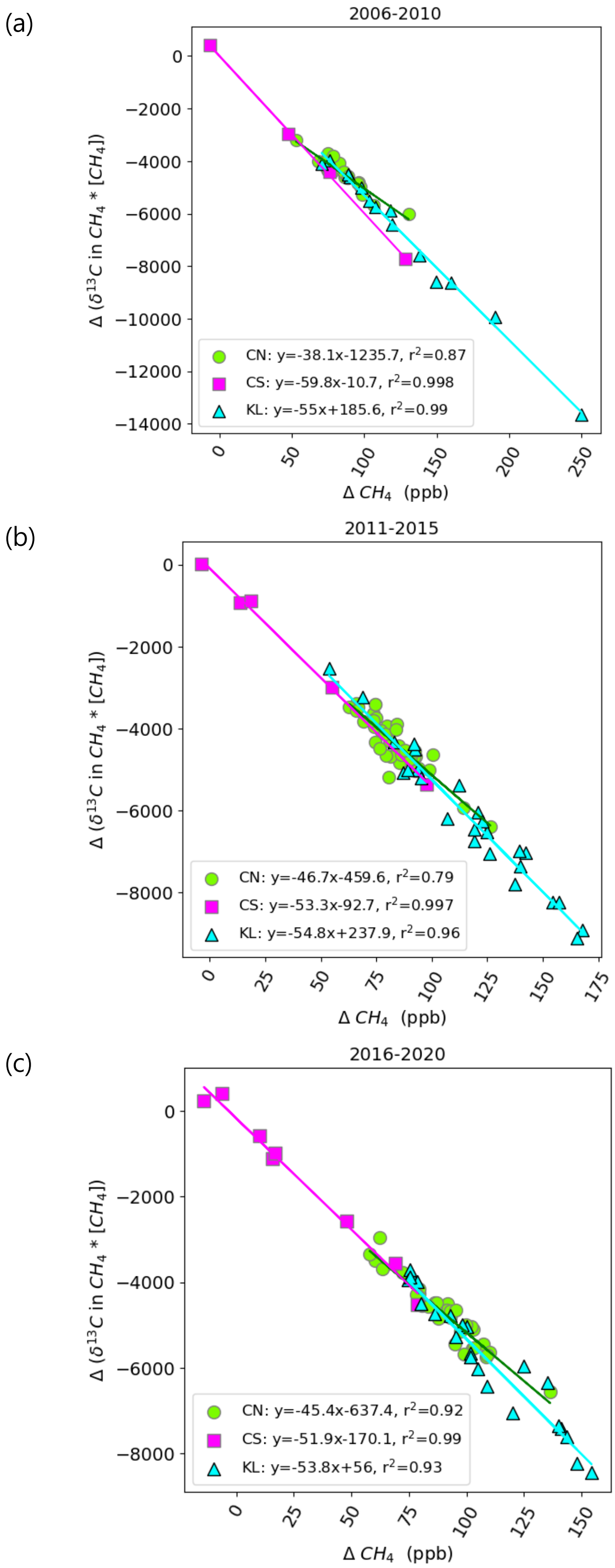


Figure S7. Miller-Tans plots showing the source signature of methane increments (TAP) into background air (Mauna-Loa) (a) from 2006 to 2010 (b) 2011 to 2015 and (c) 2016 to 2020. CN, CS and KL were three major source regions affected TAP with different slope in every 5 years.

APPENDIX B. Curve fitting method based on Thoning et al.(1989)

After the baseline selection through the 3 steps (section 2.4.2), selected data are computed by curve-fitting methods. The basic components of the curve are a second-order polynomial representing the long-term trend, and a series of harmonics representing the average seasonal cycles (Eq.S1).

$$f(t) = a_0 + a_1 t + a_2 t^2 + \Sigma_{k=1}^{4}\left[b_{2k-1}\sin\left(\frac{2\pi kt}{T}\right) + b_{2k}\cos\left(\frac{2\pi kt}{T}\right)\right]$$

Equation S1. Function fit

Where t is the time in days and T is 365.25 days.

And then calculate and filter the residuals of the selected daily data about the fit function. The method used is to transform the residuals into the frequency domain using a Fast Fourier Transform (FFT), apply a low pass filter function (Eq. 2) to the frequency data, and then transform the filtered data to the real domain using an inverse FFT.

$$H(f) = \exp\left[-\ln(2) * \left(\frac{f}{f_c}\right)^6\right]$$

Equation S2. Low pass filter function

Where f is frequency and fc is cut-off frequency. Here fc is cut-off values of 80 days for the short-term filter and 667 days for the long-term filter.

Finally determine the smoothed curve of interest by combining the function with the filtered data.

APPENDIX C. Determination of component signals

The definitions are derived from Global Monitoring Laboratory - Carbon Cycle Greenhouse Gases (noaa.gov) (https://gml.noaa.gov/ccgg/mbl/crvfit/crvfit.html, last access, 16, 2023).

Simply L3 monthly mean are calculated by average of daily smoothed data from Eq. S1 and Eq. S2. Daily smoothed data is the function fit plus the filtered residuals using the short term cut off (80 day/cycle)-1.

Long-term trend is upward growth in the data with the seasonal cycle removed. This is the polynomial part of the function fit plus the filtered residuals using the long-term cut-off value with (667 day/cycle)-1.

Seasonal amplitude is magnitude of the peak to trough of the detrended seasonal cycle. Detrended seasonal cycle can be obtained by subtracting the trend curve from the smooth curve.