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2 **Did Atmospheric CO₂ and CH₄ Observation at**
3 **Yonagunijima Detect Fossil-Fuel CO₂ Reduction due to**
4 **COVID-19 Lockdown?**

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6 **Yasunori TOHJIMA¹, Yosuke NIWA**

7 *National Institute for Environmental Studies*
8 *Tsukuba, Japan*

9

10 **Kazuhiro TSUBOI**

11 *Meteorological Research Institute*
12 *Tsukuba, Japan*

13

14 **and**

15

16 **Kazuyuki SAITO**

17 *Japan Meteorological Agency*
18 *Tokyo, Japan*

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25 1) Corresponding author: Yasunori Tohjima, National Institute for Environmental Studies,
26 Earth System Division, Environmental Chemodynamics Section, 16-2 Onogawa, Tsukuba
27 305-8506, Ibaraki, JAPAN
28 Email: tohjima@nies.go.jp
29 Tel: +81-29-850-2485
30 Fax: +81-3-6453-0612

Abstract

Synoptic-scale variabilities of atmospheric CO₂ and CH₄ observed at Yonagunijima (Yonaguni Island, YON, 24.47°N, 123.01°E) during winter (from January to March) in 1998-2020 were examined. The monthly mean variability ratios ($\Delta\text{CO}_2/\Delta\text{CH}_4$) based on correlation slopes within 24-hour time windows showed a clear increasing trend, which is mainly attributed to the unprecedented increase in the fossil fuel-derived CO₂ (FFCO₂) emissions from China. A similar increasing trend of the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio had been reported for the observation at Hateruma Island (HAT, 24.06°N, 123.81°E), located at about 100 km east of YON. However, the absolute values for YON were 34% larger than those for HAT. In addition, the monthly average in February 2020 for YON showed no marked change, while that for HAT showed an abrupt considerable decrease associated with the FFCO₂ emission decrease in China presumably caused by the COVID-19 lockdown. Investigating the diurnal variations, we found that the local influences were larger at YON, especially during daytime, than at HAT. Using nighttime data (20-6 LST) and a longer time window (84-hour), we succeeded in reducing the local influences and the resulting monthly mean $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio showed considerable similarity to that observed at HAT including the abrupt decrease in February 2020. These results convinced us that the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio could be successfully used to investigate the relative emission strength in the upwind region.

51 **Keywords** atmospheric CO₂; atmospheric CH₄; synoptic-scale variation; COVID-19

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53 **1. Introduction**

54 In December 2019, the novel coronavirus disease (COVID-19) was first reported in
55 Wuhan, China, and rapidly spread across the whole country. The government of China
56 imposed lockdown measures first in Wuhan on 23 January 2020, then extended the area
57 across the nation in February 2020. Since such lockdown measures strictly restricted
58 socioeconomic activities, a considerable reduction in fossil fuel-derived CO₂ (FFCO₂)
59 emissions was expected (Le Quéré et al., 2020). This situation raised the question of
60 whether atmospheric observations could detect the COVID-19-induced FFCO₂ reductions.

61 Substantial reductions in air pollutants like nitrogen dioxide (NO₂) were observed during
62 the lockdown period in China by using in-situ and satellite measurements (e.g., Bauwens et
63 al., 2020; Le et al., 2020). However, the corresponding change in atmospheric CO₂ mole
64 fraction was expected to be relatively small because of its rather long lifetime in comparison
65 with the above air pollutants. Satellite retrievals of the CO₂ column average (XCO₂) over
66 China were examined by several studies to detect the influence of the COVID-19 lockdown
67 (Chevallier et al., 2020; Buchwitz et al., 2021), which concluded that the detection was
68 challenging because the expected change of XCO₂ was less than the precision of the
69 satellite observations.

70 Examining synoptic-scale variations in the atmospheric CO₂ and CH₄ observed at

71 Hateruma Island (HAT, 24.06°N, 123.81°E), Tohjima et al. (2020) found that the $\Delta\text{CO}_2/\Delta\text{CH}_4$
72 variability ratio showed a marked decrease in February 2020 compared with the former 9-
73 year (2011-2019) average. Because HAT is in the downwind region of continental Asia
74 during the late autumn - early spring season because of the East Asian monsoon, correlative
75 elevations of the CO_2 and CH_4 mole fractions are often observed (Tohjima et al. 2010). A
76 previous study revealed that the gradual increase of the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio was attributed to
77 the unprecedented increase in the FFCO_2 emissions from China (Tohjima et al., 2014).
78 Consequently, Tohjima et al. (2020) concluded that the observed $\Delta\text{CO}_2/\Delta\text{CH}_4$ decrease in
79 February 2020 was related to the COVID-19 lockdown in China. Moreover, they evaluated
80 the decrease in the FFCO_2 emissions from China to be about $32 \pm 12 \%$ and $19 \pm 15 \%$ for
81 February and March, respectively, from a comparison of the observed and simulated
82 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios. However, the change in the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio associated with the
83 COVID-19 was considerably faint and was detected only in single-site observations, limiting
84 the certainty of the estimate of emission change. Therefore, to enhance the certainty, an
85 independent observation is required.

86 Yonaguni Island (YON, 24.47°N, 123.01°E), where atmospheric CO_2 and CH_4 have also
87 been observed by the Japan Meteorological Agency (JMA) since 1997 and 1998,
88 respectively, is located at about 100 km west of HAT. The observed seasonal cycles and
89 trends for HAT and YON were almost identical (Zhang, et al., 2007) because of the similarity
90 of the geographical positions. Therefore, it is expected that the CO_2 and CH_4 observations

91 at YON can be used to constrain the relative emission strengths from China. In this study,
92 we examined whether the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio observed at YON showed similar variations,
93 especially an abrupt decrease in February 2020, as the observations at HAT did.

94 **2. Data and Method**

95 2.1 Yonagunijima (YON)

96 Yonagunijima (Yonaguni Island), located at the western end of the Ryukyu Archipelago, is
97 the westernmost inhabited island of Japan. The island has a shape of an almond (longer in
98 the east-west direction), an area of about 29 km² with the highest point being 231 m, and a
99 population of about 1700. There are mountainous areas in the southern and western parts
100 of the island, covered by subtropical forests. JMA built a local meteorological station to the
101 east of the north settlement and about 800 m inland from the north coast in 1989. The station
102 is surrounded by grassland and sugarcane fields. The observation at YON is generally
103 influenced by the air masses from the Pacific region during summer and from the Asian
104 continent during winter. Such seasonality in the air mass transport also influences the
105 observation at HAT. Note that no clear diurnal patterns in wind direction and wind speed
106 were observed at YON.

107 2.2 CO₂ and CH₄ measurement systems

108 The atmospheric CO₂ and CH₄ measurement systems used at YON are briefly described
109 here; the details were presented elsewhere (e.g., Watanabe et al., 2000; Tsutsumi et al.,
110 2006). The sample air was drawn by a pump from an air intake located at the top of a mast

111 near the station building (20 m above ground level) and was delivered to the CO₂ and CH₄
112 measurement systems.

113 The CO₂ in the air sample was measured by using non-dispersive infrared analyzers
114 (NDIRs). The analytical precisions of the NDIRs were better than ± 0.02 $\mu\text{mol per mol}$ (ppm).
115 The CH₄ mole fractions were measured by a NDIR during January 1998 - December 2007
116 and by a gas chromatograph equipped with a flame ionization detector (GC/FID) after
117 January 2008. The precisions of the atmospheric CH₄ measurement systems were better
118 than ± 10 nmol per mol (ppb) for the NDIR system and ± 5 ppb for the GC/FID system.

119 The CO₂ and CH₄ measurement systems were calibrated by introducing working standard
120 gases from high-pressure cylinders, which were prepared at the laboratory in JMA
121 (Watanabe et al., 2000; Matsueda et al., 2018). The CO₂ and CH₄ mole fraction values at
122 YON were reported on the WMO scale. Note that although the CO₂ and CH₄ mole fractions
123 at HAT were reported on the NIES original scales, it was confirmed from the WMO/IAEA
124 Round Robin comparison experiment (e.g., Zhou et al. 2009) that the differences between
125 the WMO and NIES scales were kept within ± 0.15 ppm.

126 2.3 Analytical methods of the variability ratio

127 The time series of CO₂ and CH₄ at YON often showed correlative synoptic-scale variations.
128 To extract the variability ratios from such correlative variations, we used the same method
129 as Tohjima et al., (2014; 2020) used to calculate the variability ratios for the observation at
130 HAT. Since the details of the methods are given in the literature, here we give only a brief

131 description of the methods.

132 First, using a set of the hourly CO₂ and CH₄ data within a certain time window, we
133 computed a correlation coefficient (R), standard deviations for both CO₂ and CH₄ (σ_{CO_2} and
134 σ_{CH_4}), and a correlation slope by using a reduced major axis (RMA) method (Hirsch and
135 Gilroy, 1984). Shifting the time window by one hour, we repeated the above computations
136 for the entire data set. Then, setting the criteria of the correlation coefficient (R) and the CO₂
137 standard deviation (σ_{CO_2}), we discarded the correlation slopes with R and σ_{CO_2} less than the
138 criteria. Finally, using the selected correlation slopes, we calculated the monthly average
139 and the moving averages of the variability ratio. Although the time window was set to 24
140 hours in Tohjima et al. (2020), we also tried a longer time period up to 120 hours in this study.
141 Note that we used the same R=0.7 and σ_{CO_2} =0.1 ppm for the criteria as were used in a
142 previous study (Tohjima et al., 2020).

143 **3. Results and Discussion**

144 3.1 Synoptic-scale variations of the atmospheric CO₂ and CH₄

145 The time series of the hourly CO₂ and CH₄ mole fractions at YON from January to
146 February 2020 and those observed at HAT are plotted in Fig. 1. Both CO₂ and CH₄ time
147 series show similar synoptic-scale variations with periods of several hours to several days.
148 The synoptic-scale variations observed at YON were almost the same as those observed at
149 HAT except for phase differences; the temporal variations at YON generally proceed a few
150 hours ahead of those at HAT. In addition, the CO₂ short-term variations observed at YON

151 seem to be slightly larger than those at HAT.

152 We calculated the average diurnal cycles of CO₂ and CH₄ at YON and HAT for three
153 months (Jan.-Mar.) to clarify the differences (Fig. 2). The average diurnal cycle is calculated
154 as the average deviation from the corresponding daily means for the individual hours.
155 Although CO₂ diurnal cycles showed a decline during daytime at both sites, the amplitudes
156 of the decline for YON are more than twice as large as those for HAT. The declines during
157 daytime are probably attributed to the photosynthetic CO₂ sequestrations of the biosphere
158 on the islands. These results suggest that the local CO₂ fluxes more strongly influence the
159 atmospheric observation at YON than at HAT. The larger local influence at YON may be
160 attributed to the fact that the station at YON is located in a more inland area of the larger
161 island and partially that the sampling inlet is placed at a lower position.

162 Meanwhile, compared with the analytical precision of the CH₄ measurements (± 2 ppb for
163 HAT (Tohjima et al., 2002)), the ranges of the average CH₄ diurnal cycles of ± 1.5 ppb and
164 ± 1 ppb for YON and HAT, respectively, are relatively small. These results suggest that the
165 influence of the local sources on the observed atmospheric CH₄ variations is negligible for
166 both sites.

167 **3.2 Temporal change in the monthly mean $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio**

168 Monthly mean $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios based on the observation at YON in January, February,
169 and March from 1998 to 2020 are plotted in Fig. 3a together with the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for
170 HAT, reported in a previous study (Tohjima et al., 2020). As the figure shows, the $\Delta\text{CO}_2/\Delta\text{CH}_4$

171 ratios for YON are 39 mol mol⁻¹ on average larger than those for HAT (averages of 156 mol
172 mol⁻¹ for YON and 117 mol mol⁻¹ for HAT). In addition, the increasing ratios determined by
173 linear regression for the entire period (1998-2020) are larger for YON (3.2 ± 0.3 mol mol⁻¹
174 yr⁻¹) than for HAT (1.9 ± 0.2 mol mol⁻¹ yr⁻¹). The smaller increasing trend at HAT may be
175 related to the fact that the ΔCO₂/ΔCH₄ ratio for HAT reached a plateau after 2011 in
176 association with the stagnant FFCO₂ emission increase from China (see Fig.2 of Tohjima et
177 al., 2020). The increasing rate for HAT decreased from 2.7 ± 0.4 mol mol⁻¹ yr⁻¹ in 1998-2010
178 to -0.6 ± 0.7 mol mol⁻¹ yr⁻¹ after 2010, while that for YON for the later period was 3.7 ± 0.7
179 mol mol⁻¹ yr⁻¹.

180 Considering the relatively close geographical locations of YON and HAT, it is quite difficult
181 to imagine that the air masses transported from continental Asia induce such considerably
182 large differences between YON and HAT. Therefore, the larger short-term variability
183 probably caused by the local CO₂ fluxes would explain the larger ΔCO₂/ΔCH₄ ratios for YON.
184 The average diurnal cycles of CO₂ at YON (Fig. 2) show rather stable values during
185 nighttime but rapid decreases and increases during daytime, which might indicate an
186 enhancement of the short-term variability during daytime. To investigate the difference in the
187 short-term variability of CO₂ for YON between daytime and nighttime, we computed the
188 frequency distributions of the standard deviations of the CO₂ data within the 24-hour time
189 windows for the daytime (7-19LST) and nighttime (20-6LST). The result showed that the
190 peak of the frequency distribution was located at a larger standard deviation for the daytime

191 data than for the nighttime data (supplementary Fig. S1). Accordingly, there is a possibility
192 that the daytime data might enlarge the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for YON.

193 3.3 Reduction of local influences

194 To reduce the local influences on the observations at YON, we calculated the monthly
195 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio by only using nighttime data (supplementary Fig. S2). Although the
196 resulted monthly means decreased by about 20 mol mol^{-1} on average, those values were
197 still about 19 mol/mol larger than the values for HAT, suggesting the local influence was still
198 not sufficiently reduced. To further reduce the local influences, we used a rather long time
199 window for the correlation analysis. As was discussed in the previous sections, the local
200 influences appeared in the diurnal cycles, while the synoptic-scale variations usually had a
201 longer timescale of several days. Therefore, the longer time window could reduce the
202 influence of the local CO_2 fluxes on the total CO_2 variations. Thus, we calculated the monthly
203 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for YON for different time windows up to 120 hours and examined the
204 root mean square (RMS) of the differences of the monthly $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios between YON
205 and HAT (supplementary Fig. S3). The RMS showed a minimum value when an 84-hour
206 time window was used. The result using the 84-hour time window is plotted in Fig. 3b. The
207 modified $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios are similar to the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for HAT; the average
208 difference (YON-HAT) is -2 mol mol^{-1} . The increasing rates determined by the linear
209 regression were $3.7 \pm 0.7 \text{ mol mol}^{-1} \text{ yr}^{-1}$ during 1998-2010 and $0.1 \pm 0.8 \text{ mol mol}^{-1} \text{ yr}^{-1}$
210 during 2011-2020, which are also consistent with those for HAT.

211 Moreover, the modified $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for YON showed a substantial decrease in
212 February 2020. As is described in the Introduction, Tohjima et al. (2020) attributed the
213 decrease in the monthly $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio in February and March 2020 from the preceding
214 9-year averages to the decrease in the FFCO₂ emissions from China associated with the
215 COVID-19-induced lockdown. The monthly averages of the modified $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for
216 YON and the previously reported corresponding values for HAT are listed in Table 1 for
217 comparison. Note that the duration of the time window also influenced the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios
218 for HAT as suggested by Tohjima et al. (2020) and the February $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for HAT
219 decreased to 22 mol mol⁻¹ when the 84-hour time window and the nighttime data were used.
220 Even if the influence of the duration of the time window is considered, the decreases in the
221 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for YON agree well with those for HAT within the uncertainties both for
222 February and March. Although the decrease in March for YON is unclear in comparison with
223 that for HAT, using a 120-hour time window enlarges the decrease to 10 mol mol⁻¹, which
224 suggests that it is difficult to completely reduce local influences.

225 Figure 4 shows the temporal variation in the 30-day moving average of the modified
226 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for YON from January to March 2020. In the figure, the preceding 9-year
227 (2011-2019) average of the 30-day moving average for YON with the range of the
228 uncertainties (1σ) and the 30-day moving average of the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for HAT from
229 January to March 2020, which were shown in Fig. 3 of Tohjima et al. (2020), are also
230 depicted. Compared with the preceding 9-year average, the moving-averaged $\Delta\text{CO}_2/\Delta\text{CH}_4$

231 ratio for YON shows a rapid decrease between January and February, a bottom in the middle
232 of February, and an asymptotic increase toward the 9-year average. Such variations are
233 generally consistent with those for HAT. Additionally, the pattern of variations in the moving-
234 averaged $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio is similar to the estimated change in the FFCO₂ emissions from
235 China based on Le Quéré et al., (2020) which is also depicted in Fig. 4.

236 From the above results, we concluded that the atmospheric CO₂ and CH₄ observations at
237 YON could detect the signals related to the COVID-19 lockdown in China as the change in
238 the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio. This conclusion supports the idea that the atmospheric $\Delta\text{CO}_2/\Delta\text{CH}_4$
239 ratio is effective in evaluating temporal changes in the relative emission strengths in the up-
240 wind source regions.

241 **4. Summary and Conclusion**

242 To detect the signal associated with the COVID-19 lockdown, we examined the synoptic-
243 scale variability ratio of the atmospheric CO₂ and CH₄ ($\Delta\text{CO}_2/\Delta\text{CH}_4$) observed at YON during
244 the period from 1998 to 2020 by applying the analytical approach of Tohjima et al. (2020).
245 Being different from the results observed at HAT (Tohjima et al., 2020), the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio
246 was about 34% larger than that for HAT, and the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio for February 2020 did not
247 show a marked decrease. Examining the diurnal variations of CO₂ and CH₄ at YON, we
248 found that the local fluxes, probably air-to-land biosphere exchange, enhanced the CO₂
249 variability especially during daytime (7-19 LST).

250 Using the nighttime (20-6 LST) data and a longer time window of 84 hours, we were able

251 to effectively reduce the local influences on the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios; the resulting monthly
252 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios showed considerable agreement with those for HAT, including a marked
253 decrease in February 2020 in comparison with the preceding 9-year (2011-2019) averages.
254 Additionally, the 30-day moving average of the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio abruptly decreased
255 between January and February 2020, reached the bottom in the middle of February, and
256 gradually returned to the level of the former 9-year average in March. Such a decline pattern
257 is similar to the change in the FFCO₂ emissions from China estimated based on the study
258 of Le Quéré et al. (2020).

259 As is described in the Introduction, detecting the decrease in the atmospheric CO₂ related
260 to the COVID-19 outbreak is still challenging (Chevallier et al., 2020; Buchwitz et al., 2021).
261 The change in the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio at HAT associated with the COVID-19 outbreak is also
262 very faint. Therefore, the fact that a decrease in the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratio in February 2020 was
263 observed at YON convinced us of the correctness of the previous result reported by Tohjima
264 et al. (2020). This result also demonstrates the usefulness of utilizing adjacent monitoring
265 sites like YON and HAT to confirm the detection of such faint signals as shown in this study.
266 Both sites are located at the downwind side of the Asian continent that is the biggest CO₂
267 emitter in the world and hence the most important area to be monitored. Monitoring with
268 observations at YON and HAT would continue to provide valid information on emission
269 changes over the continent.

270

271 **Data Availability Statement**

272 The time series of the atmospheric CO₂ and CH₄ mole fractions at YON are available via the
273 website of WDCGG (World Data Centre for Greenhouse Gases). [WDCGG \(World Data Centre](http://www.wdcgg.com)
274 [for Greenhouse Gases\) \(kishou.go.jp\)](http://www.wdcgg.com)

275

276 **Supplement**

277 Supplement 1 shows the frequency distributions of the standard deviations for the daytime
278 and nighttime CO₂ data for YON. Supplement 2 shows the same figure as Fig. 3, but only
279 nighttime data are used to calculate the monthly average of the $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for YON.
280 Supplement 3 shows the root mean square (RMS) of the differences of the monthly
281 $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios between YON and HAT against the duration of the time window used for
282 the calculation for YON.

283

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358 Fig. 2 Average diurnal cycles of (a) CO₂ and (b) CH₄ at YON (red lines) and HAT (black
359 lines) for three months (January: triangles; February: circles; March: squares). The
360 average diurnal cycle is calculated as the average deviation from the daily means for the
361 individual hours.

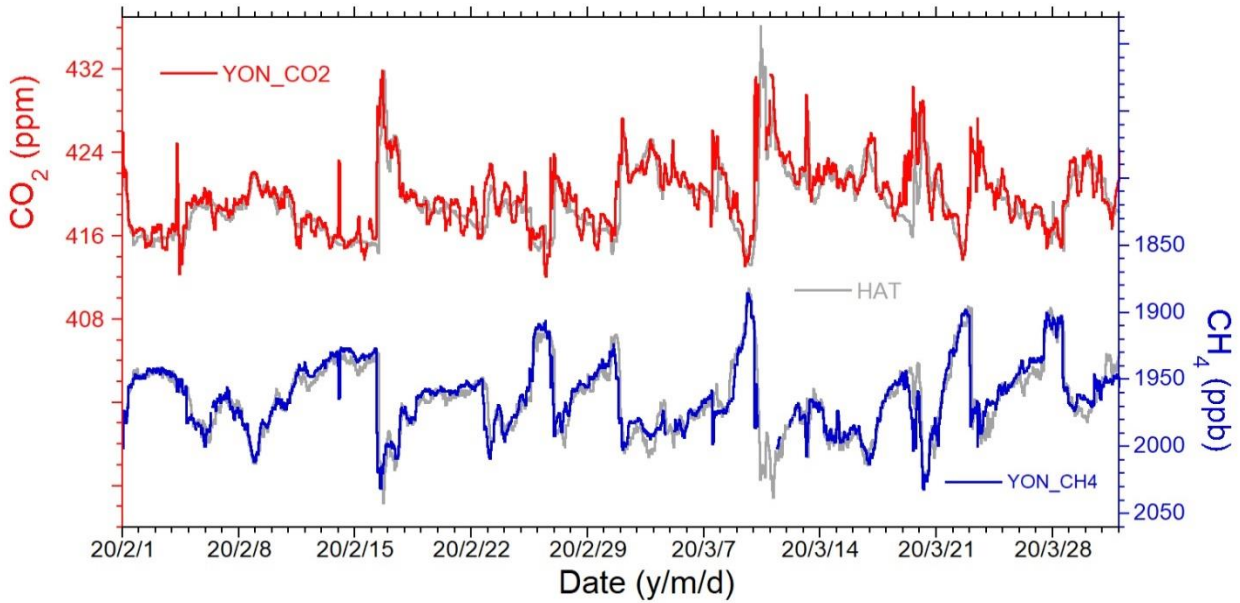
362 Fig. 3 (a) Monthly mean $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios based on the observation at YON (red closed
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365 Tohjima et al., (2020). A 24-hour time window was used to calculate the $\Delta\text{CO}_2/\Delta\text{CH}_4$
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371 to reduce the local influences (see text). The $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios for HAT were taken from
372 a previous study (Fig. 3 of Tohjima et al., 2020). The grey line with vertical bars
373 represents the preceding 9-year (2011-2019) average of the 30-day moving average for
374 YON with the range of the uncertainties (1σ). (Bottom, right Y-axis): the estimated
375 temporal change in the FFCO₂ emissions from China based on Le Quéré et al., (2020).

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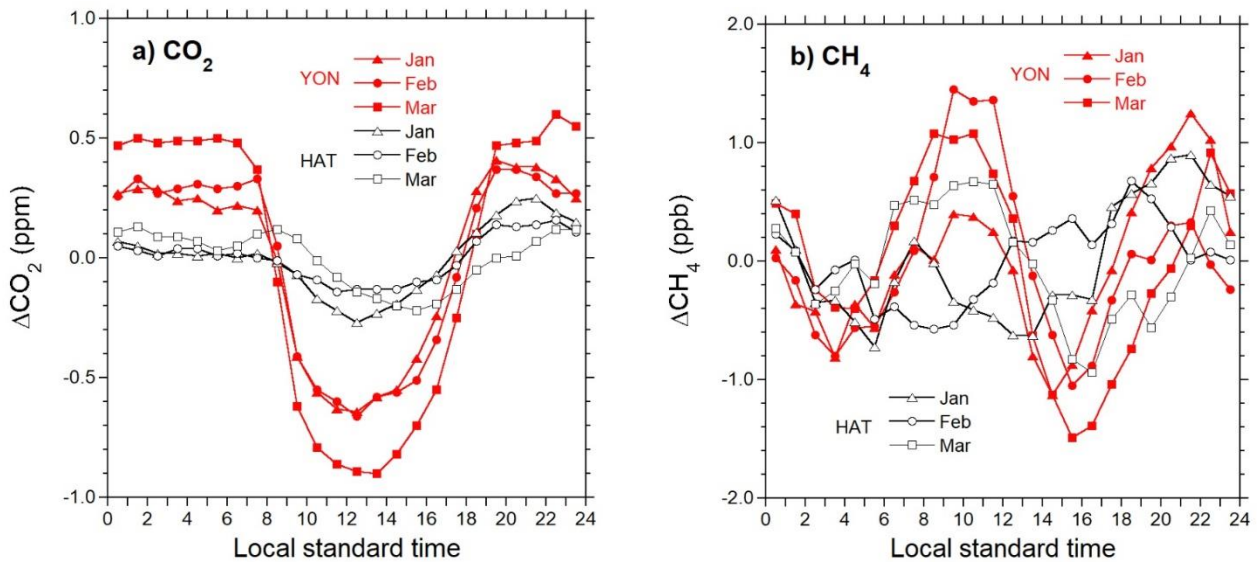
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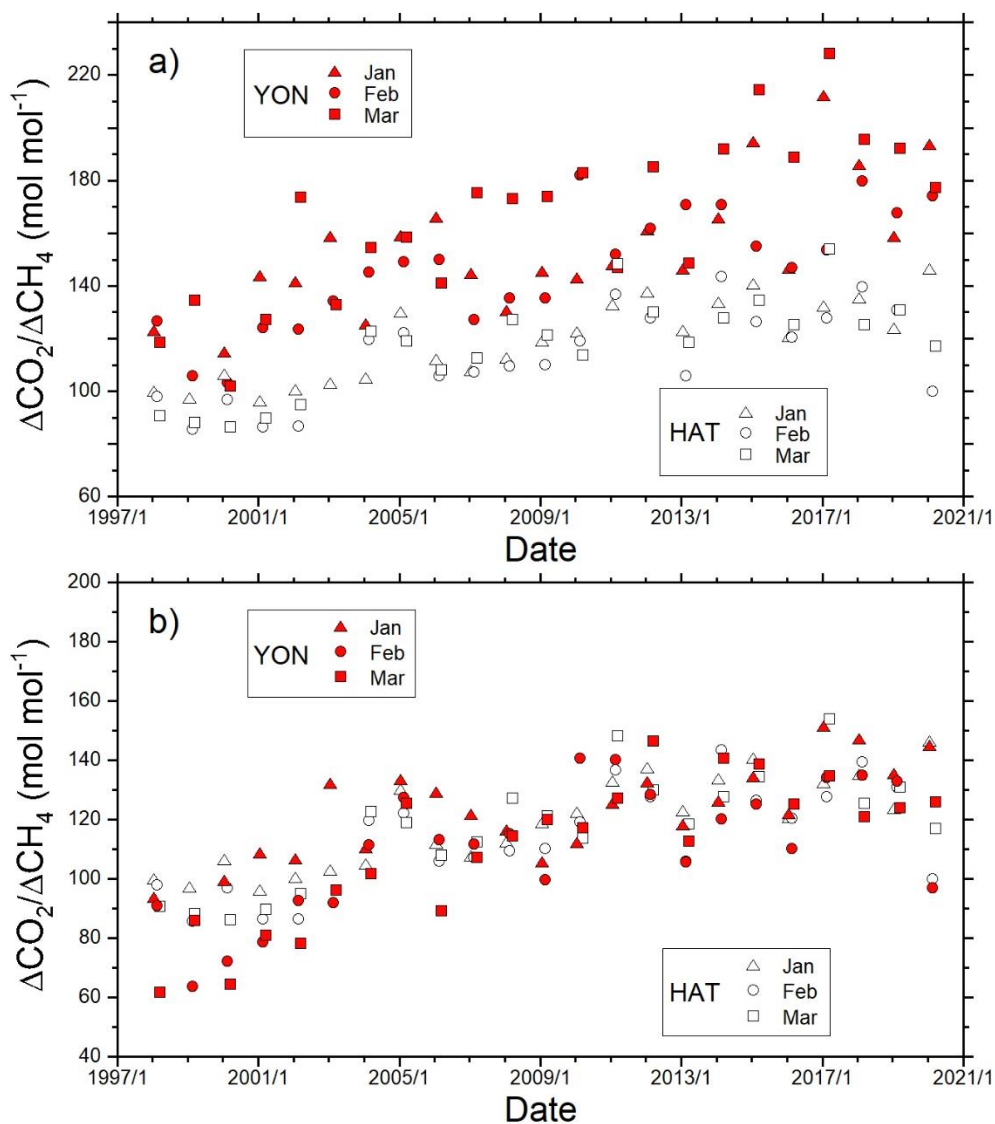
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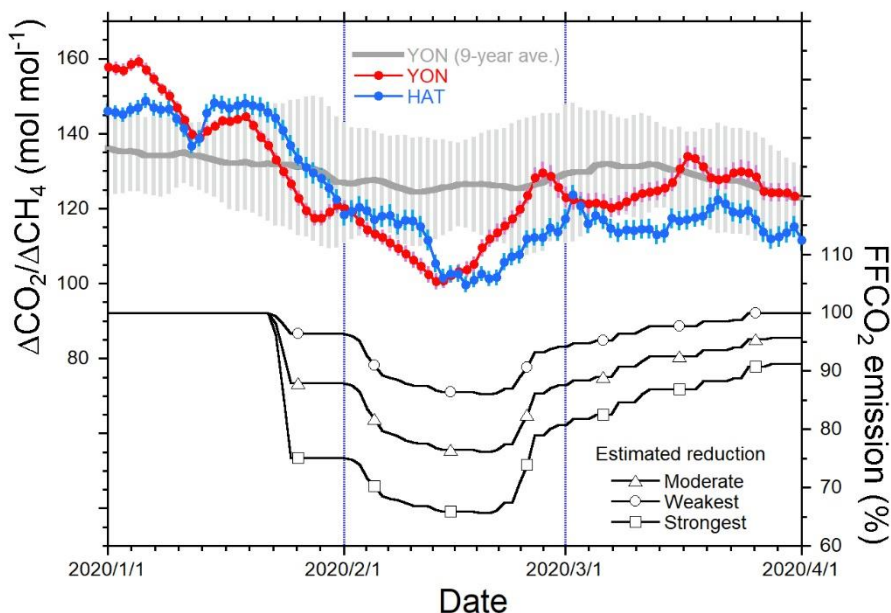
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Table 1 Estimated changes in the monthly $\Delta\text{CO}_2/\Delta\text{CH}_4$ ratios in February and March 2020 from the preceding 9-year averages for YON and HAT^a.

Site	Date	Monthly mean $\Delta\text{CO}_2/\Delta\text{CH}_4^b$	Preceding 9-year average ^c	Decrease from the 9-year average
YON	February 2020	97 ± 2	126 ± 12	28 ± 12
YON	March 2020	126 ± 2	130 ± 11	4 ± 11
HAT	February 2020	100 ± 2 ^d	129 ± 11 ^d	29 ± 11
HAT	March 2020	117 ± 2 ^d	133 ± 11 ^d	16 ± 11

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^aValues are given in mol mol⁻¹.

^bUncertainties are standard errors of the corresponding monthly averages.

^cUncertainties are standard deviations of the corresponding 9-year averages.

^dValues are taken from a previous study (see Table 1 of Tohjima et al., 2020).