



1 **Global warming will largely increase CH₄ emissions from waste treatment: insight from the**
2 **first city scale CH₄ concentration observation network in Hangzhou city, China**

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33 **Abstract:**

34 Atmospheric CH₄ is the second largest anthropogenic contributor to global warming, however its
35 emissions, components, spatiotemporal variations, and projected changes present large uncertainties
36 from city to national scales. CH₄ emissions from waste treatment account for >50% of total
37 anthropogenic CH₄ emissions at the city scale, and considering the high sensitivity of CH₄ emission
38 factors (EFs) to temperature for biological process-based sources, such as waste treatment, large bias
39 will occur when estimating future CH₄ emissions under different global warming scenarios.
40 Furthermore, the relationships between temperature and waste treatment CH₄ emissions have only been
41 determined in a few site-specific studies, and these findings lack representativeness for the whole city
42 scale, which contains various biophysical conditions and shows heterogeneous distribution. These
43 factors increase the difficulty of evaluating city-scale CH₄ emissions (especially from waste treatments),
44 and the projected changes remain unexplored. Here, we conduct the first tower-based CH₄ observation
45 network with three sites in Hangzhou city, which is located in the developed Yangtze River Delta
46 (YRD) area and ranks as one of the largest megacities in China. We found that the *a priori* total annual
47 anthropogenic CH₄ emissions and waste treatment emissions were overestimated by 36.0% and 47.1%
48 in Hangzhou city, respectively. However, the total emissions in the larger region of Zhejiang Province
49 or the YRD area was only slightly underestimated by 7.0%. Emissions from waste treatment showed
50 obvious seasonal patterns according to the air temperature. By using the constructed linear relationship
51 between monthly waste treatment CH₄ emissions and air temperature, we found that the waste
52 treatment EFs increased by 38%~50% as the temperature increased by 10°C. Together with the
53 projected temperature changes from four climate change scenarios, the global warming-induced EFs in
54 Hangzhou city will increase at rates of 2.2%, 1.2%, 0.7% and 0.5% per decade for RCP8.5, RCP6.0,
55 RCP4.5 and RCP2.6 scenarios, respectively, and by 17.6%, 9.6%, 5.6%, and 4.0% at the end of this
56 century, respectively. Additionally, the relative changes derived for the whole of China also showed
57 high heterogeneity and indicated large uncertainty in projecting future national total CH₄ emissions.
58 Hence, we strongly suggest the temperature-dependent EFs and positive feedback between global
59 warming and CH₄ emissions should be considered in future CH₄ emission projections and climate
60 change models.

61 **Keyword:** CH₄ emissions, waste treatment, observation network, global warming



62 **1. Introduction**

63 CH₄ is the second largest anthropogenic greenhouse gas, and reducing CH₄ emissions is
64 considered an effective way to mitigate future climate change on short timescales (Henne et al.,
65 2016; Lin et al., 2021). Accurate estimation of CH₄ emissions from main sources represent the
66 basis of policy making. However, recent studies have found that the total emissions, components,
67 spatiotemporal variations, and projected changes still have large uncertainties at the city scale,
68 especially for megacities in China (USPA 2013; Cai et al., 2018; Lin et al., 2021). Waste treatment
69 (mainly sewage and solid waste from landfills and incineration) is ranked as the world's third
70 largest anthropogenic source of CH₄ emissions after fuel exploitation and livestock, and these
71 emissions account for ~13% of global anthropogenic CH₄ emissions of 371 (±26) Tg a⁻¹ (Lu et al.,
72 2021). It is also ranked as the fourth largest anthropogenic source in China, the biggest
73 anthropogenic CH₄ emitting country, and accounted for ~14% of the total national anthropogenic
74 emissions of 65 (±22) Tg a⁻¹ (Saunois et al., 2020; Lu et al., 2021; Chen et al., 2022). Furthermore,
75 its contribution is larger than 50% at the city scale, where more household waste is located.
76 Therefore, the accurate quantification of waste treatment CH₄ emissions in urban areas has
77 become increasingly important.

78

79 Although some progress has been made in measuring site-scale CH₄ emissions from waste
80 treatment, the estimated emissions still show large discrepancies owing to many factors, such as
81 the amount of waste and its composition, meteorological conditions, such as temperature and
82 water content, proportion between landfills and incineration, degradable organic carbon ratio, CH₄
83 oxidation efficiency, and landfill gas collection (Masuda et al., 2018; Cai et al., 2018; Zhao et al.,
84 2019; Hua et al., 2022; Bian et al., 2022; Maasackers et al., 2022). Furthermore, CH₄ emissions
85 from sewage and landfills are caused by microbial process, especially methanogens, and their
86 emission factors (EFs) are highly sensitive to temperature. These studies were mainly conducted at
87 specific sites with various reported EFs (Du et al., 2017; 2018; Cai et al., 2014; 2018; Zhao et al.,
88 2019; NBSC, 2015; Wang et al., 2015; Florentino et al., 2010; Tolaymat et al., 2010; Hua et al.,
89 2022). The lack of detailed information for all the above factors and their uncertainties has led to
90 considerable bias in estimating CH₄ emissions for most up-to-date inventories (Höglund-Isaksson,



91 2012; USPA et al., 2013; Cai et al., 2018; Lin et al., 2021; Maasackers et al., 2022).
92
93 China is a developing country with the largest anthropogenic CH₄ emissions, its emissions are
94 expected to increase because of the projected rapid economic development, urbanization, and
95 waste generation (Cai et al., 2018). The increase in waste treatment emissions in East China was
96 also found to be the second largest sector driving national total anthropogenic CH₄ emissions since
97 2000 (Lin et al., 2021). Moreover, the mitigation potential of waste treatment in developing
98 countries is thought to be four times than in developed countries (USEPA, 2013). Therefore,
99 mitigating CH₄ emissions from waste treatment in China is a robust and cost-effective method of
100 reducing the total national anthropogenic greenhouse gas emissions.
101
102 Many previous studies have estimated the waste treatment CH₄ emissions for China by both
103 “bottom-up” and “top-down” approaches, with the results varied by 2.5-fold from 4.3 to 10.4 Tg
104 CH₄ yr⁻¹, and accounted for 8.1%~24.2% of national total anthropogenic CH₄ emissions (USEPA
105 2013; Peng et al., 2016; Miller et al., 2019; Lin et al., 2021; Lu et al., 2021; Chen et al., 2022). For
106 these “bottom-up” approach, the high uncertainties were directly attributed to the absence of many
107 small point sources and discrepancies of observed site-specific EFs, which varied largely based on
108 the climate and management technology (Zhao et al., 2019; Hua et al., 2022). As found in
109 previous studies, the most commonly used EDGAR (The Emission Database for Global
110 Atmospheric Research) inventory always uses IPCC-recommended default EF values as 15.0%
111 (Höglund-Isaksson, 2012; Lin et al., 2021; Bian et al., 2022); however, this value is approximately
112 5-7 times of EFs used in China by Zhang and Chen et al. (2014). A recent study comparing waste
113 treatment CH₄ emissions among different inventories also reported that the EDGAR v5.0 and
114 CEDS (Community Emissions Data System) inventories were 21~153% higher than other
115 inventories and EDGAR v5.0 tended to assign more emissions in urban area, especially provincial
116 capitals. In addition, emissions from wastewater have been overestimated by higher emission
117 factor or chemical oxygen demand (Peng et al., 2016; Lin et al., 2021).
118
119 For the “top-down” atmospheric inversion approach, a few studies have constrained anthropogenic



120 sources, including waste treatment, where the most widely used concentrations were satellite
121 observations (Miller et al., 2019; Lu et al., 2021; Chen et al., 2022). Satellite retrieval has the
122 advantage of easy access and global coverage. However, as already noted, the emission constraint
123 results were highly dependent on the availability of observed concentrations, which were largely
124 influenced by weather conditions and cloud coverage. As illustrated in a nearly published study by
125 Chen et al. (2022), although the number of grid cell ($0.25^\circ \times 0.3125^\circ$)-based year-round satellite
126 observations was more than 1000 in north China, the available numbers were less than 10 (and
127 even without any observations) in most part of central, western, eastern and southern China. Such
128 a sparse distribution of available data may not provide robust constraints on waste treatment
129 emissions for some Chinese cities without enough observations, especially considering that waste
130 treatment is co-located with high population density megacities in developed areas, such as eastern
131 and southern China. Furthermore, large temperature-induced monthly variations should occur in
132 waste treatment CH_4 emissions; however, almost all satellite-based inversions were conducted at
133 an annual scale without seasonal variations. Only one recent study was based on satellite
134 observations and focused on urban waste treatment CH_4 emissions, and it found that annual total
135 CH_4 emissions from four cities were 1.4 to 2.6 times larger than inventories in India and Pakistan,
136 where landfills contributed to 6~50% of total emissions and indicated a large bias in our
137 understanding of waste treatment CH_4 emissions (Maasackers et al., 2022).

138
139 The tower-based atmospheric inversion approach, which is based on hourly atmospheric
140 concentration observations within the planetary boundary layer, can be used independently to
141 constrain CH_4 emissions and their main components. Compared with “bottom-up” approach, this
142 method can avoid using the factors that lead to large uncertainties of CH_4 emissions, especially
143 from waste treatment. To the best of our knowledge, tower-based observation inversion studies
144 have not focused on waste treatment emissions at the city scale or much larger regional scales,
145 especially in China. In addition, the influences of global warming on city-scale (or higher
146 regional-scale) emissions are still unclear and have not been considered in future emission
147 projections (USEPA 2013; Cai et al., 2018). In general, previous studies that predicted future
148 waste treatment CH_4 emissions only used activity data changes without considering climate



149 change effect on its EFs. Considering the potential high sensitivity of waste treatment CH₄
150 emissions to projected global warming, how the emissions change with increasing temperature is
151 still unknown, especially within megacities, where more waste is generated and the urban heat
152 island effect will lead to a much stronger warming climate (Zhang et al., 2022).

153

154 In this study, we established three tower-based CH₄ concentration observation sites in Hangzhou
155 city, which is one of the largest megacities in China. To the best of our knowledge, this is the first
156 city-scale, tower-based CH₄ concentration observation network in China. We present our work on
157 urban CH₄ emissions inversion and aim to (1) constrain CH₄ emissions from waste treatment
158 alongside total anthropogenic emissions in Hangzhou city and (2) derive the temperature
159 sensitivity of waste treatment CH₄ emissions at the city scale and quantify the projected emission
160 changes in future climate change scenarios. One-year hourly CH₄ concentration observations from
161 December 1, 2020, to November 30, 2021, were combined with the atmospheric transport model
162 and Bayesian inversion approach to constrain the monthly CH₄ emission inventories. The
163 constructed relationship between monthly temperature and *posteriori* waste treatment CH₄
164 emissions will be used with future temperature projections to quantify how the EFs will change
165 under different global warming scenarios.

166

167 **2. Materials and Method**

168 **2.1 Tower-based CH₄ observation network and supplementary materials**

169 Hangzhou city, which has a population of 12.2 million and an area of 1.7×10^4 km² (core urban
170 area of 8.3×10^3 km²), is the capital of Zhejiang province and located in middle of East China
171 (Figure 1a). As displayed in Figures S1-S2, the East China accounted for the majority of national
172 total population and waste treatment CH₄ emissions, and Hangzhou city ranked among the top 10
173 megacities in China in terms of waste, with an annual solid waste of around 5 million tons in 2021.

174 The tower-based CH₄ concentration observation network includes three observation sites (Figure
175 1a-d): (1) Hangzhou site (120.17° E, 30.23° N, 43.2 m a.s.l.), which is located in the core urban
176 regions; (2) Linan site (119.72° E, 30.30° N, 138.6 m a.s.l.), which is located in the regional
177 background with no obvious emission sources within a 10 km radius; (3) Damingshan site



178 (119.00° E, 30.03° N, 1485.0 m a.s.l.), which is located on the top of a 1500 m mountain and
179 represents a background with much more diluted regional emission signals. The distance is around
180 50 km between Hangzhou and Linan sites and approximately 150 km between the Hangzhou and
181 Damingshan sites. These three sites represent obvious gradients from east, with densely populated
182 areas (Figure 1c-d) and anthropogenic emissions, to the west, with much weaker anthropogenic
183 influence and background conditions.

184
185 The air inlet heights are 25 m above ground at the Hangzhou site, 53 m at the Linan site, and 10 m
186 at Damingshan site. Atmospheric CH₄ concentrations at all three sites were continuously measured
187 using a cavity ring-down spectroscopy analyzer (model G2301 for Hangzhou site and G2401 for
188 Linan site and Damingshan site; Picarro Inc., Sunnyvale, CA). To obtain high-precision
189 observations, two different standard gases were measured and a linear two-point fit was used to
190 calibrate the observations, and the precision of this instrument was within 2 ppb. Additional details
191 of the observation and calibration systems are described in Fang et al., (2014; 2022). Note that
192 because of instrument issues at the Damingshan site, a data gap occurred in September and
193 October, 2021. In general, 99.4%, 99.0%, 79.3% of the hourly CH₄ observations were available
194 for the entire observation period for the Hangzhou site, Linan site and Damingshan site,
195 respectively. Meteorological observations at the Hangzhou meteorological station were used to
196 evaluate simulated meteorological fields, including air temperature at 2 m (T_{2m}), relative humidity
197 (RH), downward solar radiation (S_{\downarrow}), and wind speed (WS) at 10 m height.

198
199 Previous studies of city-scale greenhouse gas concentration observation networks have selected
200 sites at the edge of urban borders as the background in the emission inversion system (i.e.
201 Indianapolis, U.S.A., Miles et al., (2017); Los Angeles, U.S.A., Verhulst et al., (2017); Washington,
202 DC-Baltimore, U.S.A., Lopez-Coto et al., (2020); Paris, France, Lian et al., (2021)); however, we
203 used five CH₄ background sites as the potential background to be selected, including the UUM,
204 TAP, YRO, YON and WLG site (Figure 1a), which were much further than the observations at the
205 Damingshan site. This strategy is based on the following three reasons: (1) our footprint domain is
206 much larger than Hangzhou city, and these five sites are also located close to the edge of model



207 domain; (2) CH₄ concentrations within Hangzhou city will be influenced by seasonal varied
208 monsoon, and the monthly varied wind directions will lead to obvious changes in the CH₄
209 background than only at the Damingshan site; and (3) our model setups can partition CH₄
210 enhancements within Hangzhou city and other regions.

211

212 The projected climate data from four Representative Concentration Pathway (RCP) scenarios
213 (RCP8.5, RCP6.0, RCP4.5 and RCP2.6) using the MRI-CGCM3 model were downloaded from
214 the World Data Center for Climate (WDCC, <https://www.wdc-climate.de/ui/>), where the annual air
215 temperature at 2m was used from years 2021 to 2100. The most recent population density data for
216 Hangzhou city were for the year of 2019, and they were downloaded from the Chinese National
217 Resource and Environmental Science and Data Center ([http://www.resdc.cn/DOI,2017.](http://www.resdc.cn/DOI,2017.DOI:10.12078/2017121101)
218 DOI:10.12078/2017121101).

219

220 **2.2 WRF-STILT model setup**

221 The Weather Research and Forecasting (WRF, version 4.2.2)-Stochastic Time-Inverted Lagrangian
222 Transport (STILT) model will be used to simulate the hourly footprint and CH₄ enhancement (see
223 more details in Hu et al., 2019, 2021). Domain setups are displayed in Figure 1a, with the outer
224 nested domain (Domain-1, 27 km×27 km grid resolution) covering eastern and central China and
225 the inner domain (Domain-2, 9 km×9 km grid resolution) covering the YRD area. The physical
226 schemes used in the WRF model were the same as those used in our previous studies for the YRD
227 domain (Hu et al., 2019; 2021). The simulated CH₄ concentration is the sum of background and
228 enhancement, where the enhancement is calculated by multiplying all CH₄ flux by the hourly
229 footprint that represents the sensitivity of the concentration changes to its regional sources/sinks
230 with a spatial resolution of 0.1°×0.1°. To better quantify the CH₄ components at each site, CH₄
231 enhancements from different regions and sources were also tracked and separately simulated. In
232 addition, it should be noted that the CH₄ background is important in simulating CH₄
233 concentrations and atmospheric inversion. We selected the CH₄ background from the five
234 background sites based on the monthly footprint, as discussed in Section 3.1.

235

236 The most recent EDGAR v6.0 inventory, which has 20 categories, was used for the anthropogenic



237 CH₄ emissions. The main sources in Hangzhou city include solid waste landfills (SWD_LDF),
238 waste water handling (WWT), solid waste incineration (SWD_INC), fuel exploitation from coal,
239 oil, and natural gas (PRO), energy for buildings (RCO) and agricultural soils (AGS). We found
240 that emissions from SWD_LDF, WWT and SWD_INC were simply assigned to the same locations
241 in EDGAR inventory and hence were combined as waste treatment. For CH₄ emissions from
242 wetland, we used the WetCHARTs ensemble mean with a spatial resolution of 0.5° for the monthly
243 average (Bloom et al., 2017). Considering that the WetCHARTs treats rice paddies (the main
244 source of AGS) as one wetland type, the AGS in EDGAR was excluded, and we assumed that
245 WetCHARTs represent all wetland CH₄ emissions as natural wetlands and rice paddies.

246

247 2.3 Bayesian inversion framework

248 The Scale Factor Bayesian inversion (SFBI) approach was applied to interpret atmospheric CH₄
249 concentration (or enhancement) variations in terms of quantitative constraint on all CH₄ sources.
250 The relationship between the observed and simulated CH₄ concentrations (or enhancement) can be
251 expressed as follows in Equation 1:

$$252 \quad y = K\Gamma + \varepsilon \quad (1)$$

253 where y is the observed CH₄ concentration (or enhancement), K corresponds to simulated
254 enhancements from all categories, Γ is the state vector to be optimized and consists of *posteriori*
255 SFs for the corresponding categories in K , and ε is the observing system error.

256

257 The optimal solution to derive *posteriori* SFs is to minimize the cost function $J(\Gamma)$, which
258 represents the mismatch between CH₄ observations and simulations and the mismatch between
259 *posteriori* and *a priori* SFs (Miller et al., 2008; Griffis et al., 2017). The cost function $J(\Gamma)$ can be
260 expressed as follows:

$$261 \quad J(\Gamma) = \frac{1}{2} \left[(y - K\Gamma)^T S_e^{-1} (y - K\Gamma) + (\Gamma - \Gamma_a)^T S_a^{-1} (\Gamma - \Gamma_a) \right] \quad (2)$$

262 where S_e and S_a are the constructed error covariance matrices for the observations and the *a*
263 *priori* values, respectively, and S_e consists of measurement and model errors. Here, each element
264 in the *a priori* SFs Γ_a is treated as 1. Therefore, the solution for obtaining the *posteriori* SFs is to
265 solve $\nabla_{\Gamma} J(\Gamma) = 0$, and it is given as follows:



$$\Gamma_{\text{post}} = (K^T S_e^{-1} K + S_a^{-1})^{-1} (K^T S_e^{-1} y + S_a^{-1} \Gamma_a) \quad (3)$$

266
267 In the Bayesian inversion framework, we first need to estimate the error covariance matrices and
268 state vector for the *a priori* and observational data. And following our previous studies conducted
269 in East China (Hu et al., 2019; 2022). The uncertainty of 10%, 13% and 20% were assigned to the
270 measurement errors (S_{obs}), finite number of particles (500) released in the STILT model ($S_{\text{particles}}$)
271 and uncertainty in meteorological fields (S_{met}), respectively.

272

273 To provide robust constraints on CH₄ emissions, we used three cases of *a priori* uncertainty
274 combinations for different emissions in Bayesian inversion as follows: (1) the first case uses three
275 elements: wetland, waste treatment, and the rest anthropogenic sources, considering the larger
276 seasonality of waste treatment, the uncertainties of 300% were used for waste treatment and 200%
277 for other categories; (2) the second case has more detailed categories, such as wetland, waste
278 treatment, fuel exploitation, energy for building, and the rest anthropogenic sources, where an *a*
279 *priori* uncertainty of 200% was used for each category; and (3) the third case had the same
280 categories as case 1 but used a different *a priori* uncertainty for waste treatment of 200%. The
281 averages of all three cases are used as the final *posteriori* SFs, and the largest difference between
282 each of the three cases is used as the uncertainty.

283

284 **3. Results**

285 **3.1 Atmospheric CH₄ observations**

286 We first displayed the hourly CH₄ concentrations from our three tower-based sites and smoothed
287 the background at five sites using the CCGCRV fitting method, as shown in Figure 2a. The hourly
288 observations at the three towers showed similar temporal variations but with different amplitude.
289 Observations at the Hangzhou site displayed variations between 2000 ppb and 2800 ppb and were
290 much larger than those at the Linan and Damingshan sites. Their monthly averages were also
291 compared in Figure 2b, and results showed the monthly CH₄ varied between lowest 2106.3 ppb in
292 July and highest 2225.0 ppb in September (annual mean of 2159.9 ppb) at the Hangzhou site,
293 lowest 2023.3 ppb in July and highest 2132.0 ppb in September (annual mean of 2086.7 ppb) at
294 the Linan site, the lowest 1955.5 ppb in July and without observations in September at the



295 Damingshan site (annual mean of 2013.4 ppb). The similar variations among the three sites can be
296 explained by the fact that they were dominated by similar atmospheric transport processes;
297 however, their surrounding emission sources were highly different, implying that the emissions
298 from the Hangzhou site should be much larger than those from the Linan and Damingshan sites.

299

300 Because the CH₄ background is important in concentration simulation and emission inversion, we
301 also compared CH₄ background between the five sites, where the annual averages at TAP, YON,
302 RYO, WLG and UUM were 1989.8 ppb, 1850.1 ppb, 1982.7 ppb, 1973.4 ppb, and 1984.2 ppb,
303 respectively. We found that the differences were generally within 20 ppb among the TAP, RYO,
304 WLG and UUM sites (Figure 2), although a large difference was observed between the YON site
305 and the other four sites from May to August, and it reached approximately 100 ppb. Note that the
306 YON site was located to the south of the East China Sea (Figure 1a) and may be influenced by
307 monsoon with clean air flows from the South China Sea, which have much fewer CH₄ sources
308 than air flows from the Asian land area. The CH₄ background at the TAP site appeared slightly
309 higher than at the other four sites because the YON site is located on the coast of South Korea and
310 can be more easily polluted by anthropogenic emissions. Considering the large temporal
311 difference between CH₄ background sites, monthly air flows and source footprint were used to
312 identify backgrounds for our observation network, with details discussed in the Supplementary
313 Material (Section S2, Figure S3 and Table S1).

314

315 **3.2 Concentration footprint and *a priori* emissions**

316 To illustrate the potential source regions of the three sites, the annual averages of the simulated
317 footprints for each site are displayed in Figure 3a-c. The results show that their footprint
318 distributions were quite similar because of the close distance, although we also noticed that
319 obvious differences occurred in the footprint strength (i.e., the area covered by red color), with
320 Hangzhou site > Linan site > Damingshan site. The reason why the footprint at the Damingshan
321 site is the lowest is that observations were conducted at a height of 1500 m, which increased the
322 difficulty of receiving emission signals within boundary layer heights. In addition, the Hangzhou
323 site is located in the core urban area of Hangzhou city and will have a higher nighttime PBLH



324 caused by anthropogenic heat and high buildings than the grassland/farmland-dominated Linan
325 and Damingshan sites; hence, more air particles can be retained within the PBLH, which will
326 generate a stronger footprint.

327

328 The *a priori* EDGAR CH₄ emissions for total anthropogenic categories, waste treatment and their
329 proportions are further illustrated in Figure 3d-f, which reveals significant gradients from higher
330 emissions in the east to lower emissions in the west, which is consistent with our three
331 tower-based observations. The CH₄ emissions for waste treatment displayed similar spatial
332 distributions to urban land use and population density (Figure 1c-d), and waste treatment seemed
333 to emit CH₄ from area sources instead of point sources as waste treatment super plants. We should
334 note that the Chinese government constructed waste separation stations in each city with a density
335 of one station for per 150~200 households (around 450~800 people), and they can emit a large
336 amount of methane from daily biomass waste as an area source. These above analyses also imply
337 that the Hangzhou site can produce higher emissions from both waste treatment and total
338 anthropogenic emissions, which will be discussed and quantified later.

339

340 **3.3 Simulation of CH₄ concentrations and its components for three sites**

341 Comparisons between the observed and simulated daily CH₄ concentration averages are displayed
342 in Figure 4a-c and hourly concentrations are displayed in Figure S4 for the three sites. First, the
343 hourly simulations in Figure S4 showed high consistence when only comparing the temporal
344 patterns with observations, thus indicating the good performance of model transport simulations as
345 confirmed in Figure S5, for evaluating meteorological fields. However, the amplitudes displayed
346 obvious differences among the three sites for daily averages in Figure 4a-c. For the Hangzhou site,
347 the simulated CH₄ concentrations showed obvious overestimation from October to April. The
348 Damingshan site presented overestimated values, while the Linan site presented overestimated
349 values from January to April and underestimated values from May to September. Considering that
350 the source area contributions for the three sites are different, these differences among three sites
351 indicated that the bias in CH₄ emission largely varied from the Hangzhou city scale to a larger
352 regional scale.



353 To further quantify the detailed contributions from different regions and categories to each tower
354 site, CH₄ enhancements from different categories and source areas were simulated separately for
355 the three sites. As displayed in Figure 4d-e, the simulated *a priori* total enhancements at the
356 Hangzhou, Linan, and Damingshan sites were 244.3 ppb, 100.8, and 69.0 ppb, respectively. We
357 also found that contributions by waste treatments dominated the total enhancements but with
358 obvious differences among the three sites, which varied from the highest 64.2% at the Hangzhou
359 site to the lowest of 41.4% at the Damingshan site. We further calculated anthropogenic
360 contributions from Hangzhou city (excluding wetland because of the coarser spatial resolution for
361 Hangzhou city) and other provinces, which were 158.4 ppb at the Hangzhou site, 30.7 ppb at the
362 Linan site, and 10.1 ppb at the Damingshan site, respectively. These accounted for 69.3%, 34.0%,
363 and 16.9% of the total anthropogenic enhancements at the corresponding sites. These results
364 indicated that the CH₄ observations at the Hangzhou site, which is located in the core urban region,
365 were more influenced by local emissions and contained much higher enhancements than the other
366 two sites; moreover, they show that the observations at the Linan and Damingshan sites can
367 present CH₄ emissions of much larger regions, such as Zhejiang province or YRD, than Hangzhou
368 city (Figure 4e).

369

370 The seasonal-averaged diurnal variations for both the observations and simulations are also
371 displayed in Figure 5 for the three sites. Although many previous studies only used daytime
372 observations and simulations to evaluate *a priori* emissions bias and constrain emissions (Sargent
373 et al., 2018; Hu et al., 2022), these studies were based on the assumption that the diurnal scaling
374 factors used on *a priori* emissions are correct (i.e., for anthropogenic CO₂) or the emissions do not
375 have obvious diurnal variations (i.e., emissions from industries or manufacturing). As concluded
376 above, the main CH₄ component in Hangzhou city was waste treatment (Figure 3f), which should
377 be highly sensitive to temperature and have obvious diurnal patterns, with larger emissions during
378 the daytime and smaller emissions at nighttime. Its emissions will be overestimated if only
379 daytime observations and emissions were used to represent daily averages. Furthermore, we found
380 high similarities in the diurnal variations between observations and simulations for the three sites,
381 although some discrepancies were observed. For example, the observations at the Linan site were



382 generally higher than the simulations from spring to autumn for both all-day and midday averages.

383

384 Hence, our preliminary conclusions were that the *a priori* CH₄ emissions were generally
385 overestimated for Hangzhou city but underestimated in larger regions, such as Zhejiang and YRD
386 areas. We also found that simulations were higher than observations for all seasons at the
387 Damingshan site, which can be explained by the high heterogeneity around the Damingshan site,
388 where elevations changed from 0 m to 1600 m within the site located in the grid cell of 9 km (~
389 0.1°), as displayed in Figure 1b. Moreover, the mountain-valley wind and PBLH changes can only
390 be resolved with much higher spatial resolutions as < 1km. Hence, the use of coarse resolutions
391 (i.e. 9 km in this study) in the mountainous regions will bring a large bias in simulating
392 concentration and emission inversion, as was recently found in China for CO₂ as “aggregation
393 error” (Agustí-Panareda et al., 2019; Wang et al., 2022); therefore, observations at the
394 Damingshan site will not be used in emission inversions in this study.

395

396 **3.4 Constraint on anthropogenic CH₄ emissions**

397 As shown in Figures 3f and 5a and Section 3.3, simulations using *a priori* CH₄ emissions showed
398 obvious overestimations, especially from October to April at the Hangzhou site, and
399 overestimations in winter and underestimations from spring to autumn at the Linan site,
400 respectively. This bias can be attributed to *a priori* emissions or meteorological simulations. Our
401 previous studies in the YRD have evaluated the meteorological simulations using the same
402 physical parameterization schemes, which showed high consistence with the observations (Hu et
403 al., 2019; 2021; 2022; Huang et al., 2021). We also evaluated meteorological simulations with
404 observations and confirmed their good model performance (Figure S5). Furthermore, we found no
405 monthly variations in EDGAR v6.0 CH₄ emissions occurred for waste treatment, which
406 contributed 64.2% to the annual CH₄ enhancement average and was much higher in winter (Figure
407 S6). CH₄ emissions from waste treatment are contributed by a microbial process that should be
408 affected by meteorological conditions, especially seasonal temperature changes. Hence, our
409 assumption was that bias in both its seasonality and annual average led to large
410 overestimation/underestimation in the simulated CH₄ concentration. In addition, the bias in other



411 anthropogenic emissions and wetlands can also partly contribute to the bias of the simulated CH₄
412 concentration.

413

414 To quantify the bias sources and constrain the corresponding *a priori* emissions for Hangzhou city,
415 we applied the scaling factor Bayesian inversion approach with three different cases, as introduced
416 in the Methods section. Instead of using only daytime CH₄ observations to constrain *a priori*
417 emissions, we chose to use all-day hourly data at the Hangzhou site to constrain emissions for
418 Hangzhou city, which is based on the following three reasons. (1) The enhancement contributed
419 by Hangzhou city at the Hangzhou site was 69.3%, which was much larger than 34.0% and 16.9%
420 for the Linan and Damingshan sites, respectively. (2) Waste treatment dominated anthropogenic
421 CH₄ emissions in Hangzhou city because of biological processes and should be
422 temperature-dependent. The observed temperature displays obvious diurnal variations at 20 °C,
423 and the use of only daytime observations without considering diurnal CH₄ emissions will
424 introduce significant bias when using derived daytime emissions to represent all-day averages. (3)
425 Previous studies using daytime observations were mainly conducted in regions dominated by
426 industry or energy production, and these contributors have much smaller diurnal variations than
427 waste treatment.

428

429 The derived monthly *posteriori* SFs for each emission source for Hangzhou city are displayed in
430 Table 1. The results showed that the *posteriori* SFs for waste treatment were much smaller in
431 winter and higher in summer, indicating obvious seasonality. The overestimation in winter was
432 mainly contributed by waste treatment. The annual mean *posteriori* SFs for waste treatment varied
433 between 0.50 and 0.56 in all three cases, indicating overestimations in the annual average for the *a*
434 *priori* waste treatment emissions. In addition, the annual mean *posteriori* SFs varied between 0.87
435 and 0.94 for the remaining anthropogenic categories (excluding agricultural soil) and between
436 1.05 and 1.05 for wetlands (including agricultural soil and natural wetland); thus, only slight bias
437 was observed for other anthropogenic categories and wetlands.

438

439 To evaluate whether the *posteriori* SFs significantly improved CH₄ emissions, we used these SFs



440 to derive the *posteriori* emissions and re-simulated hourly concentrations shown in Figure 6 (and
441 daily averages in Figure S7). The results showed that the hourly overestimation produced using *a*
442 *priori* emissions was largely reduced by using *posteriori* emissions when compared with the
443 observations shown in Figure 6a-b, and the regression slope between daily averaged observations
444 and simulations decreased from $1.51(\pm 0.15)$ for the *a priori* simulations to $0.85(\pm 0.07)$ for the
445 *posteriori* simulations in Figure 6c. The mean bias (MB), root mean squared error (RMSE), and
446 correlation coefficient (R) between daily observations and *a priori* simulations were 64.1 ppb,
447 129.2 ppb and 0.44, respectively, and these statistics changed to -22.2 ppb, 72.3 ppb and 0.58 for
448 *posteriori* simulations. These results indicate that the *posteriori* SFs obviously decreased the bias
449 in the *a priori* emissions and were close to the observations.

450

451 Comparisons of the monthly CH₄ emissions between the *a priori* and *posteriori* waste treatment
452 and other anthropogenic sources (excluding agricultural soil) in Hangzhou city are displayed in
453 Figure 7a. For the *a priori* inventory, seasonal variations were not observed for waste treatment
454 with constant monthly emissions of 8.67×10^3 t, and other anthropogenic sources showed
455 seasonality that was much higher in winter (i.e., 5.22×10^3 t in January) than in summer (i.e.,
456 3.06×10^3 t in August). As discussed above, the constant emissions from waste treatment should
457 be incorrect because of its large temperature sensitivity, and the observed monthly temperature
458 difference between summer and winter was larger than 25°C in Hangzhou city. After applying the
459 constraint by using the observed concentration, the *posteriori* emissions for waste treatment
460 showed obvious seasonality, with the highest value in July ($7.66 \pm 0.09 \times 10^3$ t) and lowest in
461 February ($2.20 \pm 0.87 \times 10^3$ t). In addition, the other anthropogenic emissions showed much
462 smaller seasonality (highest in January of $4.18 \pm 0.69 \times 10^3$ t and lowest in August of $2.88 \pm$
463 0.15×10^3 t) than the *a priori* emissions. In general, the annual emission from waste treatment was
464 10.4×10^4 t in the *a priori* EDGAR inventory and decreased by 47.1% to $5.5 (\pm 0.6) \times 10^4$ t for the
465 *posteriori* emissions. The *a priori* emissions from other anthropogenic sources was 4.5×10^4 t and
466 only slightly decreased by 8.9% to $4.1 (\pm 0.3) \times 10^4$ t for the *posteriori* emissions. The proportion
467 of waste treatment to the total anthropogenic emissions decreased from 69.3% to 57.3%. To sum it
468 up, the annual total anthropogenic CH₄ emissions (excluding agricultural soil) decreased from



469 $15.0 \times 10^4 \text{ t}$ to $9.6 (\pm 0.9) \times 10^4 \text{ t}$, indicating an overestimation of 36.0% in Hangzhou city for the *a*
470 *priori* emissions.

471

472 However, as indicated above, the observations and simulations at the Linan site, which represents
473 a much larger region, such as Zhejiang Province or YRD area, illustrated different results in that
474 CH_4 simulations were underestimated from spring to autumn and overestimated in winter (Figure
475 4b and Figure 5e-h). Here, we used the multiplicative scaling factor (MSF) method and
476 observations at the Linan site to derive SFs on a seasonal scale (Sargent et al., 2018; He et al.,
477 2020), and 10 ppb was used as the potential CH_4 background uncertainty in winter, spring, and
478 autumn while 20 ppb was used in summer (see details in the Supplementary Material (Section
479 S2)). The derived *posteriori* SFs were $0.87 (\pm 0.08)$, $1.07 (\pm 0.11)$, $1.19 (\pm 0.24)$, and $1.16 (\pm 0.11)$
480 for winter, spring, summer, and autumn, respectively. Similar seasonal variations as those found
481 for Hangzhou city were observed, and they were $1.07 (\pm 0.14)$ of the *a priori* anthropogenic
482 emissions for the annual average. Our observations at the Hangzhou and Linan sites together
483 indicate that the *a priori* emissions are largely biased at both seasonal and annual scales, and the
484 annual anthropogenic CH_4 emissions were largely overestimated by 36.0% in Hangzhou city but
485 underestimated by 7.0% in the larger region of the Zhejiang Province or YRD area.

486

487 **3.5 Temperature sensitivity of the waste treatment CH_4 EFs and projected changes**

488 Although the derived *posteriori* monthly SFs for waste treatment reflected changes in emissions,
489 considering that the monthly activity data do not present obvious monthly changes, these SFs can
490 mainly reflect relative variations of monthly EFs and contain meteorological changes, especially
491 temperature. To evaluate the temperature sensitivity of the EFs, we first calculated the normalized
492 monthly SFs by dividing the monthly SFs by the annual averages (Table S2) and quantified the
493 relationship between the observed $T_{2\text{m}}$ and normalized SFs. The normalized SFs illustrated a
494 significant linear relationship with monthly $T_{2\text{m}}$ (Figure 7b), where the slopes imply that
495 normalized SFs (and EFs) will increase by 38–50% with a temperature increase of 10°C at the city
496 scale.

497



498 Our findings on the high sensitivity of waste treatment CH₄ emissions to temperature also
499 indicated a dramatic increase based on the projected future global warming trends. We further
500 derived the T_{2m} trends for four different RCP scenarios RCP8.0, RCP6.0, RCP4.5 and RCP2.6
501 (Figure 8a), and the results showed that T_{2m} will increase by 0.50, 0.28, 0.16°C, and 0.10°C per
502 decade for Hangzhou city, respectively. These different warming trends also indicate distinct
503 temperature-dominated influences on future CH₄ EFs and emissions from waste treatment. We
504 then used the slopes in Figure 7b and the annual temperature from 2021 to 2100 to derive relative
505 changes in EF in the future 80 years, where the observation year of 2021 was treated as the
506 baseline year. As displayed in Figure 8b, the EFs in RCP8.5, RCP6.0, RCP4.5, and RCP2.6 will
507 increase by 2.2%, 1.2%, 0.7%, and 0.5% per decade, respectively, and the CH₄ EFs for waste
508 treatment will increase by 17.6%, 9.6%, 5.6%, and 4.0% at the end of this century, respectively.

509

510 The spatial distribution of T_{2m} trends for the whole of China is also displayed in Figure S8, which
511 shows heterogeneous distributions across China for the four global warming scenarios. Because
512 East China has high population density and the majority of the national population (Figure S1),
513 and owns the largest domestic garbage-induced CH₄ emissions (Figure S2), these combined
514 factors indicate considerable CH₄ emission changes from waste treatment in such a
515 temperature-sensitive area. If we assume that the derived temperature sensitivity (which increased
516 by 44% with temperature increases of 10°C on average) is applicable for all of China, especially
517 for East China, the relative changes in waste treatment CH₄ EFs can be calculated by multiplying
518 this value by the air temperature trends. The spatial distributions of global warming-induced EFs
519 changes at the end of this century are displayed in Figure 9. For the RCP2.6 scenario, the EFs for
520 waste treatment will slightly increase by 4.0-6.5% in the north of East China and increase by
521 3.0-4.0% in south of East China. The RCP6.0 scenario also displayed heterogeneous changes in
522 East China, with the north of East China increasing by 10.5-13.0% and south of East China
523 increasing by 9.0-10.5%. Relative changes in RCP4.5 and RCP8.5 are more homogeneous for East
524 China, which indicates that EFs will significantly increase by 5.0-7.5% and 17.5-19.5%,
525 respectively. The largest changes of >20.0% will occur in West China for RCP8.5, although this
526 area has a low population density and CH₄ emissions, indicating negligible effects of global



527 warming (Figure S8). Finally, we should note that these derived relative changes are only caused
528 by global warming and the influence of activity data, management technology, and other factors is
529 beyond the scope of this study.

530

531 **4 Discussions and implications**

532 Many previous studies have compared total CH₄ emissions and their components for different
533 inventories and bottom-up methods, which illustrated large uncertainty and bias at the city scale,
534 and these biases are much larger for waste treatment (Peng et al., 2016; Saunois et al., 2020; Lin et
535 al., 2021; Bian et al., 2022). A recent bottom-up study compared wastewater CH₄ EFs in China,
536 which largely varied by four-fold in different provinces and the uncertainty in the same province
537 was even two-fold larger than its average, implying considerable bias in recent understanding of
538 waste treatment EFs at the regional scale (Hua et al., 2022). The total national emissions varied
539 between 5 and 15 Tg a⁻¹ (Peng et al., 2016; EDGAR v6). Other atmospheric inversion studies
540 have estimated China's CH₄ emissions (Hopkins et al., 2016; Hu et al., 2019; Huang et al., 2021;
541 Miller et al., 2019; Lu et al., 2021; Chen et al., 2022) and found a large bias in nation-wide emissions
542 for almost all inventories, which was mainly caused by fossil fuel exploitation, the agricultural
543 sector (livestock and rice paddies), and waste treatment. For comparisons of waste treatment
544 emissions, these satellite-based inversions also varied between 6 and 9 Tg a⁻¹ by 1.5-fold (Miller
545 et al., 2019; Lu et al., 2021; Chen et al., 2022).

546

547 The above discrepancies between “bottom-up” and “top-down” approaches indicate large
548 uncertainty in understanding China's national CH₄ emissions from waste treatment. Uncertainties
549 increase from the national to regional and city scales, and considerable bias is observed in
550 city-scale emissions for inventories. However, few studies in China and worldwide have
551 performed atmospheric inversions for city-scale waste treatment especially by using tower-based
552 observations, which can present an independent evaluation method. To the best of our knowledge,
553 only one recent research study performed an atmospheric inversion that focused on waste
554 treatment, and it used satellite-based observations to constrain emissions from four cities in India
555 and Pakistan and concluded that total CH₄ emissions were underestimated by 1.4 to 2.6 times for



556 the EDGAR inventory, where emissions from landfills accounted for 6–50% of total emissions
557 (Maasakkers et al., 2022). In our study, we found that the annual waste CH₄ emissions were
558 overestimated by 47.1% for Hangzhou city, which is different from the results in India and
559 Pakistan. These differences indicate that the bias in waste treatment CH₄ emissions vary
560 considerably in different countries and based on climate divisions. Our results highlight a large
561 knowledge gap in terms of emission mechanisms and urban waste treatment CH₄ emissions
562 estimates, especially in China.

563

564 Compared with other fossil-type sources that have much smaller monthly variations, waste
565 treatment is microbial process-based and its EFs are highly sensitive to meteorological conditions,
566 especially temperature. These factors lead to an obvious bias in waste treatment CH₄ emissions in
567 terms of the annual average and its seasonality. Although a few studies have predicted future CH₄
568 emissions from waste treatment, these studies were mainly based on activity data changes and did
569 not consider EF variations caused by future global warming trends, or were only based on
570 site-specific observations (USEPA 2013; Cai et al., 2018; Spokas et al., 2021). To the best of our
571 knowledge, no inventories have considered temperature-induced changes in its seasonal variations
572 and annual trends. Hence, how EFs change under different global warming scenarios at the city
573 scale has not been clarified.

574

575 A few observation-based measurements have been conducted for waste treatment but only at
576 specific sites with large discrepancies in EFs (Du et al., 2017; 2018; Cai et al., 2018; Zhao et al.,
577 2019; NBSC, 2015; Wang et al., 2015; Florentino et al., 2010; Tolaymat et al., 2010; Cai et al.,
578 2014; 2018). Only one previous study used year-round atmospheric CH₄ observations to constrain
579 regional scale CH₄ emissions in Nanjing city in the YRD area (Huang et al., 2021), and it revealed
580 much higher emissions of landfilling waste in summer than in winter, with the emissions in July
581 approximately four times higher in February. However, previous studies have not quantified the
582 temperature sensitivity of waste CH₄ emissions at the city scale or at much larger regional scales.
583 These two studies in different cities confirmed that temperature was the dominant factors driving
584 seasonal variations in waste treatment CH₄ emissions. Hence, our study is the first to estimate



585 city-scale waste treatment CH₄ emissions, their temperature sensitivity, and projected changes in
586 different global warming scenarios. Our findings regarding the large sensitivity of temperature
587 indicate that monthly scaling factors should be considered to better simulate atmospheric CH₄
588 concentrations.

589

590 We also note that the predictions of future climate change were mainly based on different emission
591 intensities of greenhouse gases and CH₄ contributed approximately 20% of direct anthropogenic
592 radiative forcing (Seto et al., 2014). The CH₄ emissions in different global warming scenarios
593 were mainly calculated by predicting the energy use data without considering the changes in the
594 EFs. In this study, we found that there should be a large positive feedback between global
595 warming and CH₄ emissions, especially in the RCP8.0 scenario, where global warming induced
596 emissions will increase by 17.6%. Hence, projected emissions from waste treatments and other
597 biological process-based sources together with positive feedback between temperature and their
598 emissions should be included in future climate change models. In addition, CH₄ concentration
599 simulations are essential for modeling many air pollutants (i.e., O₃, NO_x, and CO), especially in
600 the stratosphere (Isaksen et al., 2011; Kaiho et al., 2013). Considering that waste treatment CH₄
601 emissions accounted for ~25% of total anthropogenic emissions (EDGAR v6.0) in East China,
602 where severe air pollution frequently occurs, we also believe that the coupling of
603 temperature-dependent CH₄ emissions and the monthly scaling factors for CH₄ emissions can
604 improve air pollution modeling in East China.

605

606 **5 Conclusions**

607 CH₄ emissions from waste treatment ranks as one of the main global anthropogenic sources,
608 although estimations of its emissions at the city scale still have large biases and uncertainties,
609 especially in the megacities of developing countries. To better evaluate bias for city-scale
610 anthropogenic CH₄ emissions and understand the sensitivity of temperature to waste treatment
611 CH₄ emissions, we conducted three tower-based atmospheric CH₄ observation networks in
612 Hangzhou city, which is located in the developed YRD region and one of the top 10 megacities in
613 China. One-year hourly atmospheric CH₄ observations were presented from December 2020 to



614 November 2021. We then applied a scaling factor Bayesian inversion method to constrain the
615 monthly anthropogenic CH₄ emissions and components (especially for waste treatments) in
616 Hangzhou city, and used the multiplicative scaling factor method for the broader Zhejiang
617 Province and YRD area on a seasonal scale.

618

619 To the best of our knowledge, this study produced the first tower-based CH₄ observation network
620 in China. We found an obvious seasonal bias of simulated CH₄ concentrations in the core urban
621 area of Hangzhou city, and it was mainly caused by bias of waste treatment at both annual and
622 monthly scales. The derived *posteriori* CH₄ emissions displayed significant seasonal variations,
623 with a peak in summer and trough in winter that were mainly caused by waste treatment. The *a*
624 *priori* annual waste treatment CH₄ emission in Hangzhou city was 10.4×10^4 t, while the
625 *posteriori* emissions decreased by 47.1% to $5.5 (\pm 0.6) \times 10^4$ t. In addition, the total anthropogenic
626 CH₄ emissions (excluding agricultural soil) decreased from 15.0×10^4 t to $9.6 (\pm 0.9) \times 10^4$ t, thus
627 indicating an overestimation of 36.0% for the whole year of 2021. Observations at the Linan site
628 imply that the annual CH₄ emissions were slightly underestimated by 7.0% in a larger region of
629 Zhejiang Province or the YRD area, which was different from the observations at Hangzhou city.
630 Additionally, the *posteriori* monthly CH₄ emissions from waste treatment illustrated a significant
631 linear relationship with air temperature, with the regression slopes indicating an increase of 38–50%
632 when the temperature increased by 10°C. Finally, we found that the waste treatment CH₄ EFs for
633 Hangzhou city increased by 17.6%, 9.6%, 5.6%, and 4.0% at the end of this century for RCP8.0,
634 RCP6.0, RCP4.5, and RCP2.6, respectively. The derived relative changes for all of China also
635 showed high heterogeneity and indicated large uncertainty in projecting future national total CH₄
636 emissions. This study is the first to focus on the city-scale temperature sensitivity of waste
637 treatment CH₄ emissions from the perspective of the atmospheric inversion approach. Based on
638 the above results, we strongly suggest that temperature-dependent EFs should be coupled within
639 both recent CH₄ inventories and future CH₄ emission projections.

640

641 **Data availability:** The atmospheric CH₄ observations data can be requested from Cheng Hu and
642 Bing Qi. STILT model is downloaded from <http://www.stilt-model.org/>, the EDGAR inventory is
643 from <https://edgar.jrc.ec.europa.eu/>, and the projected climate data were downloaded from World



644 Data Center for Climate (WDCC, <https://www.wdc-climate.de/ui/>).

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650 **Author contribution:** Cheng Hu and Bing Qi designed the study. Cheng Hu performed the model
651 simulation, data analysis and wrote the paper; Bing Qi and Rongguang Du conducted CH₄
652 concentration observation and meteorological data collection, and all co-authors contributed to the
653 data/figures preparation and analysis.

654 **Declaration of competing interests:** The authors declare that they have no conflict of interest.

655

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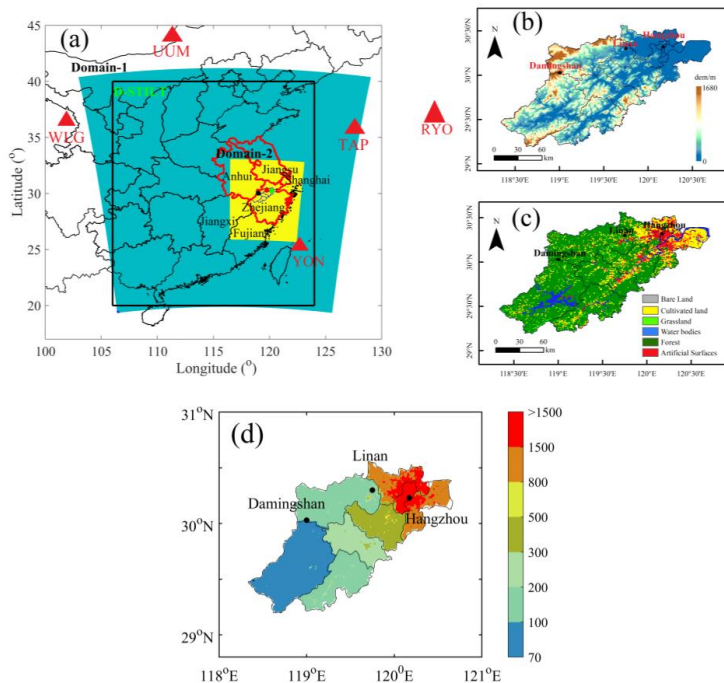
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844 Figure 1. (a) WRF-STILT model domain setup, three CH₄ concentration observation sites in
845 Hangzhou city, and five CH₄ background sites. Note that the green, red and black dots represent
846 the locations for the Hangzhou, Linan and Damingshan sites, respectively. The Yangtze River
847 Delta region is contained in the red boundary, and the back rectangle represents domain in STILT
848 model. (b) Geophysical height within Hangzhou city, (c) land surface categories in Hangzhou city,
849 and (d) population density in Hangzhou city for 2019, units: person per km².

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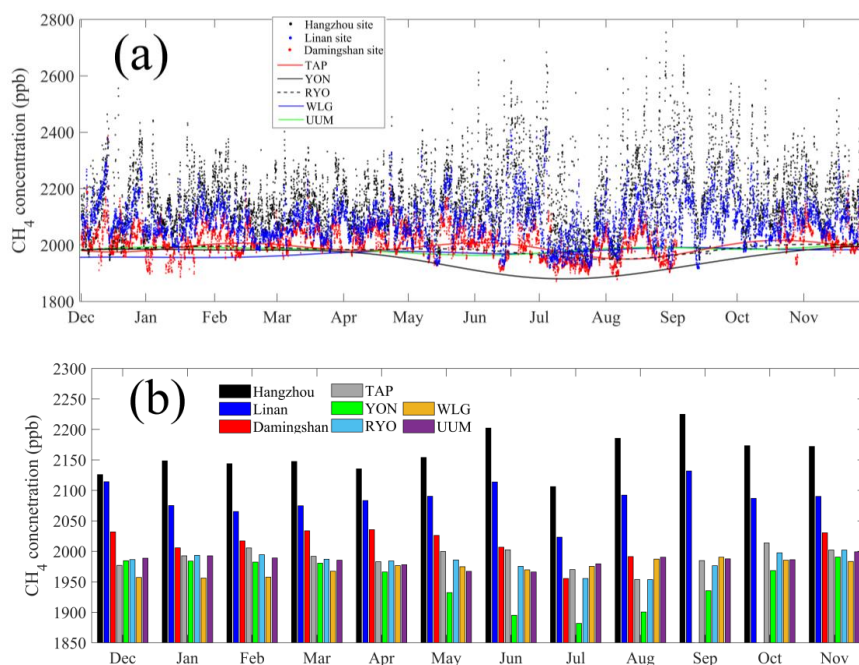
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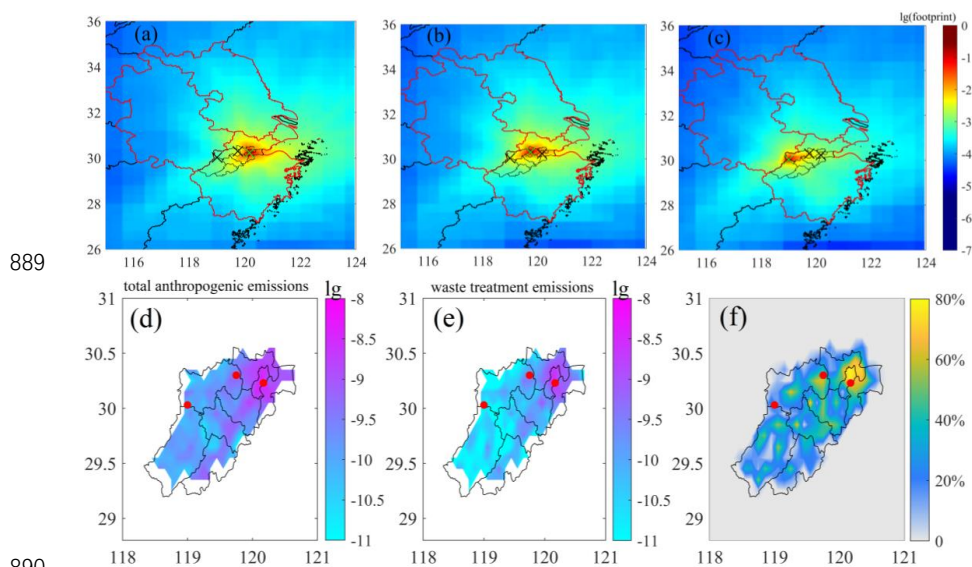
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Figure 2. (a) Hourly CH₄ concentrations at three sites within Hangzhou city, namely, Hangzhou site, Linan site, and Damingshan site, and CH₄ background fit based on the CCGCRV regression method at five background sites, namely, TAP, YON, RYO, WLG, and UUM; and (b) monthly mean CH₄ concentrations for the above eight sites. Note that the CH₄ background is smoothed by using the CCGCRV fitting method based on weekly or hourly observations, which can filter large fluctuations caused by sudden and unidentified sources

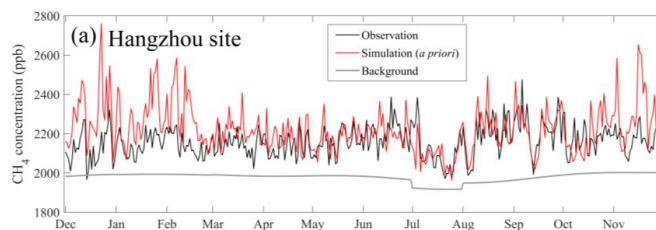


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891 Figure 3. Annual averages of the simulated footprint for the (a) Hangzhou site, (b) Linan site, and
892 (c) Damingshan site, where the green symbol “x” indicates the receptor location in each panel, (d)
893 total anthropogenic CH_4 emissions according to the EDGAR v6.0 inventory, (e) waste treatment
894 CH_4 emissions, and (f) proportion of waste treatment emissions to total anthropogenic CH_4
895 emissions. The red dot represents three sites, the unit for the footprint is $\text{ppm m}^2 \text{ s mol}^{-1}$, and the
896 unit for emissions is $\text{kg m}^{-2} \text{ s}^{-1}$.

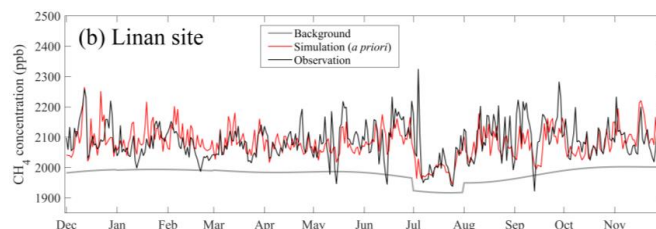
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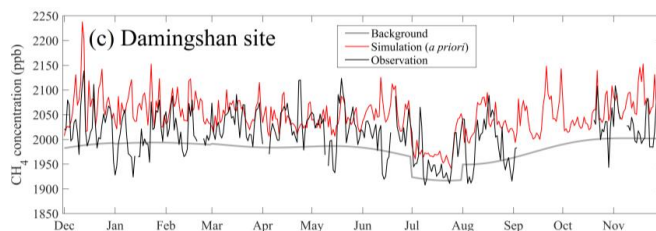
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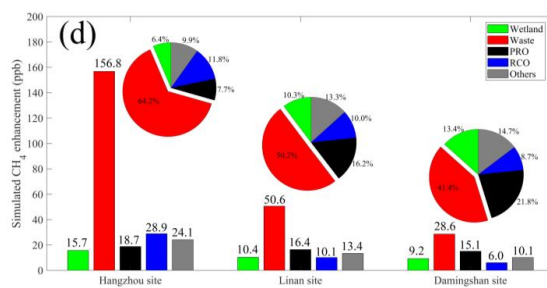
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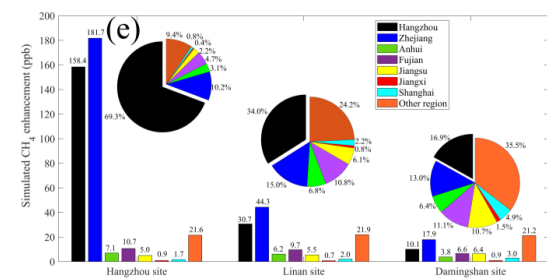


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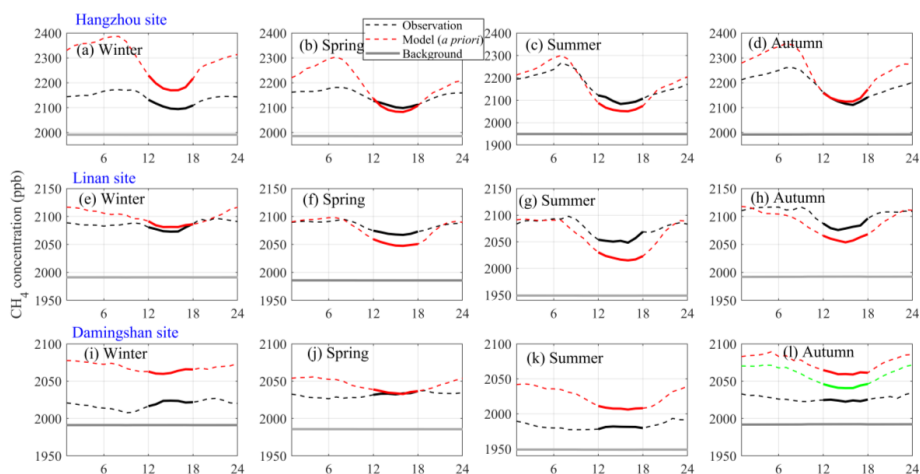
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Figure 4. Comparisons between daily CH₄ observations and simulations for the (a) Hangzhou site, (b) Linan site, (c) Damingshan site, (d) simulated CH₄ enhancements from main emission categories, and (e) simulated anthropogenic CH₄ enhancement from different regions and proportions. Note that contributions from Zhejiang Province in the pie figure (e) represent the sum of both “Hangzhou” and “Zhejiang”.



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925 Figure 5. Seasonal averaged diurnal variations for the Hangzhou site in (a) winter, (b) spring, (c)
926 summer, and (d) autumn; Linan site in (e) winter, (f) spring, (g) summer, and (h) autumn; and
927 Damingshan site in (i) winter, (j) spring, (k) summer, and (l) autumn; Note, because of the
928 two-months data gap in autumn for the Damingshan site, the green line is for all
929 September-November simulations, the red line only represents simulations for the corresponding
930 period for available observation data, and the bold lines represent data between 12:00 and 18:00.

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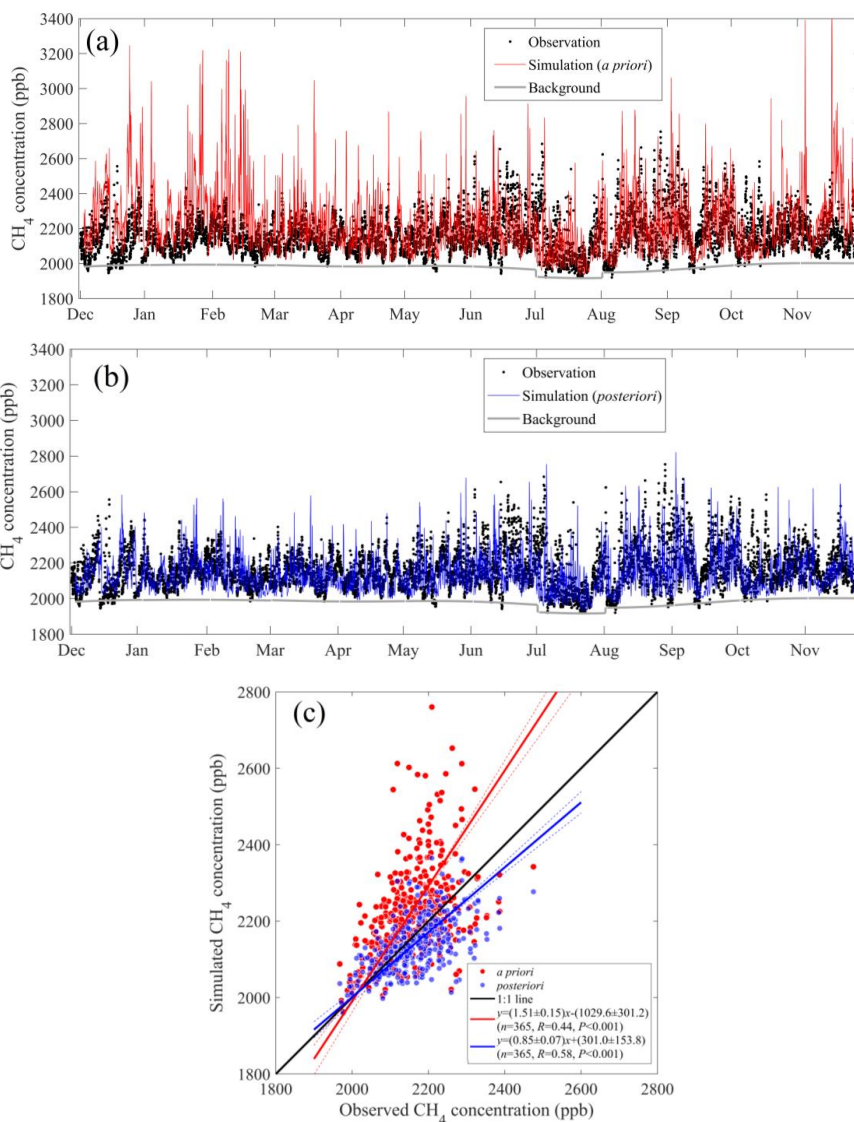
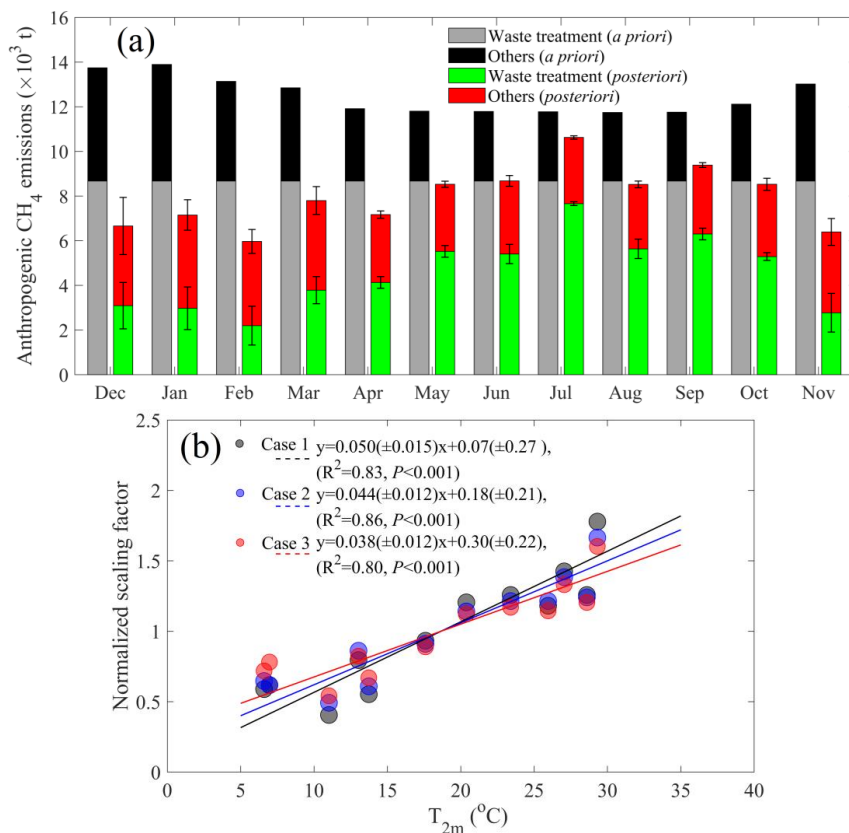


Figure 6. Comparisons of the hourly CH₄ concentrations at the Hangzhou site between observations and simulations by using (a) *a priori* and (b) *posteriori* emissions and (c) scatter plots of daily CH₄ averages by using *a priori* and *posteriori* emissions.



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967 Figure 7. (a) Monthly anthropogenic (excluding agricultural soil) CH₄ emissions for *a priori* and
968 *posteriori* emissions for Hangzhou city and (b) relationship between the monthly *posteriori* CH₄
969 emissions and temperature in three cases.

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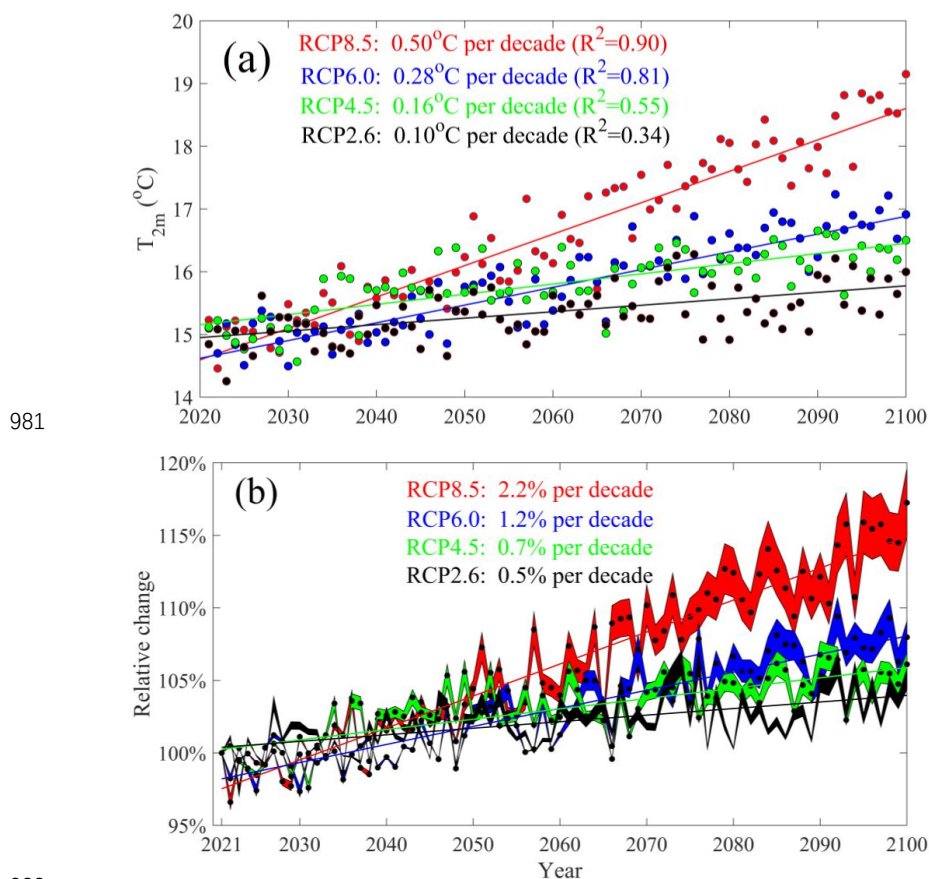
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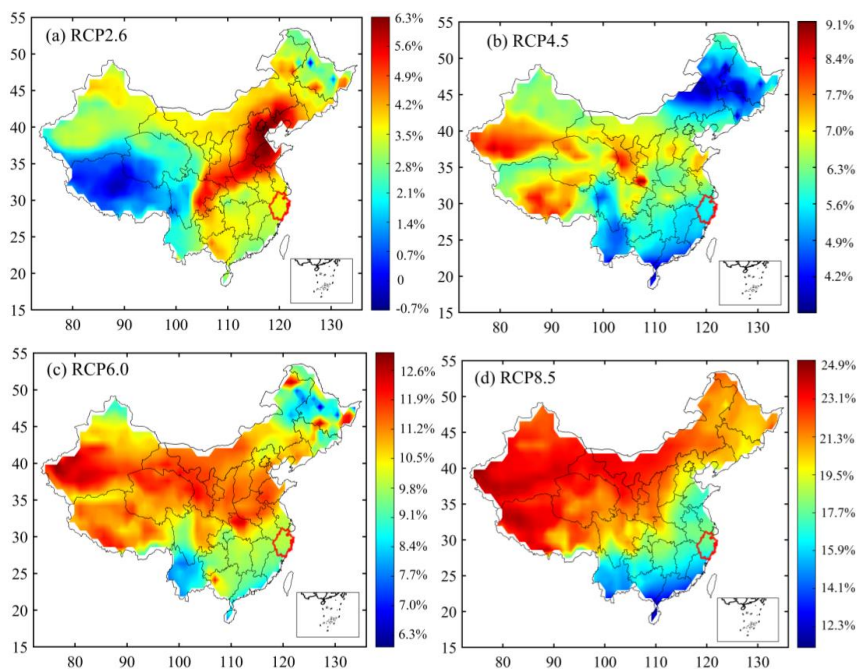


983 Figure 8. (a) Annual air temperature from 2021 to 2100 for the four different global warming
984 scenarios and, (b) projected relative change of waste treatment CH₄ emissions (or EFs). Note that
985 the shading indicates the extent of three cases.

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1002 Figure 9. Global warming-induced relative changes of the waste treatment CH₄ EFs by year for
1003 2100 for the (a) RCP2.6, (b) RCP4.5, (c) RCP6.0, and (d) RCP8.5 scenarios. Note that the red
1004 boundary is Zhejiang Province.

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1026 Table 1. *Posteriori* SFs for different categories in three cases, where wetland represents natural
1027 and agricultural wetland, Waste represents waste treatment, PRO represents fuel exploitation,
1028 RCO represents energy for building, and Others represents the remaining anthropogenic
1029 emissions.

Month	Case 1			Case 2					Case 3		
	Wetland	Waste	Others	Wetland	Waste	PRO	RCO	Others	Wetland	Waste	Others
1	1.00	0.29	0.83	1.00	0.34	0.90	0.80	0.93	1.00	0.40	0.72
2	1.00	0.20	0.89	1.00	0.26	0.97	0.83	0.93	1.00	0.30	0.77
3	1.03	0.39	1.04	1.02	0.46	1.07	0.80	0.97	1.02	0.46	0.95
4	1.10	0.46	0.96	1.08	0.48	1.01	0.95	0.93	1.08	0.49	0.91
5	1.12	0.62	0.99	1.10	0.64	1.06	0.97	0.92	1.11	0.65	0.95
6	1.22	0.59	1.09	1.18	0.64	1.05	0.97	1.03	1.18	0.64	1.05
7	1.10	0.88	0.96	1.09	0.88	1.00	1.00	0.94	1.09	0.89	0.94
8	1.05	0.62	0.95	1.01	0.66	0.99	0.97	0.95	1.01	0.67	0.91
9	1.04	0.71	1.01	1.02	0.73	0.96	0.98	1.04	1.02	0.74	0.98
10	1.06	0.60	0.94	1.06	0.61	0.92	0.96	1.00	1.06	0.62	0.90
11	1.01	0.27	0.86	1.00	0.32	0.91	0.85	0.93	1.00	0.37	0.75
12	1.00	0.31	0.70	1.00	0.33	0.75	0.79	0.91	1.00	0.43	0.58

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