1   **Observing a Volatile Organic Compound from a Geostationary Infrared Sounder:**
2   **HCOOH from FengYun-4B/GIIRS**

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13 **Key Points:**

14 - First retrieval of atmospheric HCOOH from a geostationary sounder is demonstrated

15 - HCOOH in fire plumes can be continuously tracked with FY-4B/GIIRS

16 - FY-4B/GIIRS HCOOH observations have comparable sensitivity to IASI

17 **Abstract**

18 Formic acid (HCOOH) is one of the most abundant volatile organic compounds (VOCs) in the

19 Earth's atmosphere and an important source of atmospheric acidity. Here we present the first

20 retrieval of HCOOH from a geostationary orbit using observations from the Geostationary

21 Interferometric Infrared Sounder (GIIRS) on board FengYun-4B. The results from July 2022 to

22 June 2023 show the monthly variation of the HCOOH distribution in Asia due to emissions from

23 biomass burning and biogenic sources. FY-4B/GIIRS effectively tracks HCOOH enhancements

24 from wildfires and biogenic sources both during the day and at night. Finally, inter-comparison

25 with IASI HCOOH data shows good agreement, indicating that FY-4B/GIIRS observations have

26 comparable sensitivity to IASI. This study is an important first step towards monitoring VOCs

27 from geostationary infrared sounders, including the existing and future GIIRS on board the FY-4

28 series and the forthcoming European geostationary infrared sounder (IRS) on board Meteosat

29 Third Generation (MTG).

30

31 **Plain Language Summary**

32 Formic acid (HCOOH) is one of the most abundant volatile organic compounds (VOCs) in the

33 Earth's atmosphere and an important source of atmospheric acidity. Satellite observations play an

34 indispensable role in improving our understanding of global HCOOH sources and sinks. However,

35 existing polar-orbiting satellites that are sensitive to tropospheric HCOOH only provide up to two

36 overpasses per day over the same spot, one during the day and one during the night. The diurnal

37 variations of tropospheric HCOOH are therefore under-constrained to track its evolution in the

38 lower troposphere and transport in the free troposphere. The Geostationary Interferometric

39 Infrared Sounder (GIIRS) onboard China's FengYun-4 satellite series is the world's first

40 geostationary hyperspectral infrared sounder. Using observations from FY-4B/GIIRS, this study

41 presents the first retrieval of HCOOH from a geostationary satellite instrument that captures the

42 HCOOH variation in Asia driven by biomass burning emissions and biogenic sources. In addition,

43 FY-4B/GIIRS effectively tracks HCOOH plumes in Southeast Asia and Siberia. Finally, inter-

44 comparison with IASI HCOOH data shows good agreement, indicating that FY-4B/GIIRS

45 observations have comparable sensitivity to IASI. This study is an important first step towards

46 monitoring VOCs from geostationary infrared sounders.

1 Introduction

Formic acid (HCOOH) is one of the most abundant volatile organic compounds (VOCs) in the Earth's atmosphere and an important source of atmospheric acidity. It contributes significantly to precipitation acidity by facilitating the nucleation of cloud droplets (e.g., **Andreae et al., 1988; Keene and Galloway, 1984; Keene and Galloway, 1988**). The production of HCOOH in the atmosphere is dominated by photochemical oxidation of biogenic and anthropogenic VOCs (**Millet et al., 2015**) and direct emissions from vegetation (**Keene and Galloway, 1984**), biomass and biofuel burning (**Goode et al., 2000**), and fossil fuel combustions (**Kawamura et al., 1985**). Yet, advanced global models underestimate the observed atmospheric levels of HCOOH, pointing to major gaps in our understanding of its emissions and formation mechanisms.

Over the past decade, studies have showcased the essential role of satellite observations in enhancing our understanding of global sources and sinks of HCOOH. For instance, **Stavrakou et al. (2012)** showed that a large source of HCOOH from boreal and tropical forests is misrepresented in models based on satellite observations from the Infrared Atmospheric Sounding Interferometer (IASI; **Razavi et al., 2011**). Using measurements from the same sounder, **Franco et al. (2021)** proposed an HCOOH production mechanism in which formaldehyde is efficiently converted to gaseous HCOOH via a multiphase pathway. In addition, it has been shown that the HCOOH emissions from wildfires have been largely underestimated using satellite observations (e.g., **Cady-Pereira et al. 2014; Chaliyakunnel et al., 2016; Pommier et al., 2017**).

Satellite observations of HCOOH provide global coverage from polar-orbiting nadir-looking infrared hyperspectral sounders including IASI (e.g., **Razavi et al., 2011; Franco et al., 2018**) and the Tropospheric Emission Spectrometer (TES) (e.g., **Cady-Pereira et al., 2014**). In addition, solar occultations from the limb sounders Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and Atmospheric Chemistry Experiment – Fourier Transform Spectrometer (ACE-FTS) provide measurements of the HCOOH abundance in the upper troposphere – lower stratosphere (e.g., **González Abad et al., 2009; Grutter et al., 2010**), However, existing polar-orbiting satellites that are sensitive to tropospheric HCOOH can only provide up to two overpasses per day over the same spot, with one in the daytime and the other in the nighttime. The diurnal change of tropospheric HCOOH is therefore under-constrained. Although it is relatively short-lived with a global tropospheric lifetime of 3-4 days (**Paulot et al., 2011; Stavrakou et al., 2012**), HCOOH produced or released into the free troposphere can be

78 transported over relatively long distances due to its slow photochemical loss (**Paulot et al., 2011**).

79 Hence, continuous high-temporal-resolution observations of HCOOH are required to track its

80 evolution and to characterize its intra-day variability in the troposphere.

81 The Geostationary Interferometric Infrared Sounder (GIIRS) onboard China's FengYun-4

82 satellite series is the world's first geostationary hyperspectral infrared sounder primarily designed

83 to observe the three-dimensional distributions of atmospheric water vapor and temperature. GIIRS

84 on board FY-4B (FY-4B/GIIRS) was launched in 2021 with an improved signal-to-noise ratio

85 (SNR) compared to its predecessor FY-4A/GIIRS, launched in 2016 (**Yang et al., 2017**). The high

86 spectral resolution (~0.625 cm^{-1}) and SNR of FY-4B/GIIRS have enabled 2-hourly measurements

87 of carbon monoxide and ammonia over eastern Asia (**Zeng et al., 2023a; Zeng et al., 2023b**).

88 Applying the optimal estimation method (OEM) on the FY-4B/GIIRS spectra, this study aims to

89 assess for the first time the information content within spectra recorded by a geostationary sounder

90 for retrieving diurnal HCOOH over eastern Asia. Additionally, it investigates how geostationary

91 observations can aid in tracking the long-range transport of HCOOH in the free troposphere. The

92 rest of this paper is organized as follows. An introduction to FY-4B/GIIRS and the observed

93 spectra is given in **Section 2**, and the retrieval algorithm based on optimal estimation is presented

94 in **Section 3**. The results are shown in **Section 4**, followed by discussion and conclusions in

95 **Section 5**.

96 2 FY-4B/GIIRS

97 FY-4B/GIIRS is an infrared Fourier transform spectrometer located at an altitude of

98 ~36,000 km above the Equator at 133° E overlooking eastern Asia. FY-4B/GIIRS scans the full

99 region with a 2-hour measurement cycle. In total, 12 cycles are made per day starting at 0, 2, 4, …,

100 22h UTC before September 06, 2022, and 1, 3, 5, …, 23h UTC after. In the 2-hour measurement

101 cycle, scanning the full region takes about 1.5 hour, followed by external spectral calibration for

102 about 0.5 hour. At nadir, the spatial footprint on the Earth's surface has a diameter of about 12 km.

103 FY-4B/GIIRS covers a long-wave IR band (680-1130 cm^{-1}) and a mid-wave IR band (1650-2250

104 cm^{-1}) with a spectral resolution of 0.625 cm^{-1}. Blackbody calibration experiments (**Li et al., 2022**)

105 carried out before launch showed that the typical noise equivalent differential radiance (NedR)

106 around the HCOOH absorption channel at 1105 cm^{-1} (**Figure 1(a)**) is less than 0.3 mW/(m^2·sr·cm^{-1}

107), which is about 0.2K for a blackbody at 300K. **Figure 1(b)** shows examples of brightness

108   temperature changes in the micro-window for varying HCOOH abundances (1, 2, 4 and 8 times

109   the a priori used in the retrieval, see Section 3). In the latter three cases, the HCOOH spectral

110   signature exceeds the spectral noise level in FY-4B/GIIRS. A more detailed description of FY-

111   4B/GIIRS can be found in **Zeng et al. (2023b)**. Similar to **Zeng et al. (2023a; 2023b)**, only clear-

112   sky or near-clear-sky pixels are used for the retrieval. We adopted the level-2 cloud mask (CLM)

113   data product from the Advanced Geostationary Radiation Imager (AGRI) onboard FY-4B to filter

114   out cloudy pixels. After cloud filtering and excluding data with a satellite zenith angle larger than

115   70°, the number of observations available for an entire day is typically of approximately 100,000.

116   **3 Retrieval algorithm based on optimal estimation**

117   We apply the FengYun-Geostationary Atmospheric Infrared Retrieval (FY-GeoAIR)

118   algorithm, originally developed for carbon monoxide (**Zeng et al., 2023a**), for retrieving HCOOH

119   using FY-4B/GIIRS spectra. Here, a brief introduction to FY-GeoAIR is given with descriptions

120   of changes necessary for the HCOOH retrieval. More details about the retrieval algorithm can be

121   found in **Zeng et al. (2023a)** and **Zeng et al. (2023b)**. Essentially, FY-GeoAIR consists of a

122   forward radiative transfer model (RTM) for simulating upwelling radiation in the thermal infrared

123   and an OEM-based inverse model for retrieving atmospheric parameters. In the forward RTM, the

124   upwelling radiance has four major components: the upwelling surface thermal emission, the

125   upwelling atmospheric thermal emissions, the surface-reflected downwelling atmospheric thermal

126   emission, and the solar radiation from surface reflection (**Clough et al., 2006**; **Hurtmans et al.,**

127   **2012**). Under clear sky conditions, scattering by clouds and aerosols can be ignored. In the forward

128   model, surface parameters (surface skin temperature and surface pressure) and atmospheric

129   parameters (temperature, H_2O, and ozone profiles) are extracted from the European Centre for

130   Medium-Range Weather Forecasts (ECMWF) Reanalysis v5 (ERA5; **Hersbach et al., 2020**). The

131   a priori HCOOH profiles over land and ocean are adopted from an updated version of the IASI a

132   priori (**Franco et al. 2018**), as shown in **Figure S1(a)**. The absorption coefficients for gas

133   absorption are pre-calculated as look-up tables based on the Line-By-Line Radiative Transfer

134   Model (LBLRTM v12.11; **Clough et al., 2005**). FY-GeoAIR retrieves the atmospheric layers

135   below 200 hPa. In the inverse model, the retrieval algorithm searches for a solution x that

136   minimizes the following cost function (**Rodgers, 2000**):

137
$$J(x) = [y - F(x, b)]^T S_\varepsilon^{-1}[y - F(x, b)] + (x - x_a)^T S_a^{-1}(x - x_a) \qquad (1)$$

138 where $\boldsymbol{x}$ is the state vector to be retrieved, which includes a set of parameters, including HCOOH

139 profile, H_2O profile, column scale factors for other interfering gases (ozone, carbon dioxide,

140 methanol, ammonia, and CFC-12), surface skin temperature, and scale factor for the atmospheric

141 temperature profile. $\boldsymbol{y}$ is the spectral radiance observation of the micro-window (1090-1120 cm^{-1})

142 for retrieval; $\mathbf{F}$ represents the forward RTM; $\boldsymbol{b}$ is a set of model parameters not to be retrieved,

143 such as observation geometries. $\mathbf{S}_\varepsilon$ is the measurement error covariance matrix; $\boldsymbol{x}_a$ is the a priori

144 state vector; $\mathbf{S}_a$ is the a priori covariance matrix for the state vector. The instrument noise (NedR)

145 for each spectral channel is used as the measurement noise for calculating $\mathbf{S}_\varepsilon$. Different from **Zeng**

146 **et al. (2023b)**, the spectral noise is not enlarged to force the reduced χ^2 from the cost function to

147 be close to 1. The Levenberg-Marquardt modification of the Gauss-Newton method is used to find

148 the solution for the cost function (**Rodgers, 2000)**. The one-sigma variability in the a priori

149 HCOOH profile from model simulations is about 70%, which is enlarged by a factor of 5 for

150 calculating $\mathbf{S}_a$ to relax the a priori constraint, as shown in **Figure S1(b)**.

151 As described by **Rodgers (2000)**, the Degree of Freedom for Signal (DOFS) and Averaging

152 Kernel (AK) matrix, two statistics generated from OEM, can be used to interpret the retrieval

153 results. AK is a metric that quantifies the sensitivity of the retrieval to the true state. Ideally, a

154 sufficiently good sensitivity of an observing system generates an AK close to an identity matrix.

155 In reality, due to the limited detectivity of the observing system, the AK can be very different. In

156 this case, the rows of AK represent the smoothing functions that vertically smooth the information

157 from the true state over different layers. DOFS is defined as the trace of the AK matrix, which

158 represents the number of independent information from the spectra for constraining the vertical

159 distribution of HCOOH. For example, a DOFS of 1.0 means that, given the assumed a priori

160 uncertainty, one independent piece of information from the spectral can be used to constrain

161 HCOOH vertically.

162

(a)
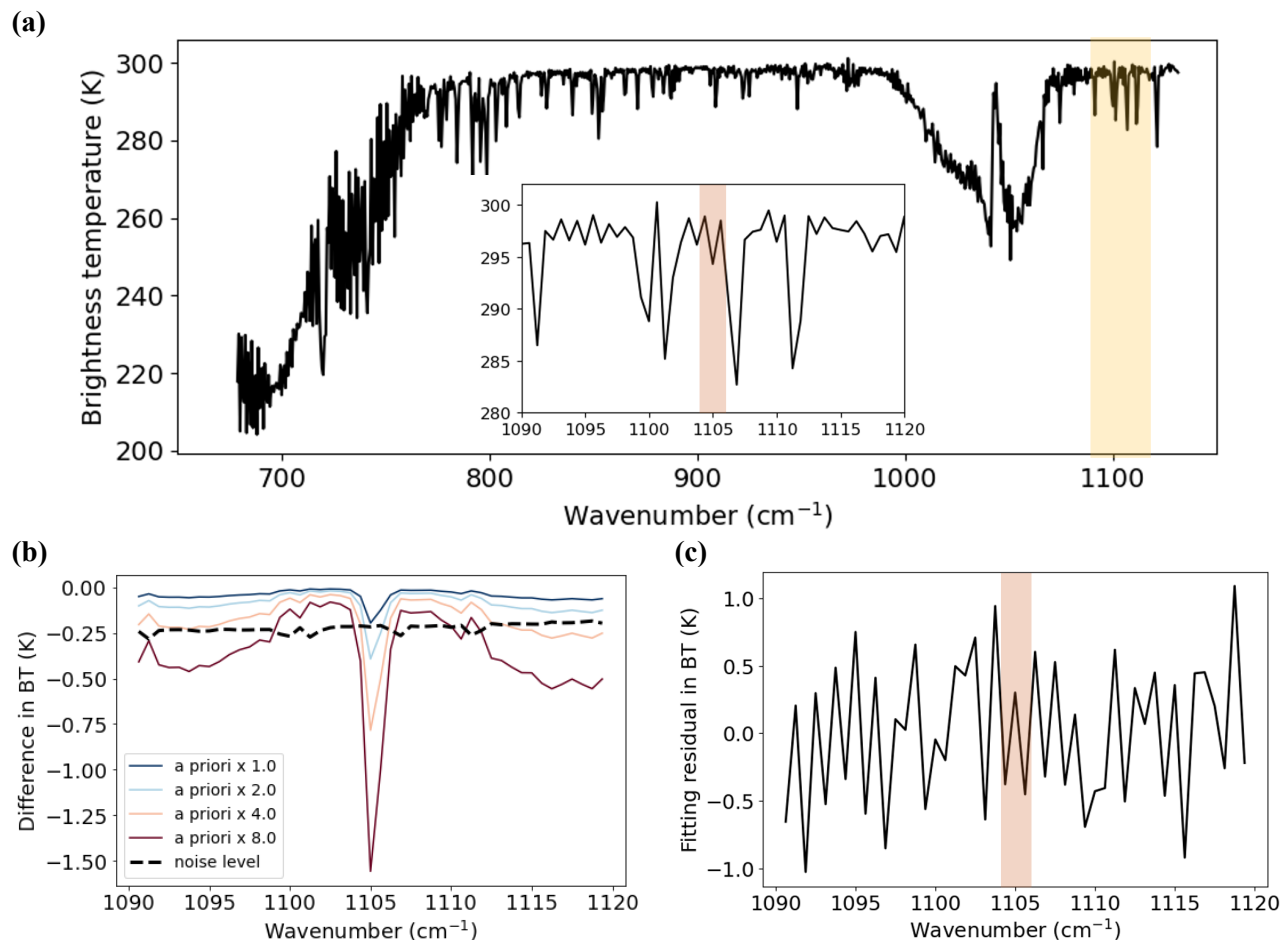

(b)

(c)

Figure 1. FY-4B/GIIRS observation spectra and the spectral window for HCOOH retrieval. (a) Example of measured longwave spectra in brightness temperature (BT) covering the HCOOH absorption micro-window centred around 1090−1120 cm⁻¹, indicated by the rectangle inset. The micro-window covers the strong absorption features of the HCOOH *v6* vibrational-rotational band; (b) Sensitivity experiment comparing absorptions of different HCOOH abundances in the 1090−1120 cm⁻¹ micro-window. The difference in BT is relative to spectra without HCOOH. The simulated spectra are generated using four different HCOOH abundances based on the a priori profile with a total column of 3.1×10^{15} molec/cm² ; the simulation is based on an observation at noon time on 11 April 2023 with thermal contrast of 13K. The noise level is derived from the spectral noise estimate in the FY-4B/GIIRS data product. (c) The spectral fit residual in BT averaged over all post-filtered retrievals on the 15ᵗʰ of all months from July 2022 to June 2023. The filters include DOFS>0.3 and the fitting residual rmse less than the monthly mean plus two sigma The standard deviations of the fitting errors are consistent across the different channels, which is about 0.4 K. The light-red band indicates the strong HCOOH absorption between 1104 and 1106 cm⁻¹.
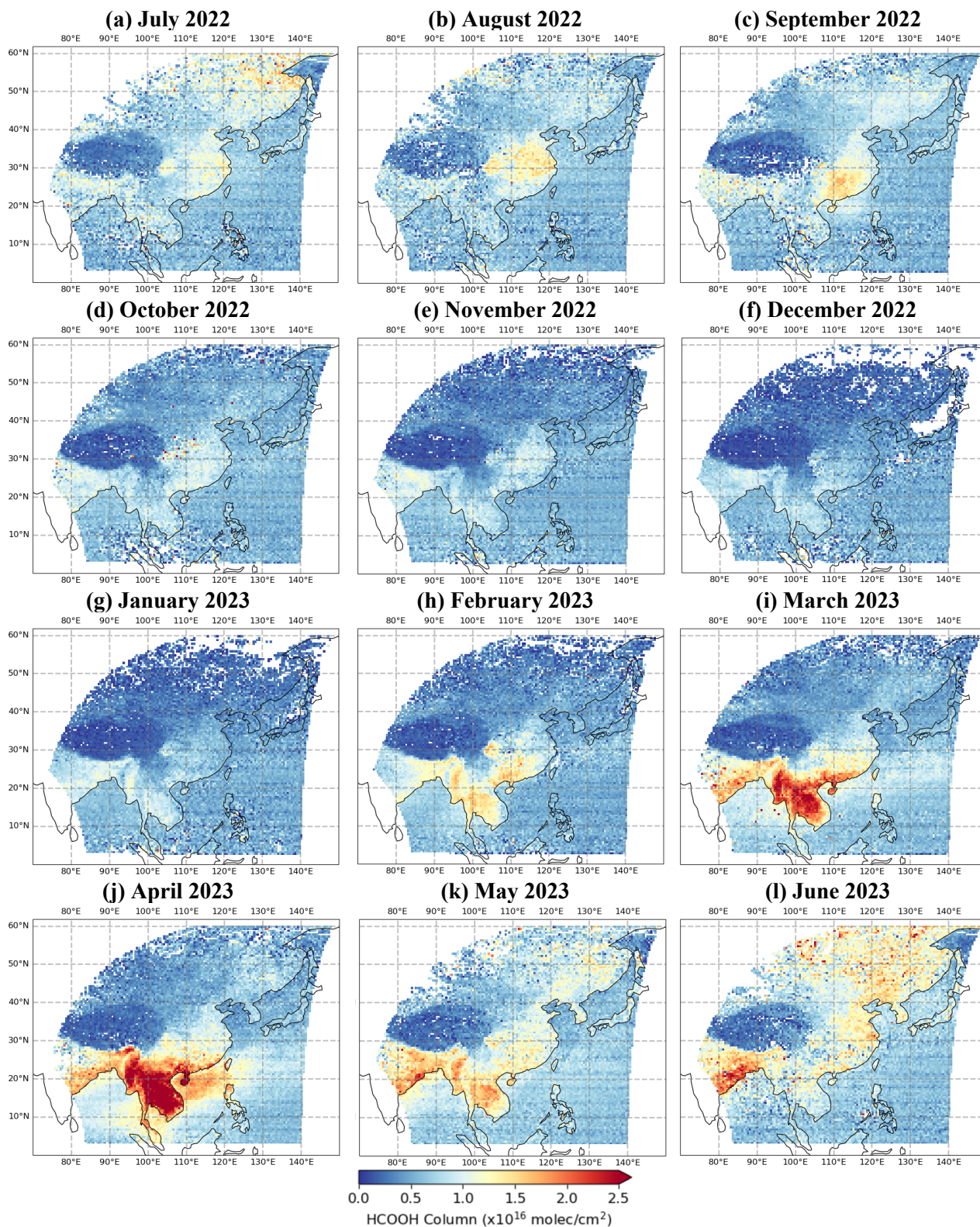
Figure 2. Monthly mean of HCOOH column retrievals at 13:00–15:00 Beijing time. These HCOOH retrievals are filtered by DOFS>0.3 and the fitting residual rmse less than the mean plus two sigma, and then gridded in 0.5° by 0.5°.
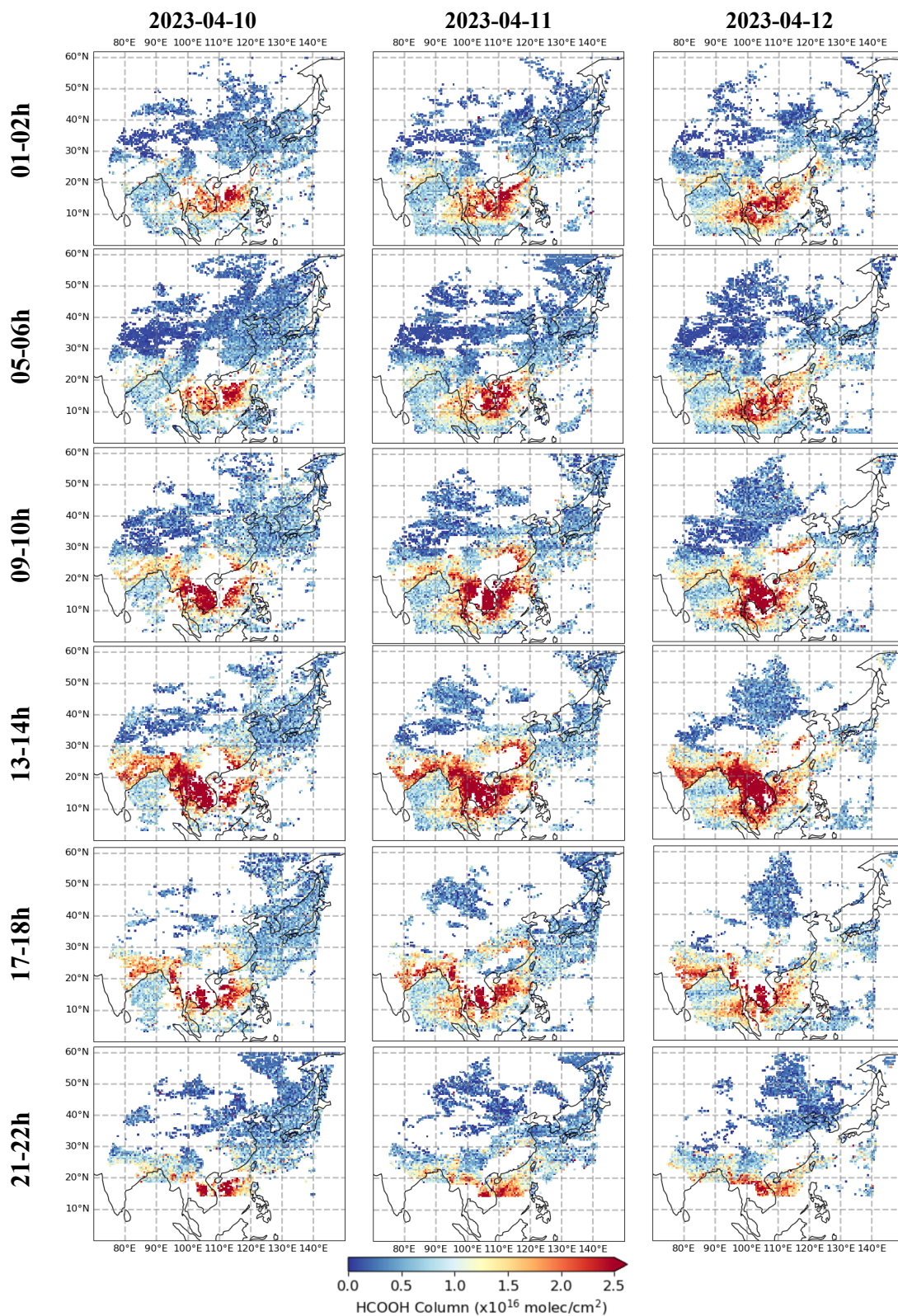
Figure 3. Example of HCOOH retrievals in each 2-hour measurement cycle from early morning to late evening on 10-12 June 2023. The data have been filtered by DOFS>0.3 and the fitting residual rmse less than the mean plus two sigma, and then gridded in 0.5° by 0.5°.

4 Results

4.1 HCOOH column retrievals and the information content

Retrieval outputs for each HCOOH observation include the HCOOH column derived from the retrieved profile, the associated posterior error covariance, the AK matrix and resulting DOFS, and the root-mean-square-error (RMSE) of the spectral fitting residual. The histograms of the fitting residual RMSE are shown in **Figures S2**. The fitting residual RMSE for all retrievals is about 0.7K on average with a standard deviation of about 0.1K. The spectral fitting residuals are shown in **Figure 1(c)**, averaged over all post-filtered retrievals on the 15th of all months from July 2022 to June 2023. Although there is a systematic pattern that can be seen in the averaged fitting residual, probably due to imperfect spectroscopy or the forward radiative transfer model, this pattern is not correlated with the absorption feature of the target gas HCOOH. To ensure high quality retrievals, the HCOOH column retrievals for each month are filtered by DOFS>0.3 and the fitting residual RMSE is less than the monthly mean plus two standard deviations.

Figure 2 shows the monthly mean of the retrieved HCOOH columns after post-filtering for the 13:00–15:00 BJT measurement cycle, when the land surface temperature and DOFS are expected to be the highest during the day, from July 2022 to June 2023. The results show the monthly changes in the HCOOH distribution over Asia, which are mainly due to the seasonality of emissions from biomass burning and biogenic sources. The largest HCOOH columns are observed in Southeast Asia in March and April, when fire emissions peak from the burning of crop residues from the so-called "slash and burn" agriculture (**Li et al., 2014; Lasko et al., 2017**). By examining the relationships between IASI-observed HCOOH, biogenic emissions, and fire radiative power, **Franco et al. (2020)** suggested that in Southeast Asia the high HCOOH emissions in spring are contributed jointly from terrestrial vegetation and wildfires. The authors indicated that photochemical production by various previously-identified sources lead to a stronger HCOOH enhancement over Southeast Asia. Over these regions, the HCOOH columns exceed 2.5×10^{16} molec/cm^2, which is more than 8 times the a priori column. In addition, relatively high abundances are observed over Siberia and the surrounding regions in June and July, probably due to a

213   combination of wildfire emissions and biogenic related emissions, as suggested by **Stavrakou et**

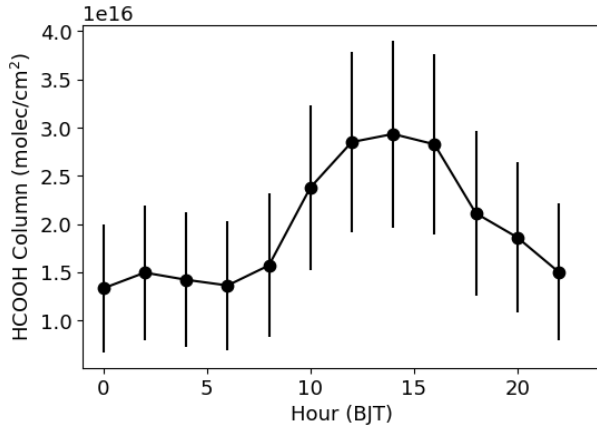214   **al. (2012) and Franco et al. (2021)**.

215   **Figures S3** and **S4** show the monthly means of HCOOH columns from, respectively, the

216   FY-4B/GIIRS retrievals covering the observation cycles at 09:00–11:00 BJT, and the daytime

217   overpasses (~9:30 am, local solar time) of IASI/Metop-B. Similar patterns in spatial distribution

218   can be observed for retrievals between FY-4B/GIIRS and IASI/Metop-B. Note that the morning

219   observations in **Figs S3 and S4** provide slightly lower abundances compared to the noontime

220   observations (**Fig. 2**), mainly due to the weaker thermal contrast (TC) conditions at the time of the

221   IASI overpass (**Clarisse et al., 2010**) and probably to the diurnal difference in production/loss

222   processes as introduced in **Millet et al. (2015)**. Here, TC is defined as the temperature difference

223   between the surface and the lower atmosphere.

224   The information content as quantified by the DOFS is summarized in **Figure S5**. The

225   DOFS is about 0.5 on monthly average with a standard deviation of 0.2, indicating that about half

226   of one piece of information is available from the spectra for HCOOH retrieval based on the

227   retrieval algorithm settings. The DOFSs over the fire plumes detected in Southeast Asia in March-

228   April are close to 1.0, indicating that the observations provide a stronger constraint on the retrieval

229   of fire-induced HCOOH emissions. As expected, we found that the DOFS is closely related to the

230   TC, which is an inherent property of thermal infrared sounding (**Clarisse et al., 2010**). Such a

231   correlation is shown in **Figure S6** for different months. Here, TC is calculated from the retrieved

232   surface temperature and the temperature of the bottom atmospheric layer. From the correlation

233   plots, there is a significant positive correlation between TC and DOFS, when TC is positive. The

234   correlation coefficients are mostly larger than 0.6. The DOFS also increases slightly when TC is

235   more negative. A similar relationship exists for the retrieval of ammonia from FY-4B/GIIRS

236   (**Zeng et al., 2023b**). In essence, for a given HCOOH abundance, large positive TC results in

237   strong HCOOH absorption spectral signatures. For large negative TC, HCOOH emission spectral

238   signatures can be detected. When TC is close to zero, corresponding to the situation where the

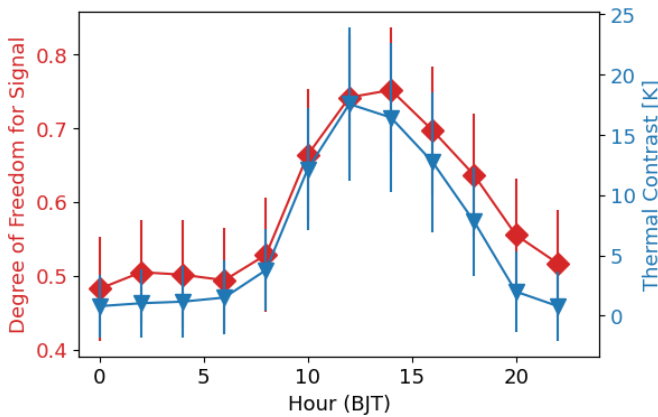239   lower atmosphere with concentrated HCOOH has a similar temperature to the surface, it is difficult

240   to decorrelate the surface layer with the rest of the lower troposphere which hampers the trace gas

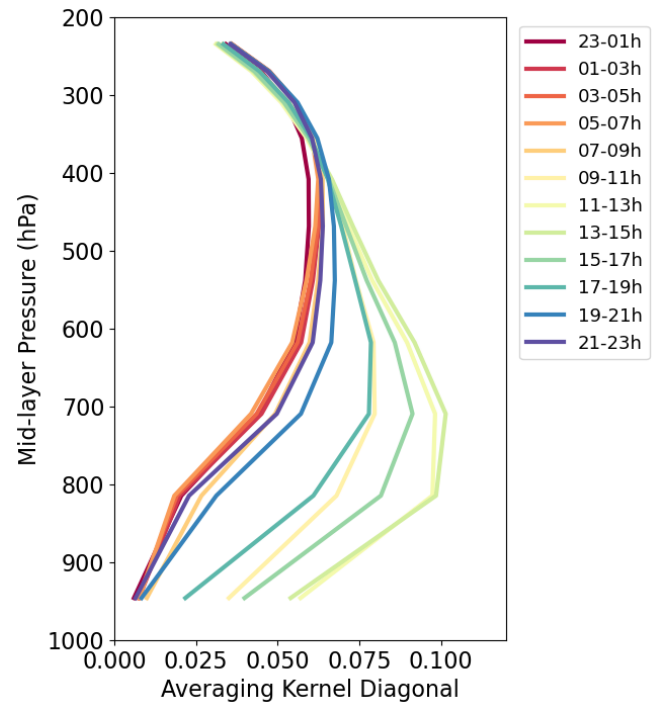241   retrieval (**Bauduin et al., 2017**).

242

(a) HCOOH columns

(c) Averaging kernel

(b) DOFS and TC

243
244   **Figure 4. The diurnal cycle in Southeast Asia. (a) Diel cycle of HCOOH columns (for every**
245   **measurement cycle of two hours) averaged for all data in April 2023 over the wildfire regions**
246   **in southeast Asia (100°E-105°E, 15°N-20°N) in the upper panel and the associated DOFS and**
247   **TC in the bottom panel. Error bars represent 1 sigma; Data have been filtered by DOFS>0.3**
248   **and the spectral fitting residual less than the sum of the mean and 2 sigma; (c) Averaged AK**
249   **diagonal vectors for every measurement cycle in April 2023 over the same region as (a)**
250

251   **4.2 Diurnal variations of HCOOH column retrievals**

252   FY-4B/GIIRS from the geostationary orbit has the advantage of observing at high temporal
253   resolution in both the daytime and nighttime, allowing the characterization of the HCOOH diel
254   cycle over East and Southeast Asia. Here, two cases are shown separately for the wildfire
255   emissions in Southeast Asia (**Figure 3**) and the biogenic emissions in Siberia (**Figure S7**),
256   separately. These events serve as good case studies to demonstrate the tracking capability of FY-
257   4B/GIIRS from geostationary orbit. From Figure 3, the strong enhancement of the HCOOH
258   columns over Southeast Asia and the transport to the surrounding oceans can be clearly observed
259   for all day and night observations. As expected, the columns are highest in the mid-afternoon and
260   lowest in the night. On average, the afternoon HCOOH columns, which peak around 14:00 BJT,
261   are twice as high as the nighttime values, as shown in **Figure 4(a)**. This diurnal cycle of HCOOH
262   columns is consistent with fire radiative power from satellite-based active fire products (**Giglio,**
263   **2007; Roberts et al., 2009**), which is associated with the diurnal changes in temperature, humidity,
264   and human activity. Similar patterns can be seen between the daytime and nighttime overpasses of
265   the IASI HCOOH retrievals, as shown in **Figure S8**.

266   However, the diel cycle detectivity of GIIRS is influenced by the diurnal variations of TC
267   over land, as has been demonstrated for other trace gases (**Zeng et al., 2023a; 2023b)**, which
268   affects the information content available from the observed spectra. This effect can be seen in the
269   HCOOH retrievals from FY-4B/GIIRS, as shown in **Figure 4(b).** The diurnal cycles of the
270   HCOOH columns and the associated DOFS and TC over the source region are highly correlated.
271   Fortunately, when the TC is low, during the night, the DOFS is still high (on average about 0.5),
272   providing a good constraint for the detection of HCOOH columns from the nighttime spectral
273   measurements. As a result, the nighttime HCOOH columns ($\sim 1.5 \times 10^{16}$ molec/cm^2) are about 5
274   times larger than the a priori total column (3.1×10^{15} molec/cm^2) used in the retrieval algorithm.
275   The change in detectivity from day to night is also reflected in the AK matrix, which indicates the
276   sensitivity of the retrieved HCOOH partial columns in each atmospheric layer to the "truth".
277   **Figure 4(c)** shows the averaged AK diagonal vectors, which quantify the detectivity of each layer
278   from the observed spectra. FY-4B/GIIRS has a higher detectivity during the day than at night.

279　From midnight (i.e., 0h) to midday (i.e., 12h), the AK values increase, and the most sensitive layer
280　shifts towards the lower atmosphere.

281　　　During the summer, large plumes of HCOOH can be observed in Siberia and the
282　surrounding areas, transported over long distances from the source regions towards the east. These
283　plumes usually escape from the boundary layer to reach the free troposphere where they undergo
284　long-range transport (**Pommier et al., 2017; Franco et al., 2020**). One of these events in early
285　June resulted in the formation of an elongated plume with a high HCOOH abundance, extending
286　from central Siberia to northern China, as is shown in **Figure S7**. The propagation and dilution of
287　the same plume of HCOOH columns can be observed from early morning to late evening on 09-
288　11 June 2023. The retrieved columns show considerable diurnal variations. The HCOOH
289　enhancements from the nighttime retrievals are less pronounced than those from the daytime
290　retrievals. With a lifetime of several days in the free troposphere, significant intraday variations of
291　the HCOOH abundance are usually not expected. However, in the case of a plume where secondary
292　formation of HCOOH associated with photochemical production along the transport is intense,
293　such a diurnal change may be possible. However, the change can also be partially attributed to the
294　diel cycle of TC. Similar patterns can be seen from the IASI HCOOH retrievals, as shown in
295　**Figure S9**.

4.3 Inter-comparison with IASI HCOOH retrievals

297　　　Independent column measurements from ground-based FTIR for validation, as in **Franco**
298　**et al. (2020)**, are unfortunately not available over this region. Instead, accuracy assessment of the
299　FY-4B/GIIRS HCOOH retrievals is performed by inter-comparison with IASI retrievals. The first
300　generation of IASI HCOOH (**Razavi et al., 2011; Pommier et al., 2016**) was retrieved using a
301　fast algorithm based on brightness temperature differences, which were then converted to HCOOH
302　columns using conversion factors derived from OEM performed over source regions. A new IASI
303　HCOOH product has been developed by **Franco et al. (2018)** using the Artificial Neural Network
304　for IASI (ANNI) retrieval framework, which is based on machine learning and a hyperspectral
305　range index that shows good sensitivity to the weak spectral absorption signature of HCOOH. IASI
306　HCOOH data have been used in combination with model simulations and have significantly

307   improved our understanding of HCOOH sources and sinks globally (e.g., **Stavrakou et al., 2012;**
308   **Pommier et al., 2016; Franco et al., 2021**).

309   In this study, we use the ANNI HCOOH v4 product from IASI/Metop-B and perform
310   collocated point-by-point comparisons. A collocated point pair between GIIRS and IASI is defined
311   as observation footprints that are less than 20 km apart in space and differ by less than 1 hour apart
312   in observation time. Theoretically, the comparison between two different instruments need to
313   reconcile the retrieval a priori profiles and the vertical sensitivity as quantified by the AK matrix.
314   Both IASI and GIIRS use the same a priori profiles, which is an advantage for the comparison. In
315   addition, **Pommier et al. (2016)** showed that the vertical sensitivity of IASI in observing HCOOH
316   peaks between 1km and 6km, which is consistent with GIIRS as shown in **Figure 4**. Therefore, in
317   this study, we carry out direct comparisons of HCOOH columns between GIIRS and IASI. The
318   comparison results are shown in **Figure 5 and Figure S10** for the day and night overpasses of
319   IASI. We specifically focus on the months when there are strong HCOOH emissions in March,
320   April, June, and July of 2023. The correlations are analyzed for three different DOFS filters of
321   GIIRS representing different levels of observational information content. Overall, the two datasets
322   show good agreement, especially for the daytime observations when there is a higher TC. A
323   persistent difference can be observed in all cases where GIIRS shows slightly higher HCOOH
324   columns compared to IASI. The RMSE is between 4.5×10^{15} and 6.0×10^{15} molec/cm^2, which may
325   be dominated by the retrieval uncertainty related to spectral noise. As expected, the agreement
326   improves as the DOFS filter becomes more stringent, suggesting that part of the systematic
327   difference is due to the lack of information content. Examples of scene-by-scene comparisons are
328   also made that compare the re-gridded IASI retrievals (in 0.5° by 0.5° grids) in a single track with
329   the re-gridded GIIRS retrievals from the closest measurement cycle. The retrieval noise is expected
330   to be reduced for the gridded averages of the HCOOH columns. **Figures S11-S12** show examples
331   of the comparison on April 10 and June 10 for scenes with significant emissions. These results
332   show that FY-4B/GIIRS overlooking East and Southeast Asia from a geostationary orbit has
333   comparable detectivity of HCOOH columns to IASI in the low Earth orbit.
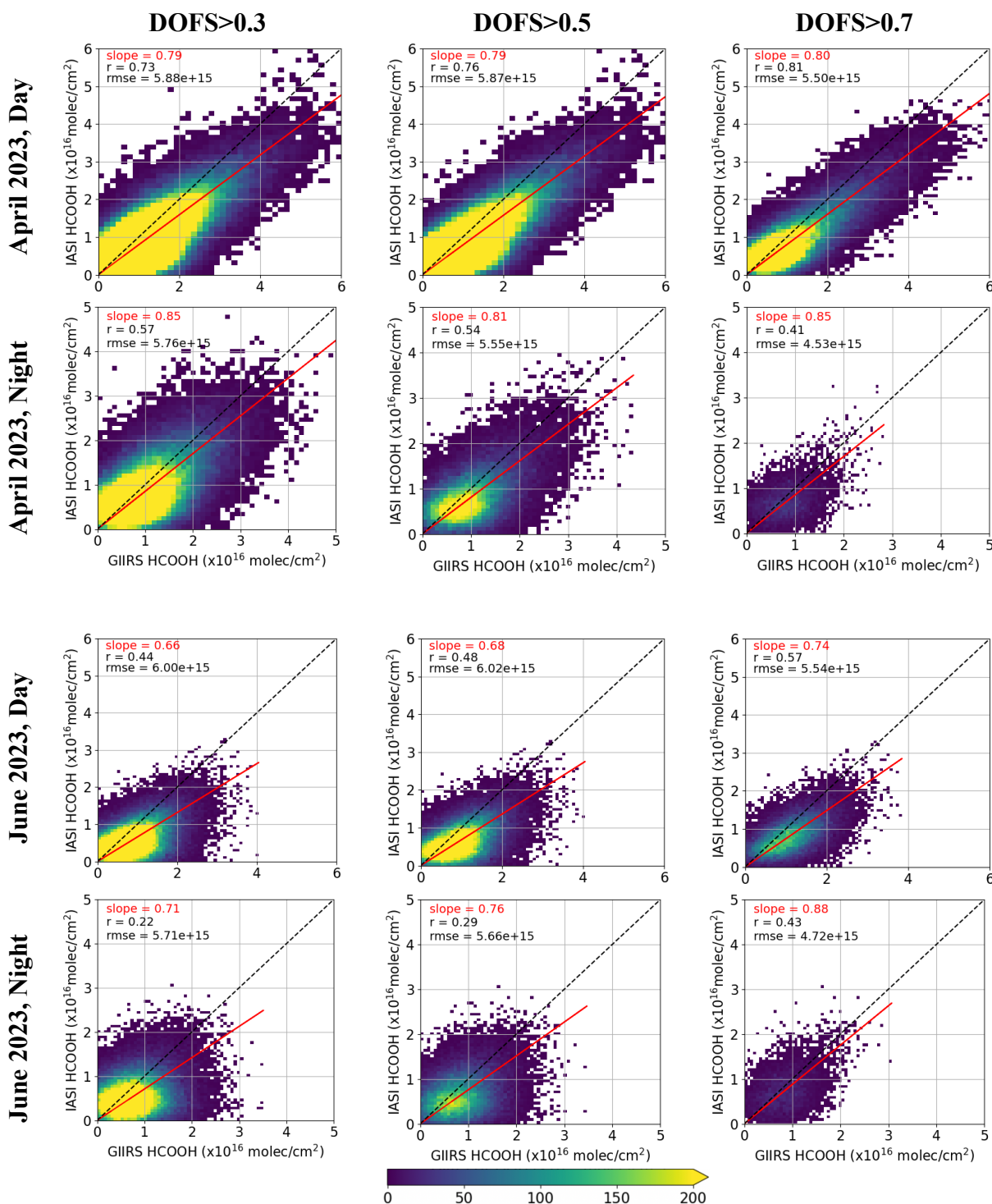
334

Figure 5. Comparison of HCOOH columns between GIIRS and IASI retrievals in April 2023 and June 2023, separately for day and night. Collocated point pairs are defined as observation footprints that are less than 20 km apart in space and differ by less than 1 hour in observation time. The black dashed line is the 1:1 line while the red line is the linear fit through the origin. The slope, correlation coefficient (r), and root-mean-square-error (rmse) are also given. These IASI HCOOH retrievals are filtered by the "postfilter" variable in the data product. GIIRS HCOOH retrievals are filtered by DOFS>0.3 and fitted residual rmse less than the mean plus two sigma.

5 Conclusions

342

343   This study presents the first results of retrieved atmospheric HCOOH retrieval from a
344 geostationary orbit using infrared hyperspectral observations from FY-4B/GIIRS. By applying the
345 FY-GeoAIR retrieval algorithm based on OEM, this study quantifies the information content of
346 the spectra in retrieving the diurnal HCOOH over Asia from July 2022 to June 2023, and
347 investigates how geostationary observations can facilitate continuous tracking of diurnal wildfire
348 emissions and the long-range transport of HCOOH. Our results show that the DOFS, which
349 represents the detectivity of HCOOH, is about 0.5 on a monthly average with a standard deviation
350 of 0.2, indicating that about half of a piece of information from the spectra is available for HCOOH
351 retrieval. In addition, the DOFS is closely related to the diurnal variation of TC, resulting in the
352 highest DOFS during the day and the lowest during the night. Nevertheless, when the TC is low
353 during the night, the DOFS is still sufficiently high to provide a constraint for the measurement of
354 HCOOH columns from wildfire emissions. In addition, from midnight (i.e., 0h) to midday (i.e.,
355 12h), the atmospheric layer with the highest detectivity, as indicated by the AK matrix, changes
356 from the upper to the lower troposphere. Finally, inter-comparison with IASI HCOOH retrievals
357 shows good agreement, indicating that FY-4B/GIIRS observations have comparable detectivity of
358 HCOOH columns to IASI in the low Earth orbit. Ideally, GIIRS HCOOH columns should be
359 compared with independent column measurements such as ground-based FTIR measurements,
360 similar to **Franco et al. (2020)**. The ground-based measurements would provide a more robust
361 validation of the GIIRS HCOOH data. There are a few FTIR stations over the GIIRS coverage
362 area. Unfortunately, they do not provide publicly available retrievals of HCOOH.

363   Tracking the abundance of HCOOH is important for understanding the complex processes
364 of HCOOH production, evolution, and loss in the atmosphere, especially for HCOOH plumes that
365 reach the free troposphere and survive for long distances. For example, GIIRS observations with
366 HCOOH-to-CO enhancement ratios can be used to study the secondary formation of HCOOH
367 along a plume. FY-4B/GIIRS in the geostationary orbit provides such important information with
368 its HCOOH retrievals every two hours from early morning to late evening. Given the diurnal
369 change in the information content available from the observed spectra, it is important to include
370 the AK matrix in further analysis such as data assimilation. Future work will focus on verifying
371 chemical transport model simulations of HCOOH with satellite observations to understand the diel

372 characteristics of sources and sinks of HCOOH in Asia, especially over major source regions, and

373 to track the evolution of HCOOH in the atmosphere.

374   This study is an important first step towards monitoring VOCs from geostationary infrared

375 sounders from a constellation of geostationary infrared sounders, including the existing and future

376 GIIRS on board the FY-4 series over Asia and the forthcoming European geostationary infrared

377 sounder (IRS) on board Meteosat Third Generation (MTG) over Europe.

378

393

394   **Open Research**

395   The HCOOH retrieval data from FY-4B/GIIRS in this study are available at

396   [**https://disk.pku.edu.cn/link/AA9FDC4AD607334B718F6A21E7870837F8%20Folder%20**

397   **Name:%20FY4B_GIIRS_HCOOH_v2**] and will be made publicly available upon publication.

398   FY-4B/GIIRS Level 1 data are publicly available from the FengYun Satellite Data Center at

399   http://satellite.nsmc.org.cn/portalsite/default.aspx; The surface emissivity datasets are

400   downloaded from the Global Infrared Land Surface Emissivity: UW-Madison Baseline Fit

401   Emissivity Database at https://cimss.ssec.wisc.edu/iremis/; The ECMWF ERA5 reanalysis

402   datasets are available from the Copernicus Climate Data Store at

403   https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels   and

404   https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels; The ECMWF

405   atmospheric composition datasets are available from the Copernicus Atmosphere Data Store at

406   https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-atmospheric-composition-forecasts.

407   The IASI/Metop-B HCOOH columns presented in this paper are publicly available on Zenodo

408   (https://doi.org/10.5281/zenodo.10894008).

Competing interest

The authors declare that they have no conflict of interest.

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