Spatial-Temporal Changes of Methane Content in the Atmosphere for Selected Countries and Regions with High Methane Emission from Rice Cultivation

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Abstract: Irrigated and flooded rice is associated with methane (CH$_4$) emissions. CH$_4$ is one of the most important anthropogenic greenhouse gases (GHG) in the atmosphere. Nowadays, mapping CH$_4$ content at a global scale is possible using satellite sensors. Sample of such a sensor is TROPOspheric Monitoring Instrument (TROPOMI) placed on the Sentinel-5 Precursor (Sentinel-5P) satellite board. In this study, the evaluation of spatial-temporal changes in CH$_4$ content in the atmosphere for selected countries and regions with high CH$_4$ emissions from rice cultivation in 2019–2021 was performed. Visual evaluation of the spatial variability on CH$_4$ content for the total study period indicates higher CH$_4$ content for almost all areas with high rice concentration. This was confirmed by positive correlations between CH$_4$ content in the atmosphere and estimated GHG emissions from croplands analyzed separately for each studied country/region. In addition, seasonal changes in CH$_4$ content in the atmosphere were observed. The lowest CH$_4$ content was observed at the beginning of the year (for the first quarter of the year) and the highest for the third quarter of the year. Moreover, a long-term increase in CH$_4$ was noticed. Regression analysis revealed that the mean increase in CH$_4$ content in most of the studied regions/countries was about 15 ppb per year. CH$_4$ content evaluated with the use of satellite data from Sentinel-5P is a reliable data source and can be used for the analysis of temporal changes at various spatial scales, including regions and countries.

Keywords: Sentinel-5P; GHG emissions; China; India; Bangladesh; Thailand; USA

1. Introduction

Rice is one of the most important crops in the world with a cultivation area covering around 150 million hectares, of which around 50% is irrigated and flooded for most of the rice-growing season [1]. In the last 50 years, the global rice harvest area has increased by 40% due to the expansion and intensification of rice cultivation which is one of the staple foods for a rapidly growing population [2]. The rice-growing area is mainly located in Southeast Asia, which accounts for 90% of the world’s rice cultivation [3,4]. In addition to China, India, and Thailand, the largest rice exporters are also the United States, where the cultivation is mainly located in the Arkansas Non-Delta, Mississippi River Delta, Gulf Coast, and Sacramento Valley of California regions [5]. The cultivation of rice is associated with increased methane (CH$_4$) emissions. The practice of flooding rice fields, leading to anaerobic conditions, promotes the formation and release of CH$_4$ [6]. The water blocks oxygen from penetrating the soil, creating ideal conditions for bacteria that emit CH$_4$.

CH$_4$ is the second most important anthropogenic greenhouse gas in the atmosphere, next to carbon dioxide (CO$_2$). Its global warming potential (GWP) over a 100-year time horizon is 25 times greater than that of CO$_2$ [7]. Global CH$_4$ emissions from rice fields are estimated to be more than 8% of total global anthropogenic CH$_4$ emissions [8]. Therefore, rice fields have a significant impact on the total global CH$_4$ emissions. Understanding
spatial distribution of CH$_4$ emissions from this crop and its changes over time is extremely important for understanding CH$_4$ emissions dynamics and for mitigating climate change.

Frequent mapping of CH$_4$ content at the global scale is possible using satellite sensors [9]. One of the most important sensors for such purpose, because of high spatial resolution and short revisit time, is the TROPOMI (TROPOspheric Monitoring Instrument). It is placed on the Sentinel-5 Precursor (Sentinel-5P) satellite board, which was launched in October 2017 [10]. Satellite data from Sentinel-5P can be transformed to the column-averaged dry-air mixing ratio of CH$_4$ in the atmosphere at spatial resolution $7 \times 7$ (to August 2019) and $7 \times 5.5$ km (from August 2019) [11]. For evaluation of CH$_4$ content, two spectral ranges are used, i.e., near-infrared (NIR): 675–775 and short-wave infrared (SWIR): 2305–2385 nm [12]. CH$_4$ content based on Sentinel-5P data is available so far only for land surfaces. For water surfaces, it will be available in a later phase. Satellite CH$_4$ content measurements can be treated as reliable data [13,14]. Sentinel-5P-derived CH$_4$ content is in good agreement with ground-based measurements from the Total Carbon Column Observing Network (TCCON), as well as with data derived from GOSAT satellite observations. TCCON and GOSAT satellite observations have been considered a source of reliable data used for the evaluation of methane content in the atmosphere for many years. Sentinel-5P is quite a new source of such data but high agreement with these two data sources allows considering these data as reliable for evaluation CH$_4$ content all over the globe. In most cases, the standard deviation of mean bias between Sentinel-5P derived CH$_4$ and TCCON is not greater than 15 ppb [15]. When comparing the CH$_4$ content obtained from Sentinel 5P vs. GOSAT, the mean difference was 13.6 ppb, the standard deviation was 19.6 ppb, and the Pearson correlation coefficient was 0.95 [16]. The relative difference between Sentinel-5P versus TCCON-derived methane is very small, less than 1% and a very high correlation coefficient confirms a very high agreement between these two sources of data.

The aim of the study was to evaluate the spatial-temporal changes in CH$_4$ content in the atmosphere for selected countries and regions with high CH$_4$ emissions from rice cultivation in 2019–2021. Using Sentinel-5T data, it can be proven that the CH$_4$ content in the atmosphere is characterized by long-lasting growth and seasonal variability.

2. Materials and Methods
2.1. Study Area

Several countries and regions within countries were selected for the study. In the first step of the analysis, countries were selected based on the high share of CH$_4$ emissions from rice cultivation among agricultural sources using the latest FAOSTAT data for 2019 [17]. The following countries were selected (from the lowest—18% to the highest share—78%): Nepal, Cote d’Ivoire, Solomon Islands, Iraq, Suriname, Dominican Republic, India, Guyana, Lao People’s Democratic Republic, Madagascar, French Guyana, Nigeria, Indonesia, Egypt, Guinea, Myanmar, China, South Korea, Sierra Leone, Gambia, Malaysia, Bangladesh, Japan, North Korea, Cambodia, Sri Lanka, Vietnam, Comoros, Thailand, Philippines. Then, those with at least 10% of the rice cultivation area of the total region/country area were selected. We also added the part of the area of the United States because, although it does not have a high share of rice emissions in the total CH$_4$ emissions. The area taken up by rice crops in the Mississippi region is above 10%. The high spatial resolution (five minutes spatial resolution—10 km × 10 km) data about rice crop share in the total land area come from the study of Ramankutty et al. [18] available at EarthStat/ [19]. They were used for the calculation of rice crop share for selected regions to evaluate the rice share by region/country.

Figure 1 presents a map with the countries and regions included in the study.
2.2. Satellite Data

Data from Sentinel-5P were used for the analyzes. The column-averaged dry-air mixing ratio of CH$_4$ (in ppb—parts per billion) data for the period from 8 February 2019 to 30 June 2021 were used. The Sentinel-5P imagery from the TROPOMI sensor processed as Level 3 (L3) product was used [20]. Datasets were downloaded from the image collection “COPERNICUS/S5P/OFFL/L3 CH4” of Google Earth Engine as georeferenced raster files (in GeoTIFF format file) at spatial resolution 7 × 7 km. Mean values of the column-averaged dry-air mixing ratio of CH$_4$ for 3-month periods were analyzed. The only exception was the first period, which was shorter (from 8 February–31 March 2019).

2.3. Data Analysis

Raster files (GeoTIFF) containing averaged values of the air-methane mixing coefficient were used for further processing. For selected countries or regions, the mean CH$_4$ content for each study period was calculated using the zonal statistics tool in QGIS 3.18 software [21]. The means were used for evaluating the seasonal (within years) and between years changes. Moreover, within countries/regions, spatial-temporal variability was evaluated based on maps for each country or region. The results for the studied countries/regions were compared to the reference areas, i.e., neighboring areas without high concentration of rice cultivation. Three areas were selected as references, i.e., India (without the states with high rice concentration), China (without the provinces with high rice concentration), and the United States (excluding region with high rice concentration).

Relationship between estimated GHG emissions from croplands [19,22] and the mean content of CH$_4$ for the total period of study (from 8 February 2019 to 30 June 2021) were evaluated using Pearson’s correlation coefficient and for selected countries/regions using linear regression analysis.

The worldwide geographical layer of GHG emissions from croplands used for the analyses is at five minutes spatial resolution (~10 km × 10 km) and is based on circa 2000 estimates of GHG emissions which are presented in CO$_2$ equivalent and include CH$_4$, CO$_2$, and N$_2$O emissions. In the case of regions with a high concentration of rice paddy, the emission is mainly determined by CH$_4$ emissions from that crop and their percentage is quite stable from 2000 until now [17]. More details about the methods used for the estimation of GHG emissions are available in the study of Carlson et al. [22] and available at EarthStat [19].
The relationships were evaluated separately for each country/region. These analyses were performed at 42-arc minutes (0.7 degrees of longitude and latitude) resolution, i.e., unit areas of about 75 × 75 km (~5600 km²). For each square, the average CH₄ content and the average estimated GHG emissions were calculated in QGIS using zonal statistics. Then, the correlations were calculated separately for each country/region in the Statistica program [23]. The operation was repeated four times with different grid shifts: offset 0, 0.35, 0.175, and 0.525. Then the average, min, and max correlation coefficients were calculated.

Only areas inside the countries/regions were included in the analyses. Units that were located next to the borders of the countries/regions were removed (criterion was at least 3500 km² of area located inside the country or region). Because of the various sizes of the countries/regions, the number of observations (n) for the analyses varied from 5 to 112. Small regions of rice cultivation were removed from these analyses because the sample size was too small. Due to various sample sizes, the significance of the relationships strongly depends on the number of observations and varies between the countries/regions.

3. Results

3.1. Methane Content by Country/Region

The mean content of CH₄ for the total period of the study (from 8 February 2019 to 30 June 2021) was quite similar for all the studied counties (Table 1).

The lowest content of CH₄ (1836 ppb) was observed for Jawa Tengah (an island which is part of Indonesia) while the highest (1904 ppb) for Jiangsu (province of China). These differences between the mean CH₄ content for the countries/regions were caused rather by worldwide CH₄ spatial variability not local emissions from rice fields. Because of that, further analyses were performed separately for each country/region to avoid the effect of worldwide CH₄ variability. Visual evaluation of the spatial variability on CH₄ content for the total period of the study based on maps presented in Figure 2 indicates higher CH₄ content for areas with high rice concentration. One of the most visible cases is the region of rice cultivation located along the Mississippi River (United States) where CH₄ content is much higher (about 50 ppb higher) in comparison to neighboring areas. However, such high CH₄ content in areas of rice concentration does not always occur.

![Figure 2](https://earthengine.google.com/) (accessed on 5 July 2021).

**Figure 2.** Mean CH₄ content for the studied period (from 8 February 2019 to 30 June 2021) based on Google Earth Engine https://earthengine.google.com/ (accessed on 5 July 2021).
Table 1. Mean CH$_4$ content for total period of the study (from 8 February 2019 to 30 June 2021) for studied countries/regions, estimated emissions of GHG from croplands, and correlations between CH$_4$ content and estimated GHG emissions by region/country. Calculated using the data from: http://www.earthstat.org/greenhouse-gas-emissions-croplands/ (accessed on 5 July 2021) and Google Earth Engine https://earthengine.google.com/ (accessed on 5 July 2021) [19].

| Country/Region          | Country  | CH$_4$ Content in Atmosphere (ppb) | Estimated GHG Emissions from Croplands (Mg CO$_2$e Per Hectare) * | Mean Correlation | Min. Correlation | Max. Correlation |
|-------------------------|----------|------------------------------------|-------------------------------------------------------------------|-----------------|-----------------|-----------------|
| Zhejiang                | China    | 1886                               | 4.12                                                              | 0.28            | 0.16            | 0.43            |
| Jiangxi                 | China    | 1887                               | 3.84                                                              | 0.02            | −0.03           | 0.06            |
| Hubei                   | China    | 1892                               | 3.09                                                              | −0.38           | −0.47           | −0.25           |
| Anhui                   | China    | 1897                               | 3.26                                                              | −0.49           | −0.53           | −0.41           |
| Jiangsu                 | China    | 1904                               | 4.04                                                              | 0.62            | 0.57            | 0.68            |
| Hunan                   | China    | 1886                               | 3.16                                                              | 0.27            | 0.20            | 0.36            |
| Guangxi Zhuang         | China    | 1889                               | 1.21                                                              | 0.43            | −0.20           | 0.76            |
| Taiwan                  | China    | 1878                               | 3.58                                                              | −0.54           | −0.61           | −0.48           |
| Guangdong               | China    | 1894                               | 2.09                                                              | −0.30           | −0.40           | −0.21           |
| Assam                   | India    | 1893                               | 2.04                                                              | 0.25            | 0.13            | 0.39            |
| Bihar                   | India    | 1903                               | 3.16                                                              | −0.53           | −0.76           | −0.41           |
| Chhattisgarh            | India    | 1889                               | 2.53                                                              | 0.37            | 0.18            | 0.68            |
| Haryana                 | India    | 1898                               | 2.42                                                              | −0.66           | −0.82           | −0.53           |
| Jharkhand               | India    | 1895                               | 1.65                                                              | 0.17            | 0.06            | 0.23            |
| Orissa                  | India    | 1893                               | 2.74                                                              | 0.35            | 0.28            | 0.44            |
| Punjab                  | India    | 1895                               | 4.21                                                              | 0.48            | 0.24            | 0.77            |
| Tamil Nadu              | India    | 1893                               | 2.00                                                              | −0.11           | −0.37           | 0.17            |
| Uttar Pradesh           | India    | 1901                               | 2.15                                                              | −0.20           | −0.25           | −0.11           |
| West Bengal             | India    | 1901                               | 3.79                                                              | 0.42            | 0.34            | 0.52            |
| Jawa Tengah (Indonesia) | Indonesia| 1836                               | 8.77                                                              | 0.14            | 0.08            | 0.17            |
| Mississippi region (USA)|        | 1884                               | 1.69                                                              | 0.36            | −0.02           | 0.68            |
| Bangladesh              |          | 1903                               | 3.56                                                              | −0.27           | −0.44           | −0.10           |
| Sri Lanka               |          | 1863                               | 5.41                                                              | 0.25            | −0.15           | 0.70            |
| Cambodia                |          | 1889                               | 2.47                                                              | 0.52            | 0.45            | 0.60            |
| South Korea             |          | 1872                               | 6.40                                                              | −0.06           | −0.22           | 0.16            |
| Myanmar/Burma           |          | 1877                               | 1.16                                                              | 0.37            | 0.35            | 0.38            |
| Nepal                   |          | 1890                               | 1.73                                                              | 0.63            | 0.22            | 0.85            |
| Philippines             |          | 1853                               | 1.76                                                              | 0.59            | 0.04            | 0.78            |
| Thailand                |          | 1889                               | 3.40                                                              | 0.42            | 0.16            | 0.53            |
| Vietnam                 |          | 1877                               | 7.51                                                              | 0.24            | 0.18            | 0.31            |

* Significant correlations at 0.05 probability level are in red font. Because of various sample sizes (N) significance of the relationships strongly depends on N used for the analyses and varies between the countries/regions. For countries/regions with larger N critical value of correlation coefficient is lower in comparison to countries/regions where N was lower. Green color indicates low content of CH$_4$ in atmosphere (ppb) and estimated GHG emissions from croplands, while red color indicates high values of these variables. Blue color indicates low values of correlation coefficient (negative correlations), while red color indicates high values (positive correlations).

3.2. Temporal Changes in CH$_4$ Content

During the period of the study, seasonal changes, as well as a long-term trend, was observed for the content of CH$_4$ in most of the countries (Table 2). All provinces of China were characterized by a very similar pattern of changes in time. Seasonal changes of CH$_4$ were characterized by the lowest content at the beginning of the year (for the first quarter of the year) and the highest for the third quarter of the year. Moreover, a long-term increase in CH$_4$ was observed. On average, in the period of the study, an increase in CH$_4$ content by 15 ppb per year was observed for the studied provinces of China. Similar seasonal and long-term changes were observed for other regions and countries in Southeast Asia but in some cases, the changes were quite different. For example, in the states Tamil Nadu and Andhra Pradesh (Southeast India), the lowest seasonal CH$_4$ content was observed for the third quarter of the year. Another example of the exception is Jawa Tengah island (Indonesia),...
where seasonal changes for CH$_4$ content were very small but long-term changes were very similar to those observed for China. In the Mississippi region (USA), the highest seasonal CH$_4$ content was observed at the end of the year (the fourth quarter of the year), while the lowest was at the beginning of the year. Long-term changes for CH$_4$ were similar for most of the regions of Southeast Asia (increase of about 16 ppb per year). Regression equations presented in Figure 3, which presents averaged CH$_4$ content for 3-month periods, confirm that the mean increase in CH$_4$ content in regions/countries of Southeast Asia is about 16 ppb per year (about 4 ppb per each 3-month period), while in the Mississippi region (United States) the mean increase was about 15.5 ppb per year (3.87 ppb per 3-month period). A similar long-term increase was observed for both parts of India, i.e., where high concentrations of rice production occur, and for other areas of India (mean increase in CH$_4$ content by about 16 ppb per year). In the case of the United States reference area, which is most of the area of the US excluding the main regions of rice cultivation, the mean yearly increase in CH$_4$ content was smaller in comparison to regions with high rice concentration (respectively 13.56 ppb vs. 15.48 ppb of CH$_4$). The increase for total world area (limited to latitudes between 60° S and 60° N) is 12.8 ppb per year (3.20 ppb per 3-month period) and is lower in comparison to areas with rice cultivation. Similar results but for 1-month periods are presented in Figure S1 and allow evaluating temporal changes of CH$_4$ content at higher temporal resolution. It allows observing the short time increase in late summer and beginning of autumn in CH$_4$ content in the regions/countries in south-eastern Asia. The most visible increase can be noticed in Bangladesh in October when maximal CH$_4$ content was observed. The temporal changes observed for 1-month periods are much lower for reference areas, i.e., for total of the United States and total world, which suggest that are strongly dependent on the rice growing period. Regression equations based on 1-month periods presented in Figure S1 suggest a similar long-term trend of CH$_4$ but slightly different in comparison to the regression equations based on 3-month periods. For example, the mean yearly increase in CH$_4$ for total world area (limited to latitudes between 60° S and 60° N) based on regression equation for 1-month periods was equal to 14.1 ppb while based on 3-month periods was slightly lower and equal to 12.9 ppb. Usually, regression equations for studied regions/countries based on 1-month periods prove a slightly higher increase in CH$_4$ in comparison to an increase in CH$_4$ based on respective regression equations for 3-month periods. This is probably caused by higher peaks (yearly maximal content of CH$_4$) for 1-month periods in comparison to 3-month periods.

![Figure 3](image-url)
Table 2. Means of CH₄ content for selected countries or regions based on Sentinel-5P data (in ppb) and regression equation presenting relationships between CH₄ content (y) and subsequent 3-month periods (x).

| Country/Region          | Country | 8 February–31 March 2019 | 1 April–30 June 2019 | 1 July–30 September 2019 | 1 October–31 December 2019 | 1 April–30 June 2020 | 1 July–30 September 2020 | 1 October–31 December 2020 | 1 January–31 March 2021 | 1 April–30 June 2021 |
|-------------------------|---------|--------------------------|----------------------|--------------------------|---------------------------|----------------------|--------------------------|--------------------------|--------------------------|----------------------|
| Atmopshere              | Blue    | 1844                     | 1843                 | 1846                     | 1853                      | 1856                 | 1862                     | 1867                     | 1871                     | 1878                 |
|                        |         |                          |                      |                          |                           |                      |                          |                          |                          |                      |
|                        | Red     | 1844                     | 1843                 | 1846                     | 1853                      | 1856                 | 1862                     | 1867                     | 1871                     | 1878                 |
|                        |         |                          |                      |                          |                           |                      |                          |                          |                          |                      |
|                        | Green   | 1844                     | 1843                 | 1846                     | 1853                      | 1856                 | 1862                     | 1867                     | 1871                     | 1878                 |
| Blue color indicates low values while red color indicates high values of CH₄ content.
3.3. Spatial-Temporal Variability in CH$_4$ Content for Selected Countries/Regions

Three countries/regions were selected for a more detailed evaluation of spatial-temporal changes in CH$_4$ content during the period of the study. Figures 4–6 present spatial-temporal variability by 3-month periods while Figures S2–S4 present maps based on a 1-month period. These countries/regions were selected based on various criteria. The first region is Bangladesh, which is characterized by a very high percentage of rice cultivation area—about 70% of the total country area. Therefore, CH$_4$ emissions from rice fields are very high. However, the highest content of CH$_4$ was observed for the area of the capital city—Dhaka (Figures 4, 7 and S2). Because of the limited availability of data for the third quarter of the year, in years 2019 and 2020, CH$_4$ content for this period can be biased, especially for the results based on 1-month periods. The reason for very limited availability is the monsoon period (from July to September) which is characterized by overcast conditions. The highest CH$_4$ content was observed for the third quarter of the year, especially for October, i.e., for the end of the rice period called “aman”, when most of the rice in Bangladesh is harvested. In most of Bangladesh’s area, the share of rice cultivation area is very high, the exceptions are two regions located next to the coast, i.e., the southwest part where mangrove forests are located and the southeast part with hill forests. In both of these areas, CH$_4$ content is lower in comparison to the CH$_4$ content in other areas of Bangladesh.

Figure 5, 7 and S3 present CH$_4$ content in the main area of rice cultivation in the United States located along the Mississippi River in the mouth of the river and in the lower course of the river (next to the border of Mississippi State with Arkansas and Louisiana). The area of the rice cultivation is interesting because in neighboring areas there is no rice and the effect of the rice cultivation on CH$_4$ content in the atmosphere can be better evaluated. The CH$_4$ content in the atmosphere, in the area of rice cultivation, is higher in comparison to the nearest surrounding area. The differences are most visible during the fourth quarter of the year (September–December) which is the harvest time of the rice, with the maximum value of CH$_4$ in October and November. In other seasons of the year, the differences are visible, but the smallest is observed during the first quarter of the year (January–March).

Spatial-temporal changes of CH$_4$ content in Punjab state (India) are presented in Figures 6, 7 and S4. The highest CH$_4$ content was observed in the third quarter of the year (in the middle of the growing period of rice) with maximum value in September, while the lowest in the first quarter of the year with minimal value in February–March. The lowest CH$_4$ content was observed in the Northeast part of Punjab where the share of the rice area is smaller in comparison to other parts of the Punjab state.

3.4. Relationships between Estimated GHG Emissions and CH$_4$ Content

Correlations between estimated GHG emissions and methane content are presented in Table 1. Most of the correlations are positive, as expected. Some of the correlations are negative but relatively weak. The correlations were calculated separately for each country/region at 42-arc minutes (0.7 degrees) spatial resolution. The strongest positive correlations were observed for Hubei and Hunan (provinces of China), Punjab (state of India), Cambodia, Philippines, and Nepal. Mean correlation coefficients for these regions/countries were between 0.48 and 0.79. For some countries/regions, the correlations were negative.
very high. However, the highest content of CH$_4$ was observed for the area of the capital city—Dhaka (Figures 4, 7 and S2). Because of the limited availability of data for the third quarter of the year, in years 2019 and 2020, CH$_4$ content for this period can be biased, especially for the results based on 1-month periods. The reason for very limited availability is the monsoon period (from July to September) which is characterized by overcast conditions. The highest CH$_4$ content was observed for the third quarter of the year, especially for October, i.e., for the end of the rice period called "aman", when most of the rice in Bangladesh is harvested. In most of Bangladesh’s area, the share of rice cultivation area is very high, the exceptions are two regions located next to the coast, i.e., the southwest part where mangrove forests are located and the southeast part with hill forests. In both of these areas, CH$_4$ content is lower in comparison to the CH$_4$ content in other areas of Bangladesh.

Figure 4. CH$_4$ content in Bangladesh in subsequent 3-month periods. Maps were prepared using data from https://earthengine.google.com/ (accessed on 5 July 2021).
Figure 4. CH$_4$ content in Bangladesh in subsequent 3-month periods. Maps were prepared using data from https://earthengine.google.com/ (accessed on 5 July 2021).

Figure 5. CH$_4$ content in Mississippi Valley (United States) in subsequent 3-month periods. Maps were prepared using data from https://earthengine.google.com/ (accessed on 5 July 2021).

**Figure 5.** CH$_4$ content in Mississippi Valley (United States) in subsequent 3-month periods. Maps were prepared using data from https://earthengine.google.com/ (accessed on 5 July 2021).
Figure 6. CH$_4$ content in Punjab state (India) in subsequent 3-month periods. Maps were prepared using data from https://earthengine.google.com/ (accessed on 5 July 2021).
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Figure 7. Mean CH$_4$ content in selected regions (Bangladesh, the United States, and Punjab state—India for all periods of the study—8 February 2019–30 June 2021). Big black dots indicate the largest cities (with population 5 million or greater) and small black dots indicate big cities (with population in range 1–5 million).

4. Discussion

The results obtained in this study proved a long-term increase in methane content in studied countries/regions. The mean increase for most of the countries/regions with high rice concentration was about 16 ppb of CH$_4$ per year. Rice is the most important crop in wetlands and pressure to grow rice is increasing. Rice is the only major cereal crop
that is grown almost exclusively as food. In view of the growing number of people on earth, world rice production will continue to increase. The results are consistent with CH$_4$ content from corresponding data from Total Carbon Column Observing Network (TCCON) locations [13]. It confirms that CH$_4$ content evaluated on the basis of satellite data from Sentinel-5P is a reliable data source and can be used for the analysis of temporal changes at various spatial scales, including regions and countries.

The increase in CH$_4$ content during the study period is much higher in comparison to previous years which was at a global scale in average in the range from about 0 to about 12 ppb per year in various periods from the 1980s [24,25]. The results of the study of [26] based on in situ measurements, for years 2014–2017, proved a mean yearly global increase in CH$_4$ from 7.0 to 12.7 ppb. A strong global increase in CH$_4$ content in the atmosphere is observed from 2007 and in years 2007–2017 was estimated at about 7 ppb per year. In recent years, the increase is even higher which is very worrying as it can significantly increase the greenhouse effect. In the study of Dalsøren et al. [26], long-term changes of CH$_4$ content during the period 1970–2012 are presented. The study proved quite a high increase in CH$_4$ content in the years 1986–1998, which was about 11 ppb per year. It indicates that the increase in CH$_4$ content is not uniform in longer periods and during the years 1970–2012, long periods with a different increase in CH$_4$ were observed.

Seasonal variability of CH$_4$ content in the study of Nisbet et al. [27]—observed at a global scale—was similar to seasonal variability in this study. The lowest CH$_4$ content was observed at the beginning of the year while the highest was observed in the middle of the year or in autumn. However, the seasonal (within year) amplitude for the countries/regions located in Southeast Asia with high rice concentration was usually higher (50–60 ppb) in comparison to amplitude observed at a global scale (about 40 ppb). A smaller seasonal amplitude of atmospheric CH$_4$ content was observed for the United States where it was equal to about 20 ppb. There are many factors that influence the seasonal variability of CH$_4$ emissions. Firstly, the presence of rice plants enhances the escape of methane from the soil. The main factors controlling the level of CH$_4$ emission during the growing season are positively related to the water level and plant cover. The main controlling factors during the set-aside period are the water level, as well as straw absorption and soil temperature.

The amplitudes observed in Southeast Asia in this study were higher in comparison to the results of Crevoisier et al. [28] for years 2007–2011 where seasonal variations were from about 15 ppb to about 40 ppb depending on the region (higher were observed for Northern Hemisphere than in Southern Hemisphere). Higher seasonal amplitudes of CH$_4$ content in regions of Southeast Asia can be connected with the growing cycle of rice because other anthropological sources of CH$_4$ emissions (e.g., landfills) are more stable in time and are not characterized by high seasonal changes. The most important rice-growing season in Bangladesh is called aman, which starts in April and ends in November or December [29]. In other countries of Southeast Asia, the most important rice vegetation period falls into a similar period, i.e., starts in spring and ends in autumn [30,31]. The highest CH$_4$ content is observed during the intensive growth of rice during aman period. A study by Adhya et al. [30] conducted in Indian conditions proved the highest CH$_4$ emission from rice crop about 2–2.5 months after rice germination or transplantation. Depending on the rice crop calendar, such high emissions are observed in various months but are most often in the monsoon season, i.e., between June and September. The highest CH$_4$ content in the atmosphere in East China falls into the third quarter of the year which is the period of the highest CH$_4$ fluxes from rice paddies [31]. Seasonal compliance of the increases in CH$_4$ content with the rice crop vegetation was also found in the studies of Zhang et al. [4] for various regions of India, China, and Bangladesh, i.e., the highest CH$_4$ content was observed during the most intensive growth of the rice.

Effect of rice cultivation on seasonal variation of CH$_4$ content in the atmosphere is confirmed by observation of the CH$_4$ content in regions where rice is not cultivated. Examples of such areas are South-Western and South-Eastern parts of Bangladesh where forests are located [32]. In these areas, CH$_4$ content is not only lower but seasonal variation
of CH\textsubscript{4} is much lower in comparison to other parts of Bangladesh where rice is cultivated. A similar pattern of seasonal changes of CH\textsubscript{4} content is observed in the Mississippi region, where higher CH\textsubscript{4} content and higher seasonal amplitudes are observed for areas with rice cultivation in comparison to neighboring areas where rice is not cultivated.

Most of the correlations evaluated in this study between estimated GHG emissions and CH\textsubscript{4} content were positively correlated, which is consistent with expectations. Positive relationships between atmospheric CH\textsubscript{4} concentration with the percentage of paddy rice croplands were observed in the study of Zhang et al. [4] for Southeast Asia which confirms that methane emissions from rice fields are substantial and methane atmospheric transport in the horizontal direction is relatively slow. The CH\textsubscript{4} content in the study of Zhang et al. [4] was for the periods 2003–2005 and 2007–2009 from SCIAMACHY satellite sensor and for years 2011–2013 from TANSO-FTS satellite sensor. However, some of the correlations were negative. An explanation of negative correlations in the case of some countries/regions such as Bangladesh, Guangdong (province of China), and Bihar (state of India) could be higher CH\textsubscript{4} content near to locations of big cities where urban and industrial emissions of CH\textsubscript{4} occur [33], while rice crop concentration is rather low. Some negative correlations are difficult to explain, e.g., for Anhui and Jiangsu (provinces of China) and Haryana (state of India).

CH\textsubscript{4} from sources of emission, e.g., rice paddies are transported in the atmosphere mainly in the latitudinal direction because of prevailing winds [34]. It is caused by high atmospheric CH\textsubscript{4} content in areas where CH\textsubscript{4} emission is very low, e.g., in the Sahara Desert, where CH\textsubscript{4} is transported from Southeast Asia. Schuck et al. [35] confirm that methane can be transported from South Asia to North Africa. It is difficult to present direct evidence of the transport but similar methane content at the same latitude in Asia and Africa confirms such transport caused by trade winds. Seasonal variation of CH\textsubscript{4} over Asia suggests that CH\textsubscript{4} content in the atmosphere is strongly influenced by local surface emissions during summer. It confirms that despite long-distance atmospheric transport of CH\textsubscript{4}, in large part it influences the content of CH\textsubscript{4} near sources of emission.

The results obtained in the study prove higher CH\textsubscript{4} content in the regions with high rice concentration in comparison to neighboring areas. However, it is difficult to distinguish precisely between CH\textsubscript{4} derived from rice crops and other CH\textsubscript{4} sources because of atmospheric transport of methane which is difficult to evaluate at high spatial and temporal resolution. However other sources of CH\textsubscript{4} (e.g., landfills) are characterized by much lower seasonal variability than CH\textsubscript{4} emissions from rice crops and this background CH\textsubscript{4} emission is quite stable in time [36,37]. Further studies on the detection of local CH\textsubscript{4} anomalies by combining satellite measurements with high-resolution forecasts [38] are necessary, together with higher temporal resolution of the corrected Sentinel 5-P imagery [39] to better evaluate CH\textsubscript{4} local and global transport and distinguish various sources of CH\textsubscript{4} emission.

5. Conclusions

CH\textsubscript{4} content in the atmosphere is characterized by long-term increases and seasonal variability. During the period of the study (2019–2021), the mean yearly increase in CH\textsubscript{4} was about 15 ppb for all the studied areas, however, large differences were observed between the countries/regions. The highest increase (about 20 ppb per year) was observed for the Southeast states of India, while the lowest increase was observed for countries located in the Southeast corner of Asia, i.e., Thailand (about 6 ppb per year), Vietnam, and Cambodia (about 12 ppb per year). For most of the other countries/regions, the mean increase in CH\textsubscript{4} was in the range from 13 to 18 ppb per year.

Seasonal changes of CH\textsubscript{4} content were characterized by the lowest CH\textsubscript{4} content at the beginning of the year while the highest content during autumn coincides in time with intensive growth of rice. It indicates that CH\textsubscript{4} emissions from paddy fields may have a significant effect on the seasonal variability of the CH\textsubscript{4} content in the atmosphere.
Relationships evaluated separately for each country/region proved in most cases positive correlations between CH$_4$ content in the atmosphere with estimated GHG emissions from croplands. This confirms that the CH$_4$ content is higher in areas with a high concentration of rice cultivation compared to neighboring areas. The main limitation of the study is the lack of possibility to distinguish precisely between emissions of CH$_4$ from paddy rice and other agricultural and non-agricultural sources of methane. Moreover, atmospheric transport of CH$_4$ is difficult to evaluate and affects the spatial variability of CH$_4$ content.

**Supplementary Materials:** The following are available online at https://www.mdpi.com/article/10.3390/atmos12111382/s1, Figure S1: Temporal changes in selected countries/regions and total world (limited to latitudes between 60° S and 60° N) of CH$_4$ content based on Sentinel-5P data (in ppb) and regression equation presenting relationships between CH$_4$ content (y) and subsequent 1-month periods (x), Figure S2: CH$_4$ content in Bangladesh in subsequent 1-month periods, Figure S3: CH$_4$ content in Mississippi valley (United States) in subsequent 1-month periods, Figure S4: CH$_4$ content in Punjab state (India) in subsequent 1-month periods.

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