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Clarification of Methane Emission Sources Using WDCGG Data: Case Study of Anmyeon-do Observatory, Korea

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ABSTRACT

Methane concentrations have been monitored at the Anmyeon-do Observatory, Korea, since 1999. In recent years, the methane concentration has increased, but the sources of this increase have yet to be identified. This study was designed to identify the major source contributing to the increase by using World Data Centre for Greenhouse Gases (WDCGG) data and the Greenhouse Gases Emission Presumption (GEP) method. The data were collected at Anmyeon-do between 2003 and 2009 (except 2008), and the analyses showed that the increase in methane concentration originated mainly in rice paddies around the observation point. The annual average methane concentration at Anmyeon-do was 1894 ppb, of which 100-103 ppb (5.3-5.4%) was shown to originate mainly from rice paddies. The seasonal fluctuation in methane concentration from May to October estimated by the GEP method was compared with experimental data of previous research conducted on rice paddies. The close match obtained through this comparison shows that the GEP method is effective. The difference in methane concentration was also analyzed in terms of land use and land cover. It was shown that although rice paddies account for only 14.7% of the area surveyed, they accounted for between 69 and 90% of the total increase in methane concentration. These results confirm that rice paddies are the main source of the increase in methane concentration observed at Anmyeon-do.

Key words: Concentration difference, Greenhouse Gases Emission Presumption (GEP) method, Methane concentration, Rice paddy, Seasonal fluctuation

1. INTRODUCTION

The World Data Centre for Greenhouse Gases (WDCGG), which is managed by the Japan Meteorological Agency (JMA) in collaboration with the World Meteorological Organization (WMO), provides high frequency, long-term methane concentration data for 130 observation points around the globe. Methane concentrations at 73 observation points where data have been collected for more than 11 years are shown in Fig. 1. The data suggest that methane concentrations are high at mid to high latitudes of the Northern Hemisphere, between N30 and N70. According to Saeki *et al.* (1988), distribution of methane concentrations on a global scale shows increasingly higher concentrations to the north. The main sources of methane emissions are located at the mid-latitudes in the Northern Hemisphere. Around the equator, however, methane is largely eliminated in areas with high concentrations of OH radicals. Methane concentrations thus decrease as air masses are carried from mid and high latitudes towards the equator. At 5 observation points (marked by arrows on the figure), the mean methane concentration for the period between 1995 and 1997 exceeded the average concentration for the corresponding latitude (marked by [A]) measured between 2005 and 2007. This indicates a high probability that these areas contain sources of methane emissions. The main land use and land cover at these 5 observation points are as follows: Anmyeon-do, rice paddies; Black Sea, sea; Hegyhatsal, field crops; Neuglobsow, natural park and lake; and Teriberka, swamp (WDCGG, 2013). Rice paddies, swamp and plants are known methane sources (IPCC, 2007; Keppler *et al.*, 2006). The Black Sea is able to generate methane because it is the world's largest anoxic basin (Schmale *et al.*, 2010). Field crops require further study to determine if they are a source of methane emissions. Rice paddies are considered to be a

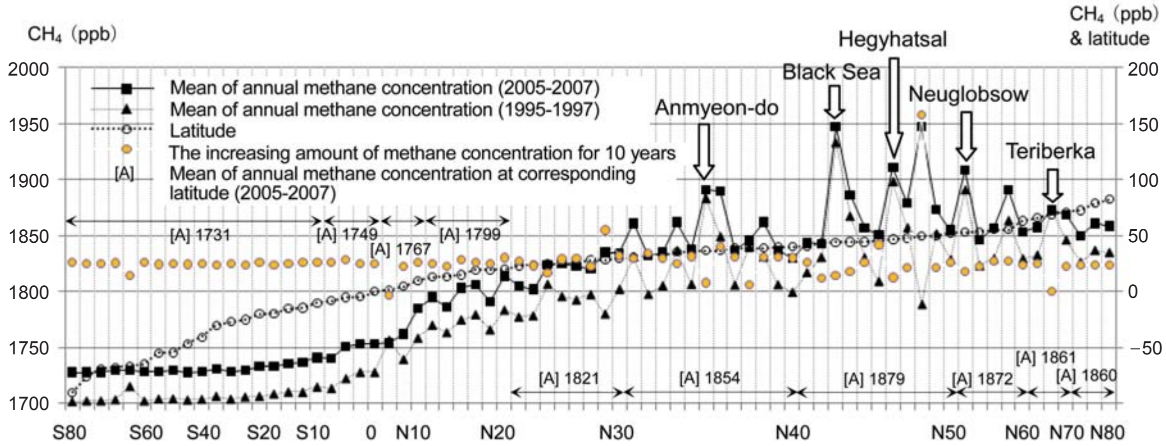

Fig. 1. Fluctuation in methane concentration according to latitude. The left vertical axis denotes the mean of the annual concentration, and the right vertical axis denotes the increase in concentration over 10 years and latitude. Seventy-three observation points where data have been collected for more than 11 years were selected from among WDCGG data (WDCGG, 2013). There are no data for Anmyeon-do between 1995 and 1997, and thus the annual average methane concentration in 1999 was utilized. The average value for between 1995 and 1997 was calculated from the following equation, $Y=A-(B-C)$, where Y is the average value between 1995 and 1997, A is the annual average in 1999, B is the annual average in 2009 and C is the average value between 2005 and 2007. The resulting value was 1868 ppb.

major source of methane emissions on a global scale, and almost 90% of the world's rice crops are produced in Asia. Anmyeon-do, the only WDCGG data site characterized by rice paddies, is therefore ideal for the evaluation of the role of land use and land cover in methane emissions.

The methane concentration at a particular site is the sum of the concentrations of existing atmospheric methane and additional methane generated at the time of observation. It is not possible to separate these two sources for independent measurement with the use of existing technologies. Furthermore, atmospheric dispersion models for estimating the concentration of atmospheric pollution depend on a variety of parameters. Therefore, there are limits to the modeling accuracy. Previous research based on WDCGG data at Anmyeon-do has identified inflow from the Asian mainland and agricultural activities in the surrounding regions as possible sources of the increase in methane concentration observed at the site (Choi *et al.*, 2006). The details of their involvement have yet to be clarified, however. As a step towards addressing this issue, Park *et al.* (2013a, b, 2012) have developed a method, the Greenhouse Gases Emission Presumption (GEP) method, which is capable of evaluating methane emissions over a wide area. By applying the GEP method, Park *et al.* (2013a, b, 2012) have conducted analyses to identify sources of methane emissions, and estimated the increase in methane concentration using WDCGG data from the Ryori, Yonagunijima, Gosan and Neuglobsow observation points.

In this paper, the GEP method is employed to analyze WDCGG data and evaluate the source of the increase in methane concentration at Anmyeon-do. Specifically, the estimated difference in methane emissions is calculated and then compared with the estimated methane emissions according to land use and land cover. In this way, this study attempts to verify the hypothesis that rice paddies are the major source of methane emissions at Anmyeon-do.

2. MATERIALS AND METHODS

2.1 Research Site

Data for this research was collected at the Anmyeon-do observation point. Observations for the WMO's Global Atmosphere Watch (GAW) program are performed at this site and hourly average data are available for a number of consecutive years. Methane concentrations have been monitored continuously since 1999. The observatory is situated at 36° 32′N 126° 19′E, at an altitude of 47 meters (Fig. 2). The observation tower is 40 meters tall. Samples are collected and analyzed every 30 minutes by gas chromatography (WDCGG, 2013).

2.2 Data

Hourly average values for methane concentration, wind speed, wind direction, relative humidity and air

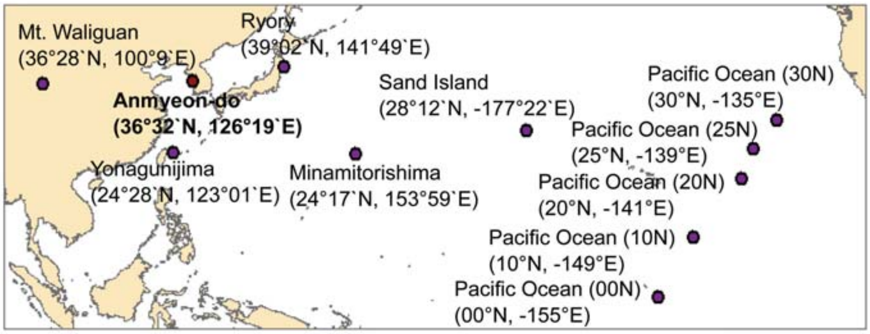

Fig. 2. Observation points in WDCGG data used for analysis.

temperature at Anmyeon-do were obtained from the WDCGG website (WDCGG, 2013). Meteorological data for the Anmyeon-do observatory were acquired for the years 2003 to 2009, but the 2008 methane concentration data were considered to be highly unusual and therefore were not used in this study. Compared to the monthly average methane concentrations for August and September of other years, the methane concentrations were significantly lower in 2008 (76 ppb and 53 ppb) (WDCGG, 2013; Korea Meteorological Administration, 2011). To compare the methane concentration data and seasonal fluctuations at Anmyeon-do with the surrounding areas, WDCGG data from the Ryori, Yonagunijima and Minamitorishima observation points (Fig. 2) were also utilized. In addition, WDCGG data from Mt. Waliguan, Sand Island and Pacific Ocean sites were obtained for a comparison of methane concentrations. Monthly averages were used since hourly averages for the latter three sites were not available.

2. 3 Analysis

2. 3. 1 Fluctuations in Methane Concentration Due to Climatic Factors and Evaluation of Methane Emission Sources

In order to determine the existence of methane emission sources and estimate fluctuations in methane concentration due to climatic factors, multiple regression analysis was performed using time-averaged value data for the period between 2003 and 2009 (except 2008). Methane concentration was used as the objective variable, and wind speed, wind direction, relative humidity and air temperature were used as the explanatory variables. The analysis was performed for three time periods: the entire year; the rice growing season (from May to October); and the paddy fallow season (from November to April). Since wind speed and wind direction were categorical data, those were regarded as dummy variables for converting qualitative variables. Wind speeds of 1.6-3.3 m/s and wind directions between 0.1° and 46° were decided to be standard variables, and were compared to the others. As this study was designed to analyze the factors governing fluctuations in methane concentration, we examined the effects of each explanatory variable on methane concentration rather than the degree of fit of the multiple regression model.

2. 3. 2 Methane Emissions and Seasonal Fluctuation Calculated from the Difference in Concentration Using the GEP Method

The GEP method employs the following equation (Park *et al.*, 2012).

Estimated difference in concentration induced
by methane emissions
=Observed concentration−Existing concentration (1)

Here, the estimated difference in concentration due to methane emissions refers to the difference in concentration caused by the emission of additional methane, which is based on the existing methane concentration contained in the atmosphere. The observed concentration is considered to be the monthly average of methane concentration for a wind speed of 0.2 m/s or less. According to the Beaufort wind speed scale used by the Japan Meteorological Agency (2013b), wind speeds of 0.0-0.2 m/s correspond to scale 0, where smoke rises straight up; wind speeds of 0.3-1.5 m/s correspond to scale 1; wind speeds of 1.6-3.3 m/s correspond to scale 2; and so on up to scale 12. In order to eliminate the influence of advection, data for wind speeds of 0.3 m/s and above were eliminated from the analysis. Furthermore, the existing concentration is considered to be the concentration of methane already present in the atmosphere at the observation point. In order to compensate for differences in methane concentration by location, these data were obtained at the same latitude. Additionally, the data utilized were from areas in which the impact of methane emission sources and advection were as low as possible.

In order to evaluate the influence of advection on methane concentration, an analysis was conducted on data for wind speeds between 0.3 and 1.5 m/s, which is the wind speed range at which the increase in methane concentration is likely to be greatest. The results are shown in Table 1, which is discussed in section 3.2. The equation employed in the analysis is as follows:

$$X_i = W_i - E_i \tag{2}$$

$$Y = \left(\sum_{i=1}^{12} X_i\right)/12 \tag{3}$$

where i is the month (January to December), W_i is the monthly average methane emission for a wind speed below a certain value, E_i is the monthly average methane emission with the existing methane concentration, X_i is the monthly average difference in methane concentration, and Y is the annual average difference in methane concentration.

As can be seen in Fig. 5, multiple emission sources were present at the sites used in this study, and the results could be influenced by the choice of a representative area. To resolve this problem, based on the fact that the WDCGG data provides hourly average values, an area with a radius of 5400 m was calculated as the representative area for data with wind speeds between 0.3 and 1.5 m/s (1.5 m/s × 60 s/min × 60 min = 5400 m). Additionally, an area with a radius of 720 m was calculated as the representative area for data with wind speeds between 0 and 0.2 m/s (0.2 m/s × 60 s/min × 60 min = 720 m).

2.3.3 Emission Quantities and Contribution Ratios of Methane Emission Sources

In order to confirm the consistency of the results obtained with the GEP method, the methane emission quantities and contribution ratios of methane emission sources were estimated and analyzed according to land use and land cover within areas with radii of 720 and 5400 m. Based on assumptions derived from previous studies on rice paddies, sea, field crops, forest, etc., methane emission quantities and contribution ratios were calculated separately for the maximum and minimum values.

3. RESULTS AND DISCUSSION

3.1 Seasonal Fluctuations in Methane Concentration and Annual Average Concentration

Methane concentrations tended to be higher in winter and lower in summer in all three surrounding areas, but not at Anmyeon-do (Fig. 3). This seasonal distribution was caused by the OH radical concentration

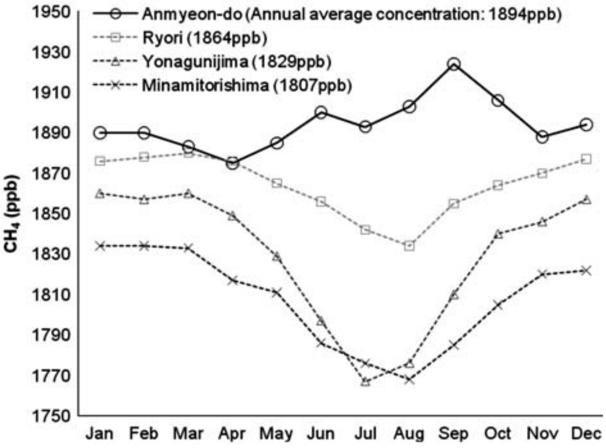

Fig. 3. Seasonal fluctuation in methane concentration. Monthly average concentration between 2003 and 2009 (except 2008).

Table 1. Results of multiple regression analysis of wind speed, relative humidity and air temperature with respect to methane concentration.

	Jan-Dec (n=19,623)	May-Oct (n=9,603)	Nov-Apr (n=10,020)
Wind speed (0.0-0.2 m/s)	0.02*	0.00	0.04***
Wind speed (0.3-1.5 m/s)	0.04***	0.02*	0.06***
Wind speed (1.6-3.3 m/s)	--	--	--
Wind speed (3.4-5.4 m/s)	−0.04***	−0.04**	−0.04***
Wind speed (5.5-10.7 m/s)	−0.20***	−0.24***	−0.15***
Wind speed (10.8-19.3 m/s)	−0.11***	−0.14***	−0.11***
Relative humidity	3.25**	0.03**	0.03**
Air temperature	0.13***	0.09***	0.03**
R^2	0.07***	0.08***	0.04***

Numbers are the standard partial regression coefficient (*$p<0.05$, **$p<0.01$, ***$p<0.001$). Hourly average values between 2003 and 2009 (except 2008), where "--" is a standard variable. Wind speed is regarded as a dummy variable.

being high in summer and low in winter due to variations in ultraviolet intensity and water vapor concentration (Japan Meteorological Agency, 2013a; IPCC, 2007). As can be seen in Fig. 4, the same pattern of seasonal fluctuation was observed for Sand Island and the Pacific Ocean. The annual average concentration at Anmyeon-do was 30 ppb higher than that at Ryori, which is located at a higher latitude. At Anmyeon-do, the absence of the low summer concentration pattern and the high annual average concentration suggested the presence of a large emission source in the vicinity of the observation point.

3. 2 Fluctuations in Methane Concentration Due to Climatic Factors and Methane Emission Sources

Results of the multiple regression analysis of wind speed, relative humidity and air temperature at Anmyeon-do are shown in Table 1. The coefficient of determination is low in all three periods (whole year, rice growing season and fallow season). Due to the large amount of data, the p value of the coefficient of determination is significant even though the coefficient of determination is low. The standard partial regression coefficient is positive at wind speeds of 1.5 m/s and lower, and the coefficient is negative for wind speeds of 3.4 m/s and higher. Accordingly, it is speculated that the increase in methane concentration was due to the influence of the wind speeds of 1.5 m/s and lower. As the standard partial regression coefficients of relative humidity and air temperature are positive and significant, the seasonal fluctuation at Anmyeon-do clearly differs from that at Ryori, Yonagunijima and Minamitorishima (Fig. 3).

The non-standard partial regression coefficient of wind direction between 46° and 91° is positive and significant (Table 2). This suggests that the increase in methane concentration originates not from China to the west, but mainly from land to the east.

3. 3 Emission Quantities and Contribution Ratios

3. 3. 1 Calculation of Existing Concentrations

Mt. Waliguan (latitude 36° 28′N and altitude 3810 m), located at almost the same latitude as Anmyeon-do (Fig. 2), is the only point on the Chinese mainland for which long-term monthly average methane concentration data can be obtained (Fig. 4). Mt. Waliguan (36N), however, shows a lower winter methane concentration than that obtained for the Pacific Ocean (25N) site. The seasonal fluctuations and summer concentrations also differ from those obtained at the other sites. The patterns observed for the mountain are most likely due to altitude and other local variables, making this site of limited value for the calculation of existing methane concentrations. This research thus adopts a different strategy for the calculation of existing methane concentrations. As can be seen in Fig. 4, methane concentrations at the 5 points shown in Fig. 2, with the exception of the Pacific Ocean site located

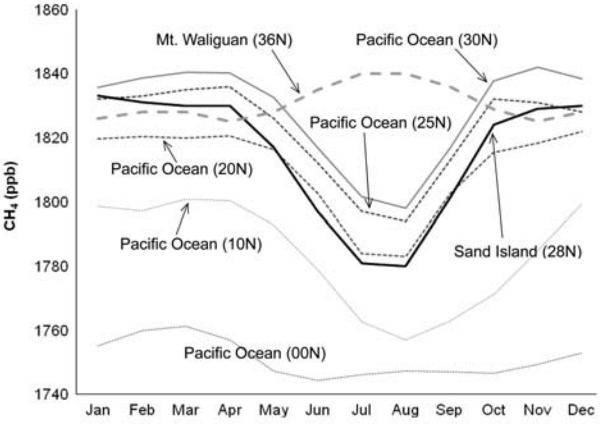

Fig. 4. Methane concentration by latitude. Monthly average between 1988 and 2007.

Table 2. Results of multiple regression analysis of wind direction with respect to methane concentration.

Wind direction	Jan-Dec (n=19,603)	May-Oct (n=9,560)	Nov-Apr (n=10,043)
Constant	1933.48***	1969.88***	1907.07***
0.1-<46°	--	--	--
46-<91°	26.91***	23.96***	18.88***
91-<136°	12.58***	−10.94*	23.67***
136-<181°	−46.64***	−85.55***	−15.65***
181-<226°	−53.23***	−94.13***	−20.48***
226-<271°	−34.49***	−68.21***	−13.06***
271-<316°	−36.33***	−65.94***	−14.81***
316-360°	−36.85***	−48.44***	−19.46***
R^2	0.09***	0.14***	0.07***

Numbers are the non-standard partial regression coefficient (*$p < 0.05$, ***$p < 0.001$). Hourly average values between 2003 and 2009 (except 2008), where "--" is a standard variable. Wind direction is regarded as a dummy variable.

Table 3. Comparison of classification results and ground truth.

Classification result	Ground truth							Total number of sample points and percentage
	Forest	Rice paddy	Field crop	Pond	Salt marsh	Urban region-1	Urban region-2	
Forest	12 (100)							12 (100)
Rice paddy		19 (95)	1 (5)					20 (100)
Field crop			13 (87)				2 (13)	15 (100)
Pond				3 (100)				3 (100)
Salt marsh				2 (33)	4 (67)			6 (100)
Urban region-1						7 (100)		7 (100)
Urban region-2							6 (100)	6 (100)

Numbers are the number of sample points and percentages. The urban region-1 indicates urban, and the urban region-2 indicates open space.

at the equator, are high in winter and low in summer. Although the methane concentration is highest at latitude 30°, it is also higher at latitude 25° than at the Sand Island observation point located at latitude 28°. Additionally, the Sand Island site is not likely to be substantially influenced by advection form the continent as it is situated in the middle of the Pacific Ocean. Consequently the Sand Island data was adopted as the baseline for calculating the existing methane concentrations in this study. However, as this site is located about 8° south of Anmyeon-do, there was a need to adjust the data. This was accomplished with the following procedure. The difference in annual average concentration is 33 ppb between the Pacific Ocean (00N) and Pacific Ocean (10N), 26 ppb between the Pacific Ocean (10N) and Pacific Ocean (20N), 18 ppb between the Pacific Ocean (20N) and Pacific Ocean (30N) and 6 ppb between the Pacific Ocean (25N) and Pacific Ocean (30N). Extrapolating from these data, 9 ppb were added to the figures for the monthly average methane concentration at Sand Island to arrive at the adjusted existing concentration values for Anmyeon-do.

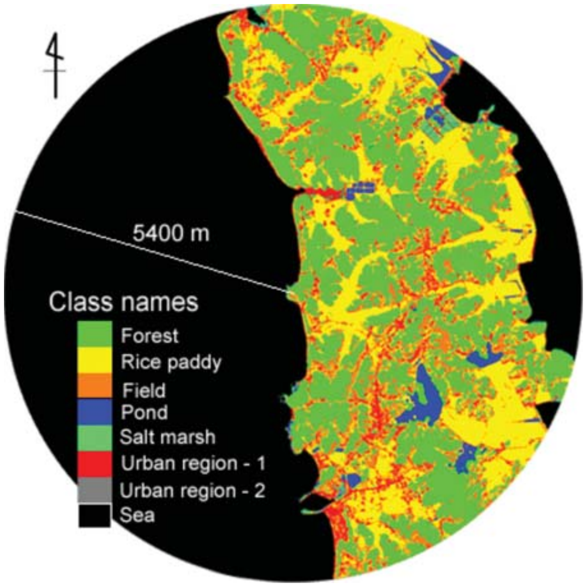

Fig. 5. Land use and land cover within a radius of 5400 m from the observation point.

3. 3. 2 Land Use and Land Cover Within a Radius of 5400 m from the Observation Point

Fig. 5 shows the land use and land cover within the study area: 53.3% of the area is sea, 18.9% forest, 14.7% rice paddies, 7.6% field crops, 3.9% urban regions, 1.2% ponds and 0.4% salt marshes. Fig. 5 is based on data received from the Advanced Land Observing Satellite (ALOS) "Daichi" (Japan). The ALOS generates images over four bands, from visible to infrared, with a ground resolution of 10 meters (Japan Aerospace Exploration Agency, 2013). The classification of land use and land cover was implemented using the Interactive Self-Organizing Data Analysis Technique A (ISODATA) (Ball and Hall, 1967). In order to verify the classification results, the sample points were selected at random with regard to the seven categories, and then those were compared with the results of field surveys, Google Earth, etc. (Table 3). Field surveys were conducted in May and August 2011. The resulting accuracies were as follows: forest, 100%; rice paddy, 95%; field crop, 87%; pond, 100%; urban region-1, 100%; and urban region-2, 100%. Since it was difficult to distinguish salt marsh form pond, the accuracy of salt marsh was low (67%). Urban region-1 indicates an urban area, and urban region-2 indicates open space. ALOS data acquired on February 11, 2007, October 1, 2008, May 19, 2009, July 7, 2010 and November 22, 2010 were used for the classification.

3. 3. 3 Methane Emissions and Seasonal Fluctuations Due to Concentration Differences

The seasonal fluctuations in monthly average methane concentrations with wind speeds of 0.0-0.2 m/s

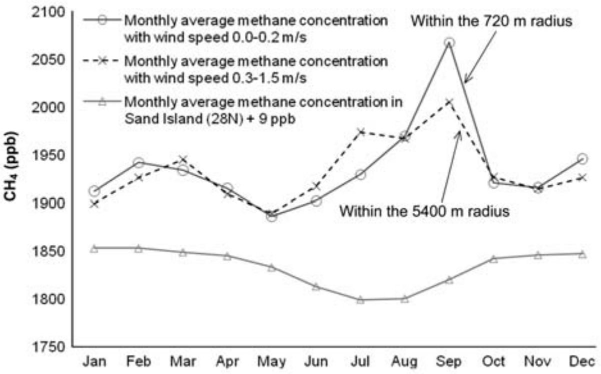

Fig. 6. Seasonal fluctuation in methane concentration. Monthly average between 2003 and 2009 (except 2008).

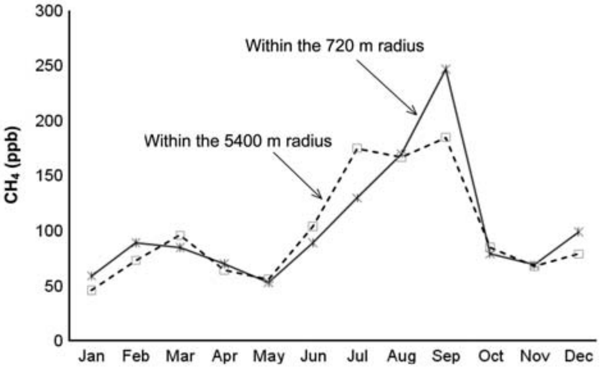

Fig. 7. Seasonal fluctuation of difference in methane concentration. Monthly average between 2003 and 2009 (except 2008).

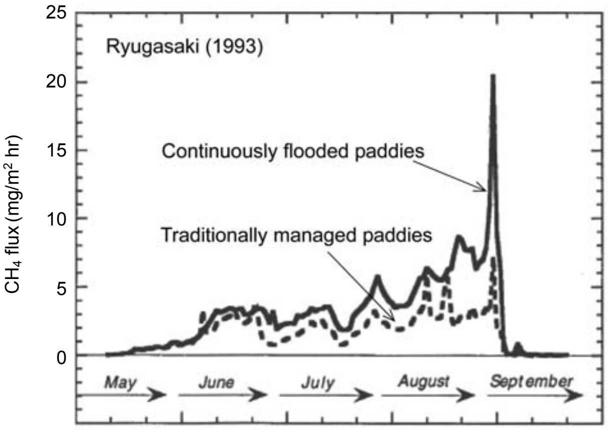

Fig. 8. Impact of paddy water management on methane emission (reproduced from Yagi (1997)).

and 0.3-1.5 m/s at Anmyeon-do, and the same monthly averages at Sand Island+9 ppb, are shown in Fig. 6. The seasonal fluctuations in concentration difference within the areas with radii of 720 and 5400 m, as calculated from Fig. 6, are shown in Fig. 7. These values were obtained by applying Eq. (2) to the existing concentration (monthly average methane concentration at Sand Island+9 ppb) and observed concentration (monthly average methane concentration for wind speeds of 0.0-0.2 m/s, n=317). The annual average difference in methane concentration, estimated using Eq. (3), was 103 ppb, which amounts to 5.4% of the annual average methane concentration of 1894 ppb. Similarly, the monthly average difference in methane concentration for wind speeds of 0.3-1.5 m/s (n=2,683) was applied to Eq. (2) to calculate the annual average difference in methane concentration within a radius of 5400 m. This figure was estimated to be 100 ppb, or 5.3% of the annual methane concentration average of 1894 ppb (Eq. (3)). Both values showed a rapid increase during the rice-growing period (May-October) at Anmyeon-do. The rice is transplanted into the paddies in mid-May to early June, and the grain heads appear in late July to mid-August (Taean-gun Agricultural Development & Technology Center, 2011). Fig. 8, based on experimental results, shows a comparison of methane emissions of traditionally managed paddies, which are drained several times during the growing season, and continuously flooded paddies that are inundated throughout the entire season. As can be seen, traditionally managed paddies show a sharp drop in methane emission levels when they are drained (Yagi, 1997). The Rural Development Administration (Republic of Korea) (2010) estimated that the area covered by continuously flooded paddies accounted for 95% of rice paddies. The experimental results for continuously inundated paddies, as seen in Fig. 8, closely resembles the emission patterns shown in Fig. 7, indicating that the GEP method is an effective tool for calculating seasonal fluctuations in methane concentrations.

3. 3. 4 Quantity of Methane Emissions and Contribution Ratios According to Land Use and Land Cover

Table 4 shows the amount of methane emissions and the contribution ratios of emission sources according to land use and land cover. Wind speed was assumed to be 0.0-0.2 m/s within a radius of 720 m and 0.0-1.5 m/s within a radius of 5400 m. The values showed a positive standard partial regression coefficient (Table 1). As can be seen, rice paddies were estimated to account for between 68 and 90% of the methane emissions. Based on land use and land cover within a radius of 5400 m, other methane sources were found to be a pond, salt marsh, pig farm and terminal sewerage treatment plant. With the exception of the pig farm, however, these all accounted for less than 1% of the

Table 4. Assumption for methane emission quantities and contribution ratios of methane emission sources according to land use and land cover within the areas with radii of 720 and 5400 m.

Classification	Area		Assumption of quantity in methane emission and contribution ratio				Calculation method
			Minimum		Maximum		
	m^2	%	tCH_4 year^{-1}	%	tCH_4 year^{-1}	%	
Within the 720 m radius							
Rice paddies	146,500	9.0	4.01	88.9	4.01	68.1	(Continuously flooded paddies: 146,500 m^2 × 95% × 0.000028 [a]) +(Traditionally managed paddies: 146,500 m^2 × 5% × 0.000016 [a])
Sea	878,999	54.0	0.01	0.3	1.32	22.3	Minimum: 878,999 m^2 × 0.0135 g [b]
Forest	390,666	24.0	0.48	10.7	0.48	8.2	Maximum: 878,999 m^2 × 1.5 g [c] 0.1074 tCH_4 year^{-1} [d] × 4.5 [e]
Field crops	138,361	8.5	0	0	0.07	1.3	Maximum: 0.0166 tCH_4 year^{-1} [d] × 4.5 [e]
Urban regions (cars)	73,250	4.5	0.01	0.1	0.01	0.1	38 cars [f] × 14,929 km [g] × 0.000010 kg/km [h]
Total	1,627,776	100	4.51	100	5.90	100	
Within the 5400 m radius							
Rice paddies	13,459,672	14.7	368.80	89.6	368.80	69.0	(Continuously flooded paddies: 13,459,672 m^2 × 95% × 0.000028 [a]) +(Traditionally managed paddies: 13,459,672 m^2 × 5% × 0.000016 [a])
Sea	48,802,759	53.3	0.66	0.1	73.20	13.7	Minimum: 48,802,759 m^2 × 0.0135 g [b] Maximum: 48,802,759 m^2 × 1,500 g [c]
Forest	17,305,293	18.9	34.03	8.3	34.03	6.3	4.8609 tCH_4 year^{-1} [d] × 7 [e]
Field crops	6,958,743	7.6	0	0	5.95	1.1	Maximum: 0.8497 tCH_4 year^{-1} [d] × 7 [e]
Ponds	1,098,749	1.2	1.24	0.3	1.24	0.2	1,098,749 m^2 × 365 days × 3.1 mg/day [c]
Urban regions (cars)	3,570,934	3.9	0.32	0.1	0.32	0.1	2,167 cars [f] × 14,929 km [g] × 0.000010 kg/km [h]
Salt marshes	36,250	0.4	0	0	0	0	
Total	91,562,400	100					
Pig farm	3,000 pigs (700 sows, 2,300 pigs)		6.06	1.5	50.76	9.5	Minimum: (Emission from rumination: 3,000 pigs × 0.0011 [h]) +(Emission from feces and urine: 3,000 pigs × 0.00092 [h]) Maximum: (Emission from rumination: 3,000 pigs × 0.0011 [a]) +(Emission from feces: 700 sows × 0.241 [a] × 0.087 [a]) +(Emission from feces: 2,300 pigs × 0.153 [a] × 0.087 [a]) +(Emission from urine: 700 sows × 0.0128 [a] × 0.087 [a]) +(Emission from urine: 2,300 pigs × 0.00694 [a] × 0.087 [a])
Terminal sewerage treatment plant	1,600 m^3/day		0.51	0.1	0.51	0.1	1,600 m^3/day × 365 days × 0.00000088 [h]
Total			411.62	100	534.81	100	

[a] Data from Ministry of Agriculture, Forestry and Fisheries (Japan) (2013). [b] Data from Kiene (1991). [c] Data form Nakamura *et al.* (1994).

[d] Emission concentration was cited with permission from Park *et al.* (2012), applied to the following equation to convert it into emission quantity.

$Y_i = (X_i[\text{ppb}] \times M[\text{g/mol}]) \div (22.4[\text{L/mol}] \times K_i)$, $K_i = (273 + T_i) \div 273$, $Y_j = (\sum_{i=1}^{n} Y_i) \div n$, $Y = Y_j \times 24$ hours × 365 days, $Z = Y \times V$, Y_i: μg/m^3, X_i: Hourly average concentration, M: Molecular weight of CH_4 (=16.04), 22.4: Molar volume of ideal gas at 0°C and 1 atmospheric pressure, K_i: Temperature coefficient, T_i: Temperature of the gas, Y_j: Hourly average emission quantity per m^3, Y: Annual average emission quantity per m^3, V: Volume assumed at a height of 1 m, Z: Annual emission quantity.

[e] Weighted value to compensate concentration difference between cumulative emission quantity and concentration from Eq. (1), including loss by advection. These values were obtained by comparing with three values ($Y \fallingdotseq 657{,}525$ μg/m^3 year^{-1}, within a radius of 720 m: 1tCH_4 year$^{-1} \fallingdotseq Y \times 1{,}627{,}776$ m^2, within a radius of 5400 m: 60 tCH_4 year$^{-1} \fallingdotseq Y \times 91{,}562{,}400$ m^2) and the above minimum emission quantity, except forests.

[f] Calculated to assume following, total number of households in Anmyeon-eup: 4,610 [i] × Within the 720 m radius: 0.82%, within the 5400 m radius: 46.99% (Area ratio within target radius against total area in Anmyeon-eup: 90.99 km^2 [i]) × 1 car per household.

[g] Data from Korea Transportation Safety Authority (2009). [h] Data from Ministry of Internal Affairs and Communications (Japan) (2010). [i] Data from Taean-gun (2013).

methane emissions. In total, methane emissions from human activity are estimated to be between 80 and 91% within a radius of 5400 m. In addition to the abovementioned sources, methane emissions from boilers and other domestic fuel consumption should also be considered (Ministry of Internal Affairs and Communications (Japan), 2010). Although the amount of fuel consumption could not be confirmed, the vehicle fuel consumption of urban regions was estimated to be less than 1%, and these sources are thus unlikely to exert a significant impact on the results shown in Table 4.

3. 3. 5 Some Issues Involving the Estimation Results for Methane Emission Quantities and Contribution Ratios

The differences in maximum and minimum estimated emissions seen in Table 4 are due to methane emissions from the sea, field crops and pig farm. The pig farm emissions could be excluded by applying the emission coefficient. With regard to the influence of the sea, in recent years large-scale reclamation work has been undertaken in coastal areas in Anmyeon-do (1980-1995, total landfill area of 15,409 ha), forming a semi-enclosed bay with numerous river inflows. The maximum quantity of methane emission was thus set at the value of Tokyo Bay, which is a similar semi-enclosed inner bay. This maximum value is almost 110 times higher than the selected minimum quantity of methane emission from the ocean. As there are no data available for methane emissions in the sea around Anmyeon-do, the reliability of the value for sea emissions could not be verified directly.

The IPCC (2007) has reported that there are no methane emissions from field crops, but there are other experimental findings that confirm methane is emitted by certain plants and from sugarcane fields (Park *et al.*, 2012; Keppler *et al.*, 2006). The amount of methane emissions caused by burning the residue of various crop species was calculated in the Greenhouse Gases Emission Estimation and Reporting Manual (Ministry of Agriculture, Forestry and Fisheries (Japan), 2013), but there are no data for emissions from the leaves of field crops cultivated at Anmyeon-do, such as red peppers, garlic and Chinese cabbage.

In addition, as yet there are no data on the seasonal fluctuations of methane emissions from ruminant livestock (Nouchi, 2006; Shibata *et al.*, 1993). This factor was thus assumed to have negligible impact on the results.

4. CONCLUSIONS

The main sources of increased methane concentrations at Anmyeon-do were evaluated by utilizing the GEP method combined with WDCGG data for the period between 2003 and 2009 (except 2008). The results showed an increase of 100-103 ppb in average annual methane concentration, which amounts to 5.3-5.4% of the annual average methane concentration of 1894 ppb, and indicated that this increase originated mainly in rice paddies near the observation point. The seasonal fluctuation (May to October) in concentration estimated by this method was compared with experimental data from rice paddies. The close match obtained in this comparison shows that the GEP method is effective. The differences in methane concentrations were also analyzed according to land use and land cover, with the results showing that although rice paddies account for only 14.7% of the surveyed area, they are estimated to account for between 68 and 90% of the total increase in methane concentration. These results confirm that rice paddies are the main source of the increase in methane concentration observed at Anmyeon-do.

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