Tellus SERIES B CHEMICAL AND PHYSICAL METEOROLOGY
PUBLISHED BY THE INTERNATIONAL METEOROLOGICAL INSTITUTE IN STOCKHOLM

Tellus (2010)

TELLUS

Continuous measurements of methane from a tower network over Siberia

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(Manuscript received 28 December 2009; in final form 6 July 2010)

ABSTRACT

We have been conducting continuous measurements of Methane (CH_4) concentration from an expanding network of towers (JR-STATION: Japan–Russia Siberian Tall Tower Inland Observation Network) located in taiga, steppe and wetland biomes of Siberia since 2004. High daytime means (>2000 ppb) observed simultaneously at several towers during winter, together with in situ weather data and NCEP/NCAR reanalysis data, indicate that high pressure systems caused CH_4 accumulation at subcontinental scale due to the widespread formation of an inversion layer. Daytime means sometimes exceeded 2000 ppb, particularly in the summer of 2007 when temperature and precipitation rates were anomalously high over West Siberia, which implies that CH_4 emission from wetlands were exceptionally high in 2007. Many hot spots detected by MODIS in the summer of 2007 illustrate that the contribution of biomass burning also cannot be neglected. Daytime mean CH_4 concentrations from the Siberian tower sites were generally higher than CH_4 values reported at NOAA coastal sites in the same latitudinal zone, and the difference in concentrations between two sets of sites was reproduced with a coupled Eulerian–Lagrangian transport model. Simulations of emissions from different CH_4 sources suggested that the major contributor to variation switched from wetlands during summer to fossil fuel during winter.

1. Introduction

Atmospheric CH_4 is the second most important anthropogenic greenhouse gas after CO_2 because of its influence on the Earth's radiation budget by infrared absorption and photochemical reactions in the atmosphere. Its concentration in the troposphere is principally determined by a balance between surface emission and destruction by hydroxyl (OH) radicals. Emission sources comprise anthropogenic activity—fossil fuel combustion, rice agriculture, livestock, landfill and waste treatment, and some biomass burning—and natural sources such as wetlands, termites and the ocean (IPCC, 2007). Imbalances induced by anthropogenic emissions after the industrial revolution produced a persistent increase in global CH_4 content followed by a period of relatively stable concentration between 1999 and 2006 (Dlugokencky et al., 2003; Rigby et al., 2008). The period of stable concentration was partly attributed to a decrease in CH_4 emissions of ~10 Tg from the region north of 50°N (the former Soviet Union) from 1990 to 1995 (Dlugokencky et al., 2003). Further research using an inverse model of atmospheric transport and chemistry attributed the stabilization of atmospheric CH_4 concentration to a steady decrease in anthropogenic CH_4 emissions between 1990 and 1999 (Bousquet et al., 2006). According to their study, anthropogenic CH_4 emissions increased after 1999, but CH_4 concentration in the atmosphere remained relatively constant because of a coincidental decrease in wetland emissions for several years after 1999. Rigby et al. (2008) reported that CH_4 concentration began to rise again at the beginning of 2007; their study was based on data from the Advanced Global Atmospheric Gases Experiment (AGAGE) and the Australian Commonwealth Scientific and Industrial Research Organisation (CSIRO). The AGAGE data of methyl chloroform indicated a small drop in OH concentration between 2006 and 2007, but the change was not statistically significant at a global level. They speculated that Siberian wetlands were the most likely source because of a striking climatic anomaly (~4 °C compared

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DOI: 10.1111/j.1600-0889.2010.00494.x

to 1961–1990) in annual mean temperature over Siberia (National Climatic Data Center, 2008). Dlugokencky et al. (2009a) examined observational data obtained from the background sites of National Oceanic and Atmospheric Administration (NOAA) and reported that the increase in atmospheric CH_4 in early 2007 persisted until 2008. They suggested that very warm temperatures at polar northern latitudes during 2007 likely enhanced emissions from northern wetlands. The $\delta^{13}C$ values in CH_4 at their northern-most site were lighter than normal in the summer of 2007, which is consistent with a wetland source.

Potential changes in natural CH_4 emissions from wetlands (Gedney et al., 2004), thawing lakes (Walter et al., 2006) and melting permafrost (Zimov et al., 2006; Khvorostyanov et al., 2008) can be induced by regional changes in temperature or precipitation patterns. This means that CH_4 measurements over Siberia are particularly crucial to estimating global CH_4 emissions; Siberia is estimated to contain approximately 132 million ha of wetlands (Sohngen et al., 2005). Studies of CH_4 behaviour and attempts to identify sources of variation have been conducted over Siberia by means of observations from aircraft (Sugawara et al., 1996; Tohjima et al., 1996, 1997; Nakazawa et al., 1997; Yamada et al., 2005) and the Trans-Siberian Railway (Bergamaschi et al., 1998; Tarasova et al., 2006). While most similar studies were limited to shorter periods or a specific season, Levin et al. (2002) and Lloyd et al. (2002) reported seasonal variation in CH_4 in 1998–2000 derived from 2 to 4-weekly vertically stratified aircraft sampling over Zotino, Siberia (60°45′N, 89°23′E). As expected, their flask samples up to 3000 m altitude displayed seasonality: maximum concentrations during autumn and winter and a minimum in summer. However, occasional high concentrations led to annual mean values higher than those of other northern European sites (Levin et al., 2002). They suggested that the CH_4 accumulation was caused by emissions from wetlands and natural gas production. Recently, Kozlova et al. (2008) presented the first results of continuous in situ measurement of CH_4 concentration at the Zotino Tall Tower Observatory (ZOTTO) in the boreal forest of central Siberia (Fig. 1). They showed CH_4 temporal variation at five heights between 4 and 300 m above ground level in November and December 2006. They found an increase in CH_4 concentration at the lower heights during periods of very low temperatures (−30 °C).

In 2002, National Institute for Environmental Studies (NIES) began a cooperative project of continuous measurement of CO_2 with a tower at Berezorechka (BRZ) in West Siberia. The number of tower sites was progressively expanded and CH_4 measurement added, and the Japan–Russia Siberian Tall Tower Inland Observation Network (JR-STATION) now consists of eight towers located in West Siberia and one tower in Yakutsk (YAK) in East Siberia (Fig. 1). Here we present a selection of the data collected from inception up until spring 2009. These data represent the most comprehensive high-frequency in situ CH_4 measurements from Siberia reported so far. We have analysed the data with the aim of describing spatial and temporal variations in CH_4 concentration. We also attempt to identify the sources responsible for variation in CH_4 concentration in the forest, steppe and wetland biomes of Siberia. Discontinuous observations such as those from flask sampling do not always show representative data of the sites because the distribution of CH_4 sources on the land surface is so heterogeneous. The advantage of tower measurements is that they continuously sample a well-mixed part of the atmosphere (mixed layer), thus allowing us to observe long-term changes in atmospheric composition.

2. Method

2.1. Site description

The towers were originally constructed and continue to be used for radio relay communication. We installed a freight container equipped with gas analysers and a data logger at the base of each tower. Atmospheric air was sampled at two levels of all towers except BRZ, which was equipped for sampling at four levels. Their location, inlet heights and other information are given in Table 1. The BRZ tower is located in the middle of a boreal forest (taiga). Although there is a small village near the tower, its population is in the order of dozens and there is no large-scale agriculture or industry within the vicinity. The

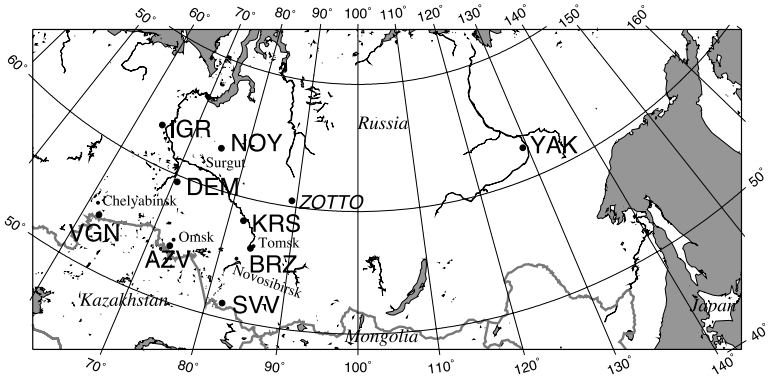

Fig. 1. Japan–Russia Siberian Tall Tower Inland Observation Network (JR-STATION) used for continuous measurement of the greenhouse gases CO_2 and CH_4. The three-letter codes are the abbreviated station names shown in Table 1. ZOTTO is the Zotino Tall Tower Observatory (Kozlova et al., 2008).

Table 1. The main features of each tower in a network of tall towers (JR-STATION) used for continuous long-term atmospheric CO_2 and CH_4 measurement over Siberia

Identifying code	Location	Latitude	Longitude	Air inlet heights (m)	Elevation at tower base (m a.s.l)[a]
BRZ	Berezorechka	56°08′56″	84°19′58″	5, 20, 40, 80	150
KRS	Karasevoe	58°14′44″	82°25′28″	35, 67	50
IGR	Igrim	63°11′25″	64°24′56″	24, 47	25
NOY	Noyabrsk	63°25′45″	75°46′48″	21, 43	100
DEM	Demyanskoe	59°47′	70°52′	45, 63	75
SVV	Savvushka	51°19′30″	82°07′40″	27, 52	400
AZV	Azovo	54°42′18″	73°01′45″	29, 50	100
VGN	Vaganovo	54°29′50″	62°19′29″	42, 85	200
YAK	Yakutsk	62°50′	129°21′	11, 70	130

[a]Approximate estimates from topographic maps.

closest large city is Tomsk (60 km northeast) with a population of approximately 0.5 million people. The KRS tower is on the shore of a 5-km diameter marshy lake in the middle of the taiga. The DEM and NOY towers are also in the middle of forest and surrounded by extensive wetlands. The IGR tower is located in the town of Igrim, which has a population of about 10 000. Igrim is located next to the Ob River and is surrounded by extensive wetlands. The SVV tower is on a hill 1 km south of a small village in the steppe region. The AZV and VGN towers are located adjacent a small town and small village in the steppe region, respectively. The closest large city to the AZV tower is Omsk (30 km northeast), which has a population of about 1 million people. For the VGN tower, the closest city is Chelyabinsk (100 km northwest), which has a population of over 1 million.

2.2. *Measurement system*

Each sample inlet was positioned approximately 3 m away from the tower at the end of an extension arm. Atmospheric air was delivered via a decabon tube by a diaphragm pump (model N86KNE, KNF, Germany) into the freight container with insulators to reduce temperature variation and dried with (1) adiabatic expansion in a glass water trap, (2) a semi-permeable membrane dryer (model PD-625–24SS, Permapure, USA) and (3) magnesium perchlorate. The dehumidified air was then introduced into a non-dispersive infrared analyser (model LI-820, LI-COR, USA; a model LI-7000 was used until September 2008 at BRZ) and a CH_4 semiconductor sensor at a constant flow rate of 35 cm^3 min^{-1} using a mass flow controller (model SEC-E40, STEC, Japan). The CH_4 semiconductor sensor based on a tin dioxide natural gas leak detector was developed by Suto and Inoue (2010) to measure atmospheric CH_4 concentration in areas lacking sufficient infrastructure to sustain a conventional measurement system, such as a large power source, carrier gas supply and temperature-stabilized laboratory. The sensor has been verified against a gas chromatograph equipped with a flame ionization detector (Suto and Inoue, 2010).

Three standard gases were prepared from pure CO_2 and CH_4 diluted with purified air, and their concentrations were determined against the NIES 95 CO_2 scale (Machida et al., 2009) and NIES 94 CH_4 scale. Each scale was established by a series of standard gases prepared by the gravimetric method. The NIES 94 CH_4 scale is higher than the NOAA 04 scale by 3.5–4.6 ppb in the range between 1750 and 1840 ppb (Zhou et al., 2009). We used in situ compressed air as a reference gas to reduce the consumption of the three standard gases because this system operates continuously and it is difficult to replace the standard gas cylinders in Siberia. To prepare the reference gas, air from the highest inlet was compressed into a 0.048-m^3 aluminum cylinder for approximately 5 h by a pump (model LOA-P103-NO, GAST, USA) to approximately 0.5 MPa, after first being passed through both a semi-permeable membrane dryer (model SWF-M06–400, AGC, Japan) and magnesium perchlorate. Two cylinders were prepared for the reference gas; one for compression and preservation and the other for continuous measurement. The cylinders were automatically exchanged when the inner pressure decreased to 0.1 MPa, which normally took about 1 week.

The air-sampling flow path was rotated every 20 min; that is, the high inlet was sampled at hh:00, the low inlet at hh:20 and the reference gas at hh:40. For each 20 min sampling period, air was pumped continuously through the sample line, stainless tube containing chemical desiccant, NDIR cell and CH_4 sensor for 17 min; for the last 3 min, the data produced by the sensors were averaged and taken as the representative data for the applicable 1 h period. Twice a day, the three standard gases were analysed over the course of an hour and the signal baseline drifts of the standard gases in 11 h were estimated with the temporal variation of the reference gas signals. The CH_4 sensor has a precision of $\pm$2.6 ppb (Suto and Inoue, 2010), which became $\pm$3 ppb when corrected with the reference gas. This system is a modified version of the method we developed for CO_2

measurement (Watai et al., 2010). The use of the reference gas reduced the consumption of standard gas in each cylinder to 0.51 m^3 yr^{-1}, meaning they only need to be refilled every 10 yr. In 2009, we introduced checks for drift in the in situ three standard gases with two standard gases from Japan; to date no drift has been detected.

Air temperature and relative humidity were measured at both levels on the tower using commercial sensors (model HMP45D, Vaisala, Finland). Wind direction and speed (model 81000, R. M. Young, USA) were determined at the high inlet. Solar radiation (model CM3, Kipp & Zonen, Holland) and precipitation (model 52202, R. M. Young, USA) were measured on top of the container laboratory.

2.3. Coupled Eulerian–Lagrangian transport model and CH_4 fluxes

Daytime CH_4 concentrations were simulated with a coupled transport model based on FLEXPART (Stohl et al., 1998), a Lagrangian particle dispersion model, to evaluate the factors controlling CH_4 variation over Siberia. FLEXPART calculates the trajectories of tracer particles using meteorological data plus random motions representing turbulence. The results presented here used 6-hourly meteorological data from the Global Forecast System (GFS) provided by the National Centers for Environmental Prediction (NCEP). The backward method was used to analyse transport pathways from potential flux regions to the receptor position (each tower site). Each simulation consisted of 10 000 particles released from the highest inlet of each tower in the period of 13:00–17:00 (note that all times mentioned in this paper refer to geographical local time at the measuring location). Released particles below 300 m in height gain CH_4 flux from the surface during 7 d backward in time (Seibert and Frank, 2004). For the initial concentration, we used the NIES offline global transport model (Maksyutov et al., 2008). We used the monthly varying CH_4 flux estimated by Patra et al. (2009). The flux is based on the Emission Database for Global Atmospheric Research (EDGAR) version 3.2 (Olivier and Berdowski, 2001) for anthropogenic CH_4 (fossil fuel, landfill and domestic animals), and the NASA Goddard Institute for Space Studies (GISS) emission (Fung et al. 1991) for natural CH_4 (wetlands and termites) and for CH_4 from rice fields. The ability of the flux to reproduce the general features of variations in CH_4 concentrations have been confirmed with data from sites of the WMO World Data Centre for Greenhouse Gases (Patra et al., 2009) and at Hateruma Island, Japan (Tohjima et al., 2010). We used the seasonally varying biomass burning flux of GFEDv2 (Randerson et al., 2006) instead of the annual-constant EDGAR3.2 biomass burning flux used in Patra et al. (2009). The base CH_4 fluxes were also used to estimate the contributions of each source to the observed temporal variations in CH_4 concentration. Chemical destruction of CH_4 by OH radicals was calculated using climatological monthly mean OH radical concentrations (Spivakovsky et al., 2000) scaled by a factor of 1.09 to reproduce realistic CH_4 increase rates observed through shipboard measurements over the Pacific Ocean (Y. Terao, private communication, 2009).

3. Results and discussions

3.1. Temporal and diurnal variation in CH_4 concentration

Following establishment of the container laboratory and CO_2 observation, measurement of CH_4 concentration began at BRZ and KRS in 2004; at IGR, NOY and DEM in 2005; at SVV and YAK in 2007 and at AZV and VGN in 2008 (Fig. 2). With the exception of a few data gaps, continuous measurements have been conducted successfully at KRS, IGR, DEM and VGN since their commissioning. The data for BRZ before 2007 are not shown because sampling problems emerged after measurement started. The CH_4 sensor at AZV did not function properly for the first 21 months after commissioning, so only a very small data record had been collected at the time of preparing this paper; we have therefore omitted this data. Sporadic gaps in the data records occur, primarily because of damage to the CH_4 sensors by lightning or water intrusion.

The CH_4 concentrations from each low inlet were generally higher than those from its high inlet mate due to CH_4 flux from the land surface. Extremely high concentrations, in the order of several ppm, were sometimes observed from both inlets, particularly at KRS, IGR, NOY and DEM, which implies that there were sporadic local CH_4 sources nearby. High CH_4 events were observed at IGR regardless of the season, and most were regarded as being subject to local influence as inferred from a data selection process that is described in more detail below. The southern towers (SVV, VGN) in steppe biome rarely produced high CH_4 events.

Remarkable diurnal variations in CH_4 concentration were observed, particularly during summer (from June to August) (for example at KRS in Fig. 3). In general, the height of the convective mixed layer varies diurnally and seasonally, with maxima in the daytime and summertime, respectively, and additionally depends on latitude. The mixed layer is also much more pronounced in inland continental locations such as Siberia, varying from 200–600 m in winter to as much as 2800 m in summer (Lloyd et al., 2002). The strong diurnal variation during summer occurred because the convective mixed layer developed during the daytime causing a decrease in CH_4 concentration due to dilution by air of low CH_4 concentration from above; after sunset, the convective mixed layer collapsed followed by CH_4 accumulation. The CH_4 gradient (concentration measured at the low inlet minus that of the high inlet) increased during the nighttime, whereas near-zero values occurred during the daytime due to turbulent mixing (Fig. 3). Methane concentrations for each hour in April, May and October displayed less variability throughout

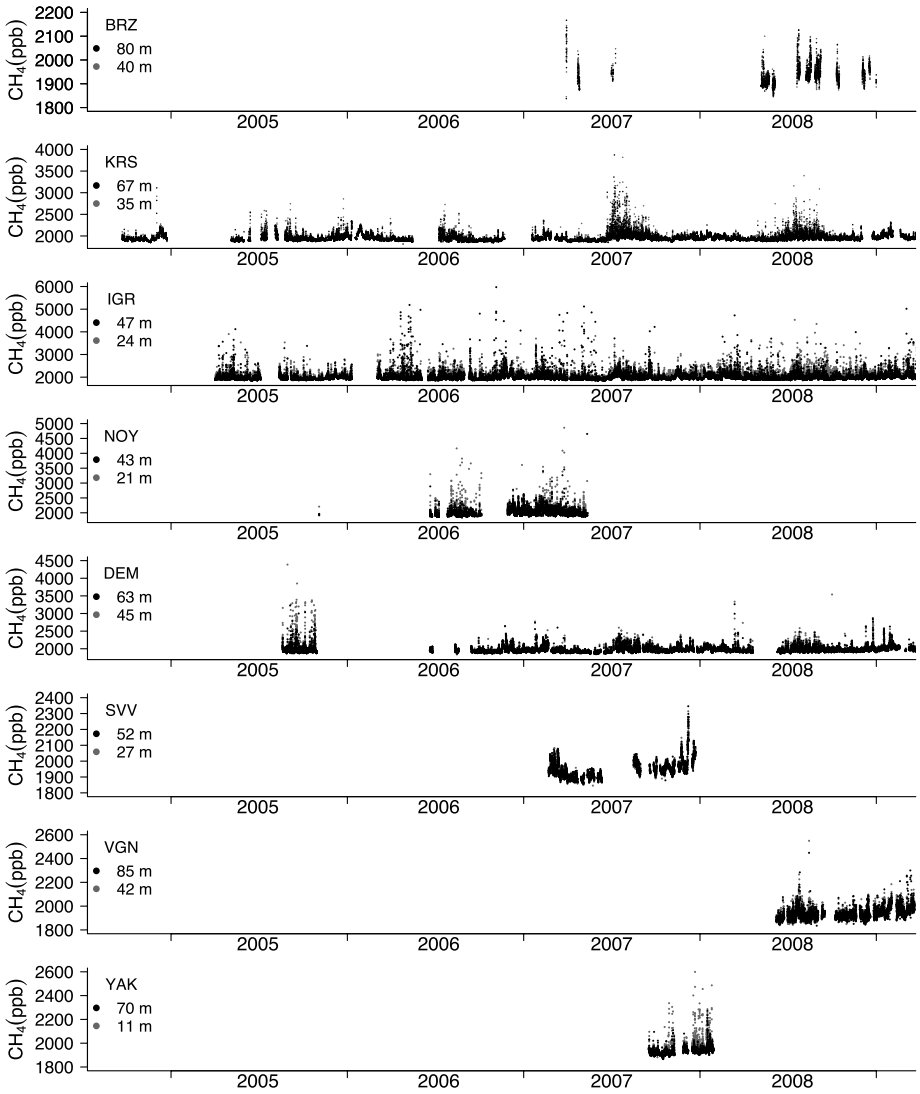

Fig. 2. Temporal variations in CH_4 concentration obtained from the JR-STATION during the period 2004 to 2009. Black circles and grey circles denote the high inlet and low inlet, respectively. Note that the range of the y-axis varies. No useable CH_4 data were obtained from the AZV tower due to technical problems, although it is now collecting data and remains a part of the JR-STATION.

the day than at other times of the year (Fig. 3). The main factor producing short-term variability was high-pressure systems during winter (see Section 3.2) and emissions from wetlands and biomass burning during summer (see Section 3.3). Hence the CH_4 concentrations were less variable during spring (April and May) and autumn (October).

We regarded data from the daytime (13:00–17:00) when the atmosphere was mixed well as representative of the wider region. Furthermore, we applied a data selection procedure using two criteria to remove local influence. First, we omitted data when the difference between the high inlet and low inlet were greater than 50 ppb. Second, from the remaining data, we retained consecutive 4-hourly data collected during daytime with a variability (maximum minus minimum) of less than 50 ppb. The first criterion was adopted because local influence is expressed more strongly in measurements at the low inlet, and a large gradient can occur even during daytime if a local influence occurs. The second criterion was used to exclude local influence from sporadic events such as leakage of natural gas that would cause spikes in the data. Airborne measurements have detected CH_4 peaks exceeding 2900 ppb at an altitude of 150 m over oil production sites and pipelines near Surgut in West Siberia (Tohjima et al., 1996). By applying our criteria, most of the highest values at IGR and NOY were excluded (Figs 4a and d). At other sites, only limited data were excluded because of rare local influence (Figs 4b–d). Many oil and natural gas pipelines cross West Siberia, and pipelines exist several km northwest of the NOY tower; extremely high CH_4 events were sometimes

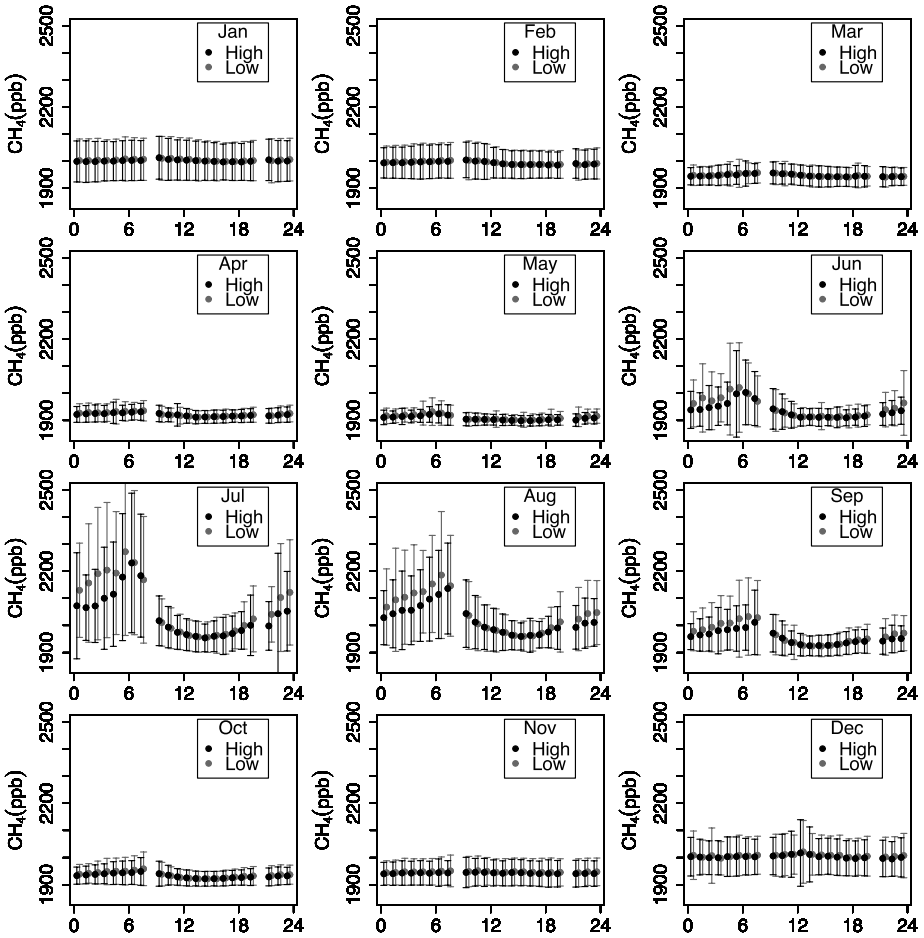

Fig. 3. Mean diurnal variation in CH_4 concentration for each month at KRS. The *x*-axis shows geographical local time of measurements, and the error bars are ± 1 *SD*. All observed data were averaged by time and month. 'High' in the legend refers to the high inlet (67 m); 'Low' refers to the low inlet (35 m). The measurement protocol involved taking one measurement from each of the two inlets and a reference gas each hour (alternating every 20 min). Every 12 h all three measurements for the hour were of standard gases; thus there are no data at 8:00 and 20:00.

observed at this tower, particularly when the dominant wind direction was from the northwest. High CH_4 events subject to leakage of CH_4 from the nearest gas pipeline or compressor station would also have been removed by our data selection criteria.

Methane concentrations from the selected daytime data displayed high values in winter (Fig. 5), which is similar to CH_4 observations at background sites such as Alert, Mace Head and Cape Grim (Rigby et al., 2008; Dlugokencky et al., 2009a). However, differences in seasonal variation exist between Siberia and background sites in the form of an obvious maximum in summer at almost all the Siberian stations. The median of monthly CH_4 values reached a minimum in May or June followed by a clear rise in July at IGR, DEM, KRS, BRZ and VGN. At background sites, minimum CH_4 values occur during summer, primarily because there is a seasonal maximum in OH concentrations. Hence, the obvious maximum observed during summer at these Siberian sites can be explained by strong CH_4 emissions from the wetlands of West Siberia. The contributions of wetlands and other possible sources to CH_4 variations are further elucidated in Section 3.4.

3.2. *Elevated CH_4 events in winter*

Events involving elevated CH_4 levels lasting several days were detected at some tower sites even after data filtering by the selection criteria. Here we focus on particular events in which elevated CH_4 concentration ($>$2000 ppb) occurred simultaneously at more than two sites for more than 2 d because such geographically widespread events were assumed to reflect synoptic weather conditions or other specific phenomena. Three periods of elevated values during winter were considered to represent specific events: (1) 2006 December 2–7, (2) 2008 January 13–17 and (3) 2009 January 26–30 (Fig. 4).

The second and third events displayed clear similarities. In the 2008 event, daytime mean CH_4 concentrations at DEM and

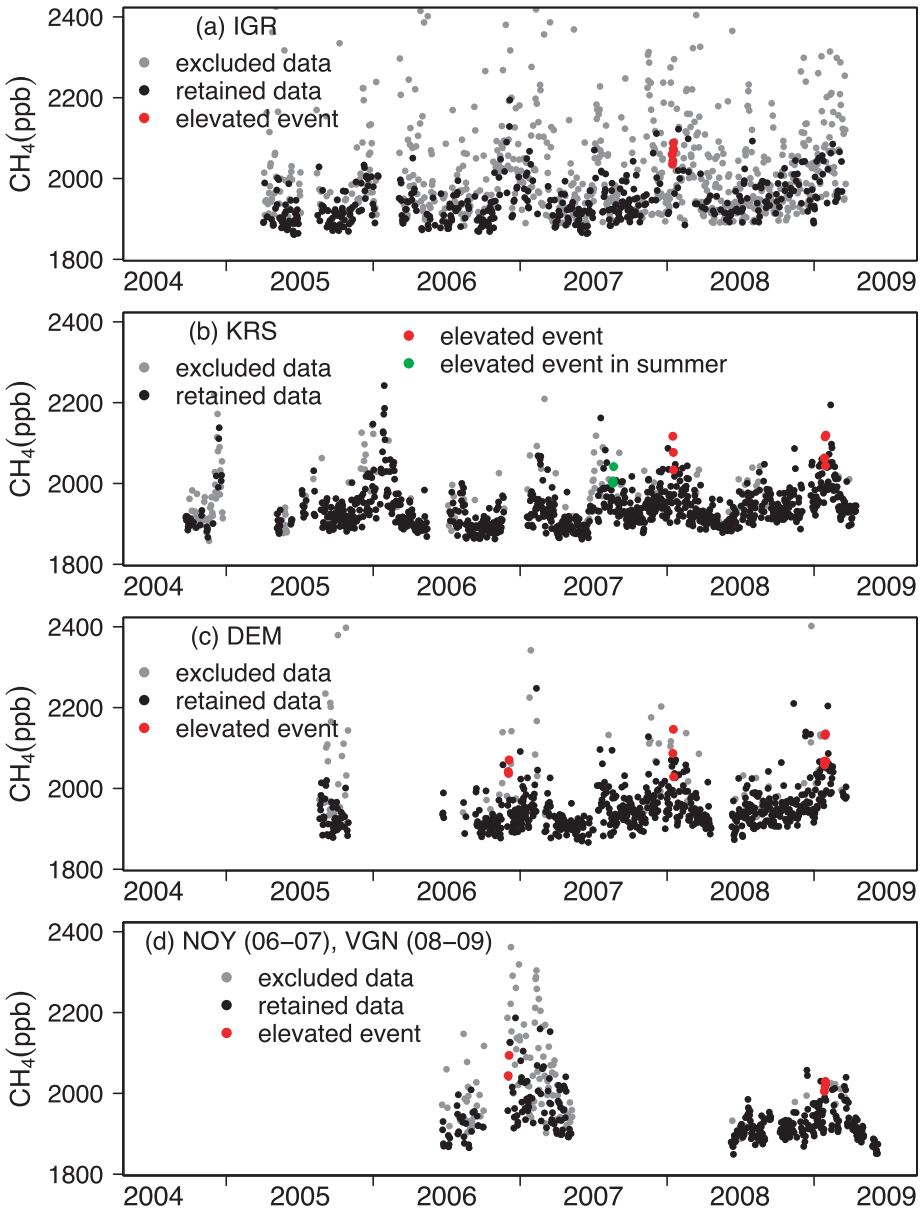

Fig. 4. Daytime mean CH_4 concentrations of excluded data (grey circles) and retained data (black circle) at (a) IGR, (b) KRS, (c) DEM and (d) NOY (2006–2007) and VGN (2008–2009). The procedure used to exclude data is described in the text. Red circles indicate data from events in which high CH_4 values were observed at different sites at the same time. Green circles indicate data from high CH_4 events during summer.

KRS on January 15, 16 and 17 were well above 2000 ppb, mostly in the order of 2100 ppb. Similar values occurred at IGR from January 13 to 17. Low temperatures ($<-20\,^{\circ}C$) had prevailed for more than 5 d before the high CH_4 event at all three sites. Even during daytime, the temperature at the low inlet was often lower than that at the high inlet at all three sites (i.e. an inversion layer had formed). Inversion layers were observed for most of the period between January 15 and 17 at DEM, January 13 and 18 at KRS, and January 12 and 17 at IGR. Inversion layers suppress vertical mixing of the lower troposphere, which allows CH_4 emitted from the land surface to accumulate. Atmospheric surface pressures were also high during this period and displayed one peak (1033–1041 hPa) on January 12 and another (1026–1033 hPa) around January 16 or 17. Sea level pressure of NCEP/NCAR Reanalysis 1 data (Kalnay et al., 1996) indicated that a high pressure system moved from the east to west around this time; the centre of the high pressure was stationed over DEM and KRS on January 16 before gradually moving to the south. High pressure causes downward flow resulting in the formation of a low boundary layer. Although the coupled transport model identified fossil fuel as a major CH_4 source during winter, measured concentrations were considerably higher than simulated

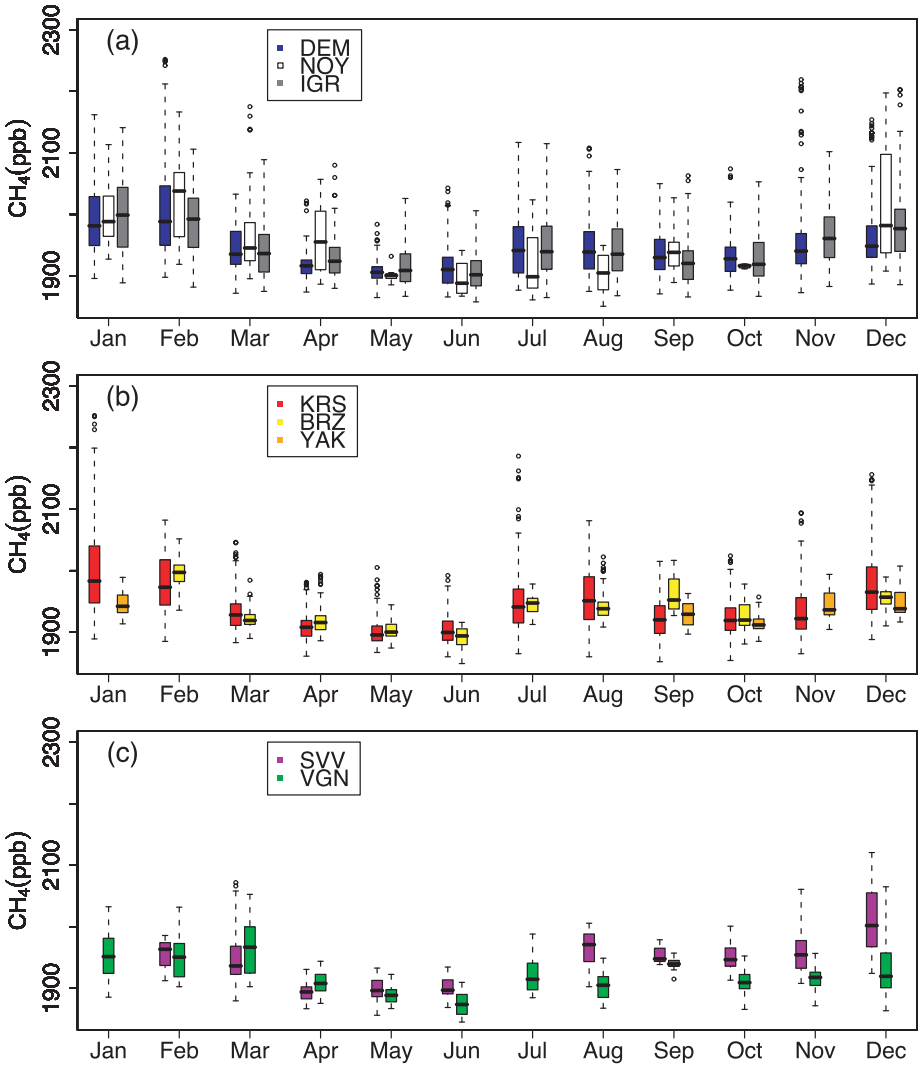

Fig. 5. Box-and-whisker diagrams of CH_4 concentration at the high inlet during 13:00–17:00 from all collected data grouped by month for (a) northern sites (DEM, NOY and IGR), (b) mid-latitude (KRS and BRZ) and eastern Siberian (YAK) sites and (c) steppe region sites (SVV and VGN). The box-and-whisker diagrams are defined as follows: the median is the thick line in the box; the bottom and top of the box are the lower and upper quartiles, respectively; the whiskers extend to the most extreme data point which is no more than two times the interquartile range from the box; individual outliers are shown as open circles outside the whiskers. Data that might have suffered from local influence were eliminated by the data selection criteria (see Section 3.1).

values (see Section 3.4). Considering that the fossil-fuel CH_4 source is not widespread over West Siberia and the emission rate is not directly related to weather conditions, we conclude that the extreme increase in CH_4 concentration at subcontinental scale occurred as a result of synoptic atmospheric conditions.

In the 2009 event, high daytime mean CH_4 concentrations in the order of 2100 ppb were detected at DEM and KRS. The mean values at IGR for this period were excluded by the data selection procedure, but were always higher than 2000 ppb. High daytime means (around 2000 ppb) were also recorded at the southern tower site (VGN) during this event. Atmospheric pressure at DEM and KRS increased from less than 1000 hPa on January 25 (DEM) and 26 (KRS) to around 1020 hPa on January 27. At the same time, temperatures dropped from –5 °C to less than –20 °C. The pressure remained high until January 31, and temperature decreased further to less than –35 °C. Temperature inversions were observed from January 28 to 29 at DEM and for much of the time between January 27 and 29 at KRS. The atmospheric pressure at VGN also increased from 995 hPa on January 25 to 1004 hPa on January 27, followed by a decrease to 994 hPa on January 29 and another increase to 1011 hPa on January 30. Since the VGN tower is located in a steppe biome at a lower latitude than DEM and KRS, the temperature at VGN was much higher than at the other two sites but showed similar temporal variation, decreasing from 1 °C on January 25 to –13 °C on January 28, and daytime inversion layers were recorded on January 24, 25,

26 and 29. Sea level pressure of NCEP/NCAR Reanalysis 1 data indicated that a low pressure system centred around 63°N 80°E on January 25 was displaced to the east by high pressure from the northwest. Having remained over IGR, DEM and KRS from January 27 to 28, the high pressure weakened once and then built up again on January 31. The similarity between the 2008 and 2009 events suggests that high-pressure systems over Siberia in winter result in the accumulation of CH_4 concentration at subcontinental scale.

In the 2006 event, elevated CH_4 concentrations (∼2040–2100 ppb) occurred at DEM and NOY on December 2–4. Atmospheric pressure at DEM remained steady at 1004–1006 hPa, and temperature decreased to less than –20 °C (minimum –27 °C) on December 2 and 3. Daytime temperature inversions were observed on December 1, 2 and 4. Mean wind speed was 4.9 m s^{-1} ($\sigma = 1.6$) from December 2 to 4. Similar weather conditions were observed at NOY from December 2 to 3 (pressure 1003–1005 hPa; temperature <–20 °C and minimum of –33 °C; and wind speed 3.1 m s^{-1}, $\sigma = 1.8$), and a strong inversion layer formed in the early morning of December 2 and persisted until the night of December 4. The elevated CH_4 events at DEM and NOY therefore appeared to be related to the prevailing weather conditions even though NCEP/NCAR Reanalysis 1 data did not display a clear high-pressure system. The relatively weak wind probably allowed strong inversion layers to form resulting in CH_4 accumulation at DEM and NOY. High CH_4 concentrations of over 2000 ppb associated with very cold temperatures, low wind and high pressure were also observed at ZOTTO during November and December 2006 (Kozlova et al., 2008).

3.3. Elevated CH_4 events in summer

Although events of high CH_4 concentrations (>2000 ppb) were occasionally observed, even during summer (from June to August), they rarely persisted for more than 2 d. Whereas no such events were detected in 2005, 2006 and 2008, several events lasting more than 2 d occurred in 2007. The longest event occurred in 2007 from August 18 to 21 when the daytime mean CH_4 concentration at KRS was 2000–2042 ppb (Fig. 4b). Daytime means of 2001 ppb were recorded at SVV on both August 18 and 19. Atmospheric pressure at KRS increased gradually from 988 hPa on August 14 to 1015 hPa on the morning of August 19, and then remained around 1014 hPa for 24 h before beginning to decrease. The daytime maximum temperature at KRS also increased over the same period, reaching over 20 °C on August 19 and 20. These weather conditions imply that a high-pressure system gradually moved in and was stationed over the KRS region on August 19 and 20. The sea level pressure of NCEP/NCAR Reanalysis 1 data corroborated this interpretation. The SVV region also appeared to be influenced by the same high pressure from August 16 to 19. Observed pressure at SVV increased from August 15 and remained high during the period. In contrast to the situation in winter, an inversion layer was observed only during the nighttime. Considering that there are also potentially strong CH_4 emissions from wetlands during summer, we cannot simply conclude that the elevated CH_4 concentrations were due to accumulations caused by synoptic conditions. However, the possibility exists that the summer events are a result of increased CH_4 emission rates from wetlands induced by the temperature change associated with high-pressure systems. High-pressure systems appear to influence CH_4 concentration at subcontinental scale during summer as well as winter, even if the mechanism is apparently different.

Other elevated CH_4 events were observed at KRS, DEM and IGR during summer 2007. They did not occur at multiple sites at the same time and thus did not appear to be directly controlled by synoptic conditions such as high-pressure systems. Methane emissions from wetlands are controlled by both soil temperature (microbial activity) and water table depth (oxidation and reduction) (Gedney et al., 2004). We calculated the regional monthly means for atmospheric temperature, temperature anomaly and precipitation rate in May, June, July and August in three latitudinal zones of 55–60°N, 60–65°N and 65–70°N between 65°E and 85°E with the data of NCEP/NCAR Reanalysis 1 for temperature and GPCP version 2.1 combined precipitation data set (Adler et al., 2003) for precipitation rate (Fig. 6). Wetlands exist extensively in these three regions. The monthly mean temperature in the 55–60°N zone displayed similar maximum values over the 5 yr, but in the 60–65°N and 65–70°N zones, they were clearly higher in July 2007 than in other months and years. The monthly mean precipitation rates in June and August 2007 were also about the highest values seen across the 5 yr. Furthermore, the highest precipitation rate for each month occurred in May 2007 in the 55–60°N and 60–65°N zones. The hot and wet summer of 2007 very likely created favourable conditions for increased CH_4 emissions from wetlands, which supports the hypothesis that this was the main cause of the elevated CH_4 events observed in 2007.

Biomass burning is known to influence the atmospheric environment over wide regions of high latitude in the Northern Hemisphere (e.g. Tanimoto et al., 2009; Warneke et al., 2009; Paris et al., 2009). We inspected fire maps detected by MODIS on board the Terra and Aqua satellites over a 10-d period (http://rapidfire.sci.gsfc.nasa.gov/firemaps/). The fire maps displayed many hot spots over West Siberia during the period from June 20 to August 8 in 2007, and the maximum activity was in the period July 10 to 19 in 2007. The elevated CH_4 events during this period in 2007 could have been at least partly explained by emissions from biomass burning. Coinciding with one such event at KRS on August 4 in 2007, the daytime mean CO_2 concentration increased to 371 ppm from 364 ppm the previous day. The summer of 2008 was relatively devoid of hot spot activity, but another very active hot spot period in 2005 between July 20 and August 8 coincided with a series of equipment malfunctions and limited CH_4 data collection at our sites. Many hot spots (but fewer than in 2007) were detected in 2006

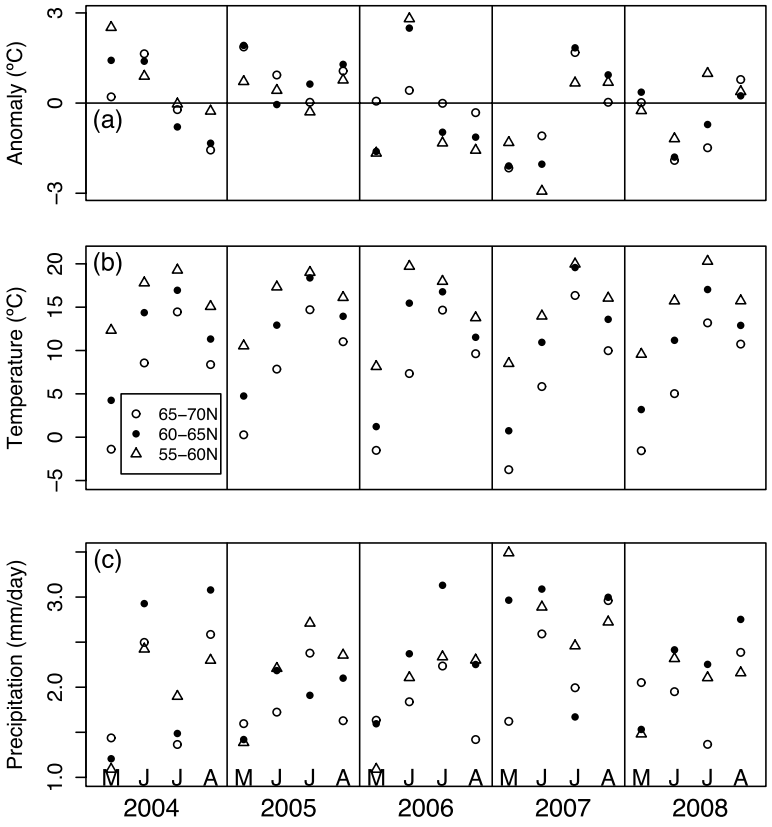

Fig. 6. Regional monthly means for (a) temperature anomaly, (b) temperature and (c) precipitation rate in May, June, July and August in three latitudinal zones of 55–60°N, 60–65°N and 65–70°N between 65°E and 85°E with the data of NCEP/NCAR Reanalysis 1 (Kalnay et al., 1996) for temperature and GPCP version 2.1 combined precipitation data set (Adler et al., 2003) for precipitation rate.

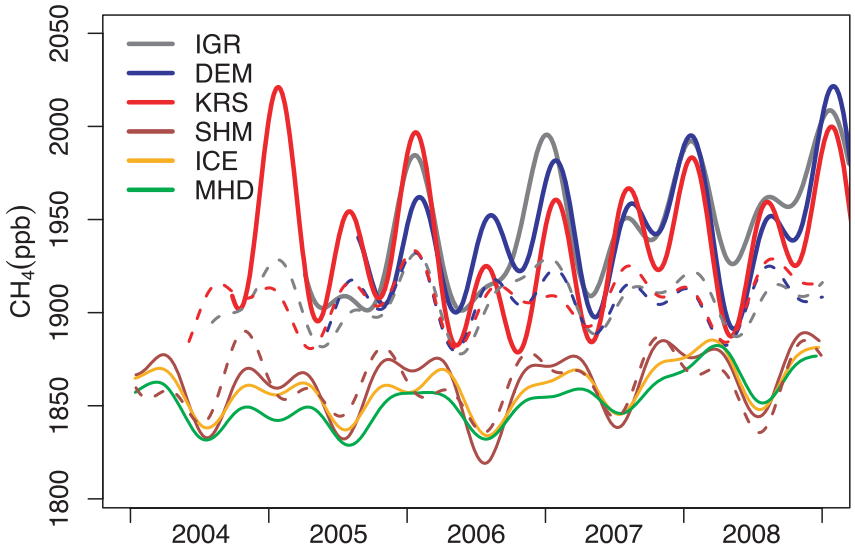

Fig. 7. Fitted curves of daytime mean CH_4 concentration at IGR, DEM and KRS and flask CH_4 concentration at the NOAA coastal sites of the same latitudinal zone (Dlugokencky et al., 2009b). The NOAA sites are Shemya Island, Alaska (SHM; 52°72′N, 174°10′E), Storhofdi, Vestmannaeyjar Iceland (ICE; 63°34′N, 20°29′W) and Mace Head, County Galway, Ireland (MHD; 53°33′N, 9°90′W). The tower data from the current study were selected according to the data selection criteria described in the text. The NOAA flask data which have 'REJECTION flag' or 'SELECTION flag' (ftp://ftp.cmdl.noaa.gov/ccg/ch4/flask/README_flask_ch4.html) were excluded and the retained data were converted to NIES 94 scale. The dotted lines are curves fitted to predicted CH_4 concentrations obtained from simulations with the coupled Eulerian–Lagrangian transport model.

between May 31 and July 29, but no elevated CH_4 events were observed at IGR, KRS, NOY and DEM during this period. However these hot spots were distributed in the southeastern region of West Siberia, so the emission plumes possibly did not reach the northern sites of IGR and NOY. Only 10 daytime means of KRS and four means of DEM remained after the data selection during this period, and their values were not particularly high (1919 $\pm$ 31 ppb at KRS and 1948 $\pm$ 28 ppb at DEM). The effect of biomass burning on CH_4 concentration at the tower sites was therefore not clearly detected in 2006 because of the small number of retained data. Furthermore, the distribution of hot spots in 2006 did appear sparser than in 2007, suggesting that the effect, if present, would not have been as pronounced as in 2007. More detailed analysis, for example using satellite data of the distribution of CO concentration, may reveal greater insights. Note also that no hot spots were detected during the time of the three elevated CH_4 events in winter.

3.4. Estimation of contributors to double maxima using simulated data

We fitted curves to the selected daytime mean CH_4 concentrations using the digital filtering technique of Thoning et al., (1989) for the data from KRS, IGR and DEM and compared them with corresponding data from three background coastal sites of NOAA (Fig. 7). The three NOAA sites, which are in the same latitudinal zone as our sites (50–70°N), display pronounced annual lows in midsummer and a broad peak during winter (Dlugokencky et al., 2009b). As mentioned before, the characteristic feature of the Siberian data is an obvious maximum during summer, which appeared in all three sites. Furthermore, compared with the NOAA data, which ranged mainly below 1900 ppb, the CH_4 concentrations at the Siberian towers were always higher, even during winter. In an attempt to identify which factors create these larger maxima in summer and winter at the Siberian tower sites, we compared the daytime means with

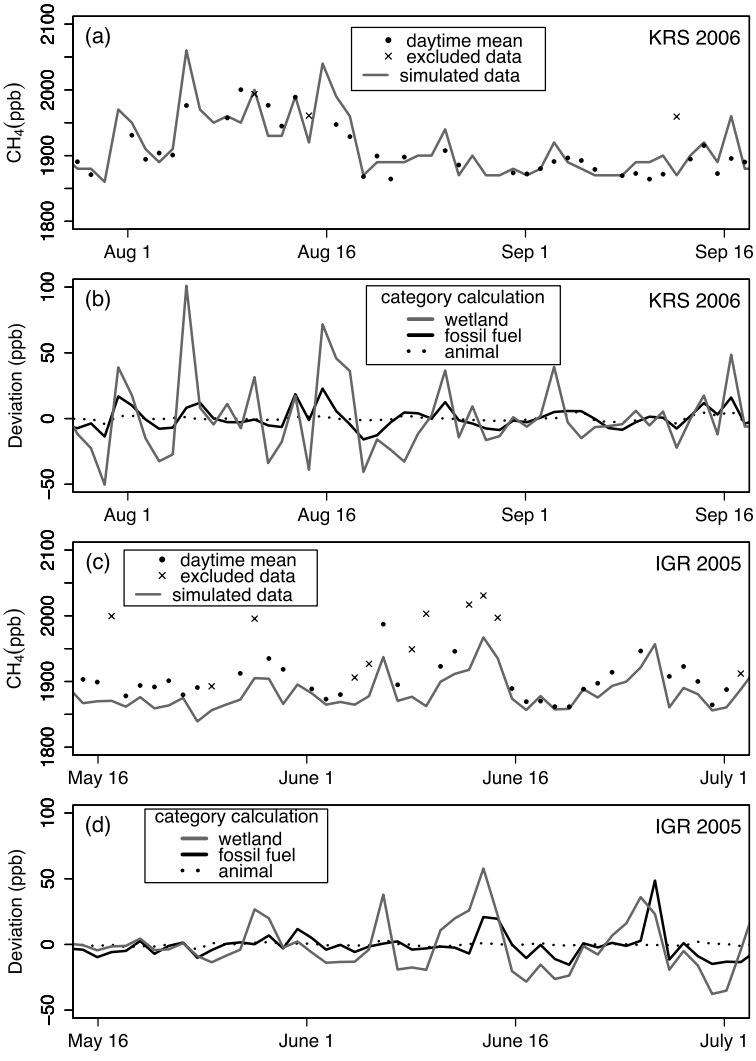

Fig. 8. Temporal variations in daytime mean and simulated data from (a) 2006 July 28 to September 17 at KRS and (c) 2005 May 15 to July 2 at IGR. Temporal variations in deviation each day from the 13-d mean (i.e. day in question and the 6 d before and after) of wetland, fossil fuel and domestic animal sources are shown for (b) KRS and (d) IGR. Deviations for other sources exhibited even less variation than that of animals and are not shown.

the results of simulations. The coupled model (FLEXPART and NIES-TM) can reproduce CH_4 variations at a coastal site (e.g. Shemya island) and exhibits a clear difference in concentrations between the coastal and Siberian sites (Fig. 7). The model also reproduced seasonal cycles that showed two maxima in summer and winter over Siberia. However the model generally underestimated CH_4 concentrations, particularly during winter.

Day-to-day variations during winter were probably not reproduced well because CH_4 accumulation caused by the strong inversion layer was not represented in our model. However, day-to-day variation during summer was reproduced well, for example, in 2006 from July 28 to September 17 at KRS (Fig. 8a) and in 2005 from May 15 to July 2 at IGR (Fig. 8c). We also used the model to calculate the contribution of each CH_4 source to the simulated temporal CH_4 variations (Figs 8b and d) using the fluxes in Patra et al. (2009) as described in chapter 2.3. The variation in CH_4 of wetland origin was clearly followed that of simulated total CH_4 concentration, which suggests that wetlands contributed to most of the day-to-day variation in CH_4 concentration in these two periods. The fossil-fuel CH_4 source (production, transport, and distribution of coal and natural gas) also displayed a similar pattern to total simulated variation but with smaller amplitude. The domestic animals and non-displayed categories (termites, rice fields, biomass burning and landfill) displayed much smaller variation. Fig. 9 shows detrend temporal variations in CH_4 concentration produced by each CH_4 source, calculated using the coupled model, for all seasons at KRS. This chart demonstrates that the summer increase in CH_4 could be mainly attributed to emissions from wetlands. On the other hand, emissions from fossil fuel contributed most to the rise in CH_4 during winter. Both the seasonal decline in OH concentrations and frequent existence of an inversion layer during winter induced CH_4 accumulation. Since CH_4 emissions from wetlands during winter are supposedly almost zero, the contribution of fossil fuel appears to have been substantial. The temporal variability in wetlands and fossil fuel was relatively small during the months when the major contributor was in transition from fossil fuel to wetlands or from wetlands to fossil fuel (April, May and October), which may account for the low hourly variability during these months in the diurnal variation in CH_4 concentration (Fig. 3). These trends were similar at other sites (not shown). The calculation of emissions from biomass burning was based on GFEDv2 (Randerson et al., 2006), which provides seasonal cycles until the year 2005. The expected increase in emissions from biomass burning during summer in 2006 and 2007 probably did not appear because the temporal variations in emissions from biomass burning after 2005 were calculated using the 2005 data. Recalculation of emissions with new spatial data of CH_4 flux from biomass burning in the later years of our data record is necessary to investigate the contribution of biomass burning to the observed high CH_4 events, particularly in the summer of 2007.

4. Conclusions

This paper presents the most comprehensive in situ, high-frequency data set to date of CH_4 concentration from the taiga, steppe and wetland biomes of Siberia. Clear diurnal variations

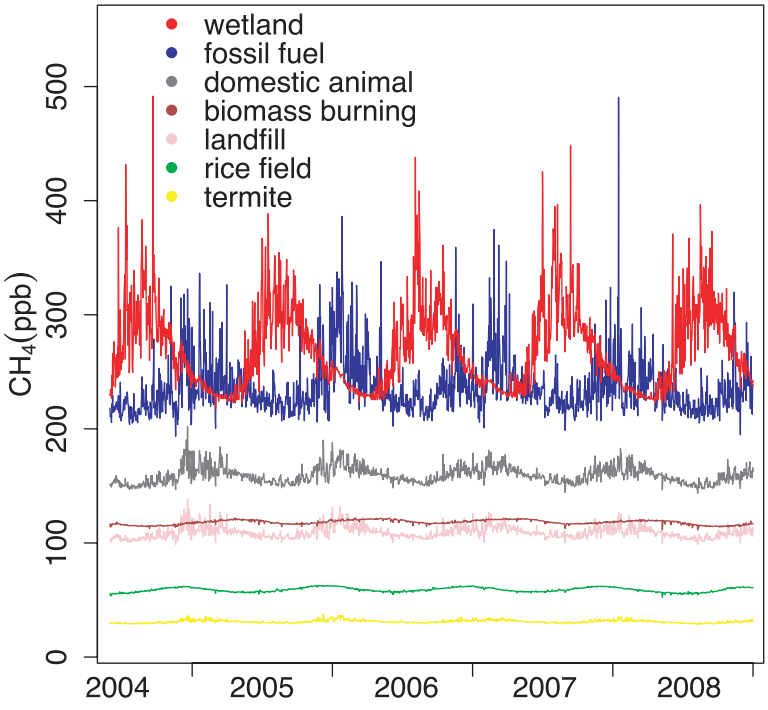

Fig. 9. Detrend temporal variations in CH_4 concentration produced by each of seven CH_4 sources (wetlands, fossil fuel, domestic animals, biomass burning, landfill, rice fields and termites) at KRS as calculated with the coupled Eulerian–Lagrangian transport model.

were observed in CH_4 concentrations, particularly during summer (from June to August) due to the diurnal change in the height of the mixed layer. Daytime CH_4 concentrations showed two maxima in summer and winter at almost all sites, and higher concentrations and bigger deviations were recorded at the northern sites, which are surrounded by extensive wetlands. Elevated CH_4 concentrations (>2000 ppb) were observed simultaneously at several sites because of governing weather conditions. High-pressure systems in winter led to the formation of a low boundary layer and caused CH_4 accumulation at subcontinental scale. The number of elevated CH_4 events in summer was greatest in 2007 when temperature and precipitation rate were the highest in the 5 yr (2004–2008) of monitoring over West Siberia, suggesting that elevated CH_4 events observed in 2007 could be mainly attributed to emissions from wetlands. However, many hot spots (fire activity) were detected over West Siberia in the summer of 2007, suggesting biomass burning may have also been a significant contributor to the high CH_4 concentrations measured. Daytime mean CH_4 concentrations over Siberia were generally higher than CH_4 values reported from flask sampling at NOAA coastal sites in the same latitudinal zone. Our coupled Eulerian–Lagrangian transport model reproduced this difference as well as the seasonal variation, displaying two maxima per year. In summer, the model also reproduced day-to-day variation well and verified that CH_4 emitted from wetlands was the predominant contributor to CH_4 variation. Partitioning CH_4 between different sources revealed that the major contributor to CH_4 variation switched from wetlands during summer to fossil fuel during winter. The JR-STATION will continue to collect data that can be applied to other tasks such as evaluating CH_4 leakage from natural gas pipelines (the CO_2 data we are collecting will assist in this regard). The calculation of a CH_4 budget at subcontinental scale will also become possible as our data record further accumulates.

4. Acknowledgments

We would like to thank Sergey Mitin (Institute of Microbiology, Russian Academy of Sciences) for administrative support and Keiichi Katsumata (NIES) for experimental assistance in maintaining the standard gases. This research was supported by the Global Environment Research Account for National Institutes of the Ministry of the Environment, Japan through its funding of the project titled: Estimation of CO_2 and CH_4 Fluxes in Siberia using Tower Observation Network.

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