Inventory of anthropogenic methane emissions in Mainland China from 1980 to 2010

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Abstract. Methane (CH_4) has a 28-fold greater global warming potential than CO_2 over one hundred years. Atmospheric CH_4 concentration has tripled since 1750. Anthropogenic CH_4 emissions from China has been growing rapidly in the past decades, and contributes more than 10% of global anthropogenic CH_4 emissions with large uncertainties in existing global

15   inventories, generally limited to country-scale statistics. To date, a long-term CH_4 emissions inventory including the major sources sectors and based on province-level emission factors is still lacking. In this study, we produced a detailed bottom-up inventory of anthropogenic CH_4 emissions from the eight major source sectors in China for the period 1980-2010. In the past three decades, the total CH_4 emissions increased from 22.2 [16.6-28.2] Tg CH_4 yr^{-1} (mean [minimum-maximum of 95% confidence interval]) to 45.0 [36.4-58.3] Tg CH_4 yr^{-1}, and most of this increase took place in the 2000s. This fast increase of

20   the total CH_4 emissions after 2000 is mainly driven by CH_4 emissions from coal exploitation. The largest contribution to total CH_4 emissions also shifted from rice cultivation in 1980 to coal exploitation in 2010. The total emissions inferred in this work compare well with the EPA inventory but appear to be 38% lower than EDGAR4.2 inventory. The uncertainty of our inventory is investigated using emissions factors collected from published literatures. We also distributed province-scale emissions into 0.5° x 0.5° maps using social-economic activity data. This new inventory could help understanding CH_4

25   budgets at regional scale and guiding CH_4 mitigation policies in China.

1 Introduction

Methane (CH_4) plays an important role on global warming as a greenhouse gas. The radiative forcing in 2011 relative to 1750 caused by anthropogenic CH_4 emissions is about 0.97 [0.74-1.20] W m^{-2}, ranging from 0.74 to 1.20 W m^{-2}, which contributes 32% of total anthropogenic radiative forcing by long-lived greenhouses gases (CO_2, CH_4, Halocarbons and N_2O)

30   since 1750 (Ciais et al., 2013). Atmospheric CH_4 concentration increased by 1080 ppb since pre-industrial times, reaching 1803 ppb in 2011 (Ciais et al., 2013). The growth of CH_4 levels in the atmosphere is largely driven by increasing

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anthropogenic emissions (e.g., Ghosh et al., 2015). Based on an ensemble of top-down and bottom-up studies, Kirschke et al. (2013) synthetized decadal natural and anthropogenic CH_4 sources for the past three decades, and reported that 50% - 65% of CH_4 emissions originate from anthropogenic CH_4 sources.

5   Between 14% and 22% of global anthropogenic CH_4 emissions in the 2000s were attributed to China (Kirschke et al., 2013). The major anthropogenic CH_4 sources in China include rice cultivation, livestock, biomass and biofuel burning, fossil fuel exploitation and combustion, and waste deposits. With rapid growth of the Chinese economy, the number of livestock has nearly tripled in the past three decades, causing an increase in CH_4 emissions from enteric fermentation and manure management (Khalil et al., 1993; Verburg and Denier van der Gon, 2001; Yamaji et al., 2003; Zhang and Chen, 2014). The

10   types of livestock (cow, cattle etc.) and their alimentation have evolved as well, and change CH_4 emissions (IPCC, 2006). The fossil fuels exploitation and consumption have increased exponentially, especially coal exploitation (e.g., Zhang et al., 2014), although large uncertainties remain in the magnitude of greenhouse gas emissions (e.g. Liu et al., 2015). On the other hand, the decrease of rice cultivation area (Verburg et al., 2001; Li et al., 2002; Kai et al., 2011) and changes in agricultural practices (Chen et al., 2013) can lead to reduced CH_4 emissions from rice paddies.

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Total methane emissions from China remain uncertain as illustrated by discrepancies between global inventories, and between bottom-up inventories and recent atmospheric-based analyses (e.g. Kirschke et al., 2013). The Emission Database for Global Atmospheric Center (EDGAR, version 4.2, http://edgar.jrc.ec.europa.eu/overview.php?v=42) reports that China has 73 Tg CH_4 yr^{-1} of anthropogenic CH_4 sources in 2008, while U.S. Environmental Protection Agency (EPA) estimates

20   that China emitted 44 Tg CH_4 yr^{-1} of anthropogenic CH_4 sources in 2010. Based on a province-level inventory, Zhang and Chen (2011) reported anthropogenic CH_4 emissions of 38.6 Tg CH_4 yr^{-1} for the year 2007. This large range of estimates (~30 Tg CH_4 yr^{-1}) is mainly caused by different emission factors (EFs) or activity data applied in these inventories (EDAGRv4.2; EPA, 2012; Zhang and Chen, 2011). Such discrepancies between inventories have been identified as limiting our ability to reduce uncertainties in the global methane budget (Dlugokencky et al., 2011; Kirschke et al., 2013; Ciais et al., 2013).

25   Atmospheric inversions also tend to infer smaller methane emissions for China than reported by EDGAR4.2, with 59 [49-88] Tg CH_4 yr^{-1} for the 2000-2009 decade in Kirschke et al. (2013) and ~40 [35-50] Tg CH_4 yr^{-1} in the inversion of Bergamaschi et al. (2013, see their Figure 5).

Global inventories generally rely on country-level socio-economic statistics, which hardly fully reflect the more local to
30   regional, possibly rapidly changing, characteristics of methane sources. This is especially the case in China where economic growth and the sources of CH_4 present large differences between provinces. To reduce uncertainties on estimates of Chinese methane emissions, it is therefore of particular importance to build a long-term consistent inventory of CH_4 emissions for each source sector based on local to regional specific EFs and activity data. This is the main goal of this study.

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A comprehensive anthropogenic CH_4 inventory for Mainland China (PKU-CH_4) was produced between 1980 and 2010, both at country and province scale, and downscaled at 0.5° spatial resolution. To do so, we compiled activity data at county or province levels for eight major source sectors: 1) livestock, 2) rice cultivation, 3) biomass and biofuel burning, 4) coal exploitation, 5) oil and natural gas systems, 6) fossil fuels combustion, 7) landfills and 8) wastewater. We also compiled

5 regional specific EFs for each source sector from published literature in English and Chinese. We then estimated annual CH_4 emissions and their uncertainty for the eight major source sectors and for total emissions. Finally, we produced annual gridded maps of CH_4 emissions at 0.5x0.5° for each source sector based on social-economics drivers (e.g., rural and urban population, coal exploitation, and Gross Domestic Product (GDP)). The database is described in section 2, methane emissions for the period 1980-2010 are presented in section 3 and discussed in section 4.

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2 Methods and Datasets

2.1 Methodology

The CH_4 emissions from livestock, rice cultivation, biomass and biofuel burning, coal exploitation, oil and natural gas systems, fossil fuels combustion, landfills and wastewater are investigated in this study. The methods of IPCC greenhouse

15 gas inventory guidelines (IPCC, 2006) were used to estimate CH_4 emissions for these nine sectors. The annual CH_4 emissions at the year t from the eight sectors are calculated by Eq. (1).

$$E(t) = \sum_S \sum_R \sum_C AD_{S,R,C}(t) \times EF_{S,R,C} \times (1 - CF_{S,R,C}(t)) \ , \quad (1)$$

Where E(t) represents the total CH_4 emissions from the eight sectors; S, R, and C indicate the index of sectors, regions/provinces and conditions, respectively; $AD_{S,R,C}(t)$ is the activity data at the year t, and $EF_{S,R,C}$ is the emission factor

20 for sector S, region R and condition C. $CF_{S,R,C}(t)$ is the correction factor at the year t for sector S, region R and condition C, which indicates the fraction of CH_4 utilized or oxidized without being released to atmosphere, such as CH_4 utilization or flaring from coal mining, CH_4 oxidation from waste. For estimation of CH_4 emissions from each source sector, the details of $AD_{S,R,C}$, $EF_{S,R,C}$ and $CF_{S,R,C}$ are introduced in the following Section 2.2.

2.2 Activity data, EFs and correction factors

25 ### 2.2.1 Livestock

CH_4 emissions from livestock are estimated as the sum of CH_4 emissions from enteric fermentation and manure management. Province-level annual census data of domestic livestock for each livestock category were collected from agriculture statistics yearbooks (CASY, 1980-2010). Livestock includes ruminants such as cattle, dairy cattle, buffalo, sheep, and goats, non-ruminant herbivores such as horses, asses, and mules, and omnivorous swine. Because seasonal births and slaughters change

30 the population of livestock, we used slaughtered population and live population at the end of the year to estimate the total

emissions from enteric fermentation. Here, average life spans in one year are 12 months for dairy cattle, 10 months for non-dairy cattle and buffalo, 7 months for sheep and goats and 6 months for swine, respectively. The EFs of enteric fermentation and manure management for each category livestock are from published studies are listed in Table 1 (IPCC, 1996, 2006; Dong et al., 2004; Khalil et al., 1993; Verburg and Denier van der Gon, 2001; Yamaji et al., 2003; Zhou et al., 2007). The

5   mean, minimum and maximum of EFs for enteric fermentation from these reported values are summarized in Table 1. For each category of livestock, separated EFs for female, youth and the rest of animals are reported when available.

Because EFs of manure management is a function of mean annual temperature under some special practice (IPCC, 2006), the EFs of manure management from default IPCC (2006) are assigned based on the mean annual temperature for each province

10 (Table 2). The uncertainty of CH_4 emissions are estimated by the range of EFs for enteric fermentation and manure management (Table 2) (IPCC, 2006). The CH_4 from manure management could be utilized by bio-digester in China, but there is limited information about CH_4 collected from bio-digesters only from manure. The correction factors are set as 0 for livestock sector, because of the limited information. We discussed the total CH_4 collected from bio-digesters with mixed crop straw, manure and waste in the discussion.

15 ### 2.2.2 Rice cultivation

CH_4 emissions from rice cultivation sector are estimated using the methodology of Yan et al. (2013). Province-level annual rice cultivation areas (early rice, middle rice and late rice) are collected from agriculture statistics yearbooks (CASY, 1980-2010). The EFs for early rice, middle rice and late rice in five regions under four different cultural conditions (with/without organic input, intermittent irrigation/continuous flooding conditions) are collected from Yan et al. (2013), which summarized

20 204 season-treatment measurements on 23 different sites (see their Table 2). We apply the EFs from Yan et al. (2003) and rice cultivation areas from yearbooks under different conditions from 1980 to 2010 to calculate CH_4 emissions from rice cultivation. 66.7% and 33.3% of rice cultivation area for intermittent irrigation and continuous flooding is assumed as in Yan et al. (2003). There is large uncertainty of rice cultivation area receiving organic input (Huang et al., 1998; Cai, 1997; Yan et al., 2003), and we assumed 50% of rice paddies received organic input (30% of rice paddies have crop straw, green manure or compost and 20% of rice paddies have animal and human waste) according Yan et al. (2003). The practices of organic

25 input have been changing with economic development and policy of agriculture and environment, and this uncertainty is discussed in the section 4.1. The growing days for early, middle, and late rice are 77, 110-130 and 93 days, respectively (Yan et al., 2003). The correction factors are set as 0 for rice cultivation sector, because no CH_4 utilization from rice paddies until now. The uncertainty of CH_4 emissions from rice cultivation is derived from the range of EFs (Yan et al., 2003).

30 ### 2.2.3 Biomass and biofuel burning

CH_4 emissions from biomass and biofuel burning mainly come from burning of firewood and straw in rural households. Province-level firewood consumption are extracted from the China Energy Statistical Yearbook (1980-2007). Because no

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firewood data is available after 2007 and firewood consumption in China is stable after 2005 (CESY, 2004-2008; Zhang et al., 2009; Zhang et al., 2014), we assumed that the consumption of firewood from 2008 to 2010 is stable and equal to the average of 2005-2007 emissions. For crops residues burning, we distinguish crops residues used as biofuels in the houses from those burnt in open fields, following Tian et al., (2011). The total crop residues are calculated as annual crops yields

5   and straw-grain ratio for major crops (rice, wheat, corn, soy, cotton and canola) in China. The crops residues burning as biomass fuels and disposed fire in open fields are separately calculated by Eq. (2).

$$RB_{crop} = \sum_c R_c \times N_c \times F \times \theta \quad , \quad (2)$$

Where RB_{crop} is the amount of burning crop residues as biomass fuel or disposed fire in open fields (Kg yr^{-1}); c is index of crop; N_c is straw-grain ratio for rice (1.0), wheat (1.4), corn (2.0), soy (1.5), cotton (3.0) and canola (3.0); F is the fraction of

10   crop residues used as biomass fuel or disposed fire in open fields (Table 2), which is determined by the province level of economic development (Tian et al., 2011); θ is burning efficiency for biomass fuel in households (100%) and fire in open fields (88.9%) (e.g., Cao et al., 2005; Tian et al., 2011).

EFs of CH$_4$ emissions from biomass and biofuel burning were collected from the scientific literature (Zhang et al., 2000;

15   Andreae et al., 2001; Streets et al., 2003; Cao et al., 2008; Tian et al., 2011). We used EFs from firewood of 2.77 ± 1.80 kg CH$_4$ t^{-1} (mean ± standard deviation), and EFs from crop residues for biomass fuel and fire in open fields of 3.62 ± 2.20 kg CH$_4$ t^{-1} and 3.89 ± 2.20 kg CH$_4$ t^{-1}, respectively (Tian et al., 2011). The uncertainty of CH$_4$ emissions (95% CI) is estimated from the range of the EFs by 1000 times of bootstrap samples.

2.2.4 Coal exploitation

20   CH$_4$ emissions from coal exploitation include fugitive CH$_4$ emissions from coal mining and post mining. In China, coal exploitation includes both underground and surface coal mines. Generally, CH$_4$ emissions per unit of coal mined from underground is much higher than that from surface (IPCC, 2006). Province-level annual coal production from underground and surface mines were collected from China Energy Statistical Yearbook and China Statistical Yearbook (1980-2010). The EFs of fugitive CH$_4$ from underground and surface mines are significantly different (Zheng et al., 2006; IPCC, 2006; Zhang

25   et al., 2014). Only 5% coal is mined from surface mines on average at country scale, with a fraction of coal mined varying from 0% for most provinces to more than 17% for Inner Mongolia and Yunnan provinces. Here, we calculated CH$_4$ emissions from both underground and surface mines. For CH$_4$ emissions from underground mines, the EFs vary among mines depending on local mines conditions such as depth of mines and methane concentration etc. Zheng et al. (2006) summarized regional EFs from coal exploitation based on measurements from ~600 coal mines in 1994 and 2000, and these

30   regional EFs correlate with properties of regional mines. For example, Southwest of China has higher EFs than other regions, because the coal mines in that region have deeper depth and higher coalbed methane, especially in Chongqing and Guizhou Province (Zheng et al., 2006; NDRC, 2014). We adopted the mean of regional EFs in China are reported in 1994 and 2000 from Zheng et al. (2006) to calculate CH$_4$ emissions from underground coal mining, and the range of the EFs as the

uncertainty (Table 2). The EFs of surface coal mines, we adopted the default value (2.5 m^3 t^{-1}) from IPCC (2006), since there is few measurements of CH$_4$ emissions from surface mines. The EF of CH$_4$ from coal post-mining including emissions during subsequent handling, processing and transportation of coal), is taken as 1.24 m^3 t^{-1} (1.18-3.00 m^3 t^{-1}), according to the weighted average of production from high- and low- CH$_4$ coal mines using IPCC (2006) default EFs for high- (3.0 m^3 t^{-1})

5   and low- (0.5 m^3 t^{-1}) CH$_4$ coal mines (Zheng et al., 2006).

Not all CH$_4$ emissions from underground coal mines are released into atmosphere as CH$_4$. A fraction of CH$_4$ from coal mines are collected for flaring or be utilized (e.g., Bibler et al., 1998; GMI, 2011). The utilization fraction of CH$_4$ from coal mines increases with economic growth and enhancement of coal safety (NDRC, 2014). For example, Zheng et al. (2006) indicates

10   that the utilization fraction of CH$_4$ from coal mines increased from 3.59% in 1994 to 5.21% in 2000. We used the utilization fraction of 3.59% before 1994 and linearly increase from 3.59% in 1994 to 9.26% in 2010 as $CF_{S,R,C}$ in Equation (1). The range of utilization (3.59% - 5.21%) is taken to calculate the uncertainty of CH$_4$ emissions from coal mining. A volumetric mass density of 0.67 Kg m^{-3} is used to convert volume of CH$_4$ emission into CH$_4$ mass.

2.2.5 Oil and natural gas systems

15   Province-level annual crude oil and natural gas production were collected from China Statistical Yearbook (1980-2010). The EFs of fugitive CH$_4$ from oil and natural gas systems in China are 0.36 kg t^{-1} for oil and 2.77 g m^{-3} for gas, respectively (Zhang et al., 1999). The uncertainty of the EFs for leakage from oil and natural gas systems in China is taken at 100% as suggested by IPCC (2006), because there is few measurements of CH$_4$ leakage from oil and natural gas systems in China.

2.2.6 Fossil fuels combustion

20   Province-level fossil fuels combustion (TJ) were collected from China Energy Statistical Yearbook (1980-2010). We used the default EFs from IPCC (2006) for CH$_4$ emissions from fossil fuels combustion, 1 Kg TJ^{-1} for coal combustion, 3 Kg TJ^{-1} for oil combustion and 1 Kg TJ^{-1} for natural gas combustion, respectively. The uncertainty of the EFs for fuels combustion is 60% (IPCC, 2006).

2.2.7 Landfills

25   Using IPCC (2006), the CH$_4$ emissions from landfills is estimated by First Order Decay (FOD) method as Eq. (3).

$$E_{Landfill}(t) = (1 - e^{-k}) \times \sum_x e^{-k \times (T_L - x)} \times MSW_L(x) \times MCF_T \times F_T \times DOC \times DOC_d \times f * (1 - O_f) \times \frac{16}{12} \quad , \quad (3)$$

Where $E_{landfill}(t)$ is CH$_4$ emissions from landfills at the year t; k is reaction constant and T_L is decay lifetime period, which are 0.3 and 4.6 years based on national inventory (NDRC, 2014); x is the year start to count. MSW_L is the total amount of municipal solid waste (MSW) treated by landfills at province scale; MCF_T is methane correction factor, which corrects CH$_4$

30   emissions from three types of landfills T (MCF_T = 1.0 for managed anaerobic landfills; MCF_T = 0.8 for deep (> 5 m) non-

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managed landfills, and $MCF_T = 0.4$ for shallow (< 5 m) non-managed landfills) (IPCC, 2006; NDRC, 2014). F_T is the fraction of MSW_L for each type landfill. We adopted the values of F_T by investigation for each province (Du, 2006, master thesis), which are shown in Table 2. DOC is fraction of degradable organic carbon in MSW, and is 6.5% in China (Gao et al., 2006). DOC_d is fraction of DOC that can be decomposed; f is fraction of CH_4 in gases of landfill gas, and O_f is oxidation factor and is set as 0.1 in this study. We adopted 0.6 for DOC_d and 0.5 for f in this study (Gao et al., 2006).

Country-total amount of MSW were collected from China Statistical Yearbook (1980-2010). Province-level amount of MSW in 1980, 1985-1988, 1996-2010 were collected from China Environmental Statistical Yearbook (1980, 1985-1988, 1996-2010). The missing province-level MSW were interpolated between periods, and the sum of province-level interpolated data keep conserved with country-total from the national yearbook. The amount of MSW treated by landfills are only available after 2003, and the rest MSW are treated compost, combustion and other processes. The fraction of MSW_L linearly decreases with GDP (R^2=0.95, P<0.001; Figure S1). We used this linear relationship to get the fractions of MSW_L before 2003, and assumed 1970s have similar MSW_L as the year of 1980. For uncertainty of CH_4 emissions from landfills, maximum CH_4 emissions with DOC_d=0.6 and f=0.6 and minimum CH_4 emissions with DOC_d=0.5 and f=0.4 were calculated.

2.2.8 Wastewater

CH_4 emissions from wastewater (domestic sewage and industrial wastewater) is estimated by Eq. (4).

$$E_{wastewater}(t) = COD(t) \times B_o \times MCF \quad , \quad (4)$$

Where $E_{wastewater}(t)$ is CH_4 emissions from wastewater treatment and discharge at the year t; $COD(t)$ is the total amount of chemical oxygen demand for wastewater at the year t; B_o is maximum CH_4 producing capacity, 0.25 kgCH_4/kgCOD; MCF is methane correction factor for wastewater. The total CH_4 emissions from wastewater include two parts: one part from wastewater treated by wastewater treatment plants (WTPs) and the other part from wastewater discharged into rivers, lakes or ocean. Here, we adopted 0.165 and 0.467 for MCF of domestic sewage and industrial wastewater treated by (WTPs), respectively (NDRC, 2014). For wastewater discharged into rivers, lakes or ocean, we adopted 0.1 for MCF (IPCC, 2006; NDRC, 2014; Ma et al., 2015).

Annual province-level amount of domestic sewage and industrial wastewater treated by WTPs or discharged into rivers, lakes or ocean were collected from China Statistical Yearbook (1998-2010). In the past three decades, China's economy grows with growth of population and the total amount of domestic sewage water exponentially increased with population (Figure S2). The COD in domestic sewage and industrial wastewater treated by WTPs increases with GDP (R^2=0.95-0.99, P<0.001; Figure S2 and S3). The fraction of discharged COD from industrial wastewater decreases with GDP (Figure S2). We used these relationship to interpolate the amount of COD in wastewater treated by WTPs and discharged into rivers, lakes or ocean before 1998, then distribute the total amount of COD into each province using the average contribution of each province to the total for the period 1980-1998.

The uncertainty of CH_4 emissions from wastewater mainly comes from the MCF term, besides the amount of COD in wastewater (IPCC, 2006; Ma et al., 2015). We assumed maximum CH_4 emissions with MCF=0.3 for domestic sewage and MCF=0.5 for industrial wastewater treated by WTPs, and minimum CH_4 emissions with MCF=0.1 for domestic sewage and

5 MCF=0.2 for industrial wastewater treated by WTPs (IPCC, 2006; Ma et al., 2015).

2.3 Maps of CH_4 emissions

In order to produce gridded emissions maps at $0.5°×0.5°$ for each source sector, we distributed the province-level CH_4 emissions using different activity data: rural or total population, GDP, agricultural GDP, crop cultivation area. First, we collected county-level rural population, total population, GDP and agricultural GDP in 2010 from statistic yearbook, and

10 converted them into 0.5º by 0.5º gridded maps. Then, these gridded maps are applied to distribute the province-level of CH_4 emissions from the eight source sectors. Because not all county data are available for every year during the period 1980-2010, we only used the activity data for 2010 (except for the average rice cultivation area in 1994-1996; Frolking et al., 2002; Qiu et al., 2003), therefore assuming that the changes in the spatial structures of the gridded maps remain limited.

15 The activity data used to distribute province-level totals vary with the sector: livestock (agricultural Gross Domestic Product, GDP), biomass and biofuel burning (rural population), fossil fuels combustion (GDP), oil and natural gas (GDP), landfills (population), wastewater (population), and coal exploitation (locations of 414 production sites in 17 provinces for years 2002, 2006, 2008 and 2009). For rice cultivation, early-, middle- and late-rice distribution maps are derived from crop maps provided by Frolking et al. (2002) and Qiu et al. (2003). We first rescaled the rice cultivation maps with annual province-

20 level rice cultivation area from agriculture statistics yearbooks to produce annual rice cultivation maps from 1980 to 2010. Then, we distributed province-level CH_4 emissions from rice cultivation on these rice cultivation area maps.

3 Results

3.1 Total and sectorial CH_4 emissions

Figure 1 shows the evolution of anthropogenic CH_4 emissions in China for the eight major source sectors and for the

25 country-total, and Table 3 lists the magnitude of CH_4 emissions and their uncertainty in 1980, 1990, 2000 and 2010. In 1980, the country-total CH_4 emissions was 22.2 [16.6-28.2] Tg CH_4 yr^{-1} (Table 3). Rice cultivation and livestock contributed 70% of anthropogenic CH_4 sources in 1980, followed by coal exploitation (15%) (Figure 1b). In the past 30 years, the CH_4 emissions doubled, reaching 45.0 [36.4-58.3] Tg CH_4 yr^{-1} in 2010 (Figure 1a). In 2010, coal exploitation became the largest contributor of Chinese CH_4 emissions (40%), followed by livestock (28%) and rice cultivation (16%) (Figure 1c). The

30 increase of CH_4 emissions between 1980 and 2010 is mainly attributed to coal exploitation (63% of the total increase) mostly after 2000, followed by livestock (27%) mostly before 2000.

Figure 2 shows the evolution of individual CH_4 sources from 1980 to 2010. Among the eight major source sectors, CH_4 emissions from seven source sectors increased from 72% to 426%, and only CH_4 emissions from rice cultivation decreased by 21 [17-23]% (Figure 2) before 2005 because of decreased rice cultivation area in this period. The increase of country-total

5   CH_4 sources accelerates after 2000 (Figure 2a). The increase of CH_4 emissions in the 2000s contributes 58% of the total increase observed between 1980 and 2010 (Table 3). The acceleration of emissions after 2000 is mainly driven by coal exploitation (Figure 2e), while CH_4 emissions from livestock, biomass and biofuel burning, landfills and rice cultivation remain stable or increased at a lower rate after 2000 resulting from the stable or slow increase in activities data in these sectors. Although CH_4 emissions from oil and gas systems, fossil fuels combustion and wastewater increased exponentially

10   after 2000, they only contributed less than 6% of the increase in total CH_4 emissions in the 2000s.

3.2 Spatial patterns of CH_4 emissions

Figure 3 shows the spatial distributions of CH_4 emissions in 2010 (Note that Figure 3a-3i have different color scales). Hotspots of CH_4 emissions are distributed mostly in the densely populated area, where we describe the emissions for South, Center and North of China country (Figure S4 shows the map these regions). These hotspots are driven by livestock, rice

15   cultivation and coal exploitation (Figure 3). North of China has high CH_4 emissions from livestock, biomass and biofuel burning, coal exploitation and wastewater. South and central of China has high CH_4 emissions from rice cultivation (Figure 3c). Southwest of China has high CH_4 emissions from rice cultivation and coal exploitation (Figure 3c and 3e). CH_4 emissions from biomass and biofuel burning, oil and gas systems, fossil fuels combustion, landfills and wastewater have one order of magnitude smaller than that from livestock, rice cultivation and coal exploitation. CH_4 emissions from biomass and

20   biofuel burning are mainly distributed in the north of China. CH_4 emissions from landfills and wastewater are mainly distributed in north and northeast of China. CH_4 leakages from oil and gas systems are located in the north part of China, where oil and gas are mostly produced (Figure 3f). CH_4 emissions from fossil fuels combustion also concentrate in east part of China (Figure 3g and 3i).

25   Figure 4 shows the spatial distribution of the changes of CH_4 emissions from 1980 to 2010. The CH_4 emissions increased in most parts of China, except in western China where there is no significant increase, and in South and Southeast of China where total emissions are decreasing (Figure 4a). The decrease in CH_4 emissions in South and Southeast of China is attributed to a decline in rice cultivation and biomass and biofuel burning emissions, which offsets the increase from other sources in these regions (Figure 4). The increase in CH_4 emissions in North and Northeast of China are attributed to livestock,

30   biomass and biofuel burning, coal exploitation, and landfills. Southwest of China has increase in CH_4 emissions from coal exploitation and landfills (Figure 4).

4 Discussion

4.1 Comparison with other inventories

Figure 2 shows the comparison of CH_4 emissions inferred in this study with EGDARv4.2 (EDGAR, http://edgar.jrc.ec.europa.eu/overview.php?v=42) and EPA (EPA, 2012) inventories. Our estimates of the total CH_4

5   emissions are very close to EPA estimates and 50-90% lower than EDGARv4.2 inventory during the period 1980-2008 (Figure 2a). The CH_4 emissions during 2000-2008 from Regional Emission inventory in Asia (REAS, http://www.nies.go.jp/REAS/) are very close to EDGARv4.2 in China (Kurokawa et al., 2013), so we only compared our estimates with EDGARv4.2 to avoid duplicated comparison. Our estimates during the 2000s are also in better agreement with atmospheric inversions for anthropogenic emissions, which consistently infer smaller emissions in China than

10   EDGAR4.2 (e.g. Bergamaschi et al., 2013, Kirschke et al., 2013). Although the magnitude of the total CH_4 emissions do not agree between EDGARv4.2, EPA and this study, the trends of the total CH_4 emissions from these three estimates are qualitatively similar, confirming the slow increase before 2000 and the acceleration thereafter (Figure 2a). However, the magnitude of the trend of anthropogenic CH_4 emissions after 2000 found in this study (1.3 Tg CH_4 yr^{-2}) and in EPA (0.7 Tg CH_4 yr^{-2}) are 54% and 75% less than in EDGAR4.2 (2.9 Tg CH_4 yr^{-2}). This discrepancy is due mostly to coal exploitation

15   (figure 2e) with smaller contributions from landfills (figure 2h) and oil and gas systems (figure 2f). The slower increase of total CH_4 emissions in China than reported by EDGARv4.2 has already be noticed (e.g. Bergamaschi et al., 2013) and is under investigation by the EDGARv4.2 team (G. Maenhout, pers. Comm.).

In the 1980s, compared with our estimate, higher emissions in EDGARv4.2 are attributed to higher estimates from rice

20   cultivation (8.9 Tg CH_4 yr^{-1}), wastewater (3.6 Tg CH_4 yr^{-1}), biomass and biofuel burning (2.5 Tg CH_4 yr^{-1}), coal exploitation (3.2 Tg CH_4 yr^{-1}), oil and gas systems (~0.7 Tg CH_4 yr^{-1}). In the 2000s, higher emissions from EDGARv4.2 are attributed to higher estimates from coal exploitation (9.1 Tg CH_4 yr^{-1}), rice cultivation (6.0 Tg CH_4 yr^{-1}), wastewater (4.6 Tg CH_4 yr^{-1}), oil and gas systems (1.7 Tg CH_4 yr^{-1}) and biomass and biofuel burning (1.0 Tg CH_4 yr^{-1}). EPA estimates of CH_4 emissions from most source sectors are in line with our estimates, except for fossil fuels combustion and wastewater (Figure 2f & 2i), due

25   mainly to the discrepancy between local and IPCC default EFs (NDRC, 2014; IPCC, 2006).

CH_4 emissions from livestock are the only one to be consistent between the three inventories (Figure 2b). Similar magnitudes of livestock emissions (~10 Tg CH_4 yr^{-1}) are also reported in previous studies (Verburg and Denier van der Gon, 2001; Yamaji et al., 2003; Zhang and Chen, 2014b). The stagnation of livestock emissions after 2000 is explained by the

30   stable domestic ruminant population (CSY, 2012). The increasing import of livestock products (e.g., meat and milk) may help to slow down the increase of domestic livestock population in the 2000s, when the demand of livestock products are increasing in China (http://faostat3.fao.org/).

Yan et al. (2003) reported 7.8 [5.8-9.6] Tg CH_4 yr^{-1} emissions from rice paddies by combining rice cultivation area in 1995 and 204 measurements of CH_4 emission rates from rice paddies with/without organic inputs and intermittent irrigation or continuous flooding. The CH_4 emissions from rice cultivation in China were reviewed by Chen et al. (2013), who found a similar number, 8.1 [5.2-11.4] Tg CH_4 yr^{-1}. NDRC (2014) reported 7.9 Tg CH_4 yr^{-1} emissions from rice paddies in 2005. Our

5 estimates of CH_4 emissions from rice paddies (8.2 [6.5-10.0] Tg CH_4 yr^{-1} during 1980-2010) is consistent with these previous estimates, while the estimates of EDGARv4.2 (15.4 Tg CH_4 yr^{-1}) is out of the range reported by Chen et al. (2013) and our estimates. The large variation of CH_4 emission rates from rice paddies in different regions and different management conditions (e.g., organic and chemical fertilizer inputs, straw application and irrigation) can significantly impact the estimates of CH_4 emissions from rice paddies (Cai et al., 2000; Zou et al., 2005; Chen et al., 2013). This could be the main

10 reason of the higher estimates in EDGARv4.2. The uncertainty of the EFs related to rice practices is still large in China. For example, the exact rice cultivation area with irrigation and rain-fed is not reported at national or province level. The area of rice cultivation received crop straw, green manure, compost and chemical fertilizer and the magnitudes of these organic and chemical fertilizer input are also uncertain (Yan et al., 2003; Chen et al., 2013). But these practices significantly impact the EFs and the total emissions (Huang et al., 1998, 2004; Cai, 2000; Zou et al., 2005). A decrease in CH_4 emissions from rice

15 cultivation is confirmed in all of these inventories, because of the decrease in rice cultivation area and northward of rice cultivation since 1970s (e.g., CASY, 2011; Chen et al., 2013). After 2003, EDGAR4.2 reports a fast increase of rice emissions, which is not found in our study (figure 2c).

For the CH_4 emissions from biomass and biofuel burning, EDGARv4.2 has a two-times larger value than EPA and our

20 estimates in the 1980s (Figure 2d). Previous studies reported 1.9-2.4 Tg CH_4 yr^{-1} emissions from biomass and biofuel burning by the same method but independent estimates of activities data (SNCCCC, 2013; Zhang and Chen, 2014a, 2014b). Tian et al. (2011) conducted emissions inventories of atmospheric pollutants from biomass and biofuel burning during the 2000s in China, and indicated that CH_4 emissions from biomass and biofuel burning increased from 1.9 Tg CH_4 yr^{-1} in 2000 to 2.2 Tg CH_4 yr^{-1} in 2007. Most of CH_4 emissions from biomass and biofuel burning in China are from firewood and straw

25 burning inside of households (Tian et al., 2011; Zhang and Chen, 2014a). The amount of firewood and straw burning have large uncertainty (Yevitch and Logan, 2003; Wang et al., 2013), especially for the time evolution of firewood and straw burning, because they are not easy to accurately deduce without information about utilization of crop residues during the last three decades when fast urbanization happened. In this study, the CH_4 emissions from manure burning in northwest of China (e.g. Tibetan Plateau) are not accounted in biomass and biofuel burning sector in order to avoid double counting as CH_4

30 emissions from manure management are integrated in the livestock sector. However, the fraction of CH_4 emissions from manure burning only account for less than 1% of CH_4 emissions from biomass and biofuel burning (Tian et al., 2011).

Our estimate of CH_4 emissions from coal exploitation (see Table 2 and Figure 2e) is consistent with previous studies and reports (e.g., CCCCS, 2000; Zheng et al., 2005; Cheng et al., 2011; NDRC, 2014; Zhang et al., 2014). For example, CH_4

emissions from coal exploitation was estimated of 8.7 Tg CH_4 yr^{-1} in 1990 (CCCCS, 2000), 6.5 Tg CH_4 yr^{-1} in 2000 (Jiang and Hu, 2005) and 12.2 Tg CH_4 yr^{-1} in 2002 (Yuan et al., 2006). According to reports of the State Administration of Coal Mine Safety (2008, 2009), CH_4 emissions from coal exploitation are 13.8 Tg CH_4 yr^{-1} in 2007 and 14.5 Tg CH_4 yr^{-1} in 2008, respectively (Cheng et al., 2011). On the one hand, the default EFs of underground coal mines (18 m^3 t^{-1} for average, 25 m^3 t^{-1} for high- and 10 m^3 t^{-1} for low- CH_4 coal mines) in IPCC (2006) are higher than the local whole-country-average EFs (21.8 m^3 t^{-1} for high- and 4.5 for low- coal mines in Zhang et al., 2014) (e.g., CCCCS, 2000; Zheng et al., 2005; Zhang et al., 2010, 2014). The higher CH_4 emissions from coal exploitation in EDGARv4.2 could thus result from their higher EFs of coal exploitation if IPCC default EFs are adopted in EDGARv4.2 (Figure 2e). On the other hand, local EFs vary by regions, because of different depths of coal mines, CH_4 concentration and coal seam permeability (e.g., Zheng et al., 2006). These regional EFs of coal mining range from ~20 m^3 t^{-1} in southwest of China and ~19 m^3 t^{-1} in northeast of China, to ~5 m^3 t^{-1} in west, east and north of China (Table 2; Zheng et al., 2006). The depths of coal mines and coalbed CH_4 concentration are regionally variable (Bibler et al., 1998). Regional EFs of coal exploitation should be considered to estimate CH_4 emission as we did in this study, resulting in lower estimates of CH_4 emissions from coal exploitation than that when applying country-average emission factor (Zhang et al., 2014). The EFs of whole-country-average therefore induces a significant bias to estimate CH_4 emissions from coal exploitation (e.g., Zhang et al., 2014). Besides the EFs, the utilization fraction of CH_4 from coal exploitation is another key parameter for estimation of CH_4 emissions (e.g., Cheng et al., 2011). This parameter increased from 3.6% in 1994 and 5.2% in 2000, based upon data of hundreds of individual coal mines (Zheng et al., 2006). The increased utilization fraction of CH_4 from coal exploitation can be an additional reason for the higher value of this source in EDGARv4.2, as we applied this increasing utilization fraction in this study although the time evolution of this parameter has large uncertainty.

Our estimates of CH_4 leakage from oil and natural gas systems are close to estimates of EPA, but much smaller than EDGARv4.2 (Figure 2f). While our estimates of CH_4 emissions from fossil fuels combustion, are close to estimates of EDGARv4.2, but much smaller than estimates of EPA (Figure 2g). NDRC (2014) reported 0.2 Tg CH_4 yr^{-1} leakage from oil and natural gas systems and 0.1 Tg CH_4 yr^{-1} emissions from fossil fuels combustion in 2005, which is consistent with our estimates. Zhang et al. (2014) reported 0.7 Tg CH_4 yr^{-1} leakage from oil and natural gas systems and 0.1 Tg CH_4 yr^{-1} emissions from fossil fuels combustion, which are in the range of our estimates. The activities data applied in these inventories are from national energy statistic data or other global statistic (e.g., CDIAC, IEA), the difference of which is less than 10% (Liu et al., 2015). Thus, the differences in these inventories could come from the uncertainty of EFs. Unfortunately, there is limited information about leakage measurements from pipelines in China, which could help reduce the uncertainty of EFs.

Gao et al. (2006) calculated 1.9-3.4 Tg CH_4 yr^{-1} emissions from Chinese landfills in 2004, using IPCC (1996) default EFs and Tier 1 mass balance method which is not suggested in IPCC (2006). NDRC (2014) reported detailed CH_4 emissions

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from landfills in 2005 (2.2 Tg CH_4 yr^{-1}) using first-order decay method in IPCC (2006) with parameters from inventory of Chinese landfills. These two estimates are consistent with our estimate (Figure 2h and Table 2). Zhang and Chen (2014) reported higher estimates (4.7 Tg CH_4 yr^{-1}) in 2008, using mas balance method with a higher MCF than this study and NRDC (2014). By first-order decay method of IPCC (2006), Li et al. (2015) calculated 3.3 Tg CH_4 yr^{-1} emissions from

5   landfills in 2011, which is the maximum estimates of this study (Figure 2h). CH_4 emissions from landfills in EDGARv4.2 are different with EPA and our estimates in the 2000s, and the trends of CH_4 emissions from landfills are different between EDGARv4.2, EPA and this study (Figure 2h). EDGARv4.2 shows an exponential increase trend of 5-8% yr^{-1} between 1980 and 2010, while EPA shows a smaller trend (<1% yr^{-1}) and this study shows an increase trend of 5-10% yr^{-1} before 2005 and stable emissions after 2005. This is because the fraction of total MSW dumped into landfills decreases with GDP (Figure S1)

10 while MSW is increasingly managed by composting and incineration (CEnSY, 2011). In this study, we considered the amount of MSW managed by landfills and province-level specific fractions of MSW treated by the three types of landfills (Table 2; Du, 2006). Our estimates of CH_4 emissions from landfills still shows large uncertainty after 2000 (20%) because of large uncertainty for fraction of degradable organic carbon in MSW, and the anaerobic conditions of different types of landfills.

15

Both EDGARv4.2 and EPA have 3-4 times higher CH_4 emissions from wastewater than our estimates (Figure 2i). NDRC (2014) reported 1.6 Tg CH_4 yr^{-1} emissions from wastewater in 2005. Zhou et al. (2012) reported 1.3 Tg CH_4 yr^{-1} emissions from wastewater in the 2000s. With the same COD data from CEnSY (2005-2010), Ma et al. (2015) adopted MCF from NDRC (2014) and EFs from IPCC (2006), and they obtained 2.2 Tg CH_4 yr^{-1} emissions from wastewater in 2010. All these

20 estimates do not consider the utilization of CH_4 from wastewater. However, Wang et al. (2011) and Cai et al. (2015) reported a tiny CH_4 emissions (<0.1 Tg CH_4 yr^{-1}) from WTPs in China, and they argued that most COD in wastewater are not removed by anaerobic biological treatments, but by oxidation exposure in WTPs. This suggests that the CH_4 emissions from wastewater could be much lower if most of wastewater is treated by oxidation exposure in WTPs. Our estimates may overestimate CH_4 emissions from wastewater, with limited information of the wastewater treatments in Chinese WTPs.

25 EDGARv4.2 and EPA probably adopted a higher MCF value for WTPs or higher discharged COD in wastewater, resulting in a higher CH4 emissions. The total COD in wastewater reported by CEnSY (2000-2010) rather than estimated by population used in this study may better represent total COD in WTPs and discharged into natural aquatic systems. In addition, the MCF values in Equation (4) for WTPs and for natural aquatic systems are the key parameters for estimating CH_4 emissions from wastewater, and need more samples in future inventory.

30 **4.2 Mitigation of CH_4 emissions in China**

The total anthropogenic CH_4 emission of China is estimated to be 38.9 [30.6-49.4] Tg CH_4 yr^{-1} on average for the 2000s decade. This large source (~7% of the global CH_4 source) offers mitigation opportunities. In the past decade, China has increased the rates of coal-mine methane (CMM) capture and utilization (Higashi, 2009). An amount of ~4 Tg CH_4 yr^{-1}

CMM is captured and ~1 Tg CH_4 yr^{-1} utilized in 2009 (Brink et al., 2013). Under the framework of Clean Development Mechanism (CDM), CH_4 utilization in Chinese CMM increased (Feng et al., 2012; SNCCCC, 2013). So did emission reductions from manure management and landfills. More than 35 million bio-digesters have been built for CH_4 utilization between 1996 and 2010, and capture annually 15 billion m^3 biogas (Feng et al., 2012). The increased utilization of CH_4 in

5   the 2000s suggests a possible overestimation of CH_4 emissions from coal exploitation and manure management in our estimates, because we assumed a conservative utilization fraction for CH_4 from coal mining and manure management (see Section 2.2).

The consumption of natural gas has exponentially grown in China (SNCCCC, 2013). The urban population using natural gas

10   from pipeline network has tripled in the 2000s, and the total length of gas pipes construction has doubled in the past five years with fast urbanization in China (CESY, 2014). Between 1980 and 2010, urban population has tripled in China, and may reach 1 billion in 2050 (UN, 2014). CH_4 leakage from natural gas distribution networks may increase this sector of CH_4 emissions in the coming decades, because of growth of urban population and increase in coverage of natural gas pipes (CESY, 2012).However, substitution of firewood and straw in China by natural gas because of decrease in rural population

15   and increase in usage of natural gas, which could reduce CH_4 emissions from biomass and biofuel burning. With population growth and sustained GDP continues in the coming decades, the CH_4 sources from livestock, MSW and wastewater are predicted to increase (e.g., https://www.globalmethane.org/; Ma et al., 2015). CH_4 emissions from rice cultivation could keep stable because almost stable rice cultivation area since 2005, but may decrease or increase from northward shift cultivation and changes in managements such as organic input and irrigation etc. CH_4 mitigation provides a co-benefit to reduce

20   greenhouse gases emissions and improve air pollution, and energy supply (Shindell et al., 2011). Thus, China has launched a national policy to reduce open burning of crop residues, which cuts down the pollution emissions as well as CH_4 (SNCCCC, 2013). China has also improved CH_4 mitigation within the Global Methane Initiative (GMI) and the framework of CDM on CH_4 mitigation on coal-mine methane, agriculture and MSW (Higashi, 2009; https://www.globalmethane.org/). All of these elements can contribute to reduce CH_4 emissions of China in the coming decades.

25   **5 Summary**

We collected province-level activity data of agriculture, energy and waste and emission factors of CH_4 from the eight major source sectors in Mainland China, and estimated annual CH_4 emissions from each source sector from 1980 to 2010. Our estimates of CH_4 emissions considered regional specific emission factors as much as possible. In the past decades, the total CH_4 emissions increase from 22.4 [16.6-28.1] Tg CH_4 yr^{-1} in 1980 to 45.4 [36.6-58.5] Tg CH_4 yr^{-1} in 2010. The largest

30   contributor to total CH_4 emissions is rice cultivation in 1980, but has been replaced by coal exploitation after year 2005. The increase of CH_4 emissions from coal exploitation and livestock drive the increase of total CH_4 emissions. We distributed the annual province-level CH_4 emissions into 0.5º x 0.5º high-resolution maps for each source sector using different social-

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economic data depending on the sector. These maps can be used as input data for atmosphere transport models, top-down inversions and Earth System Models, especially for regional studies. Our results were compared to EDGAR4.2 and EPA inventories. Good general consistency is found with EPA but our estimates is lower by 38% than EDGAR4.2 and shows slower increase in emissions after 2000 as in EPA.

5

We investigated the uncertainty of CH_4 emissions by using different EFs from published literatures. The EFs should evolve with level of development (e.g., technology for wastewater treatment, evolution of cattle types etc.), however, because of limited information about time evolution of EFs, the emission factors used in this study did not evolve with time. This may cause additional uncertainty for the time series of CH_4 inventory. Besides the uncertainty on emission factors, the activity

10   data and utilization fraction also have their own uncertainty. For example, there is 5%-10% uncertainty energy consumption data in China (Liu et al., 2014). The utilization fraction of CH_4 has limited information and would increase with technology innovation and economic growth. The uncertainty of activity data and utilization fraction China have not been fully investigated in this study, and should be examined in the future study if more information becomes available.

Data availability

15   CH4 inventory (PKU-CH4) in this study is public available on website.

Acknowledgements

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Tables

Table 1. Emission factors (EFs) of enteric fermentation collected from literature and summarized mean, min, max of EFs used in this study. The S1-S6 indicate values collected from references list in the bottom.

		EFs of Enteric fermentation $(kg\ CH_4\ head^{-1}\ yr^{-1})$								
		S1	S2	S3	S4	S5	S6	Mean	Min	Max
		Live								
Dairy cattle	Mature female	78	68	70	48	44	78	64	44	78
	Young (<1 yr)	39	68	38	48	44	40	46	38	68
	Other	52	68	57	48	44	58	54	44	68
Non-dairy cattle	Mature female	64	47	51	48	44	60	52	44	64
	Young (<1 yr)	32	47	29	48	44	35	39	29	48
	Other	66	47	53	48	44	58	53	44	66
Buffalo	Mature female	63	55	68	48	50	88	62	48	88
	Young (<1 yr)	45	55	38	48	50	48	47	38	55
	Other	66	55	57	48	50	68	57	48	68
Sheep	Mature female	14	5	7	5	5	5	7	5	14
	Young (<1 yr)	7	5	4	5	5	7	6	4	7
	Other	9	5	4	5	5	3	5	3	9
Goats	Mature female	9	5	7	5	5	5	6	5	9
	Young (<1 yr)	4	5	4	5	5	7	5	4	7
	Other	5	5	4	5	5	3	4	3	5
Swine	Not divided	1	1	1	1	1	1	1	1	1
		Slaughtered								
Cattle and buffalo		58	53					55	53	58
Sheep and goat		3	5					4	3	5
Swine		3	4					3	3	4

S1: Revised IPCC 1996 Guidelines; Dong et al., (2004)
S2: IPCC, 2006
S3: Yamaji et al., 2003
S4: Verburg & Vandergon, 2001
S5: Khalil et al. 1993
S6: Zhou et al. (2007)

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Table 2. The regional specific Emission factors (EFs) or parameters described in Section 2.2. Mean annual temperature (MAT), Emission factors (EFs) of CH4 emissions from manure management, fractions of burning crop residues, EFs of coal mining, and fractions of municipal solid waste treated by landfills (MSW$_L$) into different types of landfills.

Province	MAT (ºC)	EFs of manure management						Fraction of burning crop residues		EFs of coal mining From underground coal mines (m³ t⁻¹), data from Zheng et al., (2006)			Fractions of MSW$_L$ treated by different types of landfills (%); Data from Du (2006)		
		Dairy cattle	Non-dairy cattle	Buffalo	Sheep	Goats	Swine	Open burning	biomass fuels	Mean	1994	2000	Managed Landfills	non-managed landfills with depth > 5 m	non-managed landfills with depth < 5 m
Beijing	11.0	10.00	1.00	1.00	0.10	0.11	2.00	0.10	0.70	5.58	4.18	6.97	49.2	38.1	12.7
Tianjin	13.6	12.00	1.00	1.00	0.10	0.11	2.00	0.10	0.70	-	-	-	54.2	34.4	11.4
Hebei	9.6	9.00	1.00	1.00	0.10	0.11	2.00	0.20	0.40	5.58	4.18	6.97	41.8	43.7	14.5
Shanxi	8.8	9.00	1.00	1.00	0.10	0.11	2.00	0.20	0.45	5.58	4.18	6.97	2.0	73.5	24.5
Inner Mongolia	4.0	9.00	1.00	1.00	0.10	0.11	2.00	0.10	0.40	5.99	6.00	5.97	25.6	55.8	18.6
Liaoning	7.8	9.00	1.00	1.00	0.10	0.11	2.00	0.20	0.55	13.08	11.75	14.40	23.6	57.3	19.1
Jilin	4.7	9.00	1.00	1.00	0.10	0.11	2.00	0.30	0.30	13.08	11.75	14.40	17.4	62.0	20.6
Heilongjiang	1.4	9.00	1.00	1.00	0.10	0.11	2.00	0.30	0.55	13.08	11.75	14.40	26.3	55.3	18.4
Shanghai	16.5	15.00	1.00	1.00	0.10	0.11	3.00	0.30	0.20	-	-	-	0.9	74.3	24.8
Jiangsu	15.2	14.00	1.00	1.00	0.10	0.11	3.00	0.10	0.80	5.84	5.46	6.22	82.1	13.4	4.5
Zhejiang	16.3	15.00	1.00	1.00	0.10	0.11	3.00	0.30	0.45	5.84	5.46	6.22	33.7	49.7	16.6
Anhui	15.9	14.00	1.00	1.00	0.10	0.11	3.00	0.10	0.80	5.84	5.46	6.22	34.5	49.1	16.4
Fujian	18.5	17.00	1.00	1.00	0.10	0.11	4.00	0.30	0.30	5.84	5.46	6.22	36.8	47.4	15.8
Jiangxi	18.0	17.00	1.00	1.00	0.10	0.11	4.00	0.20	0.45	5.84	5.46	6.22	24.3	56.8	18.9
Shandong	13.5	12.00	1.00	1.00	0.10	0.11	2.00	0.20	0.45	5.58	4.18	6.97	49.5	37.9	12.6
Henan	14.6	13.00	1.00	1.00	0.10	0.11	3.00	0.20	0.30	7.51	7.19	7.83	46.5	40.1	13.4
Hubei	15.7	14.00	1.00	1.00	0.10	0.11	3.00	0.20	0.70	7.51	7.19	7.83	32.8	50.4	16.8
Hunan	16.9	15.00	1.00	2.00	0.15	0.17	3.00	0.20	0.40	7.51	7.19	7.83	62.1	28.4	9.5
Guangdong	21.3	21.00	1.00	2.00	0.15	0.17	5.00	0.30	0.55	7.51	7.19	7.83	61.8	28.6	9.6
Guangxi	20.4	20.00	1.00	2.00	0.15	0.17	4.00	0.20	0.45	7.51	7.19	7.83	27.8	54.1	18.1
Hainan	24.5	26.00	1.00	2.00	0.15	0.17	5.00	0.20	0.45	-	-	-	33.7	49.7	16.6
Chongqing	15.9	14.00	1.00	2.00	0.15	0.17	3.00	0.20	0.70	20.35	19.02	21.68	70.2	22.3	7.5
Sichuan	9.0	9.00	1.00	2.00	0.15	0.17	2.00	0.20	0.45	20.35	19.02	21.68	46.4	40.2	13.4
Guizhou	15.4	14.00	1.00	2.00	0.15	0.17	3.00	0.20	0.40	20.35	19.02	21.68	5.7	70.7	23.6
Yunnan	15.4	14.00	1.00	2.00	0.15	0.17	3.00	0.20	0.20	20.35	19.02	21.68	18.9	60.8	20.3
Tibet	-1.5	9.00	1.00	2.00	0.15	0.17	2.00	0.10	0.20	-	-	-	0.0	75.0	25.0
Shaanxi	10.8	10.00	1.00	2.00	0.15	0.17	2.00	0.20	0.45	5.99	6.00	5.97	0.0	75.0	25.0
Gansu	5.8	9.00	1.00	2.00	0.15	0.17	2.00	0.10	0.55	5.99	6.00	5.97	25.3	56.0	18.7
Qinghai	-2.0	9.00	1.00	2.00	0.15	0.17	2.00	0.10	0.80	5.99	6.00	5.97	58.8	30.9	10.3

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| Ningxia | 8.1 | 9.00 | 1.00 | 2.00 | 0.15 | 0.17 | 2.00 | 0.10 | 0.45 | 5.99 | 6.00 | 5.97 | 24.5 | 56.6 | 18.9 |
| Xinjiang | 6.0 | 9.00 | 1.00 | 2.00 | 0.15 | 0.17 | 2.00 | 0.10 | 0.20 | 5.99 | 6.00 | 5.97 | 0.0 | 75.0 | 25.0 |

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Table 3. Total CH_4 emissions from the eight major source sectors and their total in Mainland China in four snapshot years (1980, 1990, 2000 and 2010). Values are given in Tg CH_4 yr^{-1} (mean [min-max]).

	CH$_4$ sources in China (Tg CH$_4$ yr^{-1})			
	1980	1990	2000	2010
Livestock	6.2 [4.9-7.9]	9.0 [7.1-11.4]	12.5 [10.0-15.5]	12.4 [10.0-15.3]
Rice cultivation	9.3 [7.2-11.4]	9.0 [6.9-11.0]	7.8 [6.2-9.4]	7.4 [6.0-8.8]
Biomass and biofuel burning	1.5 [0.5-2.6]	2.1 [0.7-3.5]	2.1 [0.7-3.6]	2.7 [0.9-4.5]
Coal exploitation	3.4 [3.0-4.3]	6.8 [6.0-8.7]	6.0 [5.4-7.8]	17.9 [17.0-23.9]
Oil and gas systems	0.1 [0.0-0.2]	0.1 [0.0-0.2]	0.1 [0.0-0.3]	0.3 [0.0-0.7]
FF combustion	0.0 [0.0-0.0]	0.0 [0.0-0.1]	0.1 [0.0-0.1]	0.1 [0.0-0.2]
Landfills	0.4 [0.3-0.5]	0.8 [0.5-1.0]	1.6 [1.0-1.9]	2.0 [1.3-2.4]
Wastewater	1.2 [0.6-1.2]	1.2 [0.7-1.3]	1.5 [0.8-1.7]	2.3 [1.2-2.6]
Total	22.2 [16.6-28.2]	29.1 [22.0-37.1]	31.7 [24.2-40.2]	45.0 [36.4-58.3]

FF: fossil fuels

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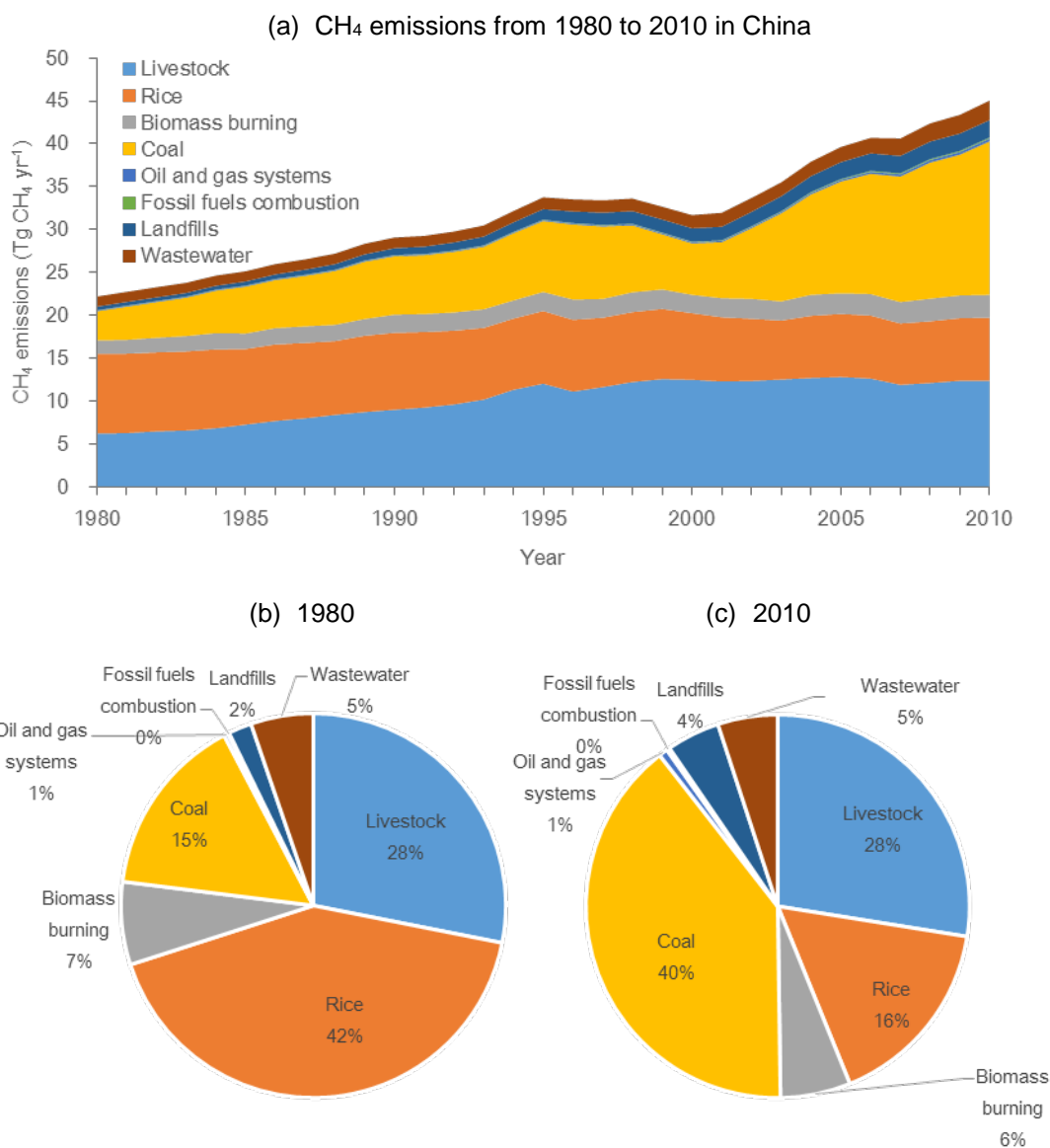and Physics
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Figures

Figure 1. (a) CH_4 emissions from the eight major source sectors during the period 1980-2010 in Mainland China. Pie diagram of CH_4 emissions (%) in (b) 1980 and (c) 2010.

5 Figure 2. (a) Annual total anthropogenic CH_4 emissions in Mainland China, and (b) – (i) CH_4 emissions from different source sectors during the period 1980-2010. The shaded area shows the 95% confidence interval of our estimates.

Figure 3. Spatial distribution of (a) total anthropogenic CH_4 emissions, and (b) – (i) CH_4 emissions from different source sectors in Mainland China in 2010. The unit of the colorbar is g CH_4 m^{-2} yr^{-1}. Note that subplots have different color scale.

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Figure 4. Spatial distribution of changes in (a) total anthropogenic CH_4 emissions, and (b) – (i) CH_4 emissions from different source sectors in Mainland China from 1980 to 2010. The unit of the colorbar is g CH_4 m^{-2} yr^{-1}. Note that subplots have different color scale.

Figure 1: (a) CH4 emissions from the eight major source sectors during the period 1980-2010 in Mainland China. Pie diagram of
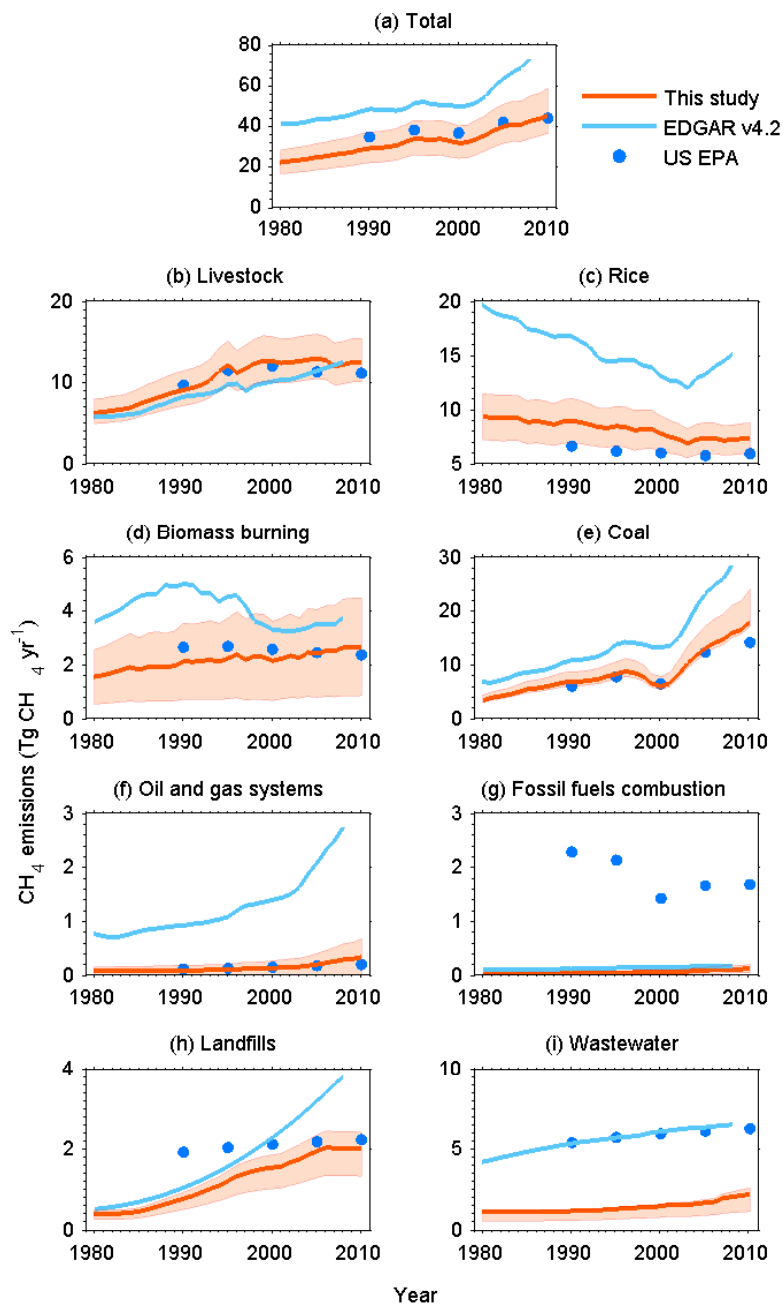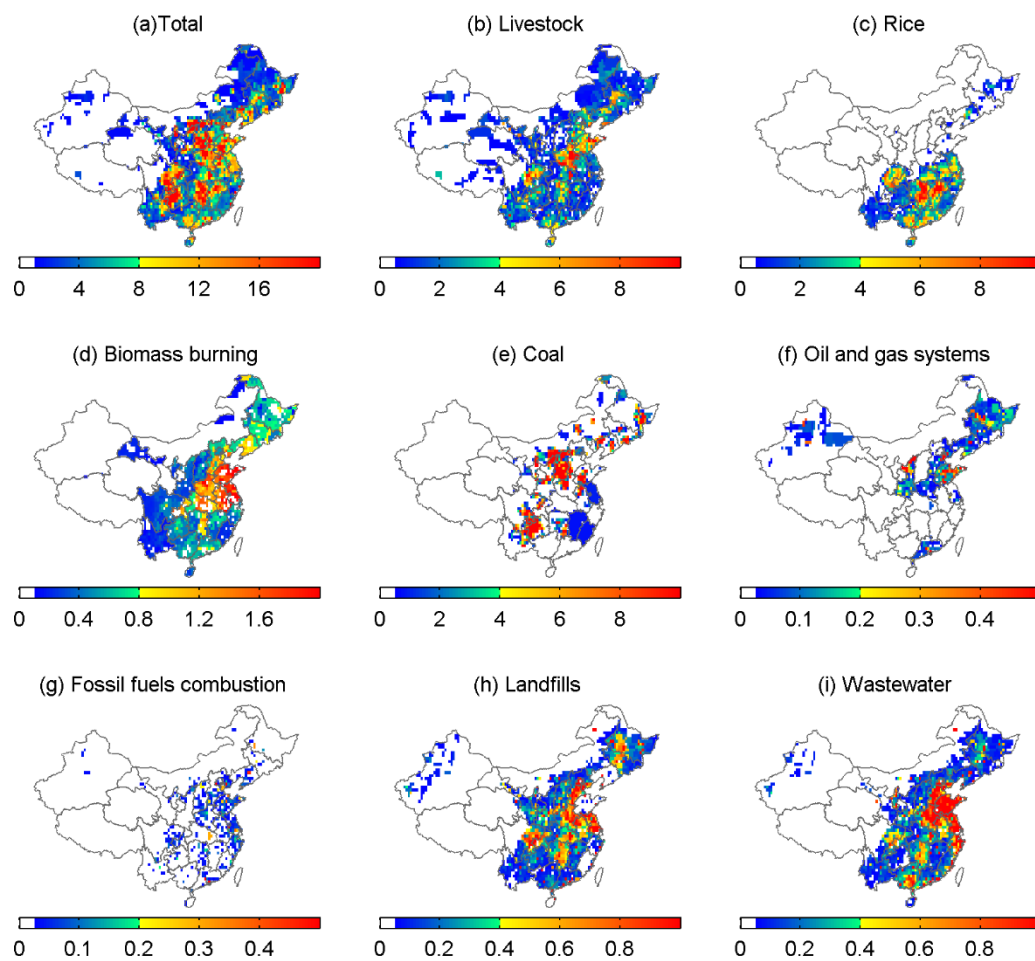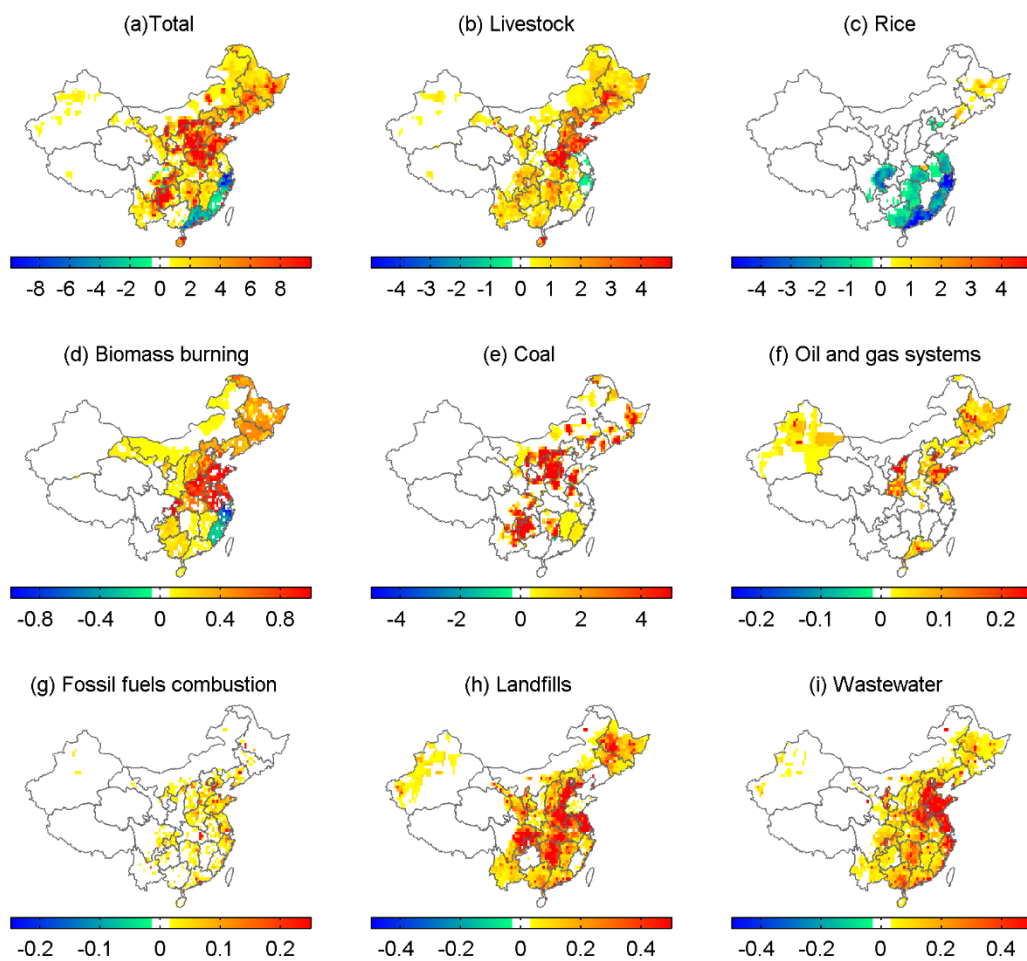CH4 emissions (%) in (b) 1980 and (c) 2010.

Figure 2. (a) Annual total anthropogenic CH₄ emissions in Mainland China, and (b) – (i) CH₄ emissions from different source sectors during the period 1980-2010. The shaded area shows the 95% confidence interval of our estimates.

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Figure 4. Spatial distribution of changes in (a) total anthropogenic CH₄ emissions, and (b) – (i) CH₄ emissions from different source sectors in Mainland China from 1980 to 2010. The unit of the colorbar is g CH₄ m⁻² yr⁻¹. Note that subplots have different color scale.