We thank the reviewers for their careful review and helpful comments. Detailed responses to each comment raised by the reviewers are given in blue as follows.

On behalf of all co-authors,

Shushi PENG

Responses to Anonymous Reviewer #1

"Inventory of anthropogenic methane emissions in Mainland China from 1980 to 2010". 2016-04-13 As the authors rightly point out, there are large discrepancies in existing bottom- up inventories of anthropogenic methane emissions for China and there is scope to improve these using more detailed information on a sector level from both international and national sources. In general, I find this paper a thorough contribution in this genre, which adds to existing inventories by the extensive use of data from national Chinese sources, thereby allowing in several sources to go beyond the use of default IPCC emission factors.

[Response] Thanks for your careful review and valuable comments.

I have, however, some concerns on the emission estimations at the sector level, which I would like addressed by the authors. I list them below by sector. Livestock:

The estimations of CH4 emissions follow standard IPCC methodology, which may explain why emissions fall within a close range of other inventories in Figure 2. I find it somewhat problematic that no adjustment has been made for the use of farm anaerobic digesters in China. I am also surprised that there would be no information available on this when China is since long a world leader on small- scale bio- digesters. I do understand that there is limited information on the number of digesters only digesting manure, since this is not very common. To get reasonable energy efficiency of the digestion, the manure needs to be mixed with at least 20 percent other organic material e.g., straw, food residuals, crop residues. Hence, you would need to look for the number of farm biogas installations that co- digest manure with other organic residues and make assumptions on the fraction of the feedstock that is manure. On p. 15 row 24 you mention that "35 million bio- digesters have been built for CH4 utilization between 1996 and 2010 and capture annually 15 bcm biogas." If the methane content of the biogas is 60%, then it means that 9 bcm CH₄ (or about 6 Tg CH₄) is captured and utilized annually. Although only some fraction of this can be referred to as methane emissions reduced compared to the practice of not treating manure in digesters, it is still likely to be a significant fraction out of the about 10 Tg CH4 estimated to be released from livestock according to Figure 2. As China is one of few countries with a widespread use of rural small- scale digesters, I find it problematic not at all accounting for this effect on methane emissions.

[Response] We agree that some of the biogas recuperation could reduce our estimate of CH4 emissions from manure management. The annual output of biogas data is available from 1996 to 2010 (Feng et al., 2012). The number of household bio-digesters increased from 4 million in the early 1980s to 6 million in 1996. If the number of bio-digesters and the annual output of biogas linearly increased from the early of 1980s to 1996, then the annual output of biogas captured increased from 0.4 Tg CH₄/yr in 1980 to 6.2 Tg CH₄/yr in 2010 (Figure R1), assuming 60% CH₄ in the biogas. However, because the fraction of manure in the mixed organic raw material (mostly mixed with manure and crop residues, or mixed municipal waste) is not clear, several scenarios are needed to estimate how much CH₄ emissions from manure management is mitigated by bio-digesters. The CH₄ production from manure is about 40% of CH₄ production of crop residues in 2012 (Yin, 2015, PhD thesis), and it is assumed that 10%, 15% and 25% of the biogas are low, medium, and high mitigation scenarios for CH4 emissions from manure management, respectively. The biogas reduced CH₄ emissions from manure management by 0.1 [0.0-0.1] Tg CH4/yr in 1980 and by 0.9 [0.6-1.6] Tg CH4/yr in 2010. Compared to the CH4 emissions from manure management without biogas mitigation in 2010 (2.3 Tg CH₄/yr), the biogas reduced ~40% [27%-68%] of CH4 emissions from manure management in 2010. We added this updated CH4 emissions from livestock with biogas accounted in the revised version.



Figure R1. The annual output of biogas (Tg CH_4/yr) from 1980 to 2010. The biogas data is assumed a linear increase from 1980 to 1996. The biogas data from 1996 to 2010 is available from Feng et al. (2012).

Rice cultivation:

The estimation of methane emissions from rice cultivation in China based on Yan et al. 2013 is in my opinion state- of- the- art.

Biomass and biofuel burning:

The estimation of methane from this sector draws on information from several national studies and appears robust. I would however like to know how these estimates compare with existing estimates from satellite images of biomass burning e.g., from GFED. To what extent are the estimates consistent/inconsistent?

[Response] We distinguished crops residues used as biofuels in the houses from those burnt in open fields in our inventory. The emissions from fire detected by satellites only include to some extent (detection of small agricultural fires being problematic) the biomass burnt in open fields. In the latest GFED4.1 products, the average CH₄ emissions including agricultural fires in China during the period 1997-2010 is 0.09 [0.04-0.18] Tg CH₄/yr (http://www.globalfiredata.org/data.html; van der Werf et al., 2010). Our estimation of the average CH₄ emissions from crop residues burnt in open fields during the same period is 0.28 [0.05-0.51] Tg CH₄/yr, which is higher than that derived from GFED4.1. But considering the uncertainty of distinguishing agricultural fire and wild fire in GFED4.1 products and the poor detection of small fires using burned area from space, our estimates are close to the total CH₄ emissions including both wild fire and agricultural fire (0.22 Tg CH₄/yr).

Because the changes in biomass burning in open fields is unknown, the fraction of biomass burnt in open fields to total crop residues is assumed to keep constant from 1980 to 2010 in our inventory. The CH₄ emissions from biomass burning in open fields in our inventory is increasing with crop residues from 1997 to 2010, but GFED4.1 based on burned area data with fixed emission factors for agricultural fires shows no trend of CH₄ emissions. This indicates that the fraction of crop residues burnt in the open fields should be changing in the past two decades, which cause further uncertainty in our inventory. We added this comparison and discussion in the revised version (Page 13, line 11-15).

Coal exploitation:

For this sector, the authors have access to extensive information about depths of coal mines in different provinces as well as the extent of surface mining as opposed to the more common underground mining. This is among the most comprehensive estimates of methane emissions from coal mining in China that I have seen. I have only one question and that is if the authors have been able to assess the prevalence of pre- mining degasification and if the effect of an increasing use of this in China (which is happening according to GMI) in the last decade has been taken into consideration? Is this part of the increased utilization of CH_4 from mines that the authors discuss?

[Response] Yes, the pre-mining degasification is considered as one way of utilization in our inventory. The increased utilization of CH₄ from coal bed methane (CBM) and coal mine methane (CMM) is accounted by the increasing utilization fraction in our study, which increased by 4% in the last decade

(from 5.2% in 2000 to 9.2% in 2010). It is assumed the utilization linearly increases from 1980 to 2010 in our inventory. In the CDM/JI pipeline database (http://www.cdmpipeline.org/overview.htm), the registered and validated projects of CBM and CMM in China started from 2004 and increased strongly in 2007/2008. The total reduction of CH₄ emissions by CBM and CMM in China derived from CDM/JI pipeline database is ~0.3 Tg CH₄/yr in 2006 and ~0.9 Tg CH₄/yr in 2010, which is close to our estimates of increased CH₄ recovery in 2006 (0.4 Tg CH₄/yr) and 2010 (0.8 Tg CH₄/yr). We added the discussion about the utilization of CBM and CMM in the revised version (Page 14, line 15-19).

Oil and natural gas systems:

I find the emission estimations of this sector the weakest point of the paper and I would like the authors to revise the emission estimations for this sector. The authors claim they are using default emission factors from IPCC (2006), but as shown in the Table below, the emission factor used for oil production is only 15% of the very low end of the IPCC default factor for oil production. For natural gas, the emission factor used is close to the very low end of the IPCC default range. I also include for comparison the corresponding emission factors used by the USA and Canada for their reporting to the UNFCCC. Just like the US and Canada, China's oil and natural gas fields are mostly on- shore and therefore likely to have relatively high emissions from unintended leakage (i.e., fugitive emissions from leakage that are not due to venting of associated petroleum gas (APG)). Moreover, NOAA estimates from satellite images of gas flares that China flares between 2 and 3 bcm of gas annually over the period 1994 to 2010. Most of this gas can be referred to flaring of associated gas primarily from oil production. Although there is not much methane being released from the flaring of associate gas as such, the flaring indicates that there is most likely also venting going on. E.g., for the Canadian province of Alberta, Johnson and Coderre (2011) estimate from measurements that out of total APG generated from conventional oil wells, 97% is recovered for reinjection or utilization, 2.1% is flared and 0.8% is vented. If we would assume similar circumstances for oil production in China as for conventional oil wells in Canada, it would mean that between 0.76 and 1.1 bcm APG is vented annually from Chinese oil production. If we assume the methane content of APG to be 85% and use the conversion factor 0.7178 kg CH₄/m3 CH₄, then China would be venting somewhere between 460 and 670 kt CH₄ annually from oil production (which is ten times higher than the authors' estimate for 1990, see Table 1). Adding emissions of unintended leakage would increase this number even further. Similar questions can be raised for the emission factor that the authors is using for gas production, transmission and distribution. It seems unreasonably low. Preferable emissions should be estimated separately for gas production, long- distance gas transmission and distribution networks.

Table 1: Methane emission factors for oil and gas systems. Note the emissions factors used in the reviewed paper for China of 0.36 kg/t for oil and 2.77 g/m3 for gas have been converted to kt CH₄/PJ to facilitate the comparison.

Ch	hina	IPCC (2006) vol.2 Tables 4.2.4 and 4.2.5		USA			Canada	
Re	eviewed							
pa	aper		NIR (2015)	UNFCCC (2	014)	NIR (201	5)	

		1000		1000	2000	2010	1000	2000	2010
		1990	Range of default ets kt CH4/PJ produced	1990	2000	2010	1990	2000	2010
Oil production (EIA, 2015)	PJ	6280		16428	13003	12244			
CH₄ from oil systems	kt CH4	54		1260	1496	1406.52			
Ef oil systems	kt CH4/PJ	0.009	Vented associated gas: 0.056 -0.39	0.077	0.115	0.115	0.25	(conventi	ional oil)
			Flaring associated gas: 0.0001 -0.0	001			0.07	-Quhi	2onv. oil)
			Unintended leakage: 0 -0.11						
			Oil refinery: 0.0006 -0.0015						
			Sum oil systems: 0.057 -0.502						
Natural gas production (EIA, 2015)	PJ	617		19335	20744	23007			
CH4 from natural gas systems	kt CH4	40		7164	7459	6412.76			
Ef gas systems	kt CH4/PJ	0.065	Vented associated gas: 0	0.371	0.360	0.279	0.15	(Qals6	
			production) Flaring associated gas: 0.0	00005	-0.00005				
			Unintended leakage: 0.01 -0.66						
			Gas transmission & storage: 0.004	-0.13					
			Gas distribution networks: 0.024 -0	0.30					
			Sum gas systems: 0.038 -1.09						

Finally, on p.9 row 15, authors mention that the province attribution of emissions from oil and gas systems has been done using GDP. There must surely be information available on the geographical distribution of oil and gas production in China. In particular for oil production, almost all emissions are released during extraction and GDP is not likely to be a good measure for the geographical attribution of these emissions.

[Response] Thank you for carefully checking fugitive emissions from oil and natural gas systems. Compared the default EFs of IPCC (2006) and EFs in Schwietzke et al. (2014a, 2014b), the EFs in the previous version have smaller value. Considering the "realistic" EFs in USA, Canada and other countries from UNFCCC (2014) and Schwietzke et al. (2014a, 2014b) suggested by the Reviewer, in the revised version, we adopted the EFs in Schwietzke et al. (2014a, 2014b) for fugitive emissions from oil and natural gas systems. For fugitive emissions from oil systems, the average EF is 0.077 kt CH₄/PJ (2.9 kg CH₄/m³ oil), and the low and high boundary of EF are 0.058 kt CH₄/PJ (2.2 kg CH₄/m³ oil) and 0.190 kt CH₄/PJ (7.2 kg CH₄/m³ oil), respectively (see Table 1 in Schwietzke et al., 2014a). These values of EF are consistent with the EFs in the table listed by the Reviewer. The fugitive CH₄ emissions from oil systems increase from 0.36 [0.27-0.98] Tg CH₄/yr in 1980 to 0.68 [0.52-1.86] Tg CH₄/yr in 2010.

For fugitive emissions from gas systems, the fugitive emissions rates (FER) of natural gas is decreasing from 1980 to 2011 (Schwietzke et al., 2014b). In China, we adopted the FER linearly decreases from 4.6% (0.81 kt CH₄/PJ) in 1980 to 2.0% (0.35 kt CH₄/PJ) in 2010. This medium FER is close to the EF in 2010 in the above table. The low and scenario of FER in China decreases from 3.9% in 1980 to 1.8% in 2010, and the high scenario of FER in China decreases from 5.7% in 1980 to 4.9% in 2010. The fugitive CH₄ emissions from gas systems increase from 0.45 [0.38-0.56] Tg CH₄/yr in 1980 to 1.27 [1.14-3.11] Tg CH₄/yr in 2010.

We agree that GDP is not likely to be a good proxy for the geographical attribution of fugitive emissions from oil and gas systems. In the revised version, we applied the spatial distribution of EDGARv42 grid maps with spatial resolution of 0.1 degree by 0.1 degree, scaled by the total emissions from oil and gas systems in each province (Schwietzke et al., 2014a). The population density, oil and gas production sites, and other proxies for transportation routes are considered in EDGARv42 grid maps for CH₄ fugitive

emissions from oil and gas systems. Thus, the spatial distributions of CH_4 fugitive emissions from oil and gas systems in the revised version include the geographical distribution of oil and gas production in China (Figure 3), which has better geographical distribution than the GDP proxy in the previous version. With all these changes, we think to have deeply revised the emissions from this sector as requested by the reviewer.

Fossil fuels combustion:

Use of default IPCC emission factors, which seems appropriate.

Landfills:

Use of FOD method, which is the recommended IPCC method. The levelling off of emissions from landfills towards the end of the period (visible in Figure 2) is explained by an increase in composting and incineration. Estimates seem consistent across mentioned studies.

Wastewater:

Estimates emissions from both domestic and industrial sources. No additional comments.

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- Yin, D. The research on regional differentiation of rural household biogas in China and the ratio of raw materials. Dissertation for Doctor Degree, Northwest A & F University, 2015.

Responses to Anonymous Reviewer #2

The paper documents an interesting and unique emissions dataset of methane for China (excluding Hong Kong and Macao) with timeseries 1980- 2010 and gridmaps at 0.5degx0.5deg. This CH₄ inventory is important input in the first place for the 2 National Communications of 10/12/2014 and of 8/11/2012 of China to UNFCCC but also for the Hemispheric Transport of Air pollution Task Force under the CLRTAP and complements there the MIX dataset, documented in Li et al. (2015, ACPD). The dataset is a bit weak on:

 the spatial distribution and could benefit of connecting with Tsinghua University (Q. Zhang) and maybe also with PKU- NH3 (X. Huang) to improve the latter.

2) the temporal resolution which would need to be for the HTAP community at least monthly. The seasonality is in particular important for agricultural sectors, which are the major sectors for CH₄. The dataset could improve on this using the temporal profiles in particular for rice cultivation from large literature by Chinese scientists.

The paper compares its inventory with other emissions inventories of USEPA and EDGARv4.2, but should extend this by considering also the national inventories reported by China in its National communications to UNFCCC. The paper also evaluates the changes of the sector- specific emissions over time, but could be completed with a real trend uncertainty and analysis of the major determinants for these trends (such as CH₄ recovery of coal mining as pushed under the CDM, change in conditions of rice cultivation, etc.).

[Response] Thanks for your review and valuable comments. We tried our best to improve our inventory of CH₄ emissions by 1) with higher spatial resolution (0.1 degree by 0.1 degree) and better proxy for the spatial distribution of CH₄ emissions from each sector, and 2) investigating/discussing uncertainty from mitigations such as bio-digesters, coal bed methane (CBM) and coal mine methane (CMM) from registered CDM database, and conditions of rice cultivation, as well as fugitive emissions from oil and gas systems. For the temporal resolution, it is very difficult to estimate the seasonality of CH₄ emissions for all the eight sectors because 1) monthly activity data is hardly available, and 2) monthly mitigation data by digesters, CBM and CMM is not available. Thus we targeted to have annual CH₄ emissions for all the eight sectors in our inventory, although monthly CH₄ emissions from one or two sectors (rice cultivation etc.) could be investigated.

General comments

The documentation of the dataset could be considerably improved by:

 Giving a full documentation of the sectors covered (maybe making use of the Common reporting format of the UNFCCC reports) and providing also info on what is not included. E.g. what is included in the gas/oil exploitation? Only gas/oil exploration and venting or also the transmission of gas/oil in pipelines, gas distribution networks (very important source, leading to hotspots in cities). What is not included in the coal exploitation? If the emissions of abandoned mines, closed mines are not estimated, this should be mentioned.

[Response] Thanks for the reminder. The fugitive emissions from oil and gas systems include emissions from venting, flaring, exploration, production and upgrading, transport, refining/processing, transmission and storage, as well as distribution networks in this study, which corresponds to IPCC subcategory 1B2. For the fugitive CH₄ from coal mines, the emissions from abandoned mines are not included in our inventory because of the limitation of unavailable data for abandoned coal mines. Except for the emissions from abandoned coal mines, emissions from mining, post-mining, and flaring defined as IPCC subcategory 1B1 are included in our inventory. We added one sentence for the abandoned mines in the revised version.

2) Giving a full documentation of the spatial distribution. References for the geo- spatial proxy datasets are missing.

[Response] We added the references of the proxy data for the spatial distribution (Table S1). In the revised version, we improved the spatial distribution with higher resolution and better proxy data as the reviewers suggested. For livestock sector, we used spatial distribution of number of animals from global livestock production systems (Robinson et al., 2011) as the proxy data. For the sector of oil and gas systems, we used the proxy data from EDGARv4.2 as Schwietzke et al. (2014a). For the other sectors, we used higher resolution of proxy data. We added these details about spatial distribution in the revised version.

 Elaborating more on the intercomparison of inventories, including the UNFCCC National Communications of China and using the uncertainty recommendations of IPCC GL (2006)
 [Response] We already included the comparison between our inventory and the values in 2005 China reported to UNFCCC (Second National Communication on Climate Change of The People's Republic of China (SNCCCC); The National Development and Reform Commission: The People's Republic of China national Greenhouse gas inventory for the year 2005 to UNFCCC, 2014. Beijing, China Environmental Press.). In the revised version, we also included the comparison between our inventory and the values with default IPCC (2006) EFs.

The content of the paper could be enriched by:

1) Addressing the seasonality, in particular of the agricultural activities. Ideally providing monthly gridmaps with full documentation of used temporal profiles.

[Response] It is very difficult to estimate the seasonality of CH_4 emissions for all the eight sectors because 1) monthly activity data is hardly available, and 2) monthly mitigation activity data is not available. Thus we targeted to have annual CH_4 emissions for all the eight sectors in our inventory, although monthly CH_4 emissions from one or two sectors (rice cultivation etc.) could be investigated.

2) When describing the emissions at province level, please mention that Macao and Hong Kong are not included. Please compare the emissions magnitude and emission trends between the different

provinces. Can there be particular shifts of emissions from one province to another be observed over time? How do the emission factors (per unit of activity) vary amongst the different provinces? Maybe also a mapping of the major emission sectors for each province might be interesting.

[Response] We already limited our inventory in "Mainland China" in our title, and our inventory only included the provinces in Mainland China. We added one table for the emissions magnitude for each province in the SI (Table S3). The emission factors for each province are shown in Table 2. The spatial patterns of each source sector are already shown in Figure 3.

3) Highlighting the fact that the database is a fully consistent bottom- up database with activity data and with recovery (correction factor), which allows to conclude for the trend analysis on the determinant factors of some CH₄ mitigation measures (e.g. CH₄ recovery of coal mining, also CH₄ recovering of the gas/oil exploitation, waste separation, ...) with the effect they had on the emissions of China. Please derive which reduction potentials further exist.

[Response] In this study, we would like to give a full documentation of bottom-up inventory for anthropogenic methane emissions in China. Although the future reduction potentials are interesting, we would like to focus on the past three decades in this study and simply mention the possible reduction potentials. Quantitatively estimates of the reduction potentials of CH₄ emissions could be investigated in future study.

4) Discussing an outlook on how to maintain and update the database, at which frequency, using which data sources.

[**Response**] This database will be regularly updated every two or three years, depending on the availability of activity data.

Specific comments

) English could be improved: p.1 113 "have", 114 "contribute"; p4, 118 "are"; p5, 118: remove
"emissions", p7, 19 there are few measurements, p12 126 "and northward of" needs to be corrected; p13, 16, "Yevich", p14, 11 "and 5.2%" should be "to 5.2%"; p.17 113 "publicly"
[Response] They are corrected.

-) abstract: please mention that it is an ANNUAL bottom- up inventory [**Response**] It is added.

-) page 2 line 22: is the 2010 number of EPA reported/calculated or projected. If it is the latter, please make the difference between reported/calculated data and projected data. Also in fig. 2, make the distinction by have e.g. open circle for projected data.

[**Response**] The EPA 2010 data are projected. Following your suggestion, we used open circle for the 2010 EPA data in Figure 2.

-) page 3: instead of mentioning "English and Chinese literature", please give the real list of references (and mention the language in the reference list).

[Response] We listed the detailed reference list for each sector in the section 2.

-) page 3: formula: what do you mean exactly with "conditions". Do you mean "technologies/ practices, modi operandi"? Moreover: why is the EF not varying in time but only the correction factor? [**Response**] The conditions have different meanings for different sectors. For example, condition means underground/surface mines for coal mines, practices/managements (organic input, continuous irrigation) for rice paddy, and burning in households/open field for biomass burning. This word ("conditions") comes from IPCC guidelines vol. 2 (2006). We described the details for EF selections for each sector in the following text. The EF should be varying in time, but little information about time evolution of EF can be available. Thus, we kept the EF unchanged from 1980 to 2010. For the correction factor, normally it correlates with the improvements of technology/practices, or economy development. We can infer the varying correction factor from these indirect indicators. In the revised version, we revised the EF is varying with time in Equation (1), but mentioned that we used constant EFs through the period 1980-2010. "Note that the EFs used in this study did not evolve with time because of limited information about time evolution of EFs." (Page3, line 27)

) page 4: "CH₄ utilization or flaring"? You mean the "CH₄ recovery instead of venting into the atmosphere"? Please use the standard reporting language (as also in the CDM)
 [Response] Yes, it is corrected.

-) page 4 – Table 1: enteric fermentation is (as described in the IPCC GL (2006)) depending for the dairy cattle on the milk production per head and for the non- dairy cattle and other cattle on the live weight per head. These details would be of interest, also complementing the info in the IPCC GL (2006). **[Response]** We agree that the enteric fermentation depends on the live weight for the non-dairy cattle and milk production for the dairy cattle. In IPCC (2006), the average milk production for the dairy cattle in Asia is 1650 kg/head/yr, and the live weight of non-dairy cattle in China is 300-400 kg/head (Table 10A.2 in IPCC, 2006), which are from the revised 1996 IPCC Guidelines for GHG inventories (IPCC, 1997). Dong et al. (2004) applied the same live weight for non-dairy cattle as IPCC (2006), but higher milk production for the dairy cattle (4000-5000 kg milk /head/yr) for the enteric fermentation, which has a higher EF of female dairy cattle (Table 1). Yamaji et al. (2003), Verburg & Vandergon (2001) and Khalil et al. (1993) referred or adjusted the EFs from previous studies (Dong et al., 2000; Ward and Johnson, 1990; Crutzen et al., 1986) or IPCC guidelines without giving the information of milk production and live weight for the EFs. In the past three decades, the EFs of enteric fermentation could change with milk production for the dairy cattle and live weight for the non-dairy cattle in China. The constant EFs through the past three decades in this study may underestimate the increasing trend of enteric fermentation, because of the increasing milk production per head of dairy cattle and increasing live weight per head non-dairy cattle during the past three decades in China. On the other hand, the increasing weight of crop production feeding system in livestock and increasing feed of treated straw or

residues would reduce the EFs of enteric fermentation (Dong et al., 2004). We discussed the information of milk production and live weight and the uncertainty of possible varying EFs for livestock in the revised version. "Besides the uncertainty of population, the EF of livestock are highly correlated to the live weight per head for meat cattle and milk production per head for dairy cattle (Dong et al., 2004; IPCC, 2006). In this study, as the previous studies, we assumed the EF did not evolve with time because of limited information about the weight distribution of livestock population types besides numbers of animals, although we assessed the uncertainty with different EFs (Table 1). On the one hand, the increasing live weight and milk production per head can increase EFs of enteric fermentation (IPCC, 2006). On the other hand, the increasing weight of crop production feeding system in livestock and increasing feed of treated crop residues can reduce the EFs of enteric fermentation (Dong et al., 2004). The possible changing EF resulting from increased live weight and milk production per head or more feed with treated crop residues should be investigated in a future study." (Page 12, line 5-15)

-) page 5: What do you mean exactly with "biomass burning"? Only small scale or also forest fires, etc. ? Moreover, in formula 2: Why do F and theta not have the index C?

[Response] Biomass and biofuel burning includes firewood and crop residues burning as biofuel in rural households, as well as disposed crop residues burning in open field. The CH₄ emissions from wildfire of natural ecosystems (forests, grasslands etc.) are not included in our inventory, because this inventory only includes the anthropogenic methane emissions. The total CH₄ emissions from wildfire of natural ecosystems in GFED4.1s during 1997 to 2010 is 0.11 Tg CH₄/yr, which is less than 5% of CH₄ emissions from biofuel burnt in households. In formula 2, theoretically, F and θ could be different for different crop residues. However, limited information of F and θ can be available. Thus, we assumed the F and θ are the same across the six types of crops in formula 2.

-) page 6: in e.g. UK we see huge differences in EF for the fugitive emissions from coal mines, because of different geological underground (based on measurements). Is Zeng et al (2006) for China, a much larger country than UK not reporting a similar large variety?

[Response] Zheng et al. summarized regional EFs for six regions (North, Northeast, Northwest, Southwest, Center and South, East of China), which are shown in Table 2. The fugitive EF for coal mines may have large variety across the country because of different coal bed methane store and different coal mining depth in the coal mines, but is not detailed in Zheng et al. (2006). We used the varied regional average EFs from Zheng et al. (2006) to estimate fugitive emissions from coal mines.

-) page 6: Have emissions estimates from abandoned mines, closed mines been omitted?

[Response] On one hand, high moisture in coal strata in China could inundate the abandoned mines, and inhibit the CH_4 emissions from abandoned mines. One the other hand, permeability of coalbed in China is small (~0.001 mD), which indicate the limited CH_4 emissions from abandoned coal mines. Because 1) the emissions from abandoned mines are less than 1% of total emissions from coal mining (NRDC, 2014), and 2) the time series of numbers and locations of the abandoned mines are unavailable (NRDC, 2014), emissions from abandoned mines are not included in our inventory. This is clarified in the

revised version (Page 6, line 27-30).

-) page 6 – Table 2: please specify the CH_4 recovery of coal mining gas in the table per province. Please add to the Table also the rice cultivation per province and reflecting as such the difference in cultivated area times the number of cropping seasons. This would be valuable information that adds to the information at Chinese province level in the IPCC GL2006)

[Response] The national CH₄ recovery of coal mining gas in 1994 and 2000 is reported in Zeng et al. (2006). The database of CDM projects only reported 13 registered coal bed/mine methane before 2009. Thus, we assumed that national average value for CH₄ recovery of coal mining gas for each province. For the rice cultivation, the total early, middle and late rice cultivation areas for each province are collected from agriculture statistics yearbooks, and vary year by year. We do not think that it is a good idea to put the yearly varying rice cultivation area of each province in Table 2, otherwise Table 2 is a too "big" to read.

-) page 7 17: please specify the EFs in kg CH_4 per TJ instead of per kton oil or per m3 gas, because the heat value can change significantly between the different types of oil and different types of gas. Please have an evaluation of the gas distribution leakage. Even though Lelieveld et al (2005, Nature) did not found large leakages from transmission pipelines, it is well- known that the gas distribution networks (especially of the old steel pipeline networks in older cities) are subject to large leakages.

[Response] For the EFs of fugitive emissions from oil and gas systems, based on the EFs in UFNCCC (2014) and Schwietzke et al. (2014a, 2014b), in the revised version, we adopted the EFs in Schwietzke et al. (2014a, 2014b) for fugitive emissions from oil and gas systems. For fugitive emissions from oil systems, the average EF is 0.077 kt CH₄/PJ (2.9 kg CH₄/m³ oil), and the low and high boundary of EF are 0.058 kt CH₄/PJ (2.2 kg CH₄/m³ oil) and 0.190 kt CH₄/PJ (7.2 kg CH₄/m³ oil), respectively (see Table 1 in Schwietzke et al., 2014a). These values are consistent with the EFs in the table listed by the Reviewer#1. The fugitive CH₄ from oil systems increase from 0.36 [0.27-0.98] Tg CH₄/yr in 1980 to 0.68 [0.52-1.86] Tg CH₄/yr in 2010.

For fugitive emissions from gas systems, we used the fugitive emissions rates (FER) to estimate the fugitive CH₄ from gas systems (Schwietzke et al., 2014a, 2014b), including venting, flaring, exploration, production and upgrading, transport, processing, transmission and storage, as well as distribution networks. The gas distribution leakage is included in our inventory. The fugitive emissions rates (FER) of natural gas is decreasing from 1980 to 2011 (Schwietzke et al., 2014b). For China, we adopted the FER linearly decreases from 4.6% (0.81 kt CH₄/PJ) in 1980 to 2.0% (0.35 kt CH₄/PJ) in 2010. The low and scenario of FER in China decreases from 3.9% in 1980 to 1.8% in 2010, and the high scenario of FER in China decreases from 5.7% in 1980 to 4.9% in 2010. The fugitive CH₄ emissions from gas systems increase from 0.45 [0.38-0.56] Tg CH₄/yr in 1980 to 1.27 [1.14-3.11] Tg CH₄/yr in 2010.

-) page 8, 12: Is the China Env. Stat. Yearbook not showing differences in practices between large versus small or young versus new cities?

[Response] No.

-) page 9, 17: please map carefully in a table for each (sub-)sector the specific proxy datasets (over time) are used; page 9, 114 why is livestock distributed with agricultural gross domestic product and GDP and not with the maps of animal numbers, as available from the geonetwork at the FAO site? Why is the oil & gas distributed with GDP, if there are data available on oil and gas exploitation from NOAA? Why considering only 414 coal exploitation sites, if Liu et al (2015, Nature) has a map of several thousand sites. The two- step distribution as described in lines 19- 20 should be used for all (sub-)sectors. **[Response]** We added the details for proxy datasets for the spatial mapping (Table S1). In the revised version, we used gridded maps of animal number as the proxy data for CH₄ emissions from livestock, instead of agricultural GDP. For the geographical attribution of fugitive emissions from oil and gas systems, instead of GDP, we applied the spatial distribution of the EDGARv42 gridded maps for fugitive emissions from oil and gas systems with spatial resolution of 0.1 degree by 0.1 degree, scaled by the total emissions from oil and gas systems in each province (Schwietzke et al., 2014a). The population density, oil and gas production sites, and other proxies for transportation routes are used in EDGARv42 to distribute those CH₄ fugitive emissions from oil and gas systems. For coal exploitation, we used data from 414 counties in the previous version, not 414 sites. In each county, there are probably several hundreds of coal production sites. To get a higher spatial resolution, we used the location of 4264 coal production sites from Liu et al. (2015) as proxy data in the revised version. We summed the total annual coal production in each grid of 0.1 degree by 0.1 degree as the weight to distribute the total CH₄ emissions from coal mines in each province.

-) page 10, 18: please carefully derive when the acceleration in CH_4 emissions start, definitely after 2000, but can we even say in 2002 when China joined the WTO?

[Response] We applied piecewise linear regression on the time series of total CH₄ emissions, and found that the acceleration in CH₄ emissions starts from 2002 (the trends of total CH₄ emissions before and after 2002 are 0.5 Tg CH₄/yr² and 1.3 Tg CH₄/yr², respectively), which is attributed to the acceleration in CH₄ emissions from coal mining after 2002 (the trend of CH₄ emissions from coal mining from 2002 to 2010 is 1.1 Tg CH₄/yr). This could be related to remarkable achievements in economic index, when China joined WTO starting from December, 2001.

-) page 12, 116: Seen the relative large variation in rice emissions over time (in EDGARv4.2 varying from 19.2 to 11.9 Tg CH₄/yr), please compare the emissions of the same years: so the 2005 value of 13.2 Tg CH₄/yr with the NDRC value of 7.9 Tg CH₄/yr and with the Chen (2013) estimate of ... in 2005. [**Response**] It is revised. Note that Chen et al. (2013) compiled all sites data measured in different years and used rice cultivation area in 2008 to estimate the methane emissions from rice cultivation.

-) page 13, 11: maybe a discrepancy can be found in the definition of "biomass burning". Please have a careful look what is included: vegetal waste burning, agricultural waste burning, crop residue burning, field burning, grassland fires, woodland fires, forest fires, ...?

[Response] Biomass and biofuel burning includes firewood and crop residues burnt as biofuel in rural households, as well as disposed crop residues burnt in open field. The CH_4 emissions from wildfire of natural ecosystems (forests, grasslands etc.) are not included in our inventory, because this inventory only includes the anthropogenic methane emissions. We added the definition of "biomass and biofuel burning" in the method section 2.2.3.

) page 14: 13: EDGARv4.2 uses the CDM of UNFCCC as input for all developing countries on coal mine gas recovery (cfr. IEA's CO2 from fuel combustion book, part III, GHG).
[Response] We also considered the increased utilization of CH₄ from coal bed methane (CBM) and coal mine methane (CMM) as accounted for by the increasing utilization fraction in our study (Page 7, lines 1-5), which increased by 4% in the last decade (from 5.2% in 2000 to 9.2% in 2010). Please also see the

above response to the CH4 recovery from coal mines.

-) page 16: please give a quantitative evaluation of the mitigation measures and an outlook on the further reduction potential based on the references. Page 16, l6: please evaluate carefully that new PVC gas distribution networks are better than the old steel networks and that new transmission pipelines (such as for the connection Russia and China) are not expected to lead to high leakages. Input on these issues can be gained also from the Chapter 5 of the AMAP report on CH_4 from Hoeglund- Isaksson et al. (2016)

[**Response**] Please see the above response to quantitatively estimate the reduction potentials. In this study, we assumed that the fugitive emissions rates (FER) from natural gas systems linearly decreased from 1980 to 2010 because of reduced unintended leakage and technically leakage control. In 2010, the FER in China is 2.0%, including production emissions and the leakage from natural gas production and distribution networks. We agree that PVC pipeline is better than the old steel pipeline. In the 2000s, the networks of natural gas distribution in cities of China are PVC pipelines. If China follow the rates of maximum technically feasible reduction potentials in Höglund-Isaksson et al. (2015), the leakage of long-distance gas transmission could be reduced by 60% and the total fugitive emissions from oil and gas systems can be reduced by 58% in 2030.

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Responses to Anonymous Reviewer #3

The paper provides a consistent time series of CH₄ emissions from China from 1980-2010. China is an important contributor to total global CH4 emissions and a better understanding of the sources and possible mitigation options is relevant for the scientific community. Methane emission inventories for China have been made before and as such the work is not novel but the compilation of the different sources and the consistent time series make it certainly worthwhile. Also, as discussed in the paper, the discrepancies between various existing estimates for China is substantial and the investigation of the causes or at least identification of sectors that are most uncertain is important for both the global and the Chinese CH₄ budget. I think that for several sources the review of emission factors and especially possible trends in these emission factors or the emission controlling variables over the time period could be more in-depth and that this could still further improve the inventory. On the other hand, an inventory includes many sources and a balance between total time spend on each category and the overall result needs to be found. I would recommend the paper for publication but would like to see several points discussed in more detail or added. If for some reason the authors find it unrealistic or over-demanding to make those changes, some argumentation why this is not feasible or out of scope should be provided. [Response] Thank you for your valuable and constructive comments. We revised the manuscript following your suggestions. The details can be found as below.

First of all, as lined out in the beginning of section 2.1., the methods of the IPCC GHG inventory guidelines were followed. The authors then search and use for several, but not all, sources more representative Chinese emission factors. I think it would be valuable to also have a full IPCC emission factor only emission calculation, next to the final result of the paper. This is 1) the easiest way to understand what the impact of the country specific emission factors (EFs) on the total Chinese emission estimate is. 2) In the comparison with the other sources such as EDGAR or EPA – again it would be very useful to know if these estimates' are in line or higher / lower than using avg IPCC EFs. Since the structure followed by the authors is based on the IPCC methodology, my feeling is making an "base-line" avg IPCC EF calculation is not a very demanding task. There are good arguments why the current approach is more accurate but it would provide a very useful benchmark for comparing the impact of more detailed information as well as in the comparison with EDGAR and EPA values. [Response] Following your suggestions, we added the estimates with IPCC default EFs (see Figure 2 and Table S1). In Figure 2, we added the lines for estimates with IPCC EFs and its high/low bound range.

An important aspect of the paper is the long time series. Something that is not well discussed is whether the activity data and emission factor data really cover the temporal changes. For example if the emission factors are based on using a certain technology but this technology was not used before 1990, the EF might not be representative for the 1980-1990 period. While there are good reasons to use it as best guess, the trend 1980-1990 is then highly uncertain and much less reliable than 1990-2010. I would like to discuss that in more detail for the CH₄ emission from rice agriculture.

[Response] Thanks. We discussed the uncertainty of changes in EFs on the trend of emissions in the revised version. For the rice paddy sector, the details about the changes in EFs can be found in the next response.

CH₄ emissions from rice agriculture

In section 2.2.2 the authors explain their approach to calculate CH₄ emissions from rice. While it is clearly acknowledged in the paper that the emission factors depend on such things as organic matter (OM) input and water management, no trends in these controlling factors are discussed. Denier van der Gon and Neue (1995) and Denier van der Gon (1999) have provided a simple, empirical impact relationship for CH4 emission from rice fields with OM input versus chemical fertilizer. A ~5 t OM/ha input creates a doubling of the CH4 emission, a 10 t OM/ha triples the CH4 emission. Peng et al use an assumption based on Yan et al (2003) that 50% of the rice paddies received organic input. While that may be the case at a certain moment in time for the trend in CH4 emission it is crucial to understand the trend in the OM input because it is such a strong driver of CH₄ emissions from rice fields. Denier van der Gon (1999) compiled the green manure statistics, fertilizer production and harvested rice area statistics in China over the period 1960-1995. Especially from the mid-1970s onwards the production of fertilizer in China grows tremendously but the harvested rice area remains the same or declines somewhat. It is a logical hypothesis that the every year increasing availability of fertilizer (urea) started replacing the much more labor-intensive use of OM incorporation. While reliable statistics for total OM use are lacking, the green manure statistics support this hypothesis. From 1980-1990 the harvest rice area slowly declines, the fertilizer production rapidly increases and the planted green manure area roughly halves. The green manure statistics are available at the regional level and show for example a much stronger impact in the Central and east China (See Figure 3 and table 1 in Denier van der Gon, 1999). The impact of less OM input in the rice field is further enhanced by the change of rice varieties from traditional to high yielding varieties. The main trait of these high yielding varieties is that they are very responsive to N fertilizer and allocate (or invest) a much smaller part of their total net primary production in the below ground root system (which will be the OM for the next growing season). This trend is described by Denier van der Gon (2000) but that paper does not give data for China nevertheless the high yielding varieties have also been introduced in China and it will also have contributed to making less OM available for CH4 production in Chinese rice soils. This reviewer would therefore argue that the trend for CH₄ from rice as shown in table 3 of the paper, strongly underestimated the trend between 1980 and certainly 1990. An educated guess would be that the year 2000 value is realistic and in line with most available estimates as discussed by the authors but the emissions from rice should show a declining trend in emission since 1980 mostly due to lower OM input into the rice cropping system which is in line with the strong growing availability of urea fertilizer. The authors could use the trends and data compiled in Denier van der Gon 1999 or references therein which would result in a CH₄ emission from rice cultivation in an estimated range of ~15 Tg/yr. As a result the trend would be rather similar to EDGAR (Fig 2 in the paper), although the absolute emission level remains

lower. Indeed, as mentioned by the authors, the increasing trend in the EDGAR estimate after 2003 is remarkable and not easily understood but that is outside of the scope of the paper.

[Response] Thank you for this constructive comment. We fully agree that the decline of rice paddy area with OM input since late of the 1970s can decrease the CH_4 emission from rice paddy. In the revised version, we tried to include the estimates of CH_4 emission from rice paddy with changing OM input.

In China, OM input includes animal and human wastes, crop straw, green manure and compost and fermented residues. As discussed in Yan et al. (2003), there is little statistic data about the fraction of rice paddy area with OM input as well as the amount of average OM input per hectare (Denier van der Gon, 1999). The only clear message is that the planted area of green manure decrease from 1976 to the late of the 1980s (Denier van der Gon, 1999; China Agricultural Statistic Yearbook), but how much green manure is grown for rice paddy is unclear. Here, several assumptions are applied to get the changes of area of rice paddy with OM input. First, it can be assumed that 100% of rice paddy received OM input before the chemical fertilizer input, since OM input has long history in China (Denier van der Gon, 1999)., Second, we assumed that the area of rice paddy with OM input linearly decreased with the amount of chemical fertilizer input, because most of OM input are labor-intensive and farmers prefer more profitable work in allocating their time rather than preparing OM input for the fields. In the year of 2000, the total chemical fertilizer consumed in China is 41.5 million ton (China Statistic Yearbook, 2001), and 50% of rice paddy with OM input suggested as Yan et al. (2003). Thus, the area of rice paddy with OM input decreased by 1.2% per million ton chemical fertilizer. From 1980 to 2000, the total chemical fertilizer utilization increased from 12.7 million ton to 41.5 million ton (all cultivation types, Figure R1), and the fraction of rice paddy area with OM input decreased from 85% in 1980 to 50% in 2000. After 2000, on one hand, the chemical fertilizer kept increasing (Figure R1); on the other hand, the practice of returning crop residues and using organic fertilizer applications are popularized again because of sustainable quality of arable land and air quality control, which can be indirectly supported by increasing number of the machines for returning crop residues in the 2000s (from 0.44 million in 2004 to 0.62 million in 2011). Thus, in absence of more detailed information we have assumed that the fraction of rice paddy with OM kept stable after 2000. Based on the changing fraction of rice paddy with OM input, CH₄ emissions from rice paddy decreased by 3.4 Tg CH₄ yr⁻¹ (44% of CH₄ emission from rice paddy), compared to 1.6 Tg CH₄ yr⁻¹ with constant fraction of rice paddy with OM input during the period 1980-2010. We used this estimate with the inferred changing fraction of rice paddy with OM input in the revised version, which could correct the underestimated trend of CH₄ emission from rice paddy between 1980s and 1990s (Figure R2). Besides the changing OM input, the fraction of rice paddy with continuous irrigation may also changes. But without information of irrigation on rice paddy, we cannot deduce the impact of possible changing irrigation on CH₄ emissions from rice paddy. We also discussed the uncertainty from practice of irrigation in the revised version.



Figure R1. The total chemical fertilizer input from 1980 to 2010 in China.



Figure R2. CH_4 emissions from rice cultivation in the previous and revised version.

CH₄ Emissions from Oil and Gas industry

Emissions from natural gas production sites are characterized by skewed distributions, where a small percentage of sites-commonly labeled super-emitters-account for a majority of emissions. (Zavala-Araiza et al., 2015). The importance of these super-emitters in the O&G sector is a rather new insight and probably not well represented in the current emission factors. It only surfaced due to large numbers of measurements that showed the "fat tail distribution" of the EFs. Therefore, I would argue that using standard emission factors may well lead to underestimation for the emissions from this sector. Moreover, the emission factors used in the paper appear really low. I would like to see a very simple "sanity check" on these numbers. When taking the total calculated CH₄ emission from the Oil and Gas industry for example in 2000 (0.1 Tg / yr) or 2010 (0.3 Tg/yr) (see Table 3 in the paper); what share is due to the gas industry and what percentage of total natural gas production is this? And does it make sense over time? At a first glance it seems a really low estimate that is presented here. To get a feeling I have taken the data from Schwietzke et al., 2015 and looked at the CH4 emissions from china from Natural gas industry only if a Fugitive Emission Rate (FER) of 1% is assumed (see figure below). This leads to a factor 2 higher emissions than reported by Peng et al. and the gap is much bigger because in the below estimate oil industry is not included whereas Peng's estimate includes both oil and gas. While this does not mean that the presented estimated in the paper is wrong, I would like to see more discussion and think that expressing the FER as a % of the production is a very useful thing to do to show that really low % are currently assumed in this paper whereas recent measurements in the US and Canada found FER's of 2-4% more realistic.

Constant global avg. Fugitive Emission Release (FER) of 1% of natural gas production only: data taken from Schwietzke et al., 2014. The figure does not include the oil sector emissions yet but these are available from Schwietzke et al and would further increase the emission estimate.



[Response] Thank you for pointing out this possible underestimated EFs for fugitive emissions from oil and natural gas systems. Comparing the default EFs of IPCC (2006) and EFs in Schwietzke et al. (2014a, 2014b), the EFs from Zhang and Chen (2010) and NDRC (2014) in the previous version have smaller values. Considering the EFs in USA, Canada and other countries from UNFCCC (2014) and Schwietzke et al. (2014a, 2014b), in the revised version, we adopted the EFs in Schwietzke et al. (2014a, 2014b) for fugitive CH4 from oil and natural gas systems (see reply to Reviewer #1). For fugitive emissions from oil systems, the average EF is 0.077 kt CH₄/PJ (2.9 kg CH₄/m³ oil), and the low and high boundary of EF are 0.058 kt CH₄/PJ (2.2 kg CH₄/m³ oil) and 0.190 kt CH₄/PJ (7.2 kg CH₄/m³ oil), respectively (see Table 1 in Schwietzke et al., 2014a). These values are consistent with the EFs in the table listed by the Reviewer#1. The fugitive CH_4 from oil systems increase from 0.36 [0.27-0.98] Tg

CH₄/yr in 1980 to 0.68 [0.52-1.86] Tg CH₄/yr in 2010. 10

For fugitive CH₄ from natural gas systems, the fugitive emissions rates (FER) of natural gas is decreasing from 1980 to 2011 (Schwietzke et al., 2014b). For China, We assumed a FER linear decrease from 4.6% (0.81 kt CH₄/PJ) in 1980 to 2.0% (0.35 kt CH₄/PJ) in 2010, which is today close to the FER (1.9%) in OECD countries in 2010. The

- 15 range of uncertainty was estimated with a scenario assuming a low FER in China decreasing from 3.9% in 1980 to 1.8% in 2010, and a scenario with high FER in China decreasing from 5.7% in 1980 to 4.9% in 2010. The fugitive CH₄ from natural gas systems increased from 0.45 [0.38-0.56] Tg CH₄/yr in 1980 to 1.27 [1.14-3.11] Tg CH₄/yr in 2010.
- The total CH₄ emissions from oil and natural gas systems increase from 0.81 [0.65-1.54] Tg CH₄/yr in 1980 to 1.95 20 [1.66-4.98] Tg CH₄/yr in 2010, which is consistent with the values from Schwietzke et al. (2014a) and is lower than EDGARv42, but higher than the values reported by NDRC (2014). In the revised version, we also applied the spatial distribution of EDGARv42 grid maps with spatial resolution of 0.1 degree by 0.1 degree, scaled by the total emissions from oil and gas systems in each province (Schwietzke et al., 2014a), which could have better geographical distribution than the GDP proxy used in the previous version.
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Inventory of anthropogenic methane emissions in Mainland China from 1980 to 2010

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Abstract. Methane (CH₄) has a 28-fold greater global warming potential than CO₂ over one hundred years. Atmospheric CH₄ concentration has tripled since 1750. Anthropogenic CH₄ emissions from China hashave been growing rapidly in the past decades, and contributescontribute more than 10% of global anthropogenic CH₄ emissions with large uncertainties in existing global inventories, generally limited to country-scale statistics. To date, a long-term CH₄ emissions inventory including the major sources sectors and based on province-level emission factors is still lacking. In this study, we produced a detailed annual bottom-up inventory of anthropogenic CH₄ emissions from the eight major source sectors in China for the period 1980-2010. In the past three decades, the total CH₄ emissions increased from 22.2 [1624.4 [18.6-28.230.5] Tg CH₄ yr⁻¹ in 1980 (mean [minimum-maximum of 95% confidence interval]) to 45.044.9 [36.6-56.4-58.3] Tg CH₄ yr⁻¹, and most in 2010. Most of this
20 increase took place in the 2000s decade with averaged yearly emissions of 38.5 [30.6-48.3] Tg CH₄ yr⁻¹. This fast increase of the total CH₄ emissions after 2000 is mainly driven by CH₄ emissions from coal exploitation. The largest contribution to total CH₄ 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 3836% and 18% lower than the EDGAR4.2 inventory.

25 emissions factors collected from <u>state-of-the-art</u> published literatures. We also distributed province-scale emissions into 0.5°<u>1</u>° x 0.5°<u>1</u>° maps using social-economic activity data. This new inventory could help understanding CH₄ budgets at regional scale and guiding CH₄ mitigation policies in China.

1 Introduction

Methane (CH₄) plays an important role on global warming as a greenhouse gas. The radiative forcing in 2011 relative to 1750 caused by anthropogenic CH₄ 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 (CO₂, CH₄, Halocarbons and N₂O) since 1750 (Ciais et al., 2013). Atmospheric CH₄ 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 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.

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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, livestock, biomass and biofuel burning, 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

- 10 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 types of livestock (cow, cattle etc.) and their alimentation have evolved as well, and change CH₄ 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)
- $15 \quad \text{and changes in agricultural practices (Chen et al., 2013) can lead to reduced CH_4 emissions from rice paddies.}$

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

- 20 CH₄ yr⁻¹ of anthropogenic CH₄ sources in 2008, while U.S. Environmental Protection Agency (EPA) estimates that China emitted 44 Tg CH₄ yr⁻¹ of anthropogenic CH₄ sources in 2010. Based on a province-level inventory, Zhang and Chen (2011) reported anthropogenic CH₄ emissions of 38.6 Tg CH₄ yr⁻¹ for the year 2007. This large range of estimates (~30 Tg CH₄ 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 inclose the global methane budget (Dlugokencky et al., 2011; Kirschke et al., 2013; Ciais et al., 2013). Atmospheric inversions also tend to infer smaller methane emissions for China than reported by EDGAR4.2, with 59 [49-88] Tg CH₄ yr⁻¹ for the 2000-2009 decade in Kirschke et al. (2013) and ~40 [35-50] Tg CH₄ yr⁻¹ in the inversion of Bergamaschi et al. (2013, see their Figure 5).
- 30 Global inventories generally rely on country-level socio-economic statistics, which hardly fully reflect the more local to regional, possibly rapidly changing, characteristics of methane sources. This is especially the case in China where economic growth and the sources of CH₄ 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 CH₄ emissions for each source sector based on local to regional specific EFs and activity data. This is the main goal of this study.

A comprehensive anthropogenic CH₄ inventory for Mainland China (PKU-CH₄) was produced between 1980 and 2010, both at country and province scale, and downscaled at 0.51° 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

5 exploitation, 5) oil and natural gas systems, 6) fossil fuels combustion, 7) landfills and 8) wastewater. We also compiled regional specific EFs for each source sector from published literature in English and Chinese. We then estimated annual CH₄ emissions and their uncertainty for the eight major source sectors and for total emissions. Finally, we produced annual gridded maps of CH₄ emissions at 0.5x0.5°1°x0.1 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 10 for the period 1980-2010 are presented in section 3 and discussed in section 4.

2 Methods and Datasets

2.1 Methodology

The CH₄ emissions from <u>8 sectors, namely</u> livestock, rice cultivation, biomass and biofuel burning, coal exploitation, oil and 15 natural gas systems, fossil fuels combustion, landfills and wastewater are investigated in this study. The methods of IPCC greenhouse gas inventory guidelines (IPCC, 2006) were used to estimate CH₄ emissions for these <u>nineeight</u> sectors. The annual CH₄ emissions at the year t from the eight sectors are calculated by Eq. (1).

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

Where E(t) represents the total CH₄ 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}(*t*) is the emission factor at the year *t* 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 CH₄ utilized or oxidized without being released to atmosphere, such as CH₄ utilization or flaring from recovery instead of venting into the atmosphere in coal mining, CH₄ oxidation from waste, or reduced emissions due to biogas utilization, for instance. For estimation of CH₄ 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. We also applied the same activity data and
- correction factors but using IPCC default EFs (Table S2) to illustrate the impact of the new EF used in this study compared to the IPCC values. Note that the EFs used in this study do not evolve with time because of the limited information available about time evolution of EFs, which is a limitation of our study.

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2.2 Activity data, EFs and correction factors

2.2.1 Livestock

CH₄ emissions from livestock are estimated as the sum of CH₄ emissions from enteric fermentation and manure management. Province-level annual census data of domestic livestock for each livestock category were collected from agriculture statistics

- 5 yearbooks (CASY, 1980-2010). Livestock includes ruminants such as cattle, dairy cattle, buffalo, sheep, and goats, nonruminant herbivores such as horses, asses, and mules, and omnivorous swine. Because seasonal births and slaughters change 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 nondairy cattle and buffalo, 7 months for sheep and goats and 6 months for swine, respectively. The EFs of enteric fermentation
- 10 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 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.
- 15 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 (Table 2). The uncertainty of CH₄ emissions are estimated by the range of EFs for enteric fermentation and manure management (Table 2) (IPCC, 2006). The CH₄ from manure management could be utilized by bio-digester on large scale in China since the 1970s, but there is limited information about CH₄ collected from bio-digesters only from manure. The
- 20 correction factors are set as 0 for livestock sector, because of the limited information. We discussed We collected the total CH₄ collected<u>emission</u> from bio-digesters with mixed crop straw, manure and waste <u>during the period 1996-2010 from Feng et al.</u> (2012). Before 1996, the annual output of biogas (i.e. avoided CH₄ emissions compared to standard manure management practice) was assumed to linearly increase from the early 1980s to 1996, based on the number of household bio-digesters that increased from 4 million in the discussion.early 1980s to 6 million in 1996 (Figure S1). Since the biogas contained CH₄ from
- 25 both manure and crop residues, it is assumed that 10%, 15% and 25% of the biogas are low, medium, and high mitigation scenarios for CH₄ emissions only from manure management, respectively (Yin, 2015, master thesis), which is removed from the total emissions from standard emissions from manure management in livestock sector. CH₄ recovery and reduced emissions due to biogas utilization with manure feedstock is thus accounted in the livestock sector.

2.2.2 Rice cultivation

30 CH₄ emissions from rice cultivation sector are estimated using the methodology of Yan et al. (20132003). 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. (20132003), which summarized 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 CH₄ emissions from rice cultivation. 66.7% and 33.3% of rice cultivation area for intermittent irrigation and continuous flooding is assumed

- 5 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 <u>in 2000</u> (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 input have been changing with economic development and policy of agriculture and environment, and this uncertainty is<u>especially with increasing chemical fertilizer input in the 1980s and 1990s (Figure S2). It is assumed that organic</u>
- 10 matter input to rice paddies linearly decreased with increasing chemical fertilizer input before 2000, and that the fraction of rice paddy with organic input decreased from 85% in 1980 to 50% in 2000 (Figure S2). After 2000, on the one hand, chemical fertilizer kept increasing (Figure S2) but, on the other hand, the practice of returning crop residues and organic fertilizer applications became popularized again because of policy about sustainable quality of arable land and air quality control in China (http://www.sdpc.gov.cn/gzdt/201511/t20151125_759543.html), which can be indirectly supported by increasing
- 15 number of the machines for returning crop residues in the 2000s (from 0.44 million in 2004 to 0.62 million in 2011). The uncertainties of rice cultivated areas receiving organic input and irrigation conditions are 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 CH₄ utilizationrecovery from rice paddies is observed until now. The uncertainty of CH₄ emissions from rice cultivation is derived from the range of EFs (Yan et al., 2003).

20 2.2.3 Biomass and biofuel burning

CH₄ emissions from biomass and biofuel burning mainly come from burning of firewood and straw in rural households. <u>In our</u> <u>inventory, this sector includes emissions from firewood and crop residues burnt as biofuel in households and from disposed</u> <u>crop residues burnt in the open fields</u>. Province-level firewood consumption are extracted from the China Energy Statistical Yearbook (1980-2007). Because no firewood data is available after 2007 and firewood consumption in China is stable after

- 25 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 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).
- 30 $RB_{crop} = \sum_{c} R_{c} \times N_{c} \times F \times \theta$, (2)

Where RB_{crop} is the amount of burning crop residues as biomass fuel or disposed fire in open fields (Kg yr¹); *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 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 CH₄ emissions from biomass and biofuel burning were collected from the scientific literature (Zhang et al., 2000;
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 CH₄ t⁻¹ (mean ± standard deviation), and EFs from crop residues for biomass fuel and fire in open fields of 3.62 ± 2.20 kg CH₄ t⁻¹ and 3.89 ± 2.20 kg CH₄ t⁻¹, respectively (Tian et al., 2011). The uncertainty of CH₄ emissions (95% CI) isarc estimated from the range of the EFs by 1000 times of bootstrap samples.

2.2.4 Coal exploitation

- 10 CH₄ emissions from coal exploitation include fugitive CH₄ emissions from coal mining and post mining. In China, coal exploitation includes both underground and surface coal mines. Generally, CH₄ 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 CH₄ from underground and surface mines are significantly different (Zheng et al., 2006; IPCC, 2006; Zhang
- 15 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 CH₄ emissions from both underground and surface mines. For CH₄ 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 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 CH₄ 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 m³ t⁻¹) from IPCC (2006), since there isare few measurements of CH₄ emissions from surface mines. The EF of CH₄ from coal post-mining including emissions during subsequent handling, processing and transportation of coal), is taken as 1.24 m³ t⁻¹ (1.18-3:001_30 m³ t⁻¹), according to the weighted average of production from high- and low- CH₄ coal mines using IPCC (2006) default EFs for high- (3.0 m³ t⁻¹) and low- (0.5 m³ t⁻¹) CH₄ coal mines (Zheng et al., 2006). Note that CH₄ emissions from abandoned mines are not included in our inventory, because 1) abandoned mines are estimated to account for less than 1% of total emissions from coal mining (NRDC, 2014), and 2) the time
 series of numbers and locations of the abandoned mines are unavailable (NRDC, 2014).

Not all CH₄ emissions from underground coal mines are released into atmosphere as CH₄. A fraction of CH₄ from coal mines are collected for flaring or be utilized by coal bed/mine methane in Clean Development Mechanism (CDM) projects (e.g.,

Bibler et al., 1998; GMI, 2011). The utilization fractionrecovery of CH₄ from coal mines increases increased with economic growth and enhancement of coal safety (NDRC, 2014). For example, Zheng et al. (2006) indicates that the utilization fractionrecovery of CH₄ from coal mines increased from 3.59% in 1994 to 5.21% in 2000. We used the utilizationrecovery 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 utilizationrecovery fraction (3.59% - 5.21%) is taken to calculate the uncertainty of CH₄ emissions from coal mining. A

5 of <u>utilizationrecovery fraction</u> (3.59% - 5.21%) is taken to calculate the uncertainty of CH₄ emissions from coal mining. A volumetric mass density of 0.67 Kgkg m⁻³ is used to convert volume of CH₄ emission into CH₄ mass.

2.2.5 Oil and natural gas systems

Province-level annual crude oil and natural gas production were collected from China Statistical Yearbook (1980-2010). The EFs of fugitive CH4 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 CH4 leakage from oil and natural gas systems in China. The EFs of fugitive CH4 from oil and natural gas systems in China are from Schwietzke et al. (2014a, 2014b), including venting, flaring, exploration, production and upgrading, transport, refining/processing, transmission and storage, as well as distribution networks in this study, which corresponds to definitions of IPCC subcategory 1B2. For the fugitive CH4 from oil systems, the

15 average EF from oil systems is taken as 0.077 kt CH₄ PJ⁻¹ (2.9 kg CH₄ m⁻³ oil), and the uncertainty of EF are 0.058-0.190 kt CH₄ PJ⁻¹ (2.2-7.2 kg CH₄ m⁻³ oil) (see Table 1 in Schwietzke et al., 2014a). For the fugitive CH₄ from natural gas systems, the fugitive emissions rates (FER) of natural gas is decreasing from 1980 to 2011 (Schwietzke et al., 2014b). We assumed a FER linear decrease from 4.6% (0.81 kt CH₄ PJ⁻¹) in 1980 to 2.0% (0.35 kt CH₄ PJ⁻¹) in 2010, which is today close to the FER (1.9%) in OECD countries in 2010. The range of uncertainty was estimated with a scenario assuming a low FER in China decreasing from 3.9% in 1980 to 1.8% in 2010, and a scenario with high FER in China decreasing from 5.7% in 1980 to 4.9% in 2010.

2.2.6 Fossil fuels combustion

Province-level fossil fuels combustion (TJ) were collected from China Energy Statistical Yearbook (1980-2010). We used the default EFs from IPCC (2006) for CH₄ emissions from fossil fuels combustion, 1 Kgkg CH₄ TJ⁻¹ for coal combustion, 3 Kgkg CH₄ TJ⁻¹ for oil combustion and 1 Kgkg CH₄ 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 CH₄ emissions from landfills is estimated by First Order Decay (FOD) method as Eq. (3).

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

Where $E_{landfill}(t)$ is CH₄ 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 CH₄ emissions from three types of landfills T ($MCF_T = 1.0$ for managed anaerobic landfills; $MCF_T = 0.8$ for deep (> 5 m) non-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

- 5 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 CH₄ 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).
- 10

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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 (R²=0.95, P<0.001; Figure \$4\$3). 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 CH₄ emissions from landfills, maximum CH₄ emissions with DOC_d =0.6 and f=0.6 and minimum CH₄ emissions with DOC_d =0.5 and f=0.4 were calculated.

2.2.8 Wastewater

20 CH₄ 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 CH₄ 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*; B₀ is maximum CH₄ producing capacity, 0.25 kgCH₄/kgCOD; MCF is methane correction factor for wastewater. The total CH₄ emissions from wastewater include two parts: one part from

- 25 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).
- 30 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 <u>\$254</u>). The COD in domestic sewage and industrial wastewater treated by WTPs increases with GDP (R²=0.95-0.99, P<0.001; Figure</p>

S2S4 and S3S5). The fraction of discharged COD from industrial wastewater decreases with GDP (Figure S2S5). 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.

5

The uncertainty of CH₄ 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 CH₄ emissions with MCF=0.3 for domestic sewage and MCF=0.5 for industrial wastewater treated by WTPs, and minimum CH₄ emissions with MCF=0.1 for domestic sewage and MCF=0.2 for industrial wastewater treated by WTPs (IPCC, 2006; Ma et al., 2015).

10 2.3 Maps of CH4 emissions

- In order to produce gridded emissions maps at 0- $\frac{5}{2}$ x0.5 for each source sector, we distributed the province-level CH₄ emissions using different activity data: rural or total population, GDP, agricultural GDP, crop cultivation area.(<u>Table S1</u>). First, we collected county-level rural population, <u>CSYRE</u>, 2010), gridded total population, GDP and agricultural GDPGDP with 1km spatial resolution in 2010 from statistic yearbook, 2005 and 2010 (Huang et al., 2014), gridded numbers of animals in
- 2005 (Robinson et al., 2011), gridded harvested area of rice (Monfreda et al., 2008), annual production of 4264 coal production sites (Liu et al., 2015), and converted/resampled them into 0.5°1° by 0.5°1° gridded maps. Then, these gridded maps are applied to distribute the province-level of CH₄ emissions from the eight source sectors: (Table S1). Because not all countyproxy data are available for every year during the period 1980-2010, we only used the activity data for 2010 (except for the average rice eultivation area in 1994-1996; Frolking et al., 2002; Qiu et al., 20032005 and 2010 (proxy data in 2005 for the years before
 2005, and proxy data in 2010 for the years between 2005 and 2010), therefore assuming that the changes in the spatial structures of the gridded maps remain limited.

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 resealed the rice cultivation maps with annual province-level rice cultivation area from agriculture statistics yearbooks to produce annual rice cultivation area maps.

3 Results

5

3.1 Total and sectorial CH₄ emissions

Figure 1 shows the evolution of anthropogenic CH₄ emissions in China for the eight major source sectors and for the countrytotal, and Table 3 lists the magnitude of CH₄ emissions and their uncertainty in 1980, 1990, 2000 and 2010. In 1980, the country-total CH₄ emissions was $\frac{22.2 [1624.4 [18.6-28.230.5]}{12.2 [1624.4 [18.6-28.230.5]}$ Tg CH₄ yr⁻¹ (Table 3). Rice cultivation and livestock contributed $\frac{7071\%}{10}$ of anthropogenic CH₄ sources in 1980, followed by coal exploitation ($\frac{1514\%}{10}$) (Figure 1b). In the past 30 years, the CH₄ emissions doubled, reaching $\frac{45.044.9}{15.6 (10.6-56.4-58.3)}$ Tg CH₄ yr⁻¹ in 2010 (Figure 1a). In 2010, coal exploitation became the largest contributor of Chinese CH₄ emissions (40%), followed by livestock ($\frac{2825\%}{10}$) and rice cultivation (16%) (Figure

1c). The increase of CH₄ emissions between 1980 and 2010 is mainly attributed to coal exploitation ($\frac{6370\%}{6370\%}$ of the total

10 increase) mostly after 2000, followed by livestock (2726%) mostly before 2000.

Figure 2 shows the evolution of individual CH₄ sources from 1980 to 2010. Among the eight major source sectors, CH₄ emissions from seven source sectors increased from 7268% to 426407%, and only CH₄ emissions from rice cultivation decreased by 21 [17-23]%34% (Figure 2) before 2005 because of decreased rice cultivation area in this period. The increase of country-total CH₄ sources accelerates after 2000 (2002 (from 0.5 Tg CH₄ yr² before 2002 to 1.3 Tg CH₄ yr² after 2002, Figure 2a). The increase of CH₄ emissions in the 2000s contributes 5863% of the total increase observed between 1980 and 2010 (Table 3). The acceleration of emissions after 2000starting from 2002 is mainly driven by coal exploitation (Figure 2a and 2e), while CH₄ emissions from livestock, biomass and biofuel burning, landfills and rice cultivation remain stable or increased at a lower rate after 20002002 resulting from the stable or slow increase in activities data in these sectors. Although CH₄ emissions from oil and gas systems, fossil fuels combustion and wastewater increased exponentially after 2002002, they

20 CH₄ emissions from oil and gas systems, fossil fuels combustion and wastewater increased exponentially after $\frac{20002002}{2002}$, the only contributed less than $\frac{613}{5}$ % of the increase in total CH₄ emissions in the 2000s.

3.2 Spatial patterns of CH4 emissions

Figure 3 shows the spatial distributions of CH₄ emissions in 2010 (Note that Figure 3a-3i have different color scales). <u>The total</u> emissions of each province in 1980, 1990, 2000 and 2010 are also listed in Table S3. Hotspots of CH₄ emissions are distributed

- 25 mostly in the densely populated area, where we describe the emissions for South, Center and North of China country (Figure S4S6 shows the map these regions). These hotspots are driven by livestock, rice cultivation and coal exploitation (Figure 3). North of China has high CH₄ emissions from livestock, biomass and biofuel burning, coal exploitation, <u>oil and gas systems</u>, <u>landfills</u> and wastewater. South and central of China has high CH₄ emissions from rice cultivation and coal exploitation (Figure 3c, and 3e). CH₄
- 30 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. CH₄ emissions from biomass and biofuel burning are mainly distributed in the north of China. CH₄ emissions from landfills and wastewater are mainly

distributed in north-and, northeast and coast of China. CH₄ leakages from oil and gas systems are located in the north part of China, where oil and gas are mostly produced (Figure 3f). CH₄ emissions from fossil fuels combustion also concentrate in east part of China (Figure 3g and 3i).

5 Figure 4 shows the spatial distribution of the changes of CH₄ emissions from 1980 to 2010. The CH₄ 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 CH₄ emissions in South and Southeast of China is attributed to a decline in rice cultivation, <u>livestock</u> and biomass and biofuel burning emissions, which offsets the increase from other sources in these regions (Figure 4). The increase in CH₄ emissions in North and Northeast of China are attributed to livestock, biomass 10 and biofuel burning, coal exploitation, and landfills and wastewater. Southwest of China has increase in CH₄ emissions from coal exploitation and landfills (Figure 4).

4 Discussion

4.1 Comparison with other inventories

- Figure 2 shows the comparison of CH₄ emissions inferred in this study with EGDARv4.2 (EDGAR, 15 http://edgar.jrc.ec.europa.eu/overview.php?v=42)-and), EPA (EPA, 2012) inventories, and estimates with IPCC default EFs (hereafter called IPCC-EF estimates, Table S2). We also make comparison of the emissions in 2005 in the text with the Second National Communication on Climate Change of The People 's Republic of China (SNCCCC) to UNFCCC (NDRC, 2012). Our estimates of the total CH₄ emissions are very close to EPA estimates and 50-9030-40% lower than EDGARv4.2 inventory during the period 1980-2008 (Figure 2a). Compared to IPCC-EF values, our estimates are consistent with it before 2000, but
- 20 ~30% lower after 2000. The CH₄ 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 EDGAR4.2 (e.g. Bergamaschi et al., 2013, Kirschke et al., 2013). Although the magnitude of the total CH₄ emissions do not agree between
- 25 EDGARv4.2, EPA and this study, the trends of the total CH₄ emissions from these three estimates are qualitatively similar, confirming the slow increase before $\frac{20002002}{2002}$ and the acceleration thereafter (Figure 2a). However, the magnitude of the trend of anthropogenic CH₄ emissions after $\frac{20002002}{2002}$ found in this study (1.3 Tg CH₄ yr²) and in EPA (0.7 Tg CH₄ yr²) are $\frac{54respectively 63\%}{1000}$ and $\frac{7580\%}{1000}$ less than in EDGAR4.2 ($\frac{2.93.5}{1000}$ Tg CH₄ yr²). This discrepancy is due mostly to coal exploitation (figure 2e) with smaller contributions from landfills (figure 2h) and oil and gas systems (figure 2f). The slower
- 30 increase of total CH₄ 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 cultivation ($\frac{8.9additional 7.3}{2}$ Tg CH₄ yr⁻¹), wastewater (($\frac{+}{3.6}$ Tg CH₄ yr⁻¹), biomass and biofuel burning (($\frac{+}{2.57}$ Tg CH₄ yr⁻¹), and coal exploitation (($\frac{+}{3.2}$ Tg CH₄ yr⁻¹), oil and gas systems (-0.7 Tg CH₄ yr⁻¹). In the 2000s, higher emissions from EDGARv4.2 are attributed to higher estimates from coal exploitation ($\frac{9.1(+8.7)}{2}$ Tg CH₄ yr⁻¹), rice cultivation (($\frac{+}{6.0}$ Tg CH₄ yr⁻¹), wastewater ($\frac{4.6(+3.8 \text{ Tg CH}_4 \text{ yr}^{-1})$, landfills ($\frac{+1.2}{2}$ Tg CH₄ yr⁻¹), oil and gas systems (1.7 Tg CH₄ yr⁻¹) and biomass and

- biofuel burning $(1.0(+1.2 \text{ Tg CH}_4 \text{ yr}^{-1})$ and oil and gas systems $(+0.8 \text{ Tg CH}_4 \text{ yr}^{-1})$. EPA estimates of CH₄ emissions from most source sectors are in line with our estimates, except for fossil fuels combustion and wastewater (Figure 2f & 2i), due mainly to the discrepancy between local and IPCC default EFs (NDRC, 2014; IPCC, 2006). <u>IPCC estimates are close to our estimates</u> in a majority of source sectors, except for higher values in coal exploitation and lower values in rice cultivation and landfills.
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Livestock. CH₄ emissions from livestock are the only one to be consistent between the threefour inventories (Figure 2b). Similar magnitudes of livestock emissions (~10 Tg CH₄ yr⁻¹) are also reported in previous studies (Verburg and Denier van der Gon, 2001; Yamaji et al., 2003; Zhang and Chen, 2014b). But our estimate in 2005 (12.4 Tg CH₄ yr⁻¹) is lower than the value (17.2 Tg CH₄ yr⁻¹) reported to UNFCCC (NDRC, 2014), which results from higher EFs of enteric fermentation for nondairy cattle (71 kg CH₄ head⁻¹ yr⁻¹) and dairy cattle (85 kg CH₄ head⁻¹ yr⁻¹) adopted by NDRC (2014). The stagnation of livestock emissions after 2000 is explained by the stable domestic ruminant population (CSY, 2012). The increasing import of livestock products (e.g., meat and milk) may helpcontribute 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/). In addition, the uncertainty of activity data could be further investigated by comparison between multiple sources, such as FAO, national statistics and province-level statistics in the future studies. Besides the uncertainty of population, the EF of livestock are highly correlated to the live weight per head (for meat cattle) and milk production per head (for dairy cattle) (Dong et al., 2004; IPCC, 2006). In this study, as in previous studies, we assumed that EF from livestock in China did not evolve with time because of limited information about the weight distribution of each livestock population type besides numbers of animals, although we estimated

25 per head may have increased EFs of enteric fermentation (IPCC, 2006). On the other hand, the increasing share of crop products / crop residues in the diet of livestock may have reduced the EFs of enteric fermentation (Dong et al., 2004). The possible changing EF resulting from increased live weight and milk production per head or more feed with treated crop residues should be investigated in future work.

an uncertainty using different EFs (Table 1). On the one hand, the (unaccounted for) increasing live weight and milk production

30 <u>Rice cultivation.</u> Yan et al. (2003) reported 7.8 [5.8-9.6] Tg CH₄ yr⁻¹ emissions from rice paddies by combining rice cultivation area in 1995 and 204 measurements of CH₄ emission rates from rice paddies with/without organic inputs and intermittent irrigation or continuous flooding. The CH₄ 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 CH₄ yr⁻¹. NDRC (2014) reported 7.9 Tg CH₄ yr⁻¹ emissions from rice paddies in 2005. Our estimates of CH₄ emissions from rice paddies (7.3 [5.9-8.2 [6.5 - 10.08] Tg CH₄ yr⁻¹ during 1980-2010 in 2005) is

consistent with these previous estimates, while the estimates of EDGARv4.2 (15.413.2 Tg CH₄ yr⁻¹ in 2005) is out of the range reported by <u>NDRC (2014)</u>. Chen et al. (2013) and our estimates. The large variation of CH₄ 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 CH₄ emissions from rice paddies (Cai et al., 2000; Zou et al., 2005; Chen

- 5 et al., 2013). This could be the main reason of the higher estimates in EDGARv4.2- and lower estimates in EPA and IPCC. 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., 2013).
- 10 1998, 2004; Cai, 2000; Zou et al., 2005). In this study, we assumed that the area of rice with organic input decreased with increasing chemical fertilizer input during the 1980s and the 1990s, and kept constant after 2000 because of both increasing chemical fertilizer input and returning crop residues in the 2000s (Figure S2). Without this assumption, the trend of CH₄ emissions from rice cultivation could be smaller. The area with continuous irrigation may have changed during the past three decades. This could also impact the trend of CH₄ emissions from rice cultivation, and need further study to get and analyse
- 15 detailed irrigation data, if available. A decrease in CH₄ emissions from rice cultivation is confirmed in all of these inventories, because of1) the decrease intotal rice cultivation area is decreasing and northward of2) rice cultivation moved northward 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).
- 20 Biomass and biofuel burning. For the CH₄ emissions from biomass and biofuel burning, EDGARv4.2 has a two-times larger value than EPA and our estimates in the 1980s (Figure 2d). Previous studies reported 1.9-2.4 Tg CH₄ yr⁻¹ emissions from biomass and biofuel burning by the same method but independent estimates of activities data (SNCCCC, 2013NDRC, 2014; 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 CH₄ emissions from biomass and biofuel burning increased
- 25 from 1.9 Tg CH₄ yr⁻¹ in 2000 to 2.2 Tg CH₄ yr⁻¹ in 2007. Compared to the Global Fire Emission Database (GFED) v4.1 products, our estimates of CH₄ emissions from crop residues burnt in the open fields (0.28 [0.05-0.51] Tg CH₄ yr⁻¹) are larger than so called agricultural fire emissions in GFEDv4.1 (0.09 [0.04-0.18] Tg CH₄ yr⁻¹). But considering the uncertainty of distinguishing agricultural fire and wild fire in GFED4.1 products and the poor detection of small agricultural fires using satellites, our estimates are close to the total CH₄ emissions including both wild fire and agricultural fire (0.22 Tg CH₄/yr) in
- 30 GFEDv4.1. Most of CH₄ emissions from biomass and biofuel burning in China are from firewood and straw burning inside of households (Tian et al., 2011; Zhang and Chen, 2014a). The amount of firewood and straw burning have large uncertainty (YevitchYevich 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. The assumed constant fraction of crop residues burnt in the open fields and in rural household in

this study may lead to overestimate CH_4 emissions from both firewood and crop residues burning. For improving air quality and reducing aerosol in the air, a ban on burning crop residues in open fields was passed in the late of 2000s. This should further reduce their contribution to CH_4 emissions in China. 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

5 CH₄ emissions from manure management are integrated in the livestock sector. However, the fraction of CH₄ emissions from manure burning only account for less than 1% of CH₄ emissions from biomass and biofuel burning (Tian et al., 2011).

Coal exploitation. Our estimate of CH₄ 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, CH₄ emissions from coal exploitation was estimated of 8.7 Tg CH₄ yr⁻¹ in 1990 (CCCCS, 2000), 6.5 Tg CH₄ yr⁻¹ in 2000 (Jiang and Hu, 2005) and 12.2 Tg CH₄ yr⁻¹ in 2002 (Yuan et al., 2006). NDRC (2014) reported 12.9 Tg CH₄ yr⁻¹ emissions from coal exploitation in 2005, which is quite close to our estimate (12.9 Tg CH₄ yr⁻¹). According to reports of the State Administration of Coal Mine Safety (2008, 2009), CH₄ emissions from coal exploitation are 13.8 Tg CH₄ yr⁻¹ in 2007 and 14.5 Tg CH₄ yr⁻¹ in 2008, respectively (Cheng et al., 2011). On the one hand, the default EFs of underground coal mines
(18 m³ t⁻¹ for average, 25 m³ t⁻¹ for high- and 10 m³ t⁻¹ for low- CH₄ coal mines in Zhang et al., 2014) (e.g., CCCCS, 2000;

- Zheng et al., 2005; Zhang et al., 2010, 2014). The higher CH₄ 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, CH₄ concentration and coal seam permeability (e.g.,
- 20 Zheng et al., 2006). These regional EFs of coal mining range from ~20 m³ t⁻¹ in southwest of China and ~19 m³ t⁻¹ in northeast of China, to ~5 m³ t⁻¹ in west, east and north of China (Table 2; Zheng et al., 2006). The depths of coal mines and coalbed CH₄ concentration are regionally variable (Bibler et al., 1998). Regional EFs of coal exploitation should be considered to estimate CH₄ emission as we did in this study, resulting in lower estimates of CH₄ 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
- 25 significant bias to estimate CH₄ emissions from coal exploitation (e.g., Zhang et al., 2014). Besides the EFs, the utilization fractionrecovery of CH₄ from coal exploitation is another key parameter for estimation of CH₄ 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 to 5.2% in 2000, based upon data of hundreds of individual coal mines (Zheng et al., 2006). In our inventory, we assumed that the recovery of CH₄ from coal exploitation kept increasing from 5.2%
- 30 in 2000 to 9.2% in 2010. This assumption is consistent with the register of validated CBM and CMM projects in China which started from 2004 and increased in 2007/2008 (http://www.cdmpipeline.org/overview.htm, CDM/JI database). The total reduction of CH₄ emissions by the implementation of CBM and CMM in China derived from the CDM/JI pipeline database is ~0.3 Tg CH₄ yr⁻¹ in 2006 and ~0.9 Tg CH₄ yr⁻¹ in 2010, which is close to our estimates of increased CH₄ recovery in 2006 (0.4 Tg CH₄ yr⁻¹) and 2010 (0.8 Tg CH₄ yr⁻¹). On the top of EFs differences, the increased recovery of CH₄ 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 recovery of CH_4 in this study although the time evolution of this parameter has large uncertainty.

- Oil and gas systems & fossil fuel combustion. Our estimates of CH₄ leakage from oil and natural gas systems are close to estimates of EPAIPCC, but much smaller than EDGARv4.2 and higher than EPA (Figure 2f). While ourOur estimates of CH₄ emissions from fossil fuels combustion, are close to estimates of EDGARv4.2 and IPCC, but much smaller than estimates of EPA (Figure 2g). NDRC (2014) reported 0.2 Tg CH₄ yr⁻¹ leakage from oil and natural gas systems and 0.1 Tg CH₄ yr⁻¹ emissions from fossil fuels combustion in 2005, which is consistent with our estimates. <u>Zhang et al. for emissions from fossil</u> fuels combustion but much smaller than our estimates for leakage from oil and natural gas systems. Zhang et al. (2014) reported
- 10 0.7 Tg CH₄ yr⁻¹ leakage from oil and natural gas systems and 0.1 Tg CH₄ yr⁻¹ emissions from fossil fuels combustion, which are in the range of our estimates. Jower than our estimates. In this study, we assumed the medium, low and high scenarios for EFs of fugitive emissions from oil and gas systems (Schwietzke et al., 2014a, 2014b), and the EFs are consistent with EFs reported in USA and Canada in the 2000s (~2%, Höglund-Isaksson et al., 2015). The EFs from oil and natural gas systems have a large spread, and source attribution to oil or natural gas production is also highly uncertain (Höglund-Isaksson et al., 2015).
- 15 2015). Changes in the natural gas production and distribution technology may change the EFs from natural gas systems (Höglund-Isaksson et al., 2015). This may partly contribute to the decreased FER in our inventory. 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.
- Landfills. Gao et al. (2006) calculated 1.9-3.4 Tg CH₄ 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 CH₄ emissions from landfills in 2005 (2.2 Tg CH₄ 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 CH₄ 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 CH₄ yr⁻¹ emissions from landfills in
- 2011, which is the maximum estimates of this study (Figure 2h). CH₄ emissions from landfills in EDGARv4.2 are different with EPA and our estimates in the 2000s, and the trends of CH₄ emissions from landfills are different between EDGARv4.2,
 20 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 <u>\$153</u>) 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.

Wastewaters. Both EDGARv4.2 and EPA have 3-4 times higher CH₄ emissions from wastewater than our estimates (Figure 2i). NDRC (2014) reported 1.6 Tg CH₄ yr⁻¹ emissions from wastewater in 2005. Zhou et al. (2012) reported 1.3 Tg CH₄ 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 CH₄ yr⁻¹ emissions from wastewater in 2010. All these estimates do not consider the utilizationrecovery of CH₄ from wastewater. However, Wang et al. (2011) and Cai et al. (2015) reported a tiny CH₄ emissions (<0.1 Tg CH₄ yr⁻¹) from WTPs in China, and they argued that most COD in wastewater

- 10 are not removed by anaerobic biological treatments, but by oxidation exposure in WTPs. This suggests that the CH₄ emissions from wastewater could be much lower if most of wastewater is treated by oxidation exposure in WTPs. Our estimates may overestimate CH₄ emissions from wastewater, with limited information of the wastewater treatments in Chinese WTPs. 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
- 15 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 CH₄ emissions from wastewater, and need more samples in future inventory.

4.2 Mitigation of CH4 emissions in China

The total anthropogenic CH₄ emission of China is estimated to be 38.95 [30.6-49.448.3] Tg CH₄ yr⁻¹ on average for the 2000s
decade. This large source (~7<u>12</u>% of the global <u>anthropogenic</u> CH₄ 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 CH₄ yr⁻¹ CMM is captured and ~1 Tg CH₄ yr⁻¹ utilized in 2009 (Brink et al., 2013). Under the framework of Clean Development Mechanism (CDM),CDM, CH₄ 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 CH₄ utilization performed between 1996 and 2010, and capture annually 15 billion m³ biogas (Feng et al., 2012). The fast increased utilization and manure management in our estimates, because we assumed a conservative utilization-or linearly increased recovery fraction for CH₄ from coal mining and manure management (see Section 2.2). In the CDM database, ~0.4 Tg CH₄ yr⁻¹ landfill gas utilization started from 2007 in China.

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The consumption of natural gas has exponentially grown in China (SNCCCC, 2013). The urban population using natural gas 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). <u>On the one hand, CH4</u> leakage from natural gas distribution networks may increase this sector of CH4 emissions in the coming decades, because of growth of urban population and increase in coverage of natural gas pipes (CESY, 2012).However, But on the other hand, new pipes will benefit of recent technologies contrary to older European, US, and Russian gas networks. Associated to the decrease of rural population, the substitution of firewood and straw in China by natural gas because of decrease in rural population and increase in usage of natural gas, which could reduce CH4 emissions from biomass and biofuel burning. With population growth and sustained GDP continues in the coming decades, the CH4 sources from livestock, MSW and wastewater are predicted to increase (e.g., https://www.globalmethane.org/; Ma et al., 2015).

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CH₄ mitigation provides a co-benefit to reduce 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 CH₄ (SNCCCC, 2013). China has also improved CH₄ mitigation within the Global Methane Initiative (GMI) and the framework of CDM on CH₄ mitigation on coal-mine methane, agriculture and MSW (Higashi, 2009; https://www.globalmethane.org/). All of these elements can contribute to reduce CH₄ emissions of China in the coming decades.

CH₄ 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.

15 https://www.globalmethane.org/). All of these elements can contribute to reduce CH₄ emissions of China in the coming decades. A more precise assessment of the reduction potential of Chinese CH₄ emissions could be further investigated in future research based on the detailed inventory reported here.

5 Summary

We collected province-level activity data of agriculture, energy and waste and emission factors of CH₄ from the eight major source sectors in Mainland China, and estimated annual CH₄ emissions from each source sector from 1980 to 2010. Our estimates of CH₄ emissions considered regional specific emission factors, activity data, and correction factors as much as possible. In the past decades, the total CH₄ emissions increase from 2224.4 [4618.6-28.1] Tg CH₄ yr⁻¹ in 1980 to 45.4 [36.6-5830.5] Tg CH₄ yr⁻¹ in 1980 to 44.9 [36.6-56.4] Tg CH₄ yr⁻¹ in 2010. The largest contributor to total CH₄ emissions is rice cultivation in 1980, but has been replaced by coal exploitation after year 2005. The increase of CH₄ emissions from coal

25 exploitation and livestock drive the increase of total CH₄ emissions. We distributed the annual province-level CH₄ emissions into 0.5°1° x 0.5°1° high-resolution maps for each source sector using different social-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%36% [30-40%] than EDGAR4.2 and shows slower increase in emissions after 2000 as in EPA.

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 <u>diddo</u> 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 data

5 and <u>utilizationrecovery</u> fraction also have their own uncertainty. For example, there is 5%-10% uncertainty energy consumption data in China (Liu et al., 20142015). The <u>utilization fractionrecovery</u> of CH₄ 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 <u>information becomesdata become</u> available.

10 Data availability

CH4 inventory (PKU-CH4) in this study is <u>public public public public</u> available on website, and the intention is to regularly update it every two or three years.

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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 l	head-1	yr-1)				
	S 1	S2	S 3	S4	S5	S 6	Mean	Min	Max		
					L	ive					
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		
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		
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		
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		
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		
Not divided	1	1	1	1	1	1	1	1	1		
					Slaug	htered	!				
	58	53					55	53	58		
	3	5					4	3	5		
	3	4					3	3	4		
	Mature female Young (<1 yr) Other Mature female Young (<1 yr) Other Mature female Young (<1 yr) Other Mature female Young (<1 yr) Other Mature female Young (<1 yr) Other Nature female Young (<1 yr) Other Nother	S1 Mature female 78 Young (<1 yr)	S1 S2 Mature female 78 68 Young (<1 yr)	S1 S2 S3 Mature female 78 68 70 Young (<1 yr)	EFs of I S1 S2 S3 S4 Mature female 78 68 70 48 Young (<1 yr)	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		

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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			EFs of coal mining Fractions of MSW _L t EFs of manure management burning crop residues drop from underground by different coal mines (m ³ t ⁻¹), types of landfills (n of EFs of coal mining n of From underground ng coal mines (m ³ t ⁻¹), idues data from Zhang et al. (2006).				(%);	
Province	MAT (°C)	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	0.05	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	0.05	0.70	-	-	-	54.2	34.4	11.4
Hebei	9.6	9.00	1.00	1.00	0.10	0.11	2.00	0.10	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	0.10	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	0.05	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	0.10	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	0.20	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	0.20	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	0.20	0.20	-	-	-	0.9	74.3	24.8
Jiangsu	15.2	14.00	1.00	1.00	0.10	0.11	3.00	0.05	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	0.20	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	0.05	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	0.20	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	0.10	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	0.10	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	0.10	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	0.10	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	0.10	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	0.20	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	0.10	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	0.10	0.45		-	-	33.7	49.7	16.6
Chongqing	15.9	14.00	1.00	2.00	0.15	0.17	3.00	0.10	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	0.10	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	0.10	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	0.10	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	0.05	0.20	-	-	-	0.0	75.0	25.0
Shaanxi	10.8	10.00	1.00	2.00	0.15	0.17	2.00	0.10	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	0.05	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	0.05	0.80	5.99	6.00	5.97	58.8	30.9	10.3

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.

Ningxia	8.1	9.00	1.00	2.00	0.15	0.17	2.00	0.05	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	0.05	0.20	5.99	6.00	5.97	0.0	75.0	25.0

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 emissions in O	China (Tg CH ₄ yr ⁻¹))
	1980	1990	2000	2010
Livestock	6.2 [4.9-7.8]	8.9 [7.0-11.2]	12.3 [9.9-15.2]	11.4 [9.3-13.7]
Rice cultivation	11.2 [9.0-13.4]	10.0 [7.9-12.0]	7.8 [6.2-9.4]	7.4 [6.0-8.8]
Biomass and biofuel burning	1.4 [0.4-2.5]	1.9 [0.5-3.3]	1.9 [0.5-3.3]	2.4 [0.6-4.2]
Coal exploitation	3.4 [3.0-3.7]	6.8 [6.0-7.5]	6.0 [5.3-6.7]	17.7 [16.7-20.3]
Oil and gas systems	0.6 [0.5-1.3]	0.7 [0.5-1.6]	0.9 [0.7-2.1]	1.6 [1.4-4.2]
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	24.4 [18.6-30.5]	30.3 [23.1-38.0]	32.0 [24.4-40.3]	44.9 [36.6-56.4]
FF: fossil fuels				

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 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(CI) of our estimates. IPCC-EF refers to the estimates using the same method but IPCC default emission factors, and 5-95% CI is based on high and low estimates of emission factors. Note that the empty circle indicates projected 2010 value in EPA.
- 10 Figure 3. Spatial distribution of (a) total anthropogenic CH₄ emissions, and (b) (i) CH₄ emissions from different source sectors in Mainland China in 2010. The unit of the colorbar is g CH₄ m⁻² yr⁻¹. Note that subplots have different color scale.

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

15 different color scale.

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