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Discrepancies and Uncertainties in Bottom-up Gridded Inventories of Livestock Methane Emissions for the Contiguous United States

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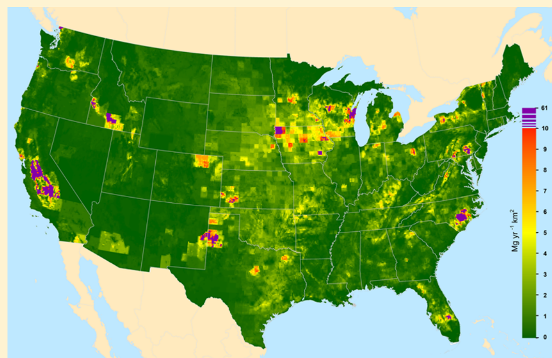
Supporting Information

ABSTRACT: In this analysis we used a spatially explicit, simplified bottom-up approach, based on animal inventories, feed dry matter intake, and feed intake-based emission factors to estimate county-level enteric methane emissions for cattle and manure methane emissions for cattle, swine, and poultry for the contiguous United States. Overall, this analysis yielded total livestock methane emissions (8916 Gg/yr; lower and upper 95% confidence bounds of ±19.3%) for 2012 (last census of agriculture) that are comparable to the current USEPA estimates for 2012 and to estimates from the global gridded Emission Database for Global Atmospheric Research (EDGAR) inventory. However, the spatial distribution of emissions developed in this analysis differed significantly from that of EDGAR and a recent gridded inventory based on USEPA. Combined enteric and manure methane emissions from livestock in Texas and California (highest contributors to the national total) in this study were 36% lesser and 100% greater, respectively, than estimates by EDGAR. The spatial distribution of emissions in gridded inventories (e.g., EDGAR) likely strongly impacts the conclusions of top-down approaches that use them, especially in the source attribution of resulting (posterior) emissions, and hence conclusions from such studies should be interpreted with caution.



INTRODUCTION

The agriculture sector is an important source of anthropogenic, non-CO_2 greenhouse gas (GHG) emissions in the United States and livestock emissions made up an estimated 48% of the 2015 agricultural GHG emissions.[1] Methane and nitrous oxide are the two most important GHGs from agricultural activities. Methane is a potent, short-lived (12.2 years[2]) GHG emitted from various sources, including fossil fuel-related activities, livestock operations, rice production, landfills, and others. According to USEPA,[1] the top three sources of anthropogenic methane in the United States are the combined energy sector (natural gas, petroleum systems, and coal mining; 40% of the total), livestock (36%), and landfills (18%).

Methane emissions from livestock operations are the result of microbial fermentation and methanogenesis in the forestomach of ruminants and similar fermentation processes in manure from both ruminant and nonruminant farm animals.[3] Methane is also produced from enteric fermentation in the digestive tract of nonruminant herbivore species, such as horses, donkeys, and mules as a result of fermentation processes in their hindgut. Hindgut fermenters, however, do not produce nearly as much methane per unit of fermented feed as ruminants (IPCC[4]).

There is a large uncertainty in both enteric and manure methane emissions from livestock. Work around the world has shown that variability in enteric methane emissions can be largely explained with variability in feed dry matter intake (DMI). Nutrient composition of the feed is also important but has a lesser impact on enteric methane production than DMI.[3,5] Meta-analyses of published literature and collaborative international databases of individual animal data have clearly shown that enteric methane prediction equations based on DMI alone predict enteric methane emissions in dairy cows with accuracy similar to more complex models.[5] The same conclusions were drawn from meta-analyses of beef cattle data by Charmley et al.[6] These analyses suggest that bottom-up enteric methane inventories can be developed using a single input variable, which is available or can be predicted using standard DMI prediction equations.

Compared with enteric methane, predicting manure methane emission is a more complex process and carries a larger uncertainty in the estimates. Manure composition, type of storage facilities, manure retention time, and environment, particularly temperature, are among the factors that affect

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Table 1. Cattle Categories, Inventories, Dry Matter Intake (DMI), and Methane Emission Factors Used To Estimate County-Level Enteric Emissions for the Continental United States (Lower and Upper 90% Confidence Bounds as a Percent of the Mean Are Shown in Parentheses)

cattle category[a]	2012 cattle inventory, × 1000 head[b]	body weight, kg[c]	predicted DMI, kg/d[d]	methane emission yield, g/kg DMI[e]	methane emission factor, g/head/d[f]
(1) beef cows	28 860	613 (18.8−19.2)[g]	9.4 (27.9−28.5)	22 (18.6−19.0)	207 (32.3−35.5)
(2) dairy cows	(9262)				
dry cows	1 762	670 (18.1−18.1)	12.7 (29.0−29.1)	22 (19.0−19.1)	280 (33.0−36.2)
lactating cows[h]	7 500	670 (18.1−17.9)	22.9 (29.4−29.0)	19 (32.7−32.2)	436 (40.8−46.5)
(3) bulls	2 125	920 (18.8−19.0)	16.2 (28.0−28.3)	22 (18.6−18.8)	356 (32.4−35.6)
(4) beef replacement heifers	5 636	406 (23.5−23.1)	8.2 (23.8−23.4)	22 (18.7−18.4)	180 (29.1−32.0)
(5) dairy replacement heifers	4 785	409 (23.7−23.8)	8.5 (23.9−24.0)	19 (32.3−32.5)	161 (38.3−42.3)
(6) cattle on feed	14 377	441 (23.5−23.7)	10.3 (20.7−20.9)	10 (41.4−41.9)	103 (44.1−48.0)
(7) heifer and steers (>500 lbs or 227 kg live weight)[i]	12 084	325 (23.6−23.8)	7.5 (23.9−24.2)	22 (18.8−19.0)	165 (28.6-31.0)
(8) calves (<500 lbs or 227 kg live weight)	14 209	123 (23.6−23.6)	3.7 (24.0−24.0)	19 (32.2−32.2)	70 (38.3−42.4)

[a]Based on NASS.[10] Beef cows are cows on pasture or rangeland; bulls are both beef and dairy bulls; heifer and steers (>500 lbs or 227 kg live weight) are both beef and dairy heifer and steers. [b]Animal inventories from the 2012 Census of Agriculture;[10] total cattle = 91 338 162; dry cows = assumed at 15% of all dairy cows. [c]References: categories 1, 3, 4, 5, 7, and 8, from USEPA;[11] category 2, from USEPA[11] and Hardie et al.;[12] category 6, from Anele et al.[13] [d]For DMI equations, see Table S1. [e]Reference: for categories 1 and 2 (dry cows), from Herd et al.;[17] for categories 2 (lactating cows), 5, and 8, from Hristov et al.;[3,18,19] for categories 3, 4, and 7, from Herd et al.;[17] and for category 6, based on refs 20−22. [f]Daily methane emissions, g/head/d = methane emission yield, g/kg DMI × DMI, kg/d. [g]95% confidence bounds, as a percent of the mean, were as follows: beef cows, 22.5−22.7%, 33.5−33.7%, 22.3−22.5%, and 37.7−43.1%; dry dairy cows, 21.2−21.1%, 33.9−33.8%, 22.2−22.2%, and 38.9−43.6%; lactating dairy cows, 21.3-21.7%, 34.4−35.1%, 38.3−39.0%, and 47.3−56.0%; bulls, 23.0−22.6%, 34.2−33.6%, 22.7−22.4%, and 38.3−42.3%; beef replacement heifers, 28.5−27.4%, 28.9−27.7%, 22.7−21.8%, and 33.9−38.5%; dairy replacement heifers, 28.4−27.8%, 28.7−28.1%, 38.8−37.9%, and 44.3−52.3%; cattle on feed, 27.8−28.0%, 24.4−24.7%, 49.0−49.4, and 52.6−58.3%; heifer and steers (>500 lbs or 227 kg live weight), 28.3−28.1%, 28.7−28.5%, 22.6−22.3%, and 34.4−38.2%; and calves (<500 lbs or 227 kg live weight), 27.8−27.9%, 28.4−28.4%, 38.0−38.0%, and 44.9−53.0% for body weight, DMI, methane yield, and methane emission factor, respectively. [h]Average daily milk yield and milk fat content specific to each state were used to calculate DMI for that state.[10] [i]Heifer and steers that are not replacement heifers or cattle on feed.

methane emissions from manure.[7] Bottom-up inventories often lack critical inputs such as manure facility type and spatial distribution or monthly variability in ambient temperature necessary to accurately predict manure emissions. As an example, manure methane emissions from a dairy cow in the USEPA[1] report vary from 20 to 30 kg/head/yr for states such as Arkansas, Pennsylvania, and Tennessee to over 200 kg/head/yr for Texas, Idaho, California, and Arizona.[1] Similarly, manure methane emissions from swine operations varied from 6 to 8 kg/head/yr (Louisiana, Florida, and Massachusetts) to above 20 kg/head/yr (California, Iowa, and Oklahoma).

Thus, the overall uncertainty in bottom-up methane emission estimates for the livestock sector represents a combination of uncertainties accumulated at the various steps of the calculation process. Gridded bottom-up inventories are used as a prior estimate in top-down inversion emission models and source attribution, emphasizing the need for representative and accurate input data for construction of bottom-up inventories. Recently, Maasakkers et al.[8] developed a gridded version of USEPA's methane inventory and concluded that total United States emission estimates are generally consistent with the Emission Database for Global Atmospheric Research (EDGAR v4.2, FT2010, available from http://edgar.jrc.ec.europa.eu/overview.php?v=42FT2010[9]), which is commonly used for inversion of atmospheric methane observations, but there are large errors in spatial allocation. Therefore, the objectives of the current analysis were, using a bottom up approach, to (1) estimate livestock (cattle, swine, and poultry) methane emissions in the contiguous United States, (2) develop a spatially explicit methane emissions inventory for the livestock sector, and (3) compare this bottom-up analysis with other existing gridded inventories.

■ MATERIALS AND METHODS

County-level annual enteric methane emissions for all states were estimated for cattle only. A total of 3063 counties in the contiguous United States were included in the cattle methane emission database. The approach in estimating enteric methane emissions was based on this generalized equation:

$$\begin{aligned}&\text{methane emission from enteric fermentation (Gg/yr)}\\&\quad= \text{cattle category-specific feed dry matter intake (DMI; kg/head/d)}\\&\qquad\times \text{cattle category-specific methane emission factor (g/kg DMI)}\\&\qquad\times 365\ \text{(d/yr)} \times \text{county cattle population by category (head)}\end{aligned}$$

Cattle inventories by county were obtained from the 2012 Census of Agriculture,[10] which is the last census data available at the time the analysis was conducted. Body weight data were derived from USEPA,[11] Hardie et al.,[12] and Anele et al.[13] as specified in Table 1. Dry matter intake was estimated based on National Research Council (NRC) prediction equations for the various categories of cattle[14−16] as specified in Table S1. Methane emission yield factors (i.e., g methane/kg DMI) were based on Herd et al.,[17] Hristov et al.,[3,18,19] Archibeque et al.,[20] Hales et al.,[21] and Freetly et al.,[22] as indicated in Table 1 footnotes. Rationale for using these emission yields is discussed in Results and Discussion. On the basis of the methane emission yields, emission factors (g methane/head/d) were calculated for each cattle category.

Manure methane emissions were estimated for cattle, swine, and poultry using animal population data as described above. Swine and poultry inventory data were also obtained from NASS.[10] Swine data were analyzed on a county level for the top five swine producing states (Iowa, Illinois, Minnesota, North Carolina, and Indiana); these five states represented 68.9% of

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Table 2. Comparison of Methane Emissions from the Livestock Sector across Alternate Bottom-up Emissions Inventories

		average annual emissions from the continental United States (Gg/year)		
emissions inventory	year	enteric fermentation	manure management	total emissions
EDGAR[9]	2010	6511[a]	2146[a]	8657
Maasakkers et al.[8]	2012	6524[a]	2505[a]	9029
USEPA[1]	2012	6433[b]	2611[c]	9044
this study	2012	6201 (15.8−16.3)[b,d]	2715 (54.4−54.4)[c,d]	8916 (19.2−19.2)[d]

[a]All livestock species. [b]Cattle only. [c]Cattle, swine, and poultry. [d]Lower and upper 90% confidence bounds as a percent of the mean are shown in parentheses; lower and upper 95% confidence bounds were enteric fermentation, 15.6−16.9%; manure management, 65.0−63.3%; and total emissions, 19.3−19.2%, respectively.

the U.S. swine population (all categories). Poultry emission data were estimated on a county-level basis for the six top producing states (Georgia, Arkansas, Alabama, North Carolina, Mississippi, and Texas); these six states represented 55.9% of the U.S. poultry population (all categories). The swine and poultry databases included 469 and 728 counties, respectively. Poultry and swine emissions for the remaining states were estimated on a state level.

Manure emission calculations were adapted from IPCC[4] Tier 2 methodology to align with factors provided in USEPA[11] (Table S2). The equation below shows the overall calculation process to derive methane emissions from manure for each animal category:

$$\begin{aligned}&\text{methane emission from manure (kg/yr)}\\&\quad= (\text{animal population} \times \text{VSE} \times B_o)\\&\qquad \times [(\text{WMS}_1 \times \text{MCF}_1) + ... + (\text{WMS}_n \times \text{MCF}_n)]\\&\qquad \times (\text{methane density})\end{aligned}$$

where animal population was from NASS;[10] VSE is volatile solids excreted by an animal (kg/head/yr) from USEPA;[11] B_o is maximum methane generation potential (m^3 methane/kg VSE) from IPCC;[4] WMS is waste management systems$_{(1-n)}$ distribution in the state (%) from USEPA;[11] and MCF is methane conversion factor for the state (%) from USEPA;[11] methane density (kg/m^3) was state-specific and based on average state temperatures from NOAA.[23] A detailed description of the manure emission calculation process is provided in Supporting Information (SI) Methods.

Enteric or manure emissions from small ruminants (sheep and goats) or equine species (horses, mules, donkeys) were not included in this analysis (see SI Results and Discussion for further discussion).

We used a Monte Carlo stochastic technique to estimate confidence bounds for inputs and emission data (Tables 1 and 2). For each source data category, a normal probability distribution was assumed. We randomly sampled these distributions, or their products as per equations 1 and 2 (see SI Methods) for each animal subcategory and the sum, where total emissions are reported. The uncertainty procedure is described in detail in SI Methods.

All GIS data processing was done using ESRI ArcGIS Desktop (version 10.4; Environmental Systems Research Institute, Redlands, CA) software. County-level total enteric and total manure methane values were allocated based upon the relative area of feed sources (from USDA-NASS CropScape data[24]) within each county to produce emission rasters in units Mg methane/yr/km^2. Areas that are unlikely to be associated with livestock activities, such as forests, urban, barren, industrial, water, wetlands, and nonagronomic crop regions were excluded. This was done for all 48 continental states for cattle (enteric and manure) and select counties for swine and poultry, as explained above. All emission rasters were projected to geographic coordinates (latitude/longitude, WGS84 datum) and resampled to 0.1 decimal degree cells aligned to facilitate comparisons with other inventories (EDGAR v4.2[9] and USEPA, i.e., Maasakkers et al.[8]) at the same spatial resolution. Gridded emissions inventories were produced for each emission raster and for all livestock sources: cattle enteric, cattle manure management, total cattle emissions, and total combined, enteric and manure emissions. Cattle, swine, and poultry manure management rasters were summed to produce a combined total manure emissions raster. The gridded inventory data can be accessed at: https://psu.app.box.com/s/xjiye6mdya3qp3mxht2d6lnrnij4ioyw.

RESULTS AND DISCUSSION

Enteric Emissions. The bottom-up approach used in this analysis was driven by the fact that DMI is the single most important factor determining enteric methane emissions in ruminant animals.[3,6] Thus, the two most critical elements of this approach were estimation of DMI (Table S1) and methane emission yields (g methane/kg DMI) for the various cattle categories.

Methane yield factors used in the analysis ranged from 10 (cattle on feed) to 22 g/kg (beef and dry dairy cows, bulls, and growing beef cattle) (Table 1). These factors represent the type of diet fed to these categories of cattle and are consistent with other studies.[3,6,20−22,25] There are differences in the emissions factors for various categories of cattle between this analysis and USEPA.[11] Feedlot cattle (i.e., cattle on feed), fed ≥85% grain-based diets have a considerably lesser enteric methane emission yield than cattle fed predominantly forage-based diets.[20,22,25−27] It is noted that the emission factor for feedlot cattle in the 2014 edition of USEPA[11] was 126 g/head/d and is similar to the emission factor used by the IPCC Tier 1 approach (126 g/head/d[4]), but is about 22% greater than our average estimate of 103 g/head/d (or around 38 kg/head/yr). The emission factor for feedlot cattle used in the current analysis is based on several studies with beef cattle fed high-grain diets.[20−22] In these studies, cattle were fed typical for the U.S. beef industry corn grain (>80% of dietary dry matter)-based diets. The emission factor for cattle on feed used in the current analysis falls within the "typical" range of enteric emissions for feedlot cattle published in the most recent edition of the Beef NRC report.[25]

For all other categories of cattle, we used methane yield factors of 19 to 22 g/kg DMI, which resulted in average enteric emission factors of 70 (calves) to 436 (lactating dairy cows) g methane/head/d. The value for calves (no distinction was made between beef and dairy calves) was considerably greater than the 33 g methane/head/d emission factor used by

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USEPA,[11] but was about half the emission factor used by IPCC[4] (i.e., 145 g/head/d), which, in our opinion is unrealistically high. The calves category in Table 1 represents mostly beef calves that are weaned around 6 to 8 mo of age and weigh around 230 kg (or 500 lbs). At that age, the rumen is fully developed and from about 4 mo of age these animals consume solid feed, mainly forage on pasture (if beef), up to 2% of their body weight, which will result in enteric methane emission yields similar to those of adult cattle fed typically all forage or forage/concentrate diets. Calves with undeveloped rumen, however, will produce much less methane. For additional discussion on this topic see SI Results and Discussion.

The emission factor for beef cows (207 g/head/d or 76 kg/head/yr), a cattle category which was the largest enteric methane emitter in the current analysis, was derived from a meta-analysis by Herd et al.[17] and is approximately 20% less than the USEPA[11] emission factor for beef cows of 260 g/head/d. Herd et al.[17] measured, in respiration chambers, enteric methane emissions from over 700 individual Angus cattle (1 or 2 yrs of age) grazed on pasture. The most recent Beef NRC report[25] indicated that enteric methane emission from beef cows on pasture, which is a typical management system for this category of cattle, varied from 87 g/head/d in the fall (low pasture availability) to 252 g/head/d in spring when forage intake from pasture was high. Thus, the emission factor for beef cows used in this analysis is representative of enteric methane emissions from common cow-calf operations in the United States.

Lactating dairy cows were the second largest enteric methane emitter category in the current analysis. The methane yield factor for this category, 19 g/kg DMI, was based on the authors' own studies with high-producing dairy cows[18] and a database of individual animal data, part of the GLOBAL NETWORK project (Hristov et al.;[19] http://animalscience.psu.edu/fnn/current-research/global-network-for-enteric-methane-mitigation; accessed September 25, 2017). The average emission factor used in the current analysis, 436 g/head/d, is about 10−11% greater than the emission factor used by USEPA[11] and about 24% greater than the one recommended for North American dairy cows by IPCC.[4] We are confident that the emission factor for lactating dairy cows used in the current study is representative of the level of DMI and milk production of dairy cows in the United States. Analysis of the GLOBAL NETWORK database gave an average enteric methane yield for dairy cows of 20.4 g/kg DMI for a combined European and North American data set (2465 individual animal observations) and 18.4 g/kg DMI for North American cows only (594 observations), which is similar to the yield used in the current analysis. Furthermore, our analysis distinguishes between emissions from dry (i.e., nonlactating) vs lactating dairy cows. Dry cows usually represent about 15% of the dairy herd and their DMI is considerably less, about half of that of lactating cows, which will result in similarly lower methane emissions, although the greater fiber content of dry cow diets is likely to offset some of the difference. Using a mechanistic model (COWPOLL), Kebreab et al.[27] predicted enteric methane emission from dry dairy cows to be from 46 (a New Mexico diet) to 58 (a Kansas diet) and 86% (a Texas diet) of the emissions from mature lactating cows. The USEPA GHG inventory does not separate dry from lactating dairy cows.

The difference in emission factors between USEPA[11] and the current analysis for other categories of cattle, such as beef replacement heifers and heifer and steers (>500 lbs or 227 kg BW), was relatively small, 6 and 0.4%, respectively. For dairy replacement heifers, the USEPA emission factor was 15% greater than the emission factor used in the current analysis.

The USEPA's methodology for estimating enteric methane emission factors is, similar to the current analysis, a bottom-up approach. The process of derivation of emission factors, however, is different between the two approaches. The USEPA approach is to (1) estimate gross energy intake using IPCC[4] Tier 2 equations, (2) determine an emission factor using the gross energy intake values and region- and cattle category-specific Y_m factors, and (3) sum the daily emissions for each animal type.[11] In contrast, the current analysis relied on estimating DMI [directly, or from Net Energy of Maintenance (NE_m) requirements] based on NRC equations for the various categories of cattle and then derived emission factors based on peer-reviewed publications and a large database of individual animal data (the GLOBAL NETWORK project). The GLOBAL NETWORK project clearly showed that simple enteric methane prediction equations based on DMI (or DMI and diet characteristics such as neutral-detergent fiber or ether extract concentration) yield prediction accuracy similar to more complex models.[5] The implication of this finding is that enteric emissions from cattle and small ruminants may be accurately predicted by using simplified equations based on DMI alone. The USEPA approach[8] and the current analysis yielded comparable total enteric emissions for the contiguous United States cattle population: 6201 Gg/year in the current analysis vs 6433 (cattle only) or 6524 Gg/year,[8] which is a 3.7 to 5.2% difference. On a state level, however, there were substantial differences between the two analyses. The largest differences between USEPA[11] and the current analysis appear to be for states with large feedlot cattle populations. For example, the 2012 USEPA[11] estimate for enteric emissions from cattle in Texas was 21% greater than the estimate from the current analysis (822.6 vs 678.7 Gg/yr, respectively). Similarly, USEPA[11] estimates for Kansas and Nebraska were 17 and 22% greater than the current analysis. At the same time, USEPA[11] 2012 estimates for states with large dairy population, such as California and Wisconsin, were lower compared with the current analysis, by 9 and 13%, respectively. These differences are primarily due to differences in methane emission factors, but some may also result from using slightly different cattle inventories; USEPA uses census and survey data, whereas our analysis is based solely on 2012 Census of Agricultural data.[10] As an example, the population of dairy cattle in 2012 in the USEPA[11] report was 13 816 000, whereas the dairy cattle population used in the current analysis (from NASS;[10] excluding Hawaii and Alaska) was 9 262 240. The population of beef cattle in the 2 data sets was 81 443 000 and 82 075 922, respectively.

The uncertainty bounds derived in the current analysis for enteric methane emission factors for the most important categories of cattle ranged from −32 to +36% (beef cows) to −41 to +47 (dairy cows) and −44 to +48% (cattle on feed; Table 1) and were considerably greater than those used by USEPA,[1] −11% (lower) and +18% (upper). Dry matter intake for all animal categories varies greatly in commercial cattle operations. In controlled experiments conducted at The Pennsylvania State University with a homogeneous group of dairy cows, DMI was on average 26 kg/d with SD = 4.2 kg/d (n = 300; A. N. Hristov, unpublished data). The bounds for methane yield ranged from approximately ±19% for beef cows

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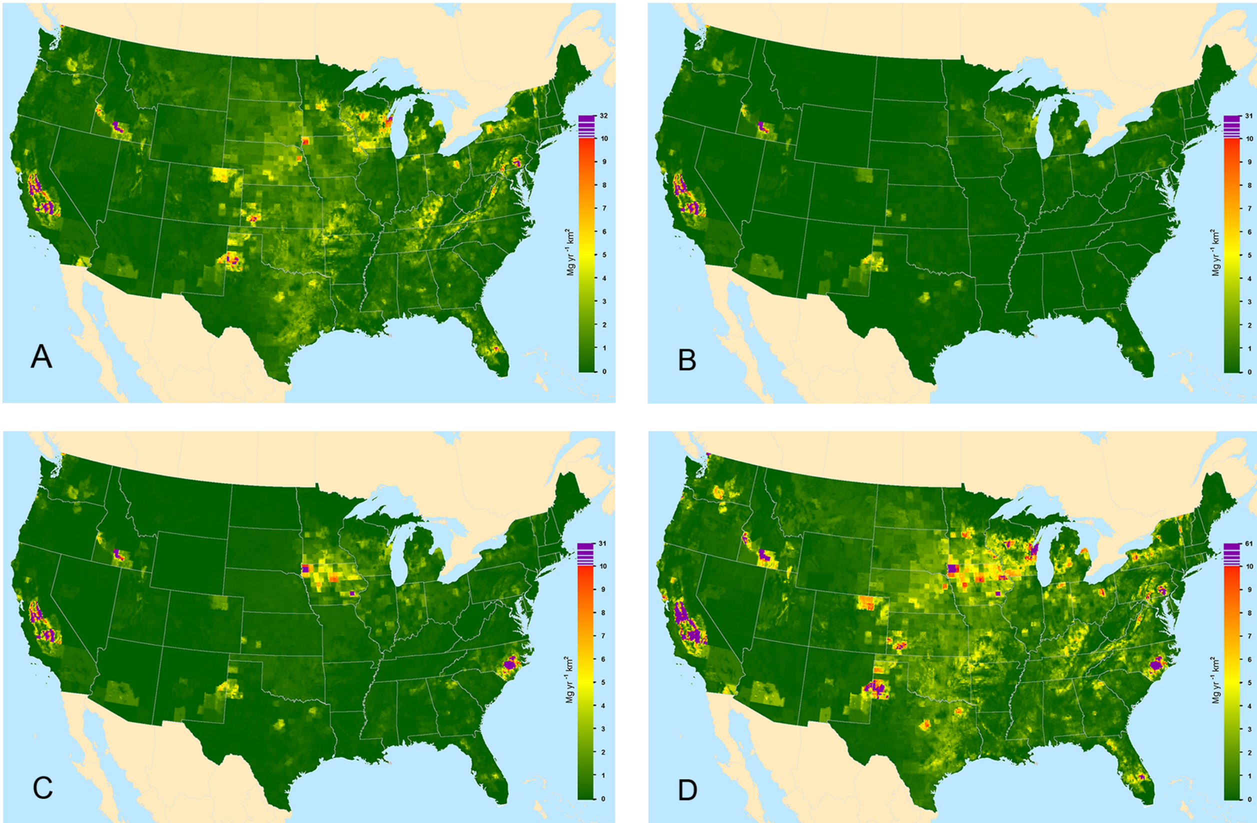

Figure 1. Gridded (0.1° × 0.1°) livestock methane emissions (Mg/yr/km^2) for the contiguous United States: enteric fermentation, cattle (panel A); manure management, cattle (panel B), manure management, cattle, swine, and poultry [panel C; swine and poultry emissions are presented on a county level for the top 5−6 producing states (see text) and on a state level for the remaining states], and cattle enteric and livestock (cattle, swine, and poultry) manure management (panel D, which is the sum of panels A and C).

to −33 and +32% for lactating dairy cows, and to −41 and +42% for cattle on feed (Table 1). On the basis of this analysis and published data, the USEPA[1] bounds for enteric methane emission appear too narrow.

Manure Emissions. Total estimated manure methane emission from cattle, swine, and poultry for the contiguous United States (2715 Gg/yr) was similar to USEPA[1] 2012 estimate (2611 Gg/yr; Table 2). This is not surprising since USEPA methodology was largely utilized in the current analysis. The largest estimated emissions were for California (421 Gg/yr), Iowa (399 Gg/yr) North Carolina (223 Gg/yr), Texas (155 Gg/yr), and Minnesota (133 Gg/yr). The uncertainty bounds for manure emissions derived in the current analysis were ±54.4% at the 90% confidence interval and −65.0 and +63.3% at the 95% confidence interval. The USEPA[1] is using considerably lower uncertainty bounds for methane emissions from manure management (−18 and +20%, lower and upper bounds, respectively). Large uncertainty in manure methane emissions has been reported in the literature (Karimi-Zindashty et al.;[28] IPCC[4] suggests ±30%) reflecting the complex nature of manure GHG emissions. A meta-analysis of on-farm studies by Owen and Silver[29] showed that anaerobic dairy lagoon methane emissions were on average 368 kg/head/yr but SD was 579 (SE = 193; n = 9) and the range of emissions was from 4 to 2814 kg/head/yr.

Many factors affect manure methane emissions, including type of animal, manure composition (its volatile solids concentration) and management system, retention time of manure in the manure system, and climate. Reliable, peer-reviewed data are lacking for many of these factors. The authors chose to utilize B_o and other factors published by the USEPA[11] because comprehensive accumulation of such a large volume of factors was beyond the scope of the current study. The authors acknowledge that some USEPA[11] factors are based on older data that may not reflect current practices or distribution of manure management systems. For instance, table values for B_o from dairy cows,[30] heifers,[31] and other cattle categories[32] are now at least 35 yrs old. Examining USEPA[1] manure emission input data for 2 states with large dairy industries, Pennsylvania and Idaho, showed that 65 vs 6% of the manure is being handled in anaerobic lagoons, respectively, with 46% of the manure in Pennsylvania being handled as daily spread. These differences in manure composition and storage facility result in a 10-fold difference in manure methane emissions from a dairy cow between the two states. It is unclear to what extent this difference is truly representative of differences in manure methane emissions between dairy operations in Pennsylvania and Idaho. Although official data does not seem to exist, based on our observations and trends for consolidation of the dairy industry in the state, it is unrealistic that manure from 46% (USEPA[1]) of the cows in Pennsylvania is hauled daily, a manure handling system that results in considerably lower methane emissions than other systems. In fact, many smaller dairies in Pennsylvania store manure in under-floor pits (so-called, gravity-flow systems) where anaerobic conditions develop rapidly and manure methane emissions can be extremely high.[33] These large discrepancies in methane emission among manure management systems is reflected in

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our uncertainty estimates and can affect the accuracy of gridded bottom-up methane emission inventories.

Another important factor controlling manure methane emissions, temperature, can vary greatly throughout the year and assuming a single annual average temperature and methane conversion factor (for cool, temperate, or warm climate; USEPA[1]) is likely also going to influence the accuracy of manure emission inventories. A recent case study compared top-down and bottom-up methane emission estimates at two California dairy farms.[34] The authors reported that open-path measurements for liquid manure storage emissions were similar to monthly USEPA estimates during the summer but not during the winter, and neither summer nor winter open-path estimates were similar to the annual USEPA estimate. This study also showed that methane emissions from manure settling basins can be considerably greater than emissions from the anaerobic lagoon and measured emissions can vary as much as 230% (0.75 to 1.72 kg/animal unit/d; Arndt et al.[34]). Extensive monitoring at a single Canadian dairy farm[35] provided manure methane emission data that were up to 60% greater than both USEPA[36] and IPCC[4] model estimates. Similarly, Owen and Silver[29] concluded from their meta-analysis that current models (IPCC[4]) underestimate methane emission from dairy manure. Another meta-analysis of data for swine systems showed large discrepancies between measured and estimated, using IPCC[4] methods, methane emissions from the buildings and lagoons.[37] Thus, it is likely that manure methane emission factors currently used by USEPA need to be updated to accurately predict total livestock GHG emissions, which emphasizes the need for current research on emission factors for manure, particularly as influenced by the manure management system. As noted by VanderZaag et al.,[38] year-round methane emission monitoring across various manure management systems and climate conditions is necessary to produce data that can be used with existing IPCC[4] Tier 2 equations.

Gridded Emission Inventory. County-level estimates of methane emissions from the livestock sector for the contiguous United States were disaggregated to a finer spatial grid (0.1° × 0.1°) and are visually shown in Figure 1. With a few exceptions, the intensive emission "hot-spots" in various regions corresponded to large cattle, particularly dairy, populations. As an example, in 2012 there were a total of 1 086 890 cattle (including 489 436 dairy cows) in Tulare and 558 926 (including 285 235 dairy cows) in Merced Counties, California, 301 982 (including 178 661 dairy cows) in Gooding County, Idaho, 400 552 (all beef cattle) in Haskell County, Kansas, 435 990 (91% beef cattle) in Castro County, Texas, and 276 729 (including 110 805 dairy cows) cattle in Lancaster County, Pennsylvania. These high animal population densities corresponded to enteric methane emissions up to 30 Mg/yr/km^2 (Figure 1, panel A). Compared with these emission hot-spots, enteric emissions were less intensive in the Midwestern states and Texas (except the Texas Panhandle, which has a high density of dairy cattle), partially reflecting predominance of feedlot cattle fed high-grain finishing diets in these areas.

The emission "hot-spots" for cattle manure (Figure 1, panel B) correspond to counties with large dairy cattle population, such as counties in California's Central Valley and Idaho's Magic Valley. In these areas manure methane emissions reached 29 to 31 Mg/yr/km^2. As already discussed, manure methane emission factors depend greatly on the type of manure management system and ambient temperature; emission factors for dairy manure are considerably greater than emission factors for manure from beef cattle. For example, the methane conversion factor for dry-lot manure systems (typical beef feedlots) is 0.01 (cool climate) to 0.05 (warm climate), whereas the conversion factor for anaerobic dairy lagoons ranges from 0.67 (Wisconsin, i.e., colder climate) to 0.80 (Florida, i.e., warmer climate);[11,4] see also Table S2. The combined manure methane emissions map for cattle, swine, and poultry (Figure 1, panel C) shows, in addition to emissions from dairy counties, emission hot-spots for counties with dense swine population in states such as North Carolina (Sampson, Duplin, Wayne, and Bladen Counties) and Iowa (Washington and Sioux Counties). Methane emissions from poultry manure are low and the contribution of states with high poultry population, such as Alabama or Georgia, to the total manure emissions is less visible in Figure 1 (panel C).

The combined (cattle enteric and manure emissions from all species) county-level methane emission map is shown in Figure 1, panel D. This map is a compilation of all enteric and manure emission data and as such shows similar trends to the maps already discussed above. Counties with the largest combined livestock methane emissions include Tulare, Merced, Stanislaus, and Kings, California (217, 123, 80, and 78 Gg methane/yr, respectively); Gooding, Idaho (75 Gg/yr); Weld, Colorado (63 Gg/yr); Kern, Fresno, and San Joaquin, California (62, 59, and 49 Gg/yr), Maricopa, Arizona (47 Gg/yr), Sampson, North Carolina (44 Gg/yr); and Yakima, Washington, and Sioux, Iowa (both at 43 Gg/yr). The spatial distribution of emissions in this study is similar to the recent analysis by Maasakkers et al.,[8] although these authors used USEPA[11] (2014 data, updated in 2016) livestock emission inventories, which are derived through a similar bottom-up methodology, but using different approach and emission factors for enteric methane.

As stated earlier, our approach in developing the county-level gridded emission inventories excluded areas that are unlikely to be associated with livestock activities, such as forests, urban, barren, industrial, water, wetlands, and nonagronomic crop regions. This approach resulted in nonuniform distribution of enteric and/or manure methane emissions for counties with large livestock populations, but diverse landscapes and more uniform distributions for counties with less diversity. For example, diverse counties such as Gooding County, Idaho, showed mean total methane emissions of 37 Mg/yr/km^2 with SD = 12.2 among individual 0.1 degree cells within the county. Other more uniform land cover counties such as Sioux, Iowa, showed much less variability among cells (mean = 20 Mg/yr/km^2; SD = 2.2). Where available, spatial distributions of individual livestock feeding operations should be used to allocate emissions to grid cells, as in the study by Cui et al.[39] for California's San Joaquin Valley.

Comparisons with Existing Bottom-up Inventories. Gridded bottom-up emission inventories are commonly used to assess the contribution of methane from different sectors within a region, whether in isolation, or in conjunction with top-down approaches that use these inventories as a prior estimate, and/or to allocate the resulting (posterior) emission estimates to these sectors.[40] The global, gridded EDGAR[9] emissions inventory includes estimates of methane emissions from enteric fermentation and manure management in the livestock sector, and has been extensively used in top-down studies. A more recent bottom-up inventory analysis suggested that global livestock methane emissions are 11% greater compared with estimates based on IPCC[4] emission factors.[41] It is interesting to

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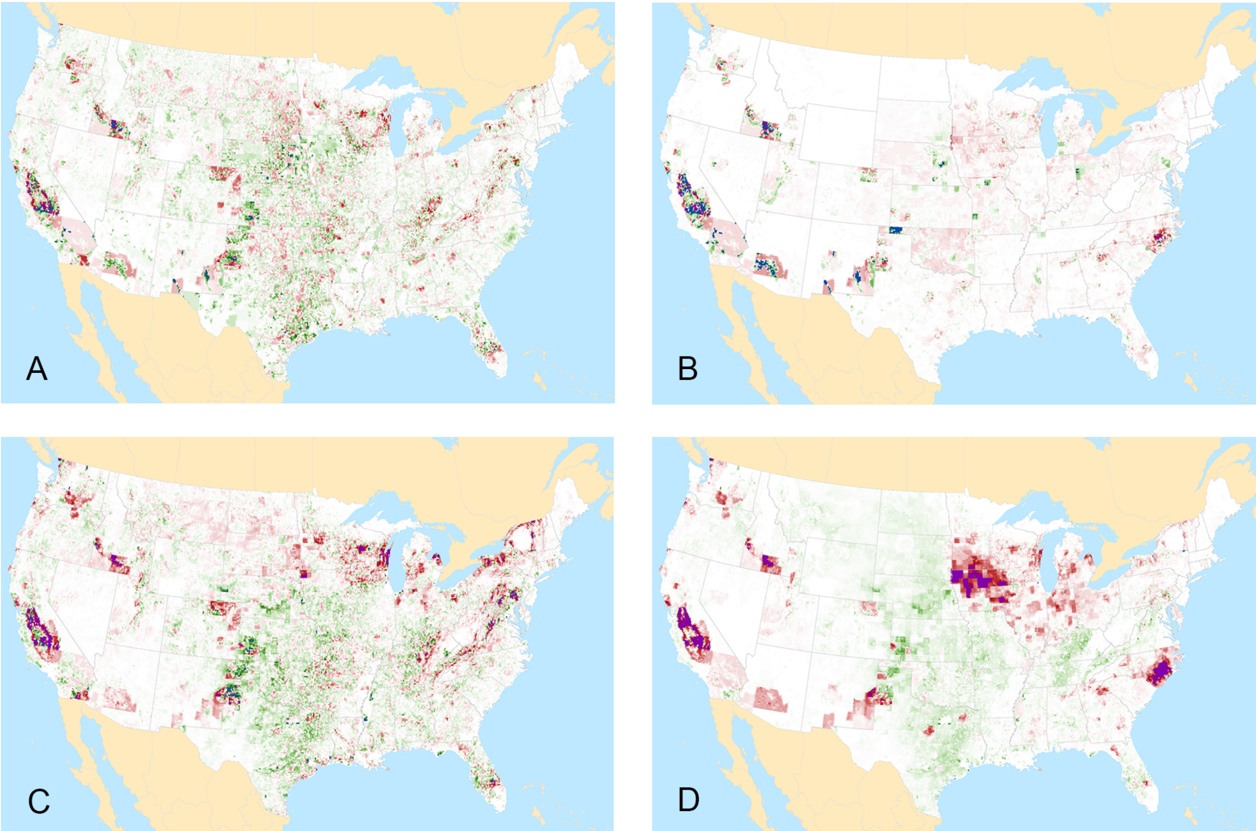

Figure 2. Gridded difference in livestock enteric and manure methane emissions (Mg/yr/km^2) between bottom-up approaches: (A) this analysis minus Maasakkers et al.,[8] enteric fermentation; (B) this analysis minus Maasakkers et al.,[8] manure management; (C) this analysis minus EDGAR[9] (http://edgar.jrc.ec.europa.eu/overview.php?v=42FT2010) enteric fermentation, and (D) this analysis minus EDGAR,[9] manure management.

point out that the Y_m factors for dairy cows used in the Wolf et al.[41] analysis were lower than IPCC[4] factors for the United States and Canada, but considerably greater for Latin America, Africa, and South Asia. Manure inputs for dairy cows, such as VSE and MCF for liquid systems, were also considerably greater than IPCC[4] inputs. Maasakkers et al.[8] used state-level USEPA emission factors and a range of other data sources to estimate a gridded inventory of methane emissions for livestock, among other sectors.[8]

The United States total methane emissions from the livestock sector estimated by EDGAR,[9] Maasakkers et al.,[8] and the USEPA[1] national GHG inventory (data for 2012) are consistent with the findings from this analysis (Table 2), within ±3%. However, this study estimates emissions from enteric fermentation to be consistently lower than that reported by the other databases; the difference with EDGAR and Maasakkers et al.[8] is partially due to the fact that the current inventory includes cattle only. Conversely, for manure management, this study estimates greater national total emissions than the other sources, for example, 27% greater than the EDGAR inventory. As discussed earlier, the differences in enteric emission estimates between the current study and Maasakkers et al.,[8] which is the national USEPA methane inventory, are mainly due to different emission factors for the various categories of cattle. For example, states with high feedlot cattle populations had lower emission values in the current report, compared with Maasakkers et al.[8] and USEPA[1] because we use a lower emission factor for feedlot cattle. In contrast, the emission factor for dairy cows used in the current report was greater than that used in Maasakkers et al.[8] and USEPA[1] and, therefore, states with large dairy cattle inventories had greater enteric methane emissions in the current report.

The county-level estimates of methane emissions from the livestock sector were disaggregated to a finer spatial grid (0.1° × 0.1°), using approaches discussed above. Figure 2 and SI Figures S3–S7 present a state level comparison, disaggregated by subsector (enteric fermentation and manure management) for EDGAR v4.2,[9] Maasakkers et al.,[8] and this analysis. The differences in spatial distribution of emissions between this analysis and the EDGAR[9] inventory were greater than differences between this analysis and Maasakkers et al.[8] (Figure 2). As an example, the difference between the current analysis and EDGAR[9] in enteric emissions for a predominantly dairy county such as Gooding, Idaho (178 661 dairy cows in 2012) was 15.6 Mg/yr/km^2 compared to a difference between the current analysis and that of Maasakkers et al.[8] of 3.2 Mg/yr/km^2. In another example, the differences in manure emissions for Sampson County North Carolina (1.9 million hogs and pigs and 11.5 million poultry in 2012) were 18.0 vs 2.4 Mg/yr/km^2, respectively. At the state level, methane emissions from livestock in Texas and California (highest contributors to the national total) in this study were 36% lower and 100% greater, respectively, than estimated by EDGAR.[9] The correlation coefficients between EDGAR[9] and this study and Maasakkers et al.[8] for the contiguous United States were 0.70 for enteric fermentation and 0.33 to 0.34 for manure management at the 0.1° × 0.1° grid scale, as presented in Table S3. The correlation between this study and Maasakkers et al.[8] were 0.90 and 0.93 for enteric fermentation and manure, respectively. Interestingly, the correlation between this study and Maasakkers et al.[8] for

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enteric fermentation emissions for Texas were high (≥0.90), suggesting that the locations of the emissions may be more consistent between these databases (driven by livestock population inventories), while the total emissions were different, indicating different emission factors.

Maasakkers et al.[8] highlighted the need for improved prior estimates (bottom-up inventories) particularly in the South Central United States, due to strong correlations between the different sectors (e.g., oil and gas and livestock) within EDGAR[9] for that region. The EDGAR inventory consistently overestimates emissions from the livestock sector in this region, relative to the current study, for enteric fermentation and manure management (Figure 2). This is counterbalanced by a consistent underestimation by EDGAR relative to this inventory in California and the Midwest region. In comparison, the largest spatial differences within states between Maasakkers et al.[8] and the current analysis were observed in Texas and California, both states with large livestock populations (Figure 2).

These differences in spatial distribution will likely have a strong impact on posterior emissions estimated by top-down approaches, within the contiguous United States, even if the overall magnitudes of these estimates are consistent. It is noted that the change of spatial repartition of prior emissions will influence state-scale estimates from inversions. The regions with the highest differences in emissions correspond to regions that include other sources of methane as well, such as the oil and gas sector, or landfills. Therefore, this may have an even greater impact on the attribution of total methane emissions to these sectors, since this is currently determined by bottom-up inventories, in the absence of other indicators such as isotope markers or ethane, propane, or higher hydrocarbons. Whereas no single bottom-up inventory can be considered entirely accurate, and there is a large uncertainty in the emission estimates (as shown in the current analysis), using a range of different estimates in conjunction may provide better uncertainty bounds that would be valuable within a top-down framework.

Data from the current analysis were also compared with bottom-up[42] and top-down[43,44] estimates for 25 counties in the Barnett Shale region of Texas (Figure S8) and livestock emission inventories from the California Greenhouse Gas Emissions Measurement (CALGEM) Project (Figure S9) http://calgem.lbl.gov/prior_emission.html; accessed September 25, 2017). For the Barnett Shale region, our total livestock emission estimates were comparable to the bottom-up estimates of Lyon et al.[42] The spatial distribution of the emissions, however, was largely different between the two analyses and we could not find a relationship between emission source (livestock, oil and gas, or other) and the discrepancy between the two estimates. Similarly, overall county emission differences between the current analysis and the CALGEM inventory were relatively small but estimates differed spatially on a subcounty scale. A more detailed discussion can be found in SI Results and Discussion.

In the current analysis, we used a unique approach for estimating enteric methane emissions, estimate uncertainties, and allocate emissions to the $0.1° \times 0.1°$ grid. The uniqueness of the enteric methane approach is that it used simple inputs such as DMI, predicted based on equations from the National Research Council for the various cattle categories, and methane yield (per kg DMI), derived from large databases or published peer-reviewed research, to estimate enteric emission factors. These emission factors can be used to produce more detailed and accurate gridded inventories for regions where farm location and other information are available. Our study also highlighted the large uncertainty in manure methane emissions and the need for accurate input data, particularly data related to type and allocation of manure management systems and flow of manure through the system.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b03332.

> Additional and detailed description of methods; discussion of calf emission factors; discussion on trends in atmospheric methane concentrations and livestock contribution; comparison of bottom-up inventories; tables detailing equations used to estimate dry matter intake; estimation of manure methane emissions, and correlations among methane emission estimates from the current analysis and literature values; figures with comparative state emissions data for the current analysis, and literature values; differences in livestock methane emissions between the current analysis and literature values for the Barnett Shale region of Texas and CALGEM (PDF)

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The authors declare no competing financial interest.

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