STRATEGIES TO REDUCE METHANE EMISSIONS FROM ENTERIC AND LAGOON SOURCES

(Contract 17RD018)

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List of Abbreviations

3NOP	3-nitrooxypropanol
BW	Body weight
CARB	California Air Resources Board
CDFA	California Department of Food and Agriculture
CP	Crude protein (% dry matter)
DDGS	Dry distillers grain with solubles
DMI	Dry matter intake (kg/d)
ECM	Energy corrected milk
GE	Gross energy
GHG	Greenhouse gases
IPCC	Intergovernmental Panel for Climate Change
LCA	Life cycle assessment
MD	Mean difference
NDF	Neutral detergent fiber (% dry matter)
OM	Organic matter)
RMD	Relative mean difference
RVE	robust variance estimation
SMD	Standardized mean difference
USDA	United States Department of Agriculture
USDA NASS	United States Department of Agriculture National Agricultural Statistics Service
USDA ERS	United States Department of Agriculture Economic Research Service

ABSTRACT

The State of California launched the short-lived climate pollutant reduction strategy (SB 1383) with the objective of decreasing methane (CH_4) emissions from livestock by 40% by 2030 from 2013 levels. Considering about 50% of CH_4 emissions in the State are attributed to enteric fermentation and manure, achieving significant CH_4 emission reduction from these sources will be critical to meeting SB1383 goals. There are numerous mitigation options described in the literature including feed and manure additives. The objective of the study was to provide quantitative analysis, evaluate feasibility, and summarize and prioritize research gaps to guide future research in the State. Specifically the current study conducted a literature review of available mitigation strategies using additives to reduce enteric and manure methane emissions including size effect and performance analyses and used life-cycle assessment tools to estimate net greenhouse gas emissions from using potential feed additives in the dairy industry. Effect size and meta-analyses were conducted to identify the additives with greatest potential for CH_4 mitigation. For feed additives, 3-nitrooxypropanol (3NOP), bromochloromethane, chestnut, coconut, distillers dried grains and solubles, eugenol, grape pomace, linseed, monensin, nitrate, nitroethane, saifoin, fumaric acid, and tannins had significant impacts on enteric emissions. For manure additives, acidification, biochar, microbial digestion, physical agents, straw, and other chemicals significantly reduced CH_4 emissions. However, there were other promising additives that need further research, including Mootral, macroalgae and SOP lagoon additive (SOP). After further analysis of variance, the most effective feed additives were 3NOP (41% in dairy and 22% reduction in beef) and nitrate (14.4% reduction). Biochar as a manure additive can be effective on compost manure (up to 82.4% reduction), but may have no impact on lagoon emissions. A life cycle assessment tool was used to estimate the net reduction in enteric CH_4 emissions by

using the feed additives 3NOP and nitrate. The overall average net reduction rate of supplementing 3NOP and nitrate were 11.7% and 4.9%, respectively. Given the toxicity concerns of nitrate, only 3NOP is recommended for use pending FDA approval. Considering California milk production of 18 billion kg in 2017, using nitrate on California dairy cows would reduce GHG emissions 1.09 billion kg CO_2e and 3NOP 2.33 billion kg CO_2e annually. Further research in the additives of Mootral, macroalage, SOP, biochar and other emerging ones is required before recommendation for use can be made.

EXECUTIVE SUMMARY

Background

About 50% of CH_4 emissions in California are attributed to enteric fermentation and manure; therefore, achieving significant methane (CH_4) emission reduction from these sources will be critical to meeting SB1383 goals. There are several strategies for reducing CH_4 emissions from enteric fermentation and manure management in the literature (e.g., Knapp et al. 2014), including diet manipulation, feed additives, anaerobic digestion and liquid-solid separation. A number of excellent reviews on enteric methane mitigation techniques have already been published. Similarly, there are a number of reviews available that summarize mitigation options to reduce CH_4 emissions from manure management. However, none of the reviews quantitatively evaluate impact of feed and manure additives in a meta-analytic and holistic manner. The overall objective of this study was to review feed and manure additives used for CH_4 emission reduction and identify those with the potential to be applied in the California livestock industry. The feed and manure additives were classified into three categories as follows. Category 1: Safe and effective for methane use, recommended when all regulatory approvals are in place. Category 2: Research to date shows this product may be effective and more research is required before it is recommended for use. Category 3: Research to date has either provided insufficient evidence to conclude that the product may be effective, or has shown that product is not effective, or has shown that the product should not be used for other reasons.

Methods

Extensive literature survey on feed and manure additives was conducted and data collected in an excel spreadsheet that includes information on methane emissions as well as dietary and other factors. Effect size estimates of mean difference (MD; i.e., mean treatment minus mean control) and standardized mean difference (SMD) were calculated using the open source statistical software R (version 3.6.1, R Foundation for Statistical Computing, Vienna, Austria). For some feed and manure additives, a meta-analysis was conducted using the robust variance estimation method to deal with unknown correlations among non-independent effect sizes. For the most promising feed additives, a life cycle assessment approach was taken in which crop production, additive production, farm operation, enteric emissions, and manure emissions were taken into account to estimate the net greenhouse gas emission in producing a kilogram of milk.

Results

A literature survey of feed additives with anti-methanogenic properties revealed over 90 potential additives. However, after analyzing their impact on CH_4 emissions only 3-nitrooxypropanol (3NOP), bromochloromethane, chestnut, coconut, DDGS, eugenol, grape pomace or marc, linseed, monensin, nitrate, nitroethane, saifoin, fumaric acid, and tannins had overall CH_4 reduction potential. Of these, only 3NOP and nitrate were considered to have the best potential outcome for mitigation. Feed additives such as Mootral, macroalgae and Agolin have also shown promise but there is limited *in vivo* work to allow full consideration. A total of 13 categories of manure additives were included for their potential to reduce emissions. In a meta-analysis, acidification, biochar, microbial digestion, physical agents, straw, and other chemicals significantly reduced CH_4 emissions. The most effective manure additive was biochar, which significantly reduced CH_4 emissions by up to 82.4%. However, further work is needed to develop a protocol on the type/dose of biochar and its effectiveness based environmental conditions. Other manure additives were not included in the analysis because only one or two experiments have been conducted (e.g. SOP; Borgonovo et al., 2019). It has a potential but needs further study. The two promising feed additives that been research extensively were further evaluated using a life cycle assessment tool to estimate their net reduction potential from dairy systems in California by considering their impact on other parts of the industry as well as environmental cost of additive production. The average net reduction rate of supplementing 3NOP and nitrate were 11.7% and 4.9%, respectively. 3NOP had a greater effect than nitrate on reducing total GHG emissions with a highest performance of 11.8%. Feeding 3NOP to only lactating cows or to the entire growth stages did not make significant difference in total GHG emissions. Considering California milk production of 18 billion kg in 2017, using nitrate on California dairy cows would reduce GHG emissions by 1.09 billion kg CO_2e and 3NOP by 2.33 billion kg CO_2e annually. Unless the toxic effect of nitrate at high doses are mitigated, nitrate is not recommended at present.

Conclusion

At the writing of the report, we recommend 3NOP to be in Category 1 with the highest potential impact pending FDA approval. Nitrate (if toxicity mitigated), Mootral, macroalgae, Agolin and grape pomace are recommended to be in Category 2 with further experiments required to verify the impact already shown in California. The rest should be in Category 3, which include additives not recommended at this time. For manure additives, biochar is in Category 1 with the caveat already mentioned above. Acidification and SOP manure additive are in Category 2, which need further study. Most of the research for biochar and straw is when used as additive to solid or semi solid manure so they should be interpreted in that context.

INTRODUCTION

Global emissions of greenhouse gases (GHG) have risen to unprecedented levels despite a growing number of policies to reduce climate change (IPCC, 2014). Anthropogenic sources account for 58% of global GHG emissions (EPA, 2011), 18% (5.0 – 5.8 Gt CO2eq /yr) of which was generated by agriculture-related activities during 2000–2010 period (Smith et al., 2014). Methane (CH_4) from enteric fermentation and manure was the largest contributor (40%) to the agricultural GHG emissions (Tubiello et al., 2013). The largest source of anthropogenic CH_4 in the US is from livestock, particularly ruminants (EPA, 2017).

The State of California launched the short-lived climate pollutant reduction strategy (SB 1383; CARB 2017) with the objective of decreasing CH_4 emissions from livestock by 40% by 2030 from 2013 levels. About 50% of CH_4 emissions in the State are attributed to enteric fermentation and manure (CARB, 2020); therefore, achieving significant CH_4 emission reduction from these sources will be critical to meeting SB1383 goals. There are several strategies for reducing CH_4 emissions from enteric fermentation and manure management in the literature (e.g., Knapp et al. 2014), including diet manipulation, feed additives, anaerobic digestion and liquid-solid separation. This proposal is focused on additives that reduce methane emissions from enteric and lagoon sources.

A number of excellent reviews on enteric methane mitigation techniques have already been published (e.g., Boadi et al., 2004; Beauchemin et al., 2009; Cottle et al., 2011; Hristov et al., 2013). Similarly, there are a number of reviews available that summarize mitigation options to reduce CH_4 emissions from manure management (e.g., Kebreab et al., 2006; Jayasundara et al., 2016). Recently, an international group of scientists (including the PI) conducted a comprehensive analysis of mitigation options for reducing enteric (Hristov et al., 2013a) and manure (Montes et al., 2013) emissions. The intention of this proposed study is not to reproduce them but to evaluate statistically the effectiveness of various mitigation techniques. Studies on novel feed additives have been published recently and continue to be reported in the literature, which may have not been included in the previously mentioned reviews. There is a need for a comprehensive review and analysis of additives that have the potential to be successful in California in mitigating emissions. The review will take a holistic approach and extend to include a life-cycle analysis (LCA) of the impact of additives. This will allow a fuller environmental impact assessment, which is associated with implementing some of the additives that have already been developed and some that are currently being tested.

The overall objective of this study is to review feed and manure additives used for methane emission reduction and identify/categorize those with the potential to be applied in the California livestock industry. The strategies will be analyzed not only for their potential to reduce emissions but also their impact, if any, on product quality and animal welfare. Analysis of additives for methane mitigation potential will take a life-cycle approach, which will be required in case production and implementation of additives will have upstream and downstream consequences that may change the net benefit. The additives will be placed into the following three categories: Category 1: Safe and effective for methane use, recommended when all regulatory approvals are in place. Category 2: Research to date shows this product may be effective and more research is required before it is recommended for use. Category 3: Research to date has either provided insufficient evidence to conclude that the product may be effective, or has shown that product is not effective, or has shown that the product should not be used for other reasons. The ultimate objective is to provide quantitative analysis, summarize and prioritize research gaps to guide future research in the State. The following specific objectives will be addressed in the current study:

1. Literature review of available mitigation strategies using additives to reduce enteric and manure methane emissions including size effect and performance analyses.

2. Prioritize research gaps and use life-cycle analysis to assess potential unintended impacts such as greater emission in sourcing the product or product development.

FEED ADDITIVES TARGETING ENTERIC METHANE EMISSIONS

A literature survey of feed additives used targeting enteric methane emissions was conducted. There were a total of 90 different feed additives collected from the literature. The counts of treatment, averages, standard deviations, minimums and maximums of Mean Difference (i.e., mean treatment minus mean control) of CH_4 production for control/treatment groups based on feed additive type is summarized in Appendix 1. Methane production and methane production per dry matter intake (DMI) were expressed in g/day and g/kg, respectively. Effect size estimates of mean difference (MD) and standardized mean difference (SMD) were calculated using the open source statistical software R (version 3.6.1, R Foundation for Statistical Computing, Vienna, Austria). Any feed additive related studies without CH_4 production information listed in the database were excluded in the further analysis. Furthermore, feed additives with only one record were excluded in further tests because lack of replications prevents the calculation of standard deviation, and *P*-values.

After the data was filtered and selected based on the criteria mentioned above, a sample *t*-test (treatment-control) was conducted based for each feed additive. Table 1 gives the *P*-values from significant *t*-test (α=0.05). As a result, feed additives including 3-nitrooxypropanol (3NOP), bromochloromethane, chestnut, coconut inclusion, DDGS concentrate, eugenol, grape pomace, linseed, monensin, nitrate, nitroethane, saifoin, fumaric acid, hydrolysable tannins, and *Sericea lespedeza* tannins significantly impacted the MD of CH_4 production. Similarly, a box and forest plots were constructed to assess the impact of feed additives on methane production (Fig. 1, 2).

Table 1. Impact of feed additives on CH_4 reductions (g/d) based on *t*-test.

Feed additive*	***P*-value**	**Feed additive**	***P*-value**
3NOP	*0.002*	Legume	0.403
Acacia mearnsii	0.057	Linseed inclusion	*0.007*
Acetate inclusion	0.084	Lotus tannins	0.989
Microbial culture	0.051	Lovastatin	0.130
Bromochloromethane	*<0.001*	Lupine seed	0.449
Calcium soap inclusion	0.248	Malic acid	0.184
Canola inclusion	0.291	Methylbutyrate inclusion	0.109
Carboxylic acid	0.636	Mimosa	0.310
Cerium chloride	0.078	Monensin	*<0.001*
Chestnut	*0.044*	Myristic acid	0.561
Chitosan	0.841	Nisin	0.205
Coconut inclusion	*0.005*	Nitrate	*<0.001*
Corn	0.183	Nitroethane	*0.045*
Cumin	0.069	Oregano	0.077
Cysteine	0.587	Polyethylene glycol	0.256
DDGS concentrate	*0.012*	Quebracho	0.109
DHA inclusion	0.372	Saifoin	*0.027*
Essential oil blend	0.172	Saponaria	0.177
Eugenol	*0.008*	*Sericea lespedeza* tannins	*0.008*
Fatty acid blend inclusion	0.101	Sorghum tannins	0.150
Fibrolytic enzyme	0.223	Soybean oil inclusion	0.749
Flaxseed inclusion	0.454	Stearic acid	0.719
Garlic	0.848	Sunflower inclusion	0.079
Glycerin	0.793	Tea saponin	0.096
Grape pomace	*0.050*	Triiodothyronine	0.695
Grass	0.676	Valonea	0.883
Hydrolysable tannins	*0.018*	Vitacogen	0.587
Iso-valerate inclusion	0.093	Yucca	0.172
Lasolocid	0.946		
Lauric	0.427		

Some data from Global Network project are included.

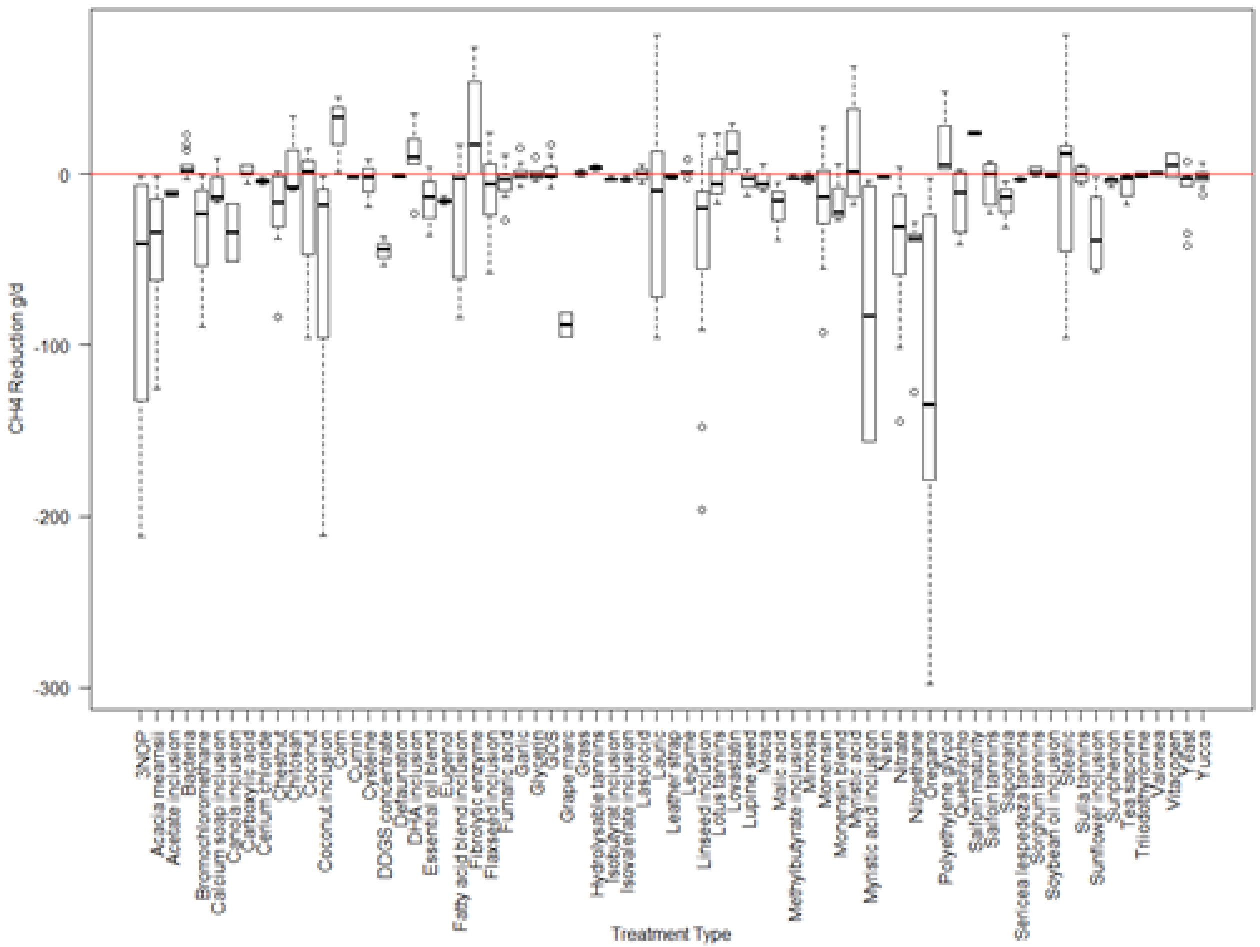
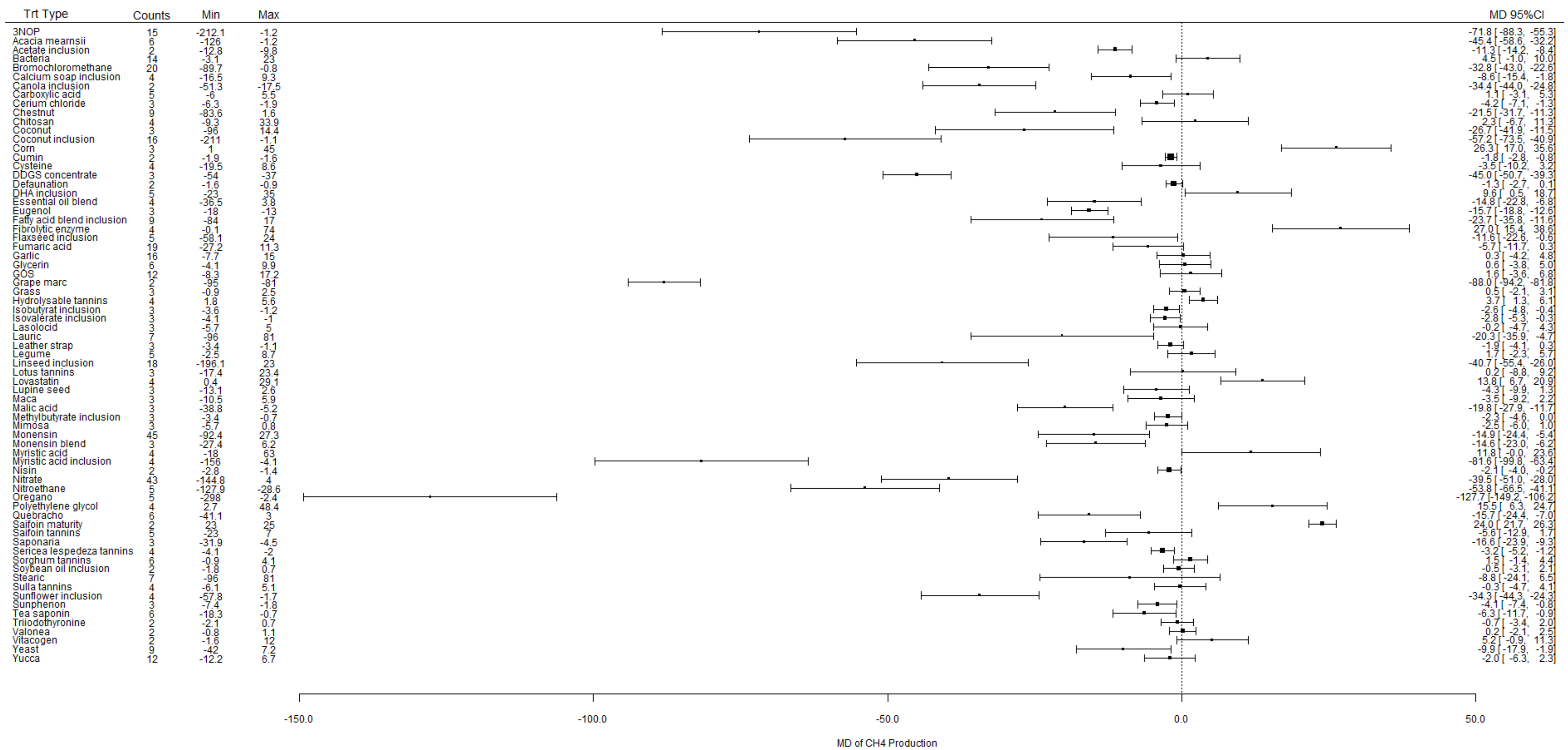


Figure 1. Boxplot of mean difference of CH_4 production. The horizontal line was the reference line of 0 g/d reduction. Some data from Global Network project are included.



Figures 2. Forest plot including the summary of valid treatment counts, lower and upper boundaries of mean difference for different feed additives with 95% credible interval. Some data from Global Network project are included.

Once the potential feed additives were identified, a secondary assessment was conducted to investigate their appropriateness for California livestock industry including cost, unintended negative consequences, availability and persistence in reducing emissions. The use of chestnut, coconut inclusion, DDGS concentrate, eugenol saifoin, fumaric acid and linseed appear to increase the cost of production as well as reduce productivity or pollutions swapping. For example increased use of DDGS may reduce methane but increases nitrogen loading, which may contribute to increased N_2O emissions. Therefore, these mitigation options were not considered further. The study by Appuhamy et al. (2013) showed that although monensin reduced methane production by about 6% in beef and 12% in dairy cattle, the effect was transient. After about 6 weeks of monensin supplementation the rumen microbes adapt to it and any benefit in reduction of methane emissions is lost. Therefore, monensin was not considered further.

Feed additives with some potential for mitigation

Secondary plant compounds

Tannins have shown promise for methane mitigation but not much work has been done in California conditions. There is a need for further investigation of the use of tannins, in particular grape pomace (or grape marc), in California, as the raw materials are easily available. UC Davis plans to conduct a trial on grape pomace. There are also feed additives that were not considered fully because of lack of studies. A feed additive based on citrus and garlic extracts, Mootral, has been studied in California. Roque et al. (2019a) showed that after 12 weeks of supplementation, Mootral reduced methane emissions by 23%. The trial was relatively short and involved 20 animals. A bigger trial with 45 beef cattle and longer period has been planned for summer 2020 at UC Davis. Following that, another trial with Mootral using dairy cattle is planned for fall 2020.

These studies will shed light to the effectiveness of Mootral under California conditions and need to be considered for use after the results are published. A research trial using Agolin has been conducted at UC Davis but results are not yet public and may be available in 2021.

Methanogenesis Inhibitors

Bromochloromethane in its pure form cannot be used as it is a banned substance under the Montreal Protocol. However, some seaweed species, particularly *Asparagopsis*, contain bromoform and bromochlormethane as active ingredients that has been shown to be effective *in vitro* (Machado et al., 2016). The first *in vivo* trial using *Asparagopsis* in cattle was conducted at UC Davis (Roque et al., 2019b) who reported up to 67% reduction in methane production in dairy cattle. The authors reported a decline in feed intake, particularly at the high level of inclusion, which might compromise milk production. Bromoform residue was not found in milk samples. A new paper was published during the final report-writing phase of this study. Kinley et al. (2020) reported that methane emissions in Brangus cattle declined 98% with inclusion of only 0.02% of *Asparagopsis taxiformis*. Additionally, they reported no reduction in feed intake or loss of productivity. Analysis of the meat from seaweed supplemented animals did not show any bromoform residue. Human consumption of high levels of bromoform could be hazardous, so the US EPA (2008) has set drinking water regulations on bromoform consumption to 80 mg/L. Another longer term study has been completed at UC Davis but results were not fully available as of the time of writing the report. Although there is no question regarding efficacy of *Asparagopsis*, some issues such as supply, cost and FDA approval remain to be solved, therefore, more research needs to be conducted to get it to market and being recommended for use in California.

Feed Additives with highest potential for mitigation

We found two feed additives that have been extensively studied (over 10 trials each). These are 3-nitrooxypropanol (3NOP), and nitrate. The rest of the section on feed additives will focus on these two. We updated a meta-analysis conducted for 3NOP and built a new meta-analysis for use of nitrate in beef and dairy cattle. We then proceeded to use a life-cycle assessment developed for California conditions (Naranjo et al. 2020) and assessed the ***net*** reduction expected if either 3NOP or nitrate were to be used. The life-cycle assessment was from cradle to farm gate so included emissions from feed production, the barn (i.e., animals, manure, electricity) and farm operations. There is a challenge of supplementing with nitrate currently due to toxicity risk. However, research is being conducted to add microbes that can help in detoxification (Latham et al., 2019) and a full analysis of nitrate use is provided below.

3-Nitrooxypropanol (3NOP)

A meta-analysis was conducted by Dijkstra et al. (2018) on effects of 3NOP. However, 4 additional papers were published that were not included in the meta-analysis. Therefore, we updated the previous meta-analysis by adding data from Martinez-Fernandez et al. (2018) (beef; 1 study), Vyas et al. (2018b) (beef; 2 studies), Kim et al. (2019) (beef; 4 studies), and Van et al. (2019) (dairy; 2 studies). The updated forest plots for Standardized Mean Difference of CH_4 production and yield are shown in Figures 3 and 4.

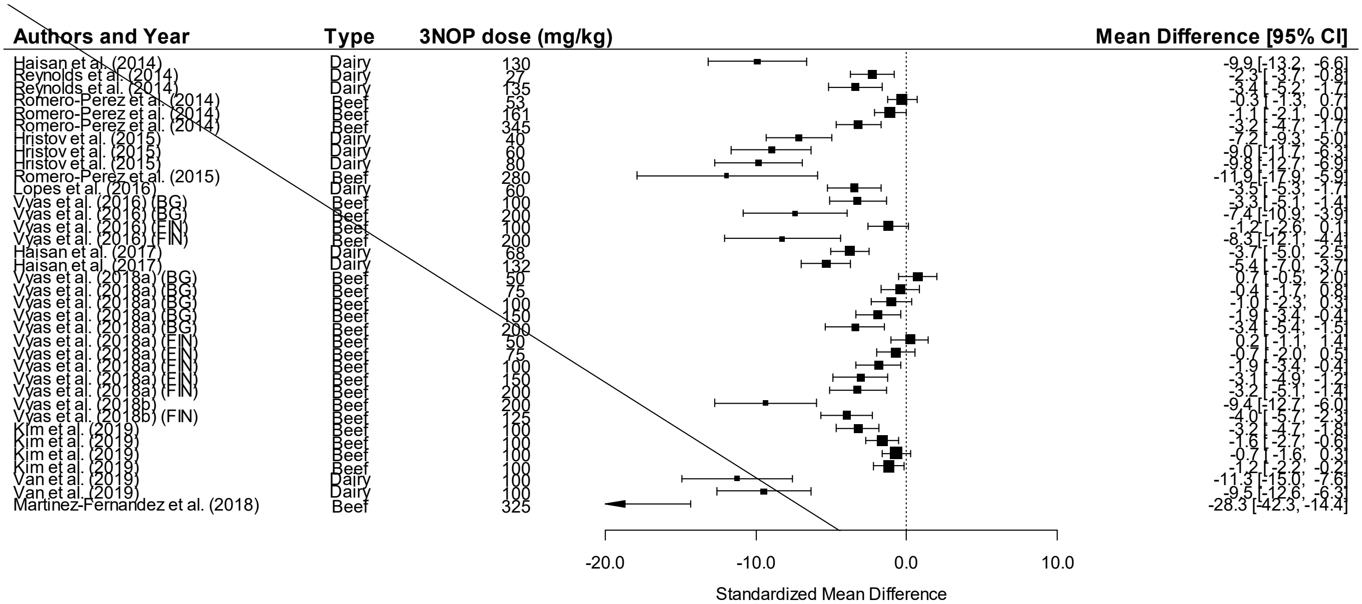


Figure 3. Forest plot showing 3-nitrooxypropanol (3NOP) dose (mg/kg of DM) and standardized mean difference (mean difference is calculated as NOP treatment mean − control treatment mean) in CH_4 production (g/d) for beef and dairy cattle studies.

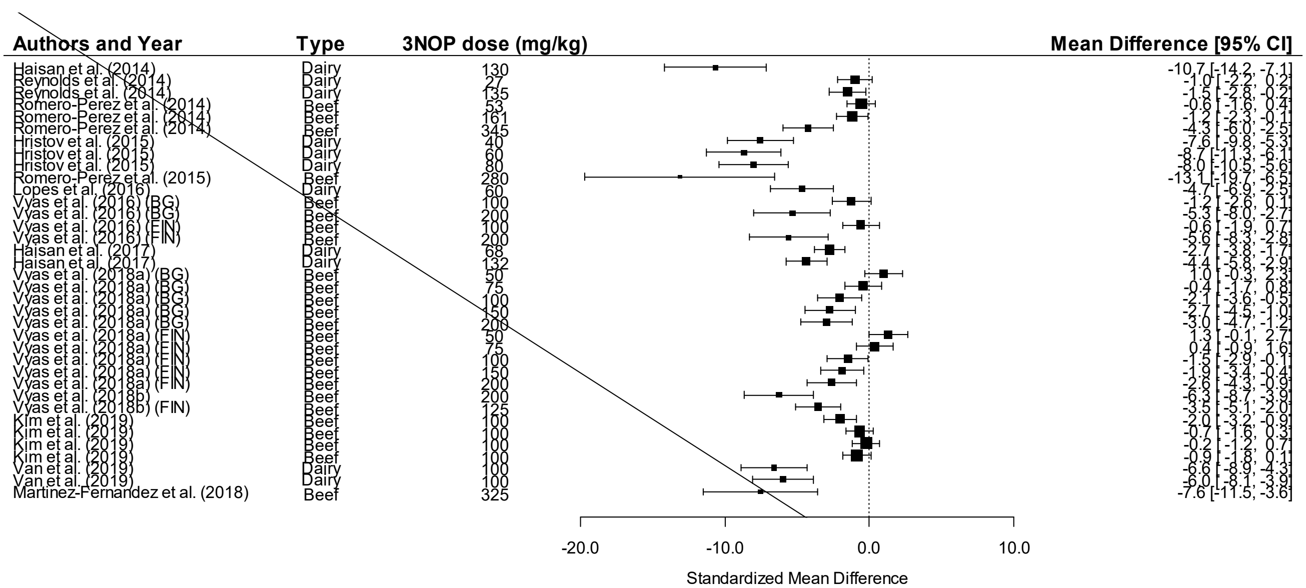


Figure 4. Forest plot showing 3-nitrooxypropanol (3NOP) dose (mg/kg of DM) and standardized mean difference (mean difference is calculated as NOP treatment mean − control treatment mean) in CH_4 yield (g/kg of DMI) for beef and dairy cattle studies.

The data was checked if it fits a normal distribution function and for outliers. A quantile-quantile plot (Q-Q plot), showed that the data was normally distributed (Figure 5), therefore, no outliers were removed before conducting the meta-analysis.

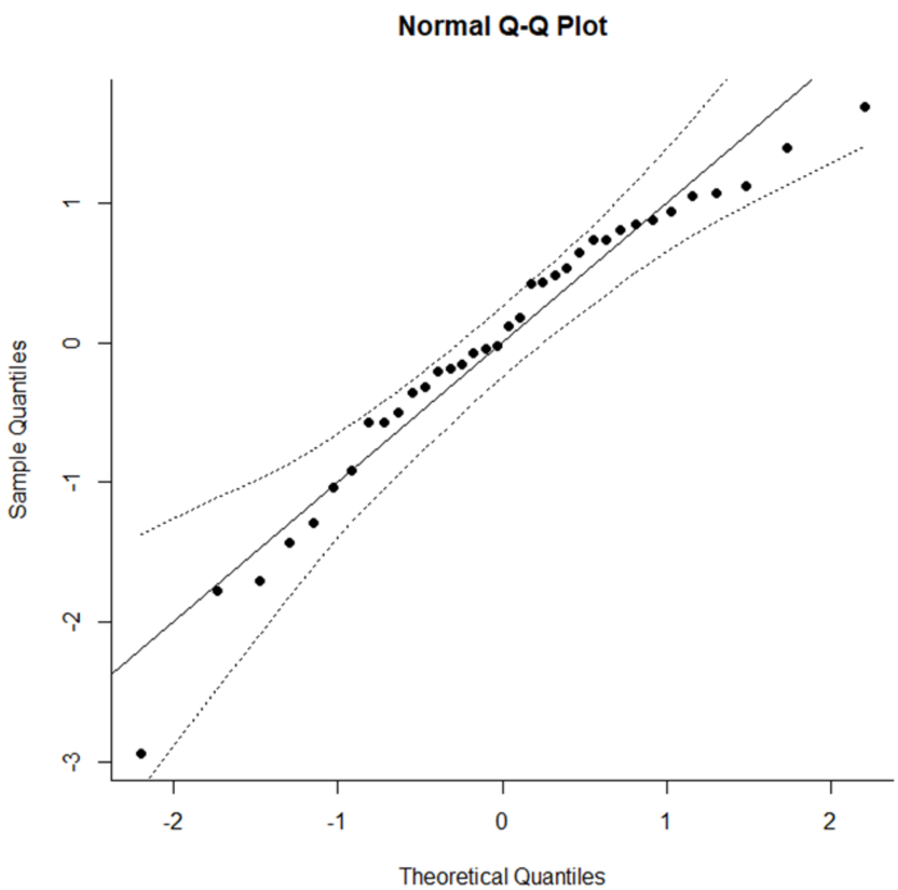


Figure 5. A quantile-quantile plot (Q-Q plot) of the data.

The results of the mixed-effect models for CH_4 production and yield was similar to the previous study which indicated effectiveness of 3NOP at mitigating CH_4 emissions (Table 2). As expected, the effect was positively associated with dose, and negatively associated with dietary fiber content. Moreover, NOP had stronger anti-methanogenic effects in dairy cattle than in beef cattle. The mean value of NOP dose was 127 mg/kg of DM which slightly increased comparing to the 123 mg/kg of DM in previous analysis. The overall mitigating effect of 3NOP was 32% at 127 mg/kg inclusion level. In dairy cattle specifically the impact was 41% reduction while in beef cattle it was 22.4% (Table 2).

Table 2. Estimates of overall 3-nitrooxypropanol (3NOP) effect size and of explanatory variables from random- and mixed-effect models for relative mean difference (MD, %) in CH_4 production (g/d) and yield (g/kg of DMI)

Variable and model	CH4 production				CH4 yield			
	Mean	SE	P-value	τ2	Mean	SE	P-value	τ2
Random-effect model								
Overall NOP effect size	-32.0	4.46	<0.001	210	-29.6	4.58	<0.001	397
Mixed-effect model, 1 explanatory variable								
Overall NOP effect size	-30.4	4.16	<0.001	331	-27.8	4.19	<0.001	305
NOP dose (mg/kg of DM)	-0.114	0.0563	0.0996		-0.128	0.0464	0.0401	
Final mixed-effect model-I								
Dairy cattle	-41.5	4.82	<0.001	128	-39.8	5.17	<0.001	162
Beef cattle	-22.8	3.68	<0.001		-19.3	3.78	0.0020	
NOP dose (mg/kg of DM)	-0.260	0.0538	0.0031		-0.265	0.0618	0.0054	
NDF content (g/kg of DM)	0.129	0.0282	0.0040		0.109	0.0310	0.0131	
If removed the 2 studies in Kim et al. (2019), final model selection did not change								
Random-effect model								
Overall NOP effect size	-32.0	4.45	<0.001	210	-29.6	4.58	<0.001	397
Mixed-effect model, 1 explanatory variable								
Overall NOP effect size	-30.6	4.17	<0.001	331	-28.0	4.19	<0.001	305
NOP dose (mg/kg of DM)	-0.114	0.0563	0.0998		-0.128	0.0464	0.0401	
Final mixed-effect model-I								
Dairy cattle	-41.0	4.83	<0.001	129	-39.5	5.20	<0.001	163
Beef cattle	-22.4	3.53	<0.001		-19.1	3.66	0.0018	
NOP dose (mg/kg of DM)	-0.258	0.0534	0.0031		-0.262	0.0613	0.0055	
NDF content (g/kg of DM)	0.127	0.0294	0.0053		0.106	0.0324	0.0178	

Nitrates

Nitrate (NO_3^-) is a strong inorganic anion and acts as an alternative hydrogen sink in rumen to potentially compete with methanogens for hydrogen utilization. Dietary nitrate is firstly reduced to nitrite (NO_2^-; $NO_3^- + H_2 \rightarrow NO_2^- + H_2O$) and then to ammonia ($NH_4^+$; $NO_2^- + 3H_2 + 2H^+ \rightarrow NH_4^+ + 2H_2O$) which is energetically more favorable than the reduction of CO_2 to CH_4 ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$) due to a higher Gibbs energy change (Villar et al., 2020). Thus, nitrate reduction is highly competitive compared with methanogenesis that leads a redirection of H flow away from CO_2 reduction, and thereby reduces enteric CH_4 production (Olijhoek et al., 2016).

Several *in vivo* studies have investigated the effects of nitrate as a CH_4 mitigation strategy in different types of ruminants such as beef steers (Hulshof et al., 2012; Troy et al., 2015; Alemu et al., 2019), dairy cows (Veneman et al., 2015; Klop et al., 2016; Meller et al., 2019) , sheep (Sar et al., 2004; van Zijderveld et al., 2010,), and goats (Zhang et al., 2019). However, the results of the trials on effectiveness of nitrate mitigation on CH_4 emissions for ruminants have been inconsistent with large variability. The studies were conducted under various dietary regimen and nitrate doses so some of the differences may be explained by dietary or other variables. For example, Guyader et al. (2015a) reported that CH_4 yield in nitrate treatment group was reduced 22% in nonlactating Holstein cows and Lee et al. (2015) showed that CH_4-mitigating effect of nitrate for beef heifers were associated with nitrate dose, and the reduction rates varied from 3.3 to 20.8%. However, van Wyngaard et al. (2019) did not find a significant effect on mitigating CH_4 emissions when dietary nitrate was fed to dairy cows grazing perennial ryegrass.

The objective of this study was to collate data on nitrate supplementation for CH_4 mitigation and quantitatively evaluate the effects of dietary nitrate for enteric CH_4 production and yield. Nitrate dose, nutrient composition of diet, dry matter intake, and cattle type may potentially explain a large proportion of the between-study variability in CH_4 mitigation effect of nitrate (Lee and Beauchemin, 2014; Dijkstra et al., 2018). Therefore, this study quantitatively analyzes explanatory variables to account for the heterogeneity observed in emission reduction due to nitrate in diet using a meta-analysis approach.

Materials and methods

Literature search was conducted using several sources including the Web of Science (Thomson Reuters Science, New York, NY), Elsevier (Elsevier, Amsterdam, the Netherlands) and Google Scholar online databases with all possible combinations of the keywords "feed additives", "nitrate", "methane" (including all variants of "CH_4" and "greenhouse gas"), "cattle" (including all variants of "dairy", "beef", "steer", "cows" and "ruminants"). The period of the study covered from 1970 to 2019. The search resulted in 45 references related to the effects of nitrate on enteric CH_4 production in cattle. All the references were scrutinized by reading the abstracts, experimental design, and results of each reference carefully. To be included in the database, the studies were required to meet the following criteria: (i) a control group treatment group that did not receive nitrate; (ii) to be conducted *in vivo* using cattle; (iii) reported CH_4 production with standard deviation, standard error or other relative data that can be used to calculate the standard error (e.g. least significant difference); (iv) described other required variables (e.g. nutritional composition) or provided enough information to estimate the variables. Of the 45 references, two were general summary papers and three articles had only abstracts available so these were excluded from the dataset. Three papers were removed because they investigated the mitigation effect on CH_4 of a mixture of nitrate and other feed additives. Five papers did not report CH_4 emissions and another five did not provide diets or dietary information useful in calculating them, therefore were not included in the database. Data from 27 articles met the criteria, however, another three articles were rejected because data were duplicates of references already included in the database. The remaining 24 articles containing 57 treatment means were selected for the final database. Of those 36 treatments were related to beef cattle (Hulshof et al., 2012; Newbold et al., 2014; Lee et al., 2015; Troy et al., 2015; Lee et al., 2017a, b; Capelari, 2018; Duthie et al., 2018; Henry et al., 2018; Tomkins et al., 2018; Alemu et al., 2019; Granja-Salcedo et al., 2019; Rebelo et al., 2019) and 21 treatments to dairy cattle (van Zijderveld et al., 2011; Guyader et al., 2015a, b; Veneman et al., 2015; Klop et al., 2016; van Wyngaard et al., 2018; Meller et al., 2019; van Wyngaard et al., 2019).

The primary response variables included the means of CH_4 production and yield in control and nitrate treatment groups. Factors having a potential to explain the variability in nitrate effect on CH_4 emissions were selected and considered in the meta-analysis. Methane production was generally reported in grams per day and CH_4 yield in grams per kilogram of DMI. If the values were reported in liters or moles per day, they were converted to grams per day assuming a volume of 22.4 L and molar weight of 16.0 g. If only one of the CH_4 production and CH_4 yield was given, the other variable was calculated as CH_4 yield = CH_4 production/DMI.

General meta-regression methods require the independency of effect sizes (i.e., the quantitative measure of the difference in magnitude in methane emission between control and treatment). However, multiple nitrate treatment groups may share a same control treatment group in some of the studies used in our database. To deal with the unknown correlations among these non-independent effect sizes, a robust variance estimation (RVE) method (Tipton, 2015) was used to conduct the meta-analysis. Studies selected in the meta-analysis were not identical in the methods and sample characteristics which may introduce variance of the true effect sizes, therefore, RVE random-effects and RVE mixed-effects models were fitted to estimate between-study variability (heterogeneity) that was assumed to be purely random (Tanner-Smith et al., 2016; Dijkstra et al., 2018). The RVE random-effects model was written as

$$y_{ij} = \beta_0 + \mu_j + e_{ij},$$

where for i =1,…, k_j, j = 1,…, m, y_{ij} is the ith effect size of jth study, β_0 is the average true effect, μ_j is the random effect at study level where $\mu_j \sim$ N (0, τ^2) and τ^2 is the between-study variance component, and e_{ij} is the residual for ith effect size in the jth study where $e_{ij} \sim$ N (0, s_i^2) and s_i^2 is the error variance component. The heterogeneity (I^2) is defined as the ratio of between-study variance (τ^2) to the total variability ($s_i^2 + \tau^2$) and an I^2 value greater than 0.5 indicates significant heterogeneity (Dijkstra et al., 2018). To examine effect size moderators and reduce heterogeneity, the RVE random-effects models can be extended to RVE mixed-effects models which include variables with the potential to account for some of the observed variability. The RVE mixed-effects model was written as

$$y_{ij} = \beta_0 + \mu_j + \mathbf{X}_{ij}\boldsymbol{\beta} + e_{ij},$$

where β_0, μ_j, and e_{ij} are as defined above, $\boldsymbol{\beta}$ = (β_1,… β_p) is a vector of unknown regression coefficients based on weighted least-squares estimates, and $\mathbf{X}ij$ is a vector of continuous or binary explanatory variables. The inverse variance weights of "correlated effects" used in RVE models were estimated following a method provided by Hedges et al. (2010):

$$\mathrm{w}_{ij} = \frac{1}{k_j(\upsilon_{.j} + \tau^2)}$$

where w_{ij} is the ith inverse variance weight in jth study, k_j is the number of effect sizes for each study j, $\upsilon_{.j}$ is the mean of within-study sampling variances (υ_{ij}) for the k_j effect sizes in jth study, and τ^2 is the between-study variance component as defined previously which describes the residual of heterogeneity that is not explained by the involved variables.

The dry matter intake (DMI), body weight (BW), roughage proportion in the diet, dietary crude protein (CP), and neutral detergent fiber (NDF), and nitrate dose were selected as potential continuous explanatory variables. Types of cattle (dairy or beef) were used as category variables. Therefore, the vector $\boldsymbol{\beta}$ can be explained as the differences in true effect sizes according to each unit changing in the continuous variables or between the two cattle types. The RVE model was first fitted with each individual variable, and two or more variables were included following a stepwise method until all explanatory variables were involved to conduct full mixed-effect models (Dijkstra et al., 2018). Only the variables showing significant effects ($P < 0.10$) were retained until the final model was selected. Multi-collinearity was investigated to examine the correlations among variables and highly correlated variables ($|r| > 0.50$) were not analyzed in the same model such as DMI and CP ($|r| = 0.59$), and CP and NDF ($|r| = 0.51$). All explanatory variables (except for cattle types) were first centered on their means. Potential variables such as gross energy (GE) content, ash content, fat content, and organic matter (OM) digestibility were also considered in data collection, however, due to the lack of information in most of the publications, they were not included in this analysis.

To prepare for the meta-analysis, effect size estimates of mean difference (MD) and standardized mean difference (SMD) were used to measure the continuous response variables of CH_4 production and yield. The MD was calculated as nitrate treatment mean minus control treatment mean and each study was weighted by its corresponding sample variation (Viechtbauer, 2010). The SMD was expressed as dividing MD by the pooled standard deviation of the two group (SMD = MD/pooled standard deviation of the 2 groups) and used to construct forest plots of response variables. The relative mean difference (RMD; RMD = MD/control treatment mean × 100%), which was a dimensionless variable, was calculated for further analyses to eliminate the large variations and different measuring scales of DMI and CH_4 production from study to study.

All statistical analyses were carried out using various packages in R (version 3.6.1, R Foundation for Statistical Computing, Vienna, Austria). The "cor" function in R (version 3.6.1) was used to test the correlation between explanatory variables. The "escalc" and "robu" functions provided by "metafor" (version 2.1-0) and "robumeta" (version 2.1) packages in R were used to calculate effect sizes (MD and SMD) and conduct RVE models, respectively.

Results and Discussion

Meta-analysis is a statistical methodology that combines quantitative findings from various studies for the main purpose of synthesizing the evidence based on the available sources (Schwarzer et al., 2015). The meta-analysis conducted in this paper aimed to evaluate the effects of nitrate as a feed additive to reduce CH_4 production and yield in dairy and beef cattle. A summary statistic of feed intake, nutrient compositions of the experimental diet, nitrate supplement, and CH_4 production is given in Table 3. The daily DMI and CH_4 production of dairy cows (16.2 ± 2.86 kg/d; 286 ± 52.1 kg/d, respectively) were greater than beef steers (9.5 ± 4.20 kg/d; 137 ± 47.2 kg/d), while the averages of supplemented nitrate dose were not significantly different between dairy (18 g/kg of DM) and beef cattle (17 g/kg of DM). On average, the effects of nitrate resulted in greater RMD in CH_4 production and yield for dairy cows (-16.7 ± 7.64%; -15.4 ± 7.66%) than those for beef steers (-12.3 ± 10.22%; 9.0 ± 11.15%). Forest plots generated with SMD for CH_4 production (Figure 6) and CH_4 yield (Figure 7) showed consistent anti-methanogenic effects in most of the studies included in this analysis. However, effect sizes were variable across studies.

At an average nitrate dose of 18 g/kg of DM, the overall CH_4 production (P < 0.001) and CH_4 yield (P < 0.001) were reduced by 14.4 ± 1.21% in dairy and 11.4 ± 1.40%, in beef cattle according to the random-effect RVE models (Table 4). Several other feed additives have also shown to reduce methane emissions but mostly at a lower effectiveness. For example, Appuhamy et al. (2013) reported monensin reduced CH_4 production by 5.6% for dairy cows and 4.6% for beef steers. Eugène et al. (2008) investigated lipid supplementation and reported it reduced CH_4 production by 9.0% in lactating dairy cows. Van Zijderveld et al. (2011) observed a 10% decrease in CH_4 emissions by supplementing mixed additives of lauric acid, myristic acid, and linseed oil in dairy cattle. However, 3NOP showed stronger antimethanogenic effects with 39% and 22% reduction level of CH_4 production in dairy and beef cattle, respectively (Dijkstra et al., 2018). Similarly, Roque et al. (2019a) reported Mootral reduced CH_4 production 23% after 12 weeks of supplementation in beef cattle.

Table 3. Summary statistics of dietary composition, feed intake, animal characteristic, and methane emission of the database.

Item	Dairy					Beef				
	Mean	Median	SD[3]	Min	Max	Mean	Median	SD[3]	Min	Max
DMI (kg/d)	16.2	17.6	2.86	10.2	19.7	9.5	8.3	4.20	6.1	22.9
Roughage proportion (% of diet DM)	61	60	10.6	50	78	62	65	21.6	10	100
NDF (g/kg of DM)	356	352	67.2	100	426	372	362	120.3	227	680
CP (g/kg of DM)	149	156	21.3	88	175	129	134	22.3	49	150
BW (kg)	466	533	187.7	117	658	430	337	147.2	283	698
Nitrate dose (g/kg DM)	18	21	4.7	5	23	17	19	5.4	5	27
CH_4 production (g/d)	286	300	52.1	175	405	137	140	47.2	71	243
MD[1] of CH_4 production (g/d)	-57	-59	26.9	-100	5	-19	-18	15.1	-43	31
RMD[2] of CH_4 production (% of control)	-16.7	-17.0	7.64	-29.8	1.3	-12.3	-11.4	10.22	-32.0	22.0
CH_4 yield (g/kg of DMI)	17.9	17.4	2.17	14.5	24.3	17.2	17.9	4.87	8.8	27.6
MD of CH_4 yield (g/kg of DMI)	-3.3	-3.3	1.86	-6.8	1.1	-1.7	-1.7	1.97	-5.7	3.3
RMD of CH_4 yield (% of control)	-15.4	-14.9	7.66	-27.6	4.7	-9.0	-9.5	11.15	-29.4	19.3

[1]MD (Mean difference) = treatment mean - control mean.

[2]RMD (Relative mean difference) = (MD/control mean) × 100%.

[3]SD = standard deviation of mean

Table 4. Estimates of overall nitrate effect from random-effect model, and of explanatory variables from mix-effect models for relative mean difference (RMD) in CH_4 production (g/d) and yield (g/kg of DMI).

Variable[1] and model	CH_4 production				CH_4 yield			
	Mean	SE	*P*-value	τ^2	Mean	SE	*P*-value	τ^2
Random-effect model								
Overall effect size	-14.4	1.21	<0.001	53.0	-11.4	1.40	<0.001	53.6
Mixed-effect model, 1 explanatory variable[2]								
Model I: Overall effect size	-14.2	1.05	<0.001	29.4	-11.5	1.30	<0.001	46.2
Nitrate dose (g/kg of DM)	-0.932	0.195	<0.001		-0.776	0.235	0.004	
Model II: Dairy cattle					-15.4	1.71	<0.001	52.6
Beef cattle					-9.03	1.90	<0.001	
Mixed-effect model, 2 explanatory variables[3]								
Model I: Dairy cattle	-14.1[4]	1.79	<0.001	29.0	-14.9[4]	1.36	<0.001	47.7
Beef cattle	-14.2	1.32	<0.001		-9.40	1.88	<0.001	
Nitrate dose (g/kg of DM)	-0.933	0.203	<0.001		-0.720	0.246	0.009	
Model II: Dairy cattle					-15.7[4]	1.50	<0.001	54.5
Beef cattle					-9.16	1.86	<0.001	
NDF content (g/kg of DM)					-0.0321	0.0164	0.083	
Model III: Nitrate dose (g/kg of DM)					-0.936	0.429	0.040	199
NDF content (g/kg of DM)					-0.0366	0.0161	0.042	
Final mixed-effect model								
Model I: Dairy cattle	-14.3[4]	1.49	<0.001	27.2	-15.2[4]	1.05	<0.001	30.7
Beef cattle	-14.0	1.35	<0.001		-9.82	1.66	<0.001	
Nitrate dose (g/kg of DM)	-1.01	0.232	<0.001		-0.967	0.229	<0.001	
NDF content (g/kg of DM)	-0.0214	0.0135	0.144		-0.0471	0.0129	0.004	

[1]The explanatory variables centered on their means (except cattle type variable): BW = 443 kg; CP content = 137 g/kg of DM; NDF content = 366 g/kg of DM; roughage proportion = 61%; DMI = 12.0 kg/d; nitrate dose = 18 g/kg of DM.
[2] Mixed-effect models with 1 explanatory variable had no significant effect on CH_4 production or CH_4 yield were not listed. Variables included: BW ($P = 0.905$), NDF ($P = 0.500$), CP ($P = 0.407$), roughage proportion ($P = 0.802$), DMI ($P = 0.994$), and cattle type ($P = 0.432$) for CH_4 production; BW ($P = 0.765$), NDF ($P = 0.112$), CP ($P = 0.537$), roughage proportion ($P = 0.342$), and DMI ($P = 0.417$) for CH_4 yield.
[3] Mixed-effect models with 2 and more explanatory variables that had no significant effect on CH_4 production or CH_4 yield were not retained.
[4] Cattle type effects for CH_4 production were not significant ($P > 0.50$); for CH_4 yield were significant ($P < 0.05$).

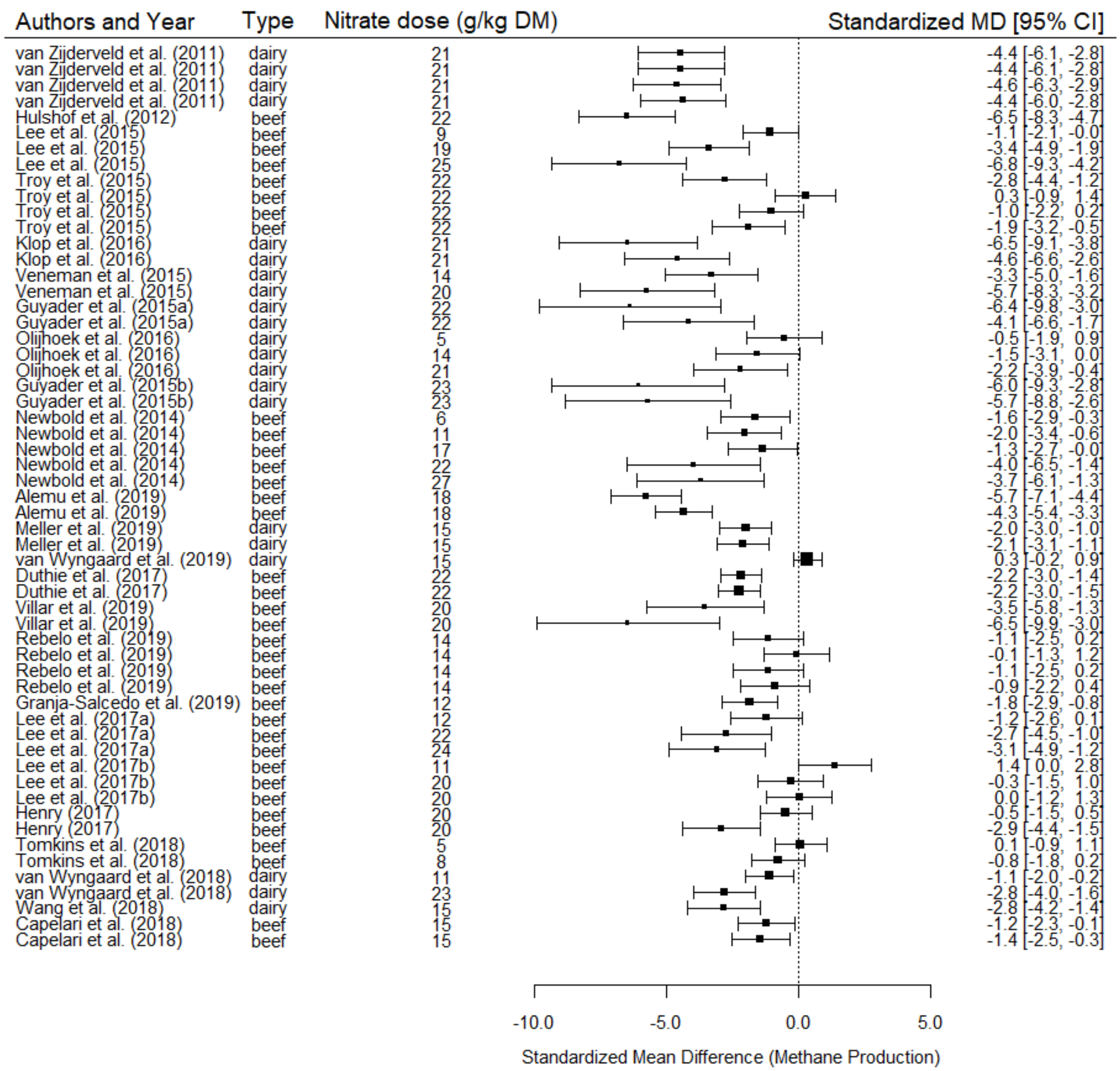


Figure 6. Forest plot showing nitrate dose (g/kg of DM) and standardized mean difference in CH_4 production (g/d) and its 95% confidence interval (CI) for beef and dairy cattle from selected studies. The dotted line represents a reference of 0 standardized mean difference. The black squares represent the power of its corresponding studies (Note: A larger box indicates a greater sample size and a smaller CI).

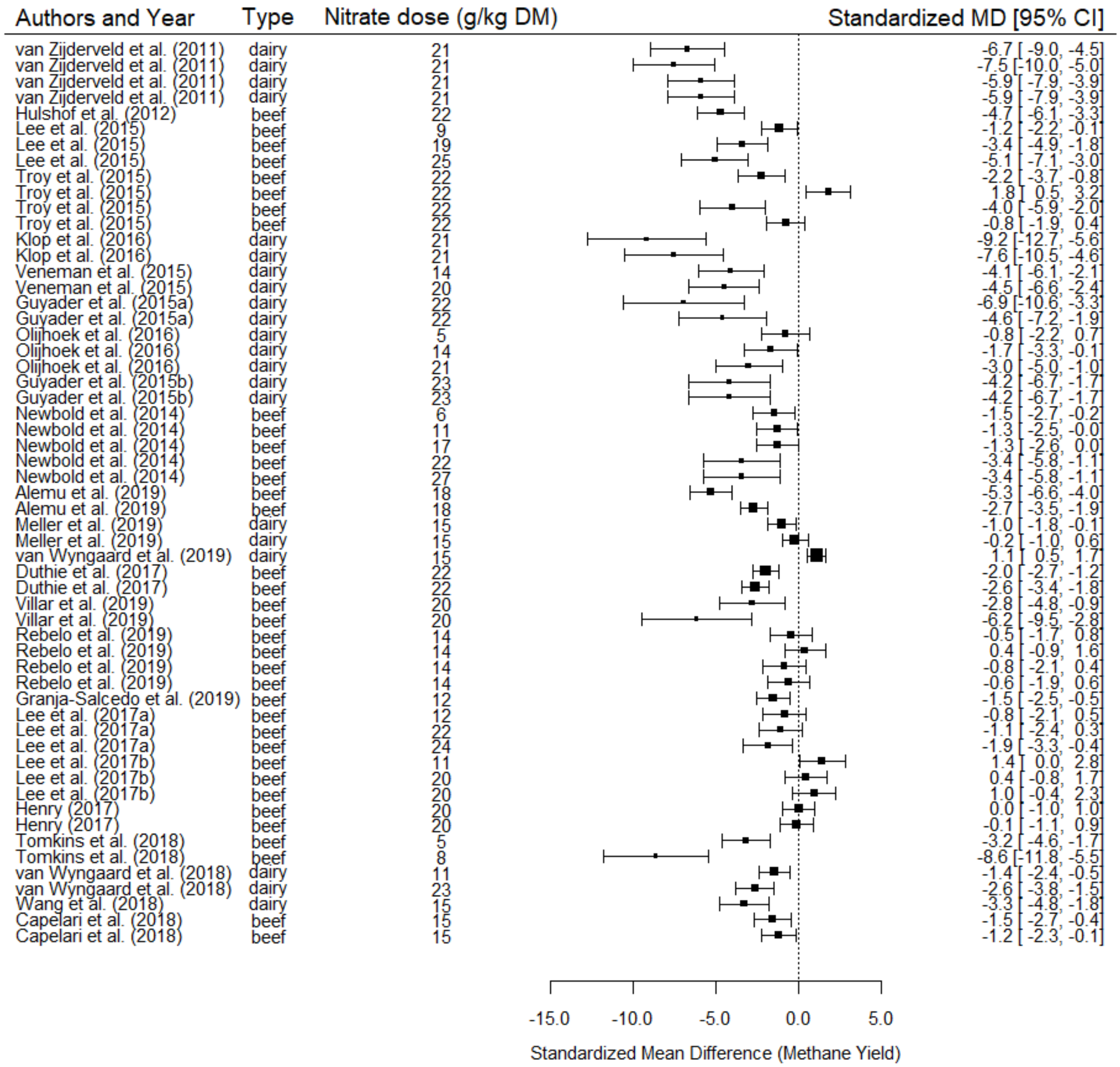


Figure 7. Forest plot showing nitrate dose (g/kg of DM) and standardized mean difference in CH_4 yield (g/kg of DMI) and its 95% confidence interval (CI) for beef and dairy cattle from selected studies. The dotted line represents a reference of 0 standardized mean difference. The black squares represent the power of its corresponding studies (Note: A larger box indicates a greater sample size and a smaller CI).

The RVE random-effect models showed that a large proportion of the total variability of nitrate effects on CH_4 production (I^2 = 69.9%) and CH_4 yield (I^2 = 99.7%) were attributed to heterogeneity. Potential explanatory variables were individually included to conduct mixed-effect RVE models to further understanding and improve the random-effect models (Table 4). The size of CH_4 production reduction was positively associated with nitrate dose ($P < 0.001$). A 10 g/kg of DM increase in nitrate dose from its mean (18 g/kg of DM), enhanced the nitrate anti-methanogenic effect of CH_4 production by 9.32 ± 1.95%. However, for RMD in CH_4 production, the categorical variable cattle type ($P = 0.432$), and continuous variables BW ($P = 0.905$), NDF content ($P = 0.500$), CP content ($P = 0.407$), roughage proportion of diet ($P = 0.802$), and DMI ($P = 0.994$) were not significant. For RMD in CH_4 yield, BW ($P = 0.765$), NDF content ($P = 0.112$), CP content ($P = 0.537$), roughage proportion of diet ($P = 0.342$), and DMI ($P = 0.417$) were not significant. But, the categorical variable cattle type ($P = 0.017$) and nitrate dose ($P = 0.004$) were significant. A 10 g/kg of DM increase in nitrate dose from its mean (18 g/kg of DM) resulted in 7.76 ± 2.35% decline in CH_4 yield (Table 4, Model I). A 10 g/kg of DM increasing in nitrate dose from its mean (18 g/kg of DM) resulted in 7.76 ± 2.35% decline of CH_4 yield (Model I). The results agree with Lee and Beauchemin (2014) in which they reported a linear reduction in CH_4 yield with increasing levels of nitrate dose. Nitrate mitigation effect on CH_4 yield in dairy and beef cattle were -15.4 ± 1.71% and -9.03 ± 1.90%, respectively, that were significantly different from each other according to Model II (Table 4). This indicates that nitrate shows a stronger impact on mitigating CH_4 yield for dairy cattle, and a higher nitrate dose is required for beef cattle to obtain the same effectiveness at reducing CH_4 yield compared that for dairy cattle. The heterogeneity was reduced by including the individual explanatory variable for both CH_4 production (τ^2 = 53.0 vs. 29.4) and CH_4 yield (τ^2 = 53.6 vs. 46.2 for Model I or 52.6 for Model II).

Adjusting the RVE mixed-effect model to use two explanatory variables, cattle type ($P <$ 0.001) and nitrate dose ($P < 0.001$; $P = 0.009$) were significantly associated with nitrate effect on CH_4 production and yield (Model I). A 10 g/kg of DM increase in nitrate dose enhanced the nitrate effect on CH_4 production by 9.33 ± 2.03% from the average of 14.1 ± 1.79% for dairy cows, and 14.2 ± 1.32% for beef steers. Similar increase in nitrate dose enhanced the nitrate effect on CH_4 yield by 7.20 ± 2.46% from the average of 14.9 ± 1.36% for dairy cows, and 9.40 ± 1.88% for beef steers. The mixed-effect model conducted with cattle type and nitrate dose slightly reduced the heterogeneity for CH_4 production (τ^2 = 29.4 vs. 29.0), however, the heterogeneity for CH_4 yield was not improved by the model (τ^2 = 46.2 vs. 47.7). When the model was adjusted for NDF content instead of nitrate dose (Model II), CH_4 yield tended to decline ($P = 0.083$) by 0.321 ± 0.164% for every 10 g/kg of DM increase in NDF content from the average in dairy (-15.7 ± 1.50%) and beef (-9.16 ± 1.86%) cattle. Although, nitrate dose ($P = 0.040$) and NDF content ($P = 0.041$) were significantly related to nitrate effect on CH_4 yield in Model III, the heterogeneity jumped from 47.7 (Model I) or 54.5 (Model II) to 199 (Model III) indicating the importance of including cattle type in the model.

The final mixed-effect models for RMD in CH_4 emissions (Table 4) included cattle type, nitrate dose and dietary NDF content. The τ^2 decreased from the random-effect model to a mixed-effect model with 1 and 2 explanatory variables, and further decreased to the final mixed-effect model with 3 explanatory variables (CH_4 production: τ^2 = 27.2 vs. 53.0; CH_4 yield: τ^2 = 30.7 vs. 53.6) but not with 2 explanatory variables for CH_4 yield (Table 4). When adjusted for the effects of nitrate dose and dietary NDF content, the anti-methanogenic effect of nitrate was similar in beef cattle (-14.0 ± 1.35%; P <0.001) compared to dairy cattle (−14.3 ± 1.49%; P <0.001) for CH_4 production. However, for CH_4 yield, with nitrate dose centered on its mean (18 g/kg of DM), and the mean NDF content of 366 g/kg of DM, the anti-methanogenic effect of nitrate was stronger in dairy cows (-15.2 ± 1.50%; P <0.001) compared to beef cattle (-9.82 ± 1.66%; P <0.001). The greater efficacy in dairy cattle may be related to the differences in the levels of feed intake (dairy: 16.2 kg/d, beef: 9.5 kg/d; Table 3. A similar difference in cattle type on efficacy of 3NOP was reported (Djikstra et al., 2018). The authors suggested that higher feed intake levels increase rumen concentrations of fermentation products, including volatile fatty acids and hydrogen and sinks of hydrogen in the rumen may be affected by hydrogen partial pressure. This will likely result in greater alternative hydrogen sinks for rumen methanogenesis. The efficacy of nitrate-N utilization may be improved, and the potential of nitrate inhibitory effect is enhanced through more completed nitrate reductions. After adjusting for cattle type and dietary NDF content in final mixed-effect models, the nitrate-induced CH_4 mitigation was 10.1 ± 2.32% (CH_4 production, P < 0.001) and 9.67 ± 2.29% (CH_4 yield, P < 0.001) per 10 g/kg of DM increase in nitrate dose from its mean (18 g/kg of DM; Table 4), which is slightly higher than the effect of nitrate dose observed in the individual and two explanatory variables mixed-effect models. In our analysis, an increase in dietary NDF content did not significantly affect the efficacy of nitrate in reducing CH_4 production (P = 0.144) but slightly increased (P = 0.004) the nitrate effect on CH_4 yield (Table 4). A 10 g/kg of DM increase in dietary NDF content from its mean (366 g/kg of DM) increased the nitrate effect on CH_4 yield by only 0.471 ± 0.129%.

ADDITIVES TARGETING MANURE METHANE EMISSIONS

Direct emissions of CH_4 and N_2O from livestock manure vary by manure treatment and storage methods. Both CH_4 and N_2O emissions can be mitigated either by reducing them during manure storage or maximizing CH_4 production and capturing the gas to produce biogas energy (USEPA, 2017). The greenhouse gas and odor emitted from manure and slurry could be directly or indirectly reduced through different technologies such as solids separation (Martinez et al., 2003; Owusu-Twum et al., 2017), dietary management strategies (Hristov et al., 2013; Lund et al., 2014; Troy et al., 2015), anaerobic digestion (Clemens et al., 2006), manure coverage (Misselbrook et al., 2016), and use of manure additives (Chen et al., 2018; Mao et al., 2018; Owusu-Twum et al., 2017; Wheeler et al., 2010; Yamulki, 2006).

Manure additives or amendments can be defined as substances that can be used to alleviate gaseous emissions associated with livestock manure handling and management. The application of manure additives is regarded as a practical and economical treatment method compared to alternative technology such as solids separation and biogas production (McCrory and Hobbs, 2001). Various types of additives have been applied on-farm and are reported in the literature over the last few decades, however, the effectiveness and performance for mitigating gas emissions of specific additives are not consistent, especially for the effects on CH_4 emissions. For example, Liu et al. (2017) and Vandecasteele et al. (2016) investigated the use of biochar and reported that it enhanced the organic matter degradation and reduced CH_4 emissions, however, Sanchez-Garcia et al. (2015) reported that there was no significant evidence showing the relevant impact of biochar on CH_4 emissions. A meta-analysis synthesizes the evidence from many available sources and combines and compares the treatment effects of individual studies by statistical methods (Dijkstra et al., 2018). The objective of this review was to investigate and quantitatively evaluate the effects of different types of manure additives on mitigating CH_4 emissions in livestock based on published literature data.

Data collection and selection

The main purposes for adding manure additives include directly reducing gas emission during storage and composting, and enhancing the gas emission to generate biogas. Literature searches of the Web of Science (Thomson Reuters Science, New York, NY) and Google Scholar online databases were conducted using the combination of search terms “manure additives”, “methane” or “CH_4”, “greenhouse gas”, “reduce” or “reduction”, “mitigate” or “mitigation”, “amend” or “amendment”. The covered period was from 2000 to 2019. A total of 42 papers were collected after the initial searching. All the references were carefully scrutinized by reading the abstracts, experimental design, and results. For inclusion in the database, the studies were required to include: (i) a control group, (ii) the CH_4 emissions reported with mean, standard deviation or standard error, and sample size and (iii) at least one type of additive was added directly into the manure for CH_4 emission reduction purposes. Studies related to increasing biogas generation by adding manure additives were not included because the objective of the manure additives was not to reduce emissions. There were 27 references remaining in the database after filtering by the criteria mentioned above. The manure additives were firstly categorized into general groups based on their function which were “acidification”, “adsorbent”, “biochar”, “biological material”, “C/N content”, “disinfection”, “essential oils”, “humate”, “microbial digestion”, “oxidizing agent”, “physical material”, “straw”, and for those that did not fit the above categories were put into “other chemicals” (Agyarko-Mintah et al., 2017; Berg et al., 2006; Chen et al., 2018; Chen et al., 2017; Chowdhury et al., 2014a; Chowdhury et al., 2014b; Hao et al., 2005; He et al., 2019; Jia et al., 2016; Liu et al., 2017; Luo et al., 2013; Mao et al., 2018; Martinez et al., 2003; Misselbrook et al., 2016; Owusu-Twum et al., 2017; Petersen et al., 2012; Regueiro et al., 2016; Samer et al., 2014; Shah & Kolar, 2012; Sommer & Moller, 2000; Sonoki et al., 2011; Vandecasteele et al., 2016; Wang et al., 2014; Wang et al., 2018; Wheeler et al., 2010; Yamulki, 2006; Zhang et al., 2017). A full list of studies investigated is given in Appendix 2. Statistical analysis and meta-analysis were both conducted based on the first level of classification due to the insufficient database for each type of second classified level. The manure came from various species of animals and were not sorted by species to obtain enough sample sizes for different manure additives.

Statistical analysis

All statistical analyses were conducted using R statistical software (version 3.1.1, R Foundation for Statistical Computing, Vienna, Austria). A statistical summary of the whole dataset was conducted based on the calculated CH_4 reduction rate using the dplyr package in R. Each of the manure additive groups were subjected to significance test ($\alpha = 0.05$) to determine if they were effective in reducing manure CH_4 emissions. The manure additives that significantly reduce emissions were then included in further meta-analysis.

The response variable was the mean CH_4 production. However, different papers reported the CH_4 production in various units and different scales, such as, "g/m^2 per d", "g/m3", "g/d", "g/kg total solid", and "g/t fresh weight". The CH_4 emissions were recorded either as daily average or in cumulative total through the experimental period. To make the emission data comparable and eliminate bias caused by different units, CH_4 emission reduction rate or relative mean difference (Eq. 1) was calculated. The relative mean difference (MD) was calculated as follows:

$$\text{Relative MD} = (\text{Treatment mean} - \text{Control mean})/(\text{Control mean})\% \quad (1)$$

The meta-analytical metric included the data of study information, and sample sizes, means of CH_4 emission, standard deviations of treatment group and control group. Due to differences in units of measurements, the standardized mean difference, which is a dimensionless effect measure was calculated (Eq. 2) using the meta package in R statistical software. The default version of standardized mean difference in meta package is Hedges's (g) mean difference which is based on the pooled sample variance and a correction factor for bias (Schwarzer et al., 2015).

$$SMD = \left(1 - \frac{3}{4n-9}\right)\frac{MD}{SD_{pool}} \quad (2)$$

where MD is the mean difference, SDpool is the pooled standard deviation, and n is the total sample size of treatment and control on which SDpool is based.

Model fitting

Each group of manure additives described above may contain several different chemicals with similar function (Appendix 2). Therefore, a random-effect model that allows the variance of true effect sizes within each subgroup was used. The random-effect model was fitted to estimate the variance of the distribution of true effect sizes—between-study variance (τ2) and heterogeneity (I2) using the following equation:

$$Y_i = \mu + \zeta_i + \varepsilon_i \quad (3)$$

where Y_i is observed effect, μ is true effect size, ζ_i is true variation in effect sizes, and εi is sampling error. The between-study random effect term ζ_i has the expression of between-study variance Var (ζ_i) = τ^2 and the sampling error term εi has the expression of sample variance Var (ε_i) = s_i^2. The heterogeneity (I^2) is determined as τ^2 divided by the sum of sample variance and between-study variance ($s_i^2 + \tau^2$) and the I^2 greater than 0.5 indicates significant heterogeneity in general (Dijkstra et al., 2018).

The between-study variability can be modeled either using separate estimates of τ^2 for different subgroups, or a pooled estimate of τ^2 for all subgroups. If the true value of τ^2 varies from one subgroup to another, which is the most likely situation in this analysis, a random-effect model with separate estimates of τ^2 of subgroups should be selected (Borenstein et al., 2011). However, since there was only a few effective studies within some of the subgroups, the separate estimates of τ^2 may be imprecise and the pooled τ^2 is preferable under this situation (Borenstein et al., 2011).

Effect sizes of manure additives

Meta-analyses aim to synthesize evidence from many possible sources, by comparing and combining findings from several studies using statistical methods (Madden and Paul, 2011). The meta-analysis in this review summarizes the effects of manure additives and their potential to reduce CH_4 production in relative terms. Using manure additives to mitigate CH_4 emissions during manure storage has not been as widely applied compared to feed additives, therefore, the number of publications that report on manure additives to control CH_4 emission is much smaller. To increase the sample size for meta-analysis, the manure additives had to be classified into several categories based on their function as mentioned above.

The relative MD of CH_4 emission (%) for each manure additive treatment and the means and standard deviation of CH_4 reduction rates for each type of manure additives were analyzed. The significance test based on grouped manure additives and corresponding *P*-values are listed in Table 5. A summarized box-plot of CH_4 reduction rate for different manure additives is given in Figure 8.

The number of treatments varied considerably for different types of manure additives. Some of the groups such as humate, physical agent, straw and other chemicals contained less than 5 studies each, while acidification and biochar contained over 20 treatments each. Acidification of livestock slurry is considered when the manure additive contains acidic materials that are added to the manure during the storage to lower the pH and inhibit gaseous emissions, including CH_4. It contained different types of acidic component such as aluminum sulfate, sulfuric acid, food industrial waste, phosphogypsum, wood vinegar, etc. The effect of acidification on mitigating NH_3 emission has been widely investigated, and several of the recent observations indicated that CH_4 emissions were also reduced by manure acidification (e.g., Misselbrook et al., 2016; Petersen et al., 2012; Wang et al., 2014). Acidification was the most frequently studied manure additive in our database. Biochar is produced from the thermal decomposition of biomass and it has been applied as manure additive to livestock manure for CH_4 mitigation (Godlewska et al., 2017). Biochar is a cost-effective material with many benefits on manure composting such as enhancing the composting process, improving transformation of nutrient, and reducing the GHG and NH_3 emissions (Mao et al., 2018). In recent years, several types of biochar (cornstalk, bamboo, woody, layer manure, charcoal, holm oak, poultry litter, rice hull, coir and greenwaste biochars) and their effect on gas emissions have been investigated (Agyarko-Mintah et al., 2017; Chen et al., 2017; Chowdhury et al., 2014b; He et al., 2019; Jia et al., 2016; Sanchez-Garcia et al., 2015). A total of 24 valid studies of biochar manure additives were involved in the analysis of CH_4 reduction rate.

Not all manure additives had positive effects on mitigating CH_4 emissions (Figure 8). The relative MD for C/N content, disinfection, masking agent, and oxidizing agent were all greater than zero which indicated the CH_4 emissions of those manure additives treatment groups had increased compared to their control groups even though the increases were not statistically significant ($P > 0.05$) (Table 5). Moreover, the standard deviations of their means were relatively high indicating large variations in mitigation potential of the manure additives. These groups of manure additives were excluded in further analysis.

All other manure additives showed mitigating effects with negative means of CH_4 reduction rate (relative MD). Particularly, the reduction rates of biological mixer, physical agent, straw, and other chemicals for all included studies were less than zero (Max ≤ 0). However, the database contained small sample sizes compared with other categories (N = 3, 3, 4, 2, respectively) (Table 5). The manure additive categories of acidification, biochar, microbial digestion, physical agent, straw, and other chemicals significantly lowered CH_4 reduction rates ($P < 0.05$) and only these manure additives were selected and investigated in the meta-analysis in the next step. There were 6 references containing a total of 18 studies for the 3 manure additives (acidification, biochar and straw) to evaluate the CH_4 emission effect. The studies involved various animal species including swine, poultry, and cattle. The CH_4 emissions reported were either in cumulative or average values during the experimental period with different units. Since the data for each species and manure additives was limited, the species were not evaluated as an effect factor in meta-analysis.

Table 5. Summary statistics of CH_4 reduction rate for different types of manure additives.

Type of Manure Additives	N[a]	Mean[b]	SD	Min	Max	P^c
Acidification	37	-58.9%	30.8%	-98.1%	12.5%	<0.001
Adsorbent	8	-8.8%	15.8%	-34.4%	14.3%	0.160
Biochar	24	-41.3%	51.6%	-85.0%	169.8%	<0.001
Biological mixer	3	-21.5%	37.3%	-64.6%	0.0%	0.423
C/N content	7	32.6%	149.6%	-50.2%	370.0%	0.585
Disinfection	6	124.0%	135.6%	-5.3%	328.6%	0.075
Masking agent	12	221.2%	405.7%	-12.9%	1360.0%	0.086
Humate	3	-8.4%	26.1%	-34.0%	18.2%	0.635
Microbial digestive	12	-33.3%	36.5%	-100.0%	10.5%	0.009
Oxidizing agent	14	60.8%	150.0%	-33.3%	542.9%	0.153
Physical agent	3	-35.6%	9.8%	-46.3%	-27.0%	0.024
Straw	4	-60.1%	26.7%	-100.0%	-45.0%	0.020
Other chemicals	2	-50.0%	5.4%	-53.8%	-46.2%	0.049

[a]N is number of treatments used for the analyses

[b]Mean is the mean reduction rate of CH_4

[c] P-value with $\alpha = 0.05$

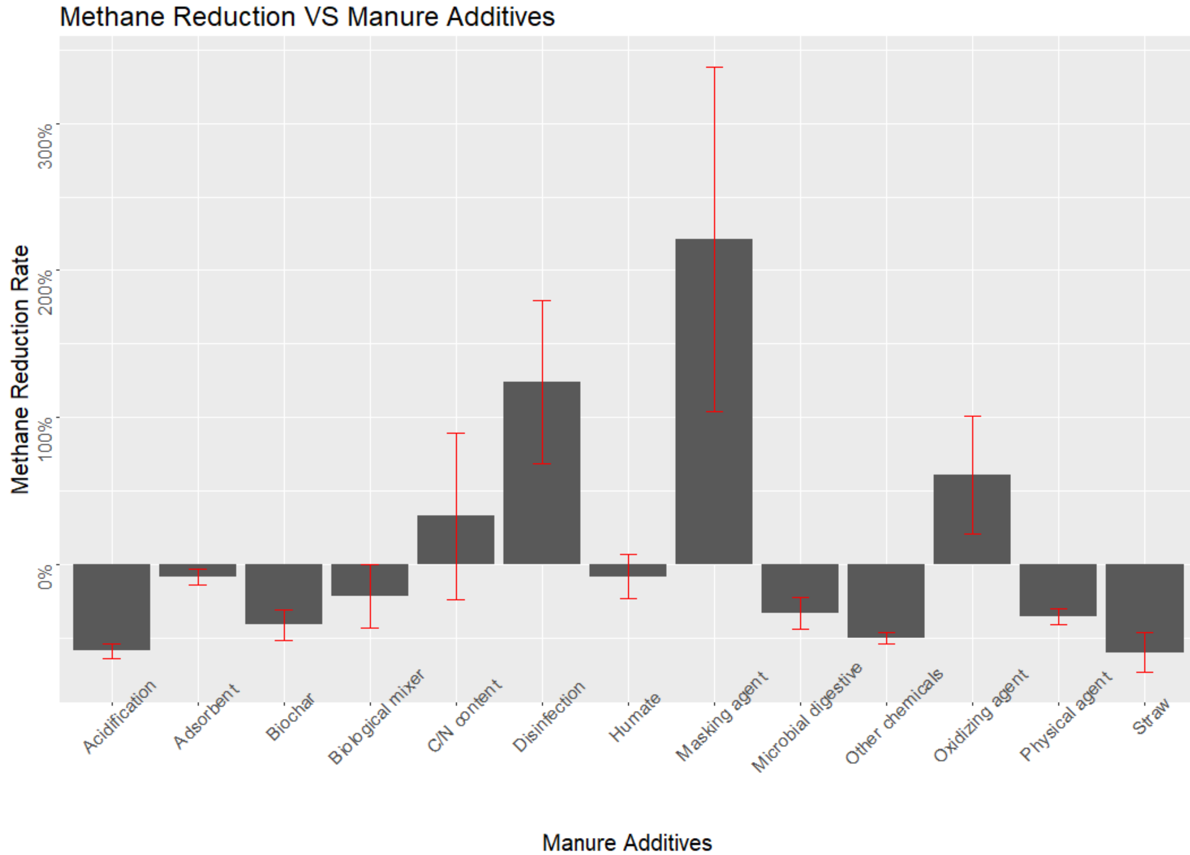


Figure 8. Box-plot of CH_4 reduction rate for different types of manure additives.

Effects of manure additives from random-effects models

The assumption of random-effects model for meta-analyses is that the true effects among all the population of studies are normally distributed and the null hypothesis is that the mean of all relevant true effects is zero (Borenstein et al., 2011). The CH_4 emissions from the 18 studies were significantly reduced by 66.3% on average (Table 6) which were consistent with the SMD from random-effects meta-analysis (P = 0.028). This overall effect indicated that the CH_4 emissions from manure storage could be mitigated by biochar, acidification, and straw. Moreover, the effect of each subgroup manure additives on mitigating CH_4 emissions was also significant with the average reduction rates of 82.4%, 78.1%, and 47.7%, respectively ($P < 0.05$). The most effective manure additive was biochar followed by acidification and straw according to relative MD analysis. Biochar as a manure additive also showed the greatest effect (SMD = -2.72), followed by straw (SMD = -1.86) and acidification (SMD = -1.31) based on the SMD of random-effects model. The SMD estimates with 95% CI according to each subgroup effects is presented using forest plot (Figure 9). The total observations of acidification subgroup were much larger compared to the other additives. The 95% CI of manure additives effects in acidification subgroup were all positive, however, the SMDs of studies from Regueiro et al. (2016) and Samer et al. (2014) varied considerably between -0.6 and -37.0. The total weights of acidification subgroup accounted for over 60% among all manure additives in the overall effect estimates, while the biochar and straw subgroups accounted for 28% and 11.9%, respectively (Table 5). This unbalanced distribution of studies' weight in the overall effect size might generate bias among different manure additives.

Table 6. Effect size and heterogeneity estimates based on overall and subgroup random-effect models.

Variable and model	Relative MD		SMD and Heterogeneity					
	MD ± SE[a]	P[b]	SMD	Weight (%)	P[b]	τ^2	I^2 (%)	P[b]
REM[c]								
Overall effect	-0.663 ± 0.006	<0.001	-1.732	100	0.028	19.4	21.8	0.195
REM[c]--Subgroup								
Acidification	-0.781 ± 0.025	<0.001	-1.311	60.1		21.8	0.0	Between group: 0.564
Biochar	-0.824 ± 0.020	<0.001	-2.721	28.0		21.8	23.8	
Straw	-0.477 ± 0.023	0.030	-1.862	11.9		21.8	0.0	

[a] SE = Standard error corresponding to number of studies for each group

[b] P-value with $\alpha = 0.05$

[c] REM = random-effect model

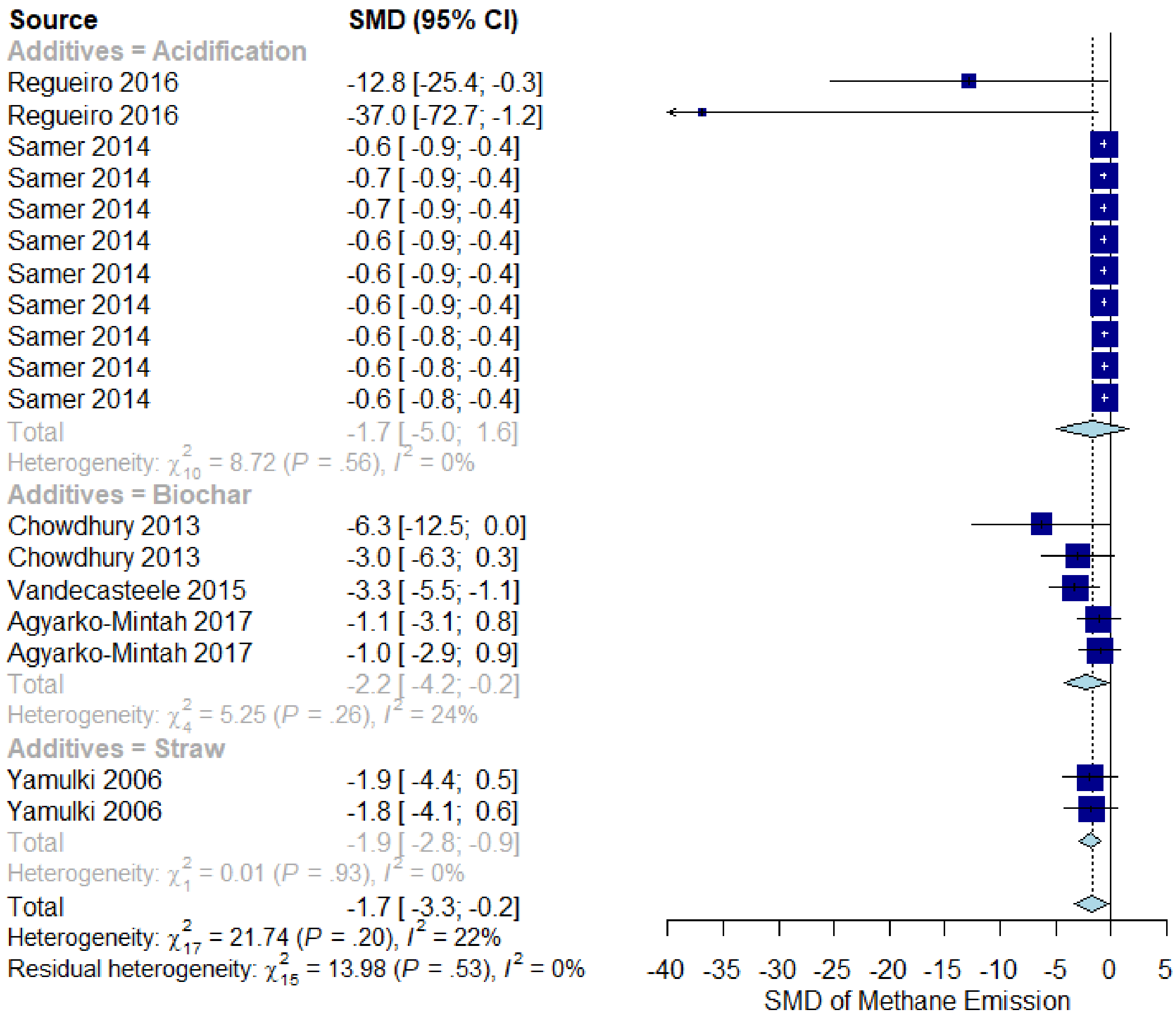


Figure 9. Forest plot showing standardized mean difference, and 95% confidence interval for three selected manure additives.

Heterogeneity test

The heterogeneity of the overall random-effects model was quantified using τ^2 and I^2. The effects of manure additives were associated with non-significant heterogeneity across all the three manure additives with only 21.8% of the total variability of the effect of manure additives in mitigating CH_4 was due to heterogeneity. As mentioned in model fitting section, a pooled τ^2 is a more precise method to conduct the random-effects model because the sample size of useful studies for manure additives was small. Therefore, the three subgroups shared the same τ^2 (21.8; Table 6). The between-study variability was not significant among the three manure additives (P = 0.564) and for the subgroups test, the heterogeneity of acidification and straw subgroups were both zero, which suggested little heterogeneity. The effects of biochar were associated with 23.8% of heterogeneity, however, this variability due to the heterogeneity was not significant (P = 0.26; Figure 9). Therefore, the heterogeneity test indicated the random-effects model was appropriate to use to quantify effect of manure additives CH_4 reduction. Mitigating CH_4 emissions by applying certain manure additives was an effective method but depends on the type of additives.

Analysis of manure type, additive type, and characteristic of treatment manure

The composting manure from livestock were categorized into manure type (cattle, poultry, swine, mixture of different manure types) and manure additives described in Table 7 and their characteristics of composting manure including pH, C/N ratio, and moisture content were generally analyzed. The initial means of pH (7.2-7.9), C/N ratio (11-18), and moisture content (58.0-75.5) for different manure types not varied in large ranges, however, the differences of means between additive types were visible. Most of the raw manure were weakly alkaline, but the averaged pH for manure composting with acidic additives (6.2 ± 1.33) was lower than other types due to the reaction of acidification. The mean of C/N ratio for biochar was relatively greater (23.6 ± 9.17) because most of the biochar contained and was made by high carbon materials such as bamboo biochar, charcoal, and cornstalk (Chowdhury et al., 2013; Chen et al., 2017; Liu et al., 2017). The highest mean C/N ratio was observed in cattle manure, and the lowest was in poultry manure, with the moderate one in swine manure which associated with the N content in the raw material. These findings showed consistency with Cao et al. (2019). Moisture content usually associated with the porosity of the material and Richard et al. (2002) indicated that moisture content for an optimum performance of composting process may vary widely between 50 to over 70% based on different raw material and composted times. All of the averaged moisture contents for different manure types and additive types were within the range of 50-75%, except for the adsorbent and biological.

A general linear regression analysis for CH_4 mitigating rate [(CH_4 emission in treatment group – control group)/control group] response to pH, C/N ratio, moisture content, manure type (cattle, poultry, and swine), and additive type (acidification, biochar, biological, physical, and C/N content) were conducted (Table 8) with a partial data from Appendix 2 which included 11 articles with 37 studies that contained completing information of all variables and CH_4 emissions (Hao et al., 2005; Yamulki, 2006; Chowdhury et al., 2013; Luo et al., 2013; Samer et al., 2014; Vandecasteele et al., 2016; Agyarko-Mintah et al., 2017; Chen et al., 2017; Liu et al., 2017; Owusu-Twum et al., 2017; Zhang et al., 2017). Moisture content significantly related to the CH_4 mitigation rate, and higher moisture content enhanced the overall reduction of CH_4 emissions ($P = 0.01$), however, the CH_4 reduction rate was not significant affected by pH ($P = 0.731$) and C/N ($P = 0.218$) in a linear manner (need to find a reasonable explanation). Manure types ($P > 0.531$) did not make significant impacts on CH_4 mitigation from composting process, however, some additives types showed significant differences from others, such as acidification vs. biological ($P = 0.001$), acidification vs. physical ($P = 0.017$), and biochar vs. biological ($P = 0.013$).

Table 7. Summary of livestock manure type, additives type, number of observations (N) and the characters of manure pH, C/N ratio, moisture content reported in mean and standard deviation.

Item		pH	N	C/N	N	Moisture content (%)	N
Manure Type	Cattle	7.2 ± 0.46	35	18 ± 4.08	8	75.5 ± 19.34	11
	Poultry	7.9 ± 0.66	8	14.6 ± 4.54	6	62.8 ± 13.45	8
	Swine	7.7 ± 0.30	10	17.8 ± 0.41	4	58.0 ± 14.12	9
	Mixture	NA	NA	11.0	1	69.0	1
Additives Type	Acidification	6.2 ± 1.33	33	20.7 ± 3.71	22	73.4 ± 19.74	33
	Adsorbent	7.2 ± 1.24	2	NA	NA	36.2	1
	Biochar	7.9 ±0.68	16	23.6 ±9.17	15	58.8 ±12.21	18
	Biological	6.9 ± 0.03	2	16.4 ± 0.00	2	43.4 ± 21.42	3
	C/N content	8.0 ± 0.13	4	14.2 ± 3.23	8	64.1 ± 2.49	10
	Disinfection	7.8 ± 0.30	3	NA	NA	NA	NA
	Masking agent	7.3 ± 0.11	6	NA	NA	NA	NA
	Humate	7.4 ± 0.02	3	NA	NA	NA	NA
	Microbial digestive	7.4 ± 0.54	5	NA	NA	NA	NA
	Oxidizing agent	7.0 ± 0.17	7	NA	NA	NA	NA
	Physical agent	8.6 ± 0.15	3	18.7 ± 1.51	3	63.0 ± 1.66	3
	Straw	8.1 ± 0.14	2	17.3 ± 2.89	3	61.7 ± 3.35	3
	Other chemicals	NA	NA	NA	NA	69.0	1

Table 8. Linear regression analysis of response CH4 mitigation rate vs. pH, C/N content, and moisture content.

Factor[1]	Mean[2]	SE[3]	P-value
CH_4 mitigation rate (%)	-32.2	46.77	0.496
pH	1.46	4.201	0.731
C/N	0.945	0.7516	0.218
Moisture content (%)	-0.802	0.2916	0.010

[1]Manure type: $P > 0.531$; Additive type: acidification vs. biochar $P = 0.055$, acidification vs. biological $P = 0.001$, acidification vs. physical $P = 0.017$; biochar vs. biological $P = 0.013$; biological vs. C/N content $P = 0.064$; all others $P > 0.100$.
[2]Mean of n = 37.
[3]SE = standard error.

Conclusions

Studies investigating manure additives for reducing CH_4 emission during storage and composting are scarce. Manure additives that include acidification, biochar, microbial digestion, physical agent, straw, and other chemicals significantly reduced CH_4 emissions from manure. In general, higher moisture contents in raw composting manure could enhance the CH_4 mitigation rates, however, the pH, and C/N content were not linearly related to CH_4 mitigation. Adding biochar, acids, and straw to manure could mitigate CH_4 emissions by 82.4%, 78.1%, and 47.7%, respectively. However, the data for straw is quite small so it should not be taken out of context as it may introduce a source of carbon into lagoons. The meta-analysis conducted with selected additives indicated manure additives were an effective method to reduce CH_4 emission, with biochar being the most effective. However, further studies of manure additives on CH_4 mitigation are required to support a more accurate quantitative analysis and potential impacts to water quality and crop yield after land application. Most of the research for biochar and straw is when used as additive to solid or semi solid manure so they should be interpreted in that context.

NET REDUCTIONS IN GREENHOUSE GASES FROM FEED ADDITIVES IN CALIFORNIA

A review of feed additives that can potentially be used in California revealed that 3NOP and nitrate may have the potential to be used as there is enough evidence of their effectiveness. Several other additives including Mootral, macroalgae and Agolin also have the potential but further studies are required to determine levels of effectiveness, safety and adequate sourcing. There was only one publication dealing with Mootral in California. Therefore, this section aims to estimate the net GHG emissions in California dairy system based on supplementation of 3NOP and nitrate to the basal diet. The following narrative will be submitted for publication to *California Agriculture*.

Materials and methods

The study was based on a life cycle assessment (LCA) conducted for the dairy industry in California (Naranjo et al., 2020). The feed ingredients used by Naranjo et al. (2020) were adjusted and recalculated using NRC (2001). The impact of producing the feed additives 3NOP and nitrate was integrated in the LCA model. Energy corrected milk (ECM) was used as the functional unit and all emissions were calculated and standardized to 1 kg of ECM.

The milk production supply chain in California from cradle to farm gate was considered the system boundary of the LCA including production of the feed additives. Specifically, these include: crop production, feed additives production, farm management, enteric methane, and manure storage. The system boundary considered emissions associated with on-farm activities, pre-farm production, and transportation of major productions up to the animal farm gate. Emissions for further activities after the products left the farm gate were not accounted in the system because they were considered to be treated in the same way for all scenarios.

Mitigation scenarios

Data sources collected from USDA National Agricultural Statistical Service (USDA-NASS) and Economic Research Service (USDA-ERS), California Department of Food and Agriculture (CDFA), peer-reviewed literature and other published resources, and databases generated from GaBi 6 software were summarized and used based on the priority of data accuracy (Naranjo et al., 2020). The GHG emissions from each process in the LCA were estimated based on the average conditions (Model 2) for dairy cattle in California as described by Naranjo et al. (2020).

The control scenario used representative diets for the California dairy cows collected from the reports by CDFA. Averaged data from 2013 to 2015 represented the diets for year 2014 in the current analysis. Within each reference year, the diets for dairy cows at different growth stages including calf up to 1 year, heifer, pregnant heifer, close-up heifer, high lactating cow, and dry cow were weighted based on a whole production cycle. We assume 4 lactations to be the average life span of a California dairy cow. The crop production for control scenario included the activities related to producing feed, and use of land, water, fertilizers, pesticides and herbicides. Additionally, energy used for machine operation, irrigation, and transportation was included. Data from USDA-NASS Quick Stats (USDA–NASS, 2017), USDA farm and ranch irrigation reports (USDA 2013), California specific agricultural reports (Burt et al., 2003; Johnson and Cody, 2015), USDA-ERS reports (USDA–ERS, 2011), University of California crop cost and return studies (UC Agricultural Issues Center, 2016), and values published in literatures (Liedke and Deimling, 2015) were used to estimate the emissions during the crop production. Enteric CH_4 emissions, farm management, energy and water used for producing crop, feeding cattle, cooling livestock facilities, animals, and milk, sanitation, cleaning, and dealing with onsite waste were according to

Naranjo et al. (2020). Similarly, manure methane and nitrous oxide (N_2O) emissions were based on methodology described by Naranjo et al. (2020).

Two scenarios were developed to estimate net mitigation effect of supplementing 3NOP to typical dairy diet in California. In scenario 1, all dairy cows were simulated to consume a diet that contains 3NOP only during lactation. In scenario 2, 3NOP was supplemented to the diet at all growing stages within a life cycle. The basal diets were same as in the control scenario and 3NOP was supplemented at a rate of 127 mg/kg DM in both scenarios.

Nitrate as a non-protein nitrogen source for cattle is usually used to replace other non-protein N sources such as urea (Velazco et al., 2014; Rebelo et al., 2019). Urea is not typically used as a nitrogen source in California representative diets, so nitrate was simulated to partially replace dietary true protein in diets to keep similar N supply for all nitrate scenarios. In nitrate scenario 1, all dairy cows were simulated to consume a diet that contained nitrate only during lactation. Nitrate was supplemented to dairy cows at all stages in nitrate scenarios 2 and 3. In nitrate scenario 2, high protein meal (e.g. corn gluten, soybean meal, and DDGS) was replaced by dietary nitrate on an equivalent N basis with no adjustment for DMI. In nitrate scenario 3, DMI was adjusted using low protein meal (e.g. corn grain, and wheat silage) to the control levels after replacing high protein meal with nitrate additives. Nitrate was supplemented to dairy cattle at a rate of 17.7 g/kg of DM for all the 3 nitrate treatment scenarios.

Emission associated with production and use of additives

3-Nitrooxypropanol

The carbon footprint of emissions associated with 3NOP production were assumed to be 52 kg CO_2e/kg 3NOP produced (DSM Nutritional Products, Ltd., pers. comm.). Moreover, with the improvement of process optimization, the carbon footprint of 3NOP could drop to 35 kg CO_2e/kg 3NOP (DSM Nutritional Products, Ltd., pers. comm.). The total GHG emissions from 3NOP production were estimated using both of the factors and the results were reported as mean with standard error to evaluate the effect of 3NOP emissions factors on total emissions. The transportation of 3NOP was calculated based on shipping from the producer (DSM Nutritional Products, Ltd., registered in Ontario, CA) to dairy farms in California by truck. The average distance used to estimate the emissions related to 3NOP transportation was weighted according to the milk production amount in California counties in 2014 (CDFA, 2014).

The magnitude of enteric CH_4 emission reduction as a result of supplementing 3NOP was calculated based on an updated version of a meta-analysis conducted by Dijkstra et al. (2018) on the anti-methanogenic effects of 3NOP. Four more recent references related to 3NOP effect on CH_4 emissions were added to the previous analysis to extend the accuracy and robustness of the meta-analytical model. The updated database included treatment means from Martinez-Fernandez et al. (2018) (beef; 1 treatment), Vyas et al. (2018) (beef; 2 treatments), Kim et al. (2019) (beef; 4 treatments), and van Wesemael et al. (2019) (dairy; 2 treatments). The final mixed-effect models for CH_4 production in the updated meta-analysis indicated effectiveness of 3NOP at mitigating CH_4 production was positively associated with 3NOP dose, and negatively associated with NDF content. Similar to the previous study, supplementation of 3NOP had stronger anti-methanogenic effects in dairy cows compared to beef cattle, at a slightly greater magnitude of mitigation. The following equations were used to calculate the mitigation effect of 3NOP that includes dose, NDF content and either dairy (Equation 1) or beef (Equation 2):

$$\text{Enteric methane reduction rate (\%)} = -41.5 - (0.260 \times \text{3NOP dose}) + (0.129 \times \text{NDF content})$$

Equation 1

Enteric methane reduction rate (%) = − 22.8 – (0.260 × 3NOP dose) + (0.129 × NDF content)

Equation 2

The equations were centered on the mean values of 127 mg 3NOP /kg DM and 326 g NDF /kg DM. Therefore, the methane reduction rates were adjusted for each cattle type when the NDF content in the 3NOP supplemented scenarios varies from the default centered value. The NDF contents for different growing stages of dairy cows in California used in this study were calculated using NRC (2001) based on ingredients supplied (Table 9). In 3NOP scenario 1, enteric CH_4 emitted from lactating cows was reduced by 38.8%, which includes adjustment for NDF content (Table 9). In scenario 2, if the cows were not lactating, the emission reduction rate was assumed to be similar to beef cattle so Equation 2 was applied. The enteric CH_4 reduction rates for heifer, pregnant heifer, close up heifer, high lactating cow, and dry cow were 11.1%, 1.1%, 10.3%, 38.8%, and 4.0%, respectively (Table 9).

The GHG emissions from the farm management and manure management processes in the LCA for 3NOP scenarios were same as for the control scenario because we assumed no residues and by-products from the 3NOP production process. Nkemka et al. (2019) confirmed that there was no residual effect on anaerobic digestion of the manure from beef cattle fed diets supplemented with 3NOP.

Table 9. Enteric methane reduction rates and total emissions per life cycle at different dairy growing stages for control and treatment scenarios.

Cattle Stage	Control		3NOP 1[a]		3NOP 2[a]		Nitrate 1		Nitrate 2		Nitrate 3	
	Reduction (%)	CH_4 (kg/lifetime)	Reduction (%)	CH_4 (kg/lifetime)	Reduction (%)	CH_4 emission (kg/lifetime)	Reduction (%)	CH_4 (kg/lifetime)	Reduction (%)	CH_4 (kg/lifetime)	Reduction (%)	CH_4 (kg/lifetime)
Calf	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Heifer	0	10.6	0	10.6	-11.1	9.4	0	10.6	-15.4	9.1	-15.4	9.6
Pregnant	0	73.8	0	73.8	-1.1	72.9	0	73.8	-15.4	62.3	-15.4	66.0
Close up	0	9.2	0	9.2	-10.3	8.2	0	9.2	-15.4	7.6	-15.4	7.8
lactating	0	575.8	-38.8	352.4	-38.8	352.4	-15.4	481.7	-15.4	481.7	-15.4	488.5
Dry cow	0	60.9	0	60.9	-4.0	58.5	0	60.9	-15.4	50.0	-15.4	52.0

aNDF content (g/kg DM) in diets for 3NOP scenarios: 250 (Calf up to 1 year), 419 (Heifer), 496 (Pregnant heifer), 425 (Close up heifer), 349 (High lactating cow), and 474 (Dry cow).

Nitrate

Nitrate was assumed to be supplemented to dairy diets as Calcium nitrate ($Ca(NO_3)_2$). Brentrup et al. (2016) reported carbon footprint associated with $Ca(NO_3)_2$ production were estimated to be 1.76 kg CO_2e/kg $Ca(NO_3)_2$ in USA and 0.67 kg CO_2e/kg $Ca(NO_3)_2$ produced in Europe. Total emissions associated with $Ca(NO_3)_2$ production were calculated using both carbon footprint values for USA and Europe, and the emissions from nitrate production process are reported as the mean with standard deviation. Emissions related to transportation of $Ca(NO_3)_2$ was calculated based on the shipping distance between supplier and dairy farms in California. Several chemical companies supply $Ca(NO_3)_2$ within California and the plant with the minimum travel distance (by truck) to each county was assumed as its $Ca(NO_3)_2$ supplier. The overall average distance was weighted based on the milk production in California counties in 2014 (CDFA, 2014) and used for emission calculations related to chemical transportations. Feed production for different nitrate treatment scenarios were recalculated based on the replacement of high protein meals by dietary nitrate to provide equivalent N as compared to the diets for control scenario at each growing stage using NRC (2001) software.

The anti-methanogenic effects of nitrate were calculated based on equations developed by Feng et al. (2020 unpublished). Meta-analytical results indicated nitrate effect on enteric CH_4 production to be significantly affected by nitrate dose. However, there was no difference in effectiveness in dairy and beef cattle. The reduction rate for enteric CH_4 emissions is estimated by the meta-analytical model as given in Equation 3.

$$\text{Enteric methane reduction rate (\%)} = -14.6 - (0.808 \times \text{nitrate dose}) \qquad \text{Equation 3}$$

The equation is centered on mean nitrate dose of the database, which was 17.7 g/kg of DM. We kept the average as the dose of nitrate supplementation in the scenarios evaluated in this study.

We assumed there were no residues and by-products from nitrate production and the total GHG emissions from farm management process for nitrate treatment scenarios including on-farm energy and water usage were not affected by nitrate additives. Methane emissions from manure storage were calculated as a function of VS (Nielsen et al., 2013) which was associated with NDF content, CP content and DMI (Appuhamy et al., 2016). As the dietary ingredients and DMI for nitrate scenarios varied with the adjustment of nitrate additives, the total GHG emissions from manure management were recalculated based on the different nitrate feeding scenarios.

Results and discussion

3-Nitrooxypropanol

The GHG emissions from crop production, farm management, enteric CH_4 and manure storage for control scenario were 0.174, 0.0608, 0.432, and 0.457 kg CO_2e per kg of ECM produced in California, respectively (Figure 10). Total GHG emissions from crop production, farm management, and manure storage were not affected by feeding 3NOP to dairy cows. The mean GHG emissions related to production of 3NOP in scenario 1 was 3.23 g CO_2e/kg ECM which was lower than 3.92 g CO_2e/kg ECM in scenario 2 because 3NOP was only fed to lactating cows in scenario 1. Enteric CH_4 emissions were 0.298 and 0.295 kg CO_2e/kg ECM for 3NOP scenarios 1 and 2, respectively, which were reduced by 31.0% and 31.7% compared to the control scenario, respectively, due to the inhibition effect of 3NOP on CH_4 production. Accounting for 3NOP production, the net enteric methane emission reduction was 30.3% in scenario 1 and 30.8% in scenario 2.

The total GHG emissions for control and 3NOP treatment scenarios 1 and 2 were 1.12, 0.993 and 0.991 kg CO_2e/kg ECM, respectively (Figure 10). Feeding 3NOP to dairy cows resulted in a net reduction of total GHG emission of 11.3% in 3NOP scenario 1 and 11.5% in 3NOP scenario 2 compared to the control scenario. Using 3NOP for dairy cows at all growing stages only further reduced 0.2 percentage points more compared to limiting 3NOP supplementation during lactation.

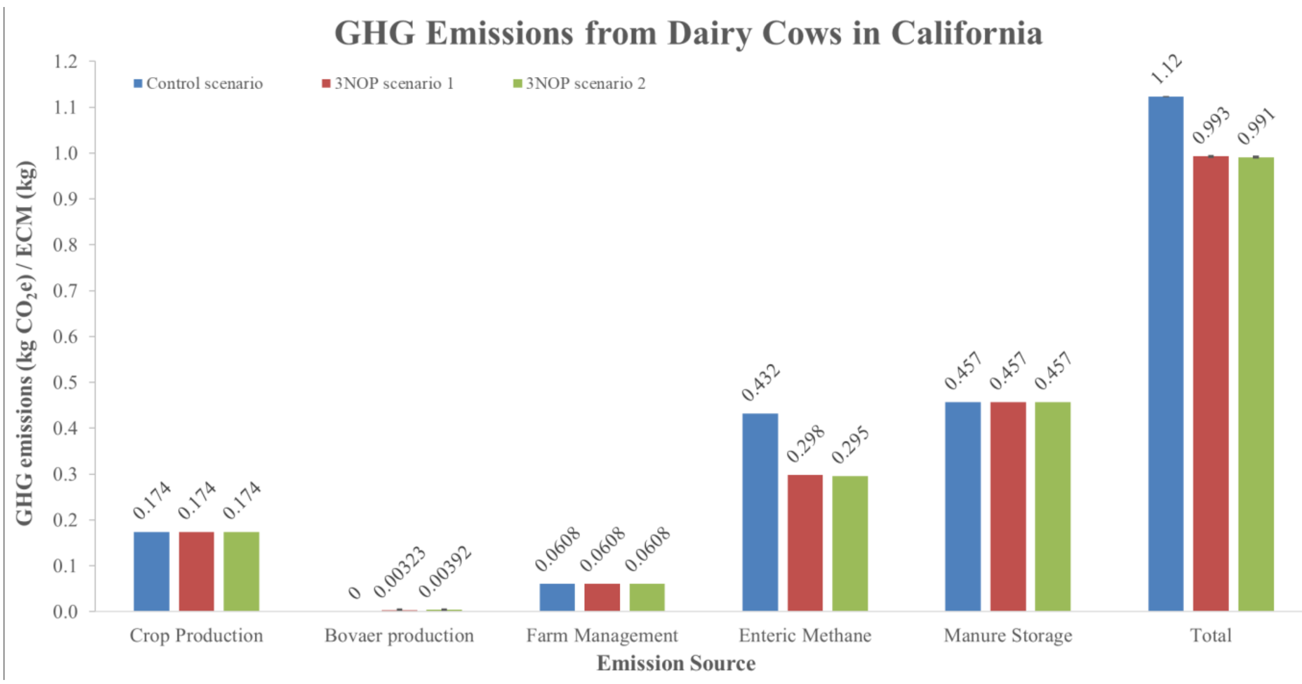


Figure 10. Comparison of global warming potential (GWP) by emission source for control and 3NOP scenarios 1 and 2 in California dairy cows.

The GHG emissions associated with 3NOP production for scenarios 1 and 2 were 3.86 and 4.69 g CO_2e/kg ECM, respectively, assuming 3NOP carbon footprint of 52 kg CO_2e/kg and 2.60

and 3.16 g CO_2e/kg ECM, respectively, using manufacturer reported values of 35 kg CO_2e/kg 3NOP. This indicates that with the improvement of manufacturing process, the GHG emissions from 3NOP production can be reduced by 32.6%, improving net impact of 3NOP in reducing enteric emissions.

Nitrate

The total GHG emissions and estimates of the various components in dairy cattle supplemented with nitrate is given in Figure 11. In nitrate scenario 1, the mean GHG emissions associated with nitrate production was 0.0182 kg CO_2e/kg ECM and 0.0219 kg CO_2e/kg ECM in nitrate scenarios 2 and 3 due to differences in the phases of dairy production that nitrate was included. The error bars for nitrate production in Figure 11 showed the deviations of GHG emissions estimated with different carbon footprint of $Ca(NO_3)_2$ production in USA and Europe. According to Brentrup et al. (2016) the difference was mainly due to a catalyst technology developed in Europe. The GHG emissions calculated with carbon footprint value for $Ca(NO_3)_2$ in USA were 0.0255 kg CO2e/kg ECM for nitrate scenario 1, and 0.0307 kg CO2e/kg ECM for nitrate scenarios 2 and 3. Using the European carbon footprint (0.67 kg CO2e/kg $Ca(NO_3)_2$ produced), the GHG emissions from nitrate production was 0.0109 kg CO2e/kg ECM for nitrate scenario 1, and 0.0131 kg CO2e/kg ECM for nitrate scenarios 2 and 3. The GHG emissions from nitrate production decreased 57.3% based on European values compared to those in USA.

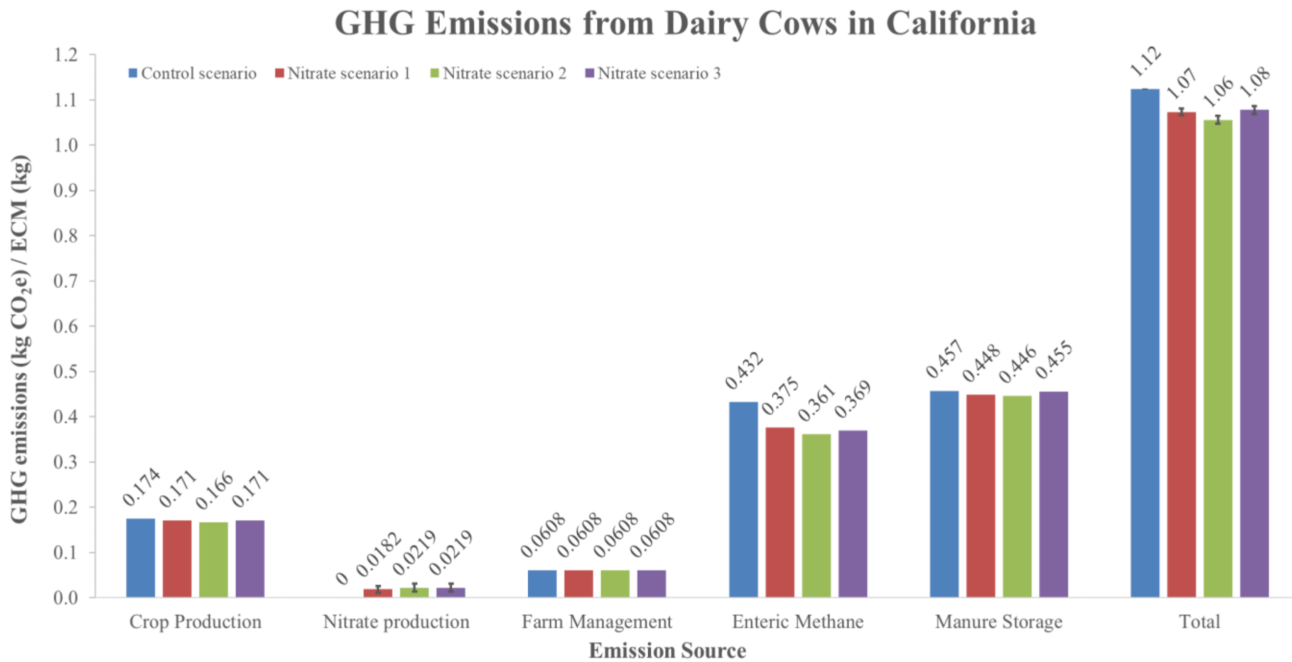


Figure 11. Comparison of global warming potential (GWP) by emission source for control and nitrate scenarios of dairy cows in California.

The GHG emissions related to crop production was 0.174 kg CO_2e/kg ECM for the control scenario, and reduced to 0.171, 0.166, and 0.171 CO_2e/kg ECM for nitrate scenarios 1, 2, and 3, respectively, which was mainly caused by the decline in the amount of protein that was replaced by nitrate. The DMI for scenario 3 was adjusted back to the control level, and therefore the GHG emissions from crop production in nitrate scenario 3 was 0.005 CO_2e/kg ECM greater than in scenario 2. The GHG emissions from manure storage were 0.457, 0.448, 0.446, 0.455 kg CO2e/kg ECM in control and three nitrate scenarios, respectively. The differences of GHG emissions from manure management among nitrate scenarios were associated with the variations in dietary NDF content, CP content, and DMI of adjusted diets. Enteric CH_4 emissions from nitrate scenarios 1 to 3 were 0.375, 0.361, and 0.369 kg CO_2e/kg ECM respectively, which were reduced by 13.2%, 16.4%, and 14.6% respectively, compared to CH_4 emissions from control scenario (0.432 CO_2e/kg

ECM) based on values calculated for CH_4-mitigating effect of dietary nitrate (Table 9). The net reduction enteric methane emission (including nitrate production) is calculated to be 8.98, 11.35 and 9.51% for nitrate scenarios 1 to 3, respectively. The GHG emissions from farm management were the same for control and all nitrate scenarios which was 0.0608 kg CO_2e/kg ECM (Figure 11).

The total GHG emissions for control scenario was 1.12 kg CO_2e/kg ECM, while with supplementing dietary nitrate to dairy cows in California, the total GHG emissions were 1.07, 1.06, and 1.08 kg CO_2e/kg ECM respectively in nitrate scenarios 1, 2, and 3. Therefore, the total GHG emissions for three nitrate scenarios were reduced by 4.5%, 5.4%, and 3.6% from the control scenario. The net reductions of total GHG emissions for nitrate scenarios 1, 2, and 3 were 0.05, 0.06, 0.04 kg CO_2e/kg ECM, respectively (Figure 11). Nitrate scenario 2 showed the greatest net reduction of total GHG emissions which reduced 0.9% and 1.8% more of total GHG emissions compared to scenarios 1 and 3, respectively.

Comparison of 3-nitrooxypropanol and nitrate additives

Total GHG emissions from control scenario were lower than values published in several previous studies. For example, Gerber et al. (2011) reported the GHG emissions in North America to be 1.20 kg CO_2e/kg ECM and Thoma et al. (2013) reported 1.23 kg CO_2e/kg ECM. In Canada, Alvarez-Hess et al. (2019) reported 1.21 kg CO_2e/kg ECM, but in two Australian dairy farms, the authors reported 1.09 and 0.97 kg CO_2e/kg ECM, respectively, which were slightly lower than the value estimated in the present study. Emissions from manure storage accounted for 40.6% to 46.1% of the total GHG emissions, which contributed the largest amount to total GHG emissions in all scenarios. Enteric CH_4 emissions from control scenario accounted for 38.4% of the total GHG emissions but the proportions of enteric CH_4 emissions dropped and varied between 29.8%

(3NOP, scenario 2) to 35.0% (nitrate scenario 1). Crop production emitted 15.5% to 17.6% of total GHG emissions and the significant decrease in enteric CH_4 emissions resulted in a proportional increase of GHG emissions of crop production in 3NOP scenarios. Only 0.3% to 2.1% of emissions were attributed to feed additives production in supplemental scenarios. The GHG emissions associated with farm management were same for all scenarios.

Although both 3NOP and nitrate additives decreased the total GHG emissions, the mitigating effect of 3NOP was greater than nitrate reaching a highest reduction rate of 11.8% (3NOP scenario 2). The average net reduction rate of GHG emissions for 3NOP was 11.7% and supplementing 3NOP to dairy cows only during lactations or to the entire growing herds had a minor difference in the total GHG emissions. The mean net reduction rate of GHG emissions in dairy cows feeding nitrate was 4.9%. The greatest net GHG emissions achieved with nitrate was 6.1% with supplementation of nitrate to dairy cows in all growing stages. These results partially agreed with Alvarez-Hess et al. (2019) who reported that the GHG emissions went down from 1.13 kg CO_2e/kg ECM to 1.10 kg CO_2e/kg ECM (a reduction of 2.65%) when nitrate was fed to lactating cows only at a rate of 21 g/kg DM. The GHG emissions from groups supplemented with 3NOP at 86 mg/kg DM were between 0.83 and 1.03 kg CO_2e/kg ECM in dairy farms in Australia and Canada (Alvarez-Hess et al., 2019).

The carbon footprint of nitrate is greater than that of 3NOP and it is fed at a rate of an average 17.7 g/kg DM compared to an average of 127 mg/kg DM for 3NOP. Therefore, much higher quantities for nitrate are required for methane mitigation resulting in about 5.6 times GHG emission from production of the additive. Moreover, nitrate toxicity caused by the high methemoglobin levels in ruminants fed in greater quantities is a concern and currently not recommended as methane mitigating feed additives to cattle (Bruning-Fann and Kaneene, 1993; Lee and Beauchemin, 2014).

The impact of manure additives can be added to the effect of feed additives. When biochar, acids, and straw are used alongside 3NOP the potential combined effect would be 20 to 34% from the whole dairy production system in CA.

Conclusions

This LCA was conducted based on dairy cows in California and evaluated the mitigation effect of two promising feed additives—3NOP and nitrate, on total GHG emissions. The average net reduction rate of supplementing 3NOP and nitrate were 11.7% and 4.9%, respectively. 3NOP had a greater effect than nitrate on reducing total GHG emissions with a highest performance of 11.8%. Feeding 3NOP to only lactating cows or to the entire growth stages did not make significant difference in total GHG emissions. Considering California milk production of 18 billion kg in 2017, using nitrate on California dairy cows would reduce GHG emissions 1.09 billion kg CO_2e and 3NOP 2.33 billion kg CO_2e annually.

SUMMARY

This study evaluated strategies to reduce methane emission from enteric and lagoon sources with emphasis on California conditions. A considerable amount of literature is available on feed additives but studies on manure additives are much more scarce. Through a literature review, a large amount of feed additives were considered, but only about 17% of those evaluated through effect size analysis had a statistically significant mitigating impact on methane emissions. The majority of those were found to either increase cost, reduce productivity or increase an alternative pollutant at the expense of methane mitigation. Therefore, only 3NOP and nitrate were identified as those with the highest potential. An updated meta-analysis for effectiveness of 3NOP showed 41% reduction in dairy cattle and 22.4% in beef cattle. A new meta-analysis for nitrate showed 14.4% reduction in mitigating methane with no differences between dairy and beef cattle. In both cases dosage of feed additives was related to further reduction in emissions.

Manure additives that include acidification, biochar, microbial digestion, physical agent, straw, and other chemicals significantly reduced CH_4 emissions from manure. In general, higher moisture contents in raw composting manure could enhance the CH_4 mitigation rates, however, the pH, and C/N content were not linearly related to CH_4 mitigation. Adding biochar, acids, and straw to manure could mitigate CH_4 emissions by 82.4%, 78.1%, and 47.7%, respectively. The meta-analysis conducted with selected additives indicated manure additives were an effective method to reduce CH_4 emission, with biochar being the most effective. However, further studies of manure additives on CH_4 mitigation are required to support a more accurate quantitative analysis. A life cycle assessment was conducted based on dairy cows in California and evaluated the mitigation effect of 3NOP and nitrate on total GHG emissions.

The average net reduction rate of supplementing 3NOP and nitrate were 11.7% and 4.9%, respectively. 3NOP had a greater effect than nitrate on reducing total GHG emissions with a highest performance of 11.8%. Feeding 3NOP to only lactating cows or to the entire growth stages did not make significant difference in total GHG emissions. Given the toxicity concerns of nitrate, only 3NOP is recommended for use pending FDA approval. However, further research is highly recommended for Mootral, macroalage and grape pomace to establish efficacy and solve related issues as there were only one or two studies conducted relevant to California conditions.

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Appendix 1. Number of treatment, mean, standard deviation, minima and maxima of mean difference of methane production for control and treatment groups based on feed additive type

Treatment Type	Col/Trt	counts	meanMD	sdMD	maxMD	minMD
3NOP	Control	8	0.0	0.00	0.0	0.0
3NOP	Treatment	15	-71.8	71.13	-1.2	-212.1
Acacia mearnsii	Control	5	0.0	0.00	0.0	0.0
Acacia mearnsii	Treatment	6	-45.4	45.06	-1.2	-126.0
Acetate inclusion	Control	2	0.0	0.00	0.0	0.0
Acetate inclusion	Treatment	2	-11.3	2.13	-9.8	-12.8
Antibloat	Control	1	0.0	NA	0.0	0.0
Antibloat	Treatment	1	-103.2	NA	-103.2	-103.2
Bacteria	Control	14	0.0	0.00	0.0	0.0
Bacteria	Treatment	14	4.5	7.81	23.0	-3.1
Bromochloromethane	Control	10	0.0	0.00	0.0	0.0
Bromochloromethane	Treatment	20	-32.8	27.07	-0.8	-89.7
Calcium soap inclusion	Control	2	0.0	0.00	0.0	0.0
Calcium soap inclusion	Treatment	4	-8.6	12.04	9.3	-16.5
Camelina inclusion	Control	1	0.0	NA	0.0	0.0
Camelina inclusion	Treatment	1	-120.0	NA	-120.0	-120.0
Canola inclusion	Control	2	0.0	0.00	0.0	0.0
Canola inclusion	Treatment	2	-34.4	23.90	-17.5	-51.3
Carboxylic acid	Control	5	0.0	0.00	0.0	0.0
Carboxylic acid	Treatment	5	1.1	4.70	5.5	-6.0
Cerium chloride	Control	1	0.0	NA	0.0	0.0
Cerium chloride	Treatment	3	-4.2	2.18	-1.9	-6.3
Chestnut	Control	7	0.0	0.00	0.0	0.0
Chestnut	Treatment	9	-21.5	27.05	1.6	-83.6
Chicory	Control	1	0.0	NA	0.0	0.0
Chicory	Treatment	1	-2.4	NA	-2.4	-2.4
Chitosan	Control	2	0.0	0.00	0.0	0.0
Chitosan	Treatment	4	2.3	21.10	33.9	-9.3
Chloroform	Control	1	0.0	NA	0.0	0.0
Chloroform	Treatment	1	38.0	NA	38.0	38.0
Coconut	Control	2	0.0	0.00	0.0	0.0
Coconut	Treatment	3	-26.7	60.35	14.4	-96.0
Coconut inclusion	Control	9	0.0	0.00	0.0	0.0
Coconut inclusion	Treatment	16	-57.2	68.93	-1.1	-211.0
Corn	Control	1	0.0	NA	0.0	0.0

Corn	Treatment	3	26.3	22.74	45.0	1.0
Cumin	Control	1	0.0	NA	0.0	0.0
Cumin	Treatment	2	-1.8	0.27	-1.6	-1.9
Cysteine	Control	4	0.0	0.00	0.0	0.0
Cysteine	Treatment	4	-3.5	11.67	8.6	-19.5
DDGS concentrate	Control	1	0.0	NA	0.0	0.0
DDGS concentrate	Treatment	3	-45.0	8.54	-37.0	-54.0
Defaunation	Control	2	0.0	0.00	0.0	0.0
Defaunation	Treatment	2	-1.3	0.51	-0.9	-1.6
DHA inclusion	Control	3	0.0	0.00	0.0	0.0
DHA inclusion	Treatment	5	9.6	21.38	35.0	-23.0
Essential oil blend	Control	4	0.0	0.00	0.0	0.0
Essential oil blend	Treatment	4	-14.8	16.58	3.8	-36.5
Eucalyptus	Control	1	0.0	NA	0.0	0.0
Eucalyptus	Treatment	1	-7.2	NA	-7.2	-7.2
Eugenol	Control	1	0.0	NA	0.0	0.0
Eugenol	Treatment	3	-15.7	2.52	-13.0	-18.0
Fatty acid blend inclusion	Control	4	0.0	0.00	0.0	0.0
Fatty acid blend inclusion	Treatment	9	-23.7	38.30	17.0	-84.0
Fibrolytic enzyme	Control	3	0.0	0.00	0.0	0.0
Fibrolytic enzyme	Treatment	4	27.0	35.22	74.0	-0.1
Flavomycin	Control	1	0.0	NA	0.0	0.0
Flavomycin	Treatment	1	-1.9	NA	-1.9	-1.9
Flavonoids	Control	1	0.0	NA	0.0	0.0
Flavonoids	Treatment	1	-2.2	NA	-2.2	-2.2
Flaxseed inclusion	Control	5	0.0	0.00	0.0	0.0
Flaxseed inclusion	Treatment	5	-11.6	31.31	24.0	-58.1
Fumaric acid	Control	13	0.0	0.00	0.0	0.0
Fumaric acid	Treatment	19	-5.7	9.46	11.3	-27.2
Garlic	Control	13	0.0	0.00	0.0	0.0
Garlic	Treatment	16	0.3	5.31	15.0	-7.7
Glycerin	Control	2	0.0	0.00	0.0	0.0
Glycerin	Treatment	6	0.6	5.00	9.9	-4.1
GOS	Control	12	0.0	0.00	0.0	0.0
GOS	Treatment	12	1.6	6.99	17.2	-8.3
Grape marc	Control	1	0.0	NA	0.0	0.0
Grape marc	Treatment	2	-88.0	9.90	-81.0	-95.0
Grass	Control	3	0.0	0.00	0.0	0.0
Grass	Treatment	3	0.5	1.79	2.5	-0.9
Hydrolysable tannins	Control	2	0.0	0.00	0.0	0.0
Hydrolysable tannins	Treatment	4	3.7	1.55	5.6	1.8
Isobutyrat inclusion	Control	1	0.0	NA	0.0	0.0

Isobutyrat inclusion	Treatment	3	-2.6	1.27	-1.2	-3.6
Isovalerate inclusion	Control	1	0.0	NA	0.0	0.0
Isovalerate inclusion	Treatment	3	-2.8	1.60	-1.0	-4.1
Lasolocid	Control	3	0.0	0.00	0.0	0.0
Lasolocid	Treatment	3	-0.2	5.38	5.0	-5.7
Lauric	Control	5	0.0	0.00	0.0	0.0
Lauric	Treatment	7	-20.3	63.05	81.0	-96.0
Leather strap	Control	1	0.0	NA	0.0	0.0
Leather strap	Treatment	3	-1.9	1.28	-1.1	-3.4
Legume	Control	5	0.0	0.00	0.0	0.0
Legume	Treatment	5	1.7	4.17	8.7	-2.5
Linoleic inclusion	Control	1	0.0	NA	0.0	0.0
Linoleic inclusion	Treatment	1	-2.5	NA	-2.5	-2.5
Linseed inclusion	Control	14	0.0	0.00	0.0	0.0
Linseed inclusion	Treatment	18	-40.7	55.95	23.0	-196.1
Lotus tannins	Control	3	0.0	0.00	0.0	0.0
Lotus tannins	Treatment	3	0.2	20.94	23.4	-17.4
Lovastatin	Control	2	0.0	0.00	0.0	0.0
Lovastatin	Treatment	4	13.8	13.25	29.1	0.4
Lupine seed	Control	3	0.0	0.00	0.0	0.0
Lupine seed	Treatment	3	-4.3	8.03	2.6	-13.1
Maca	Control	3	0.0	0.00	0.0	0.0
Maca	Treatment	3	-3.5	8.46	5.9	-10.5
Malic acid	Control	2	0.0	0.00	0.0	0.0
Malic acid	Treatment	3	-19.8	17.21	-5.2	-38.8
Methylbutyrate inclusion	Control	1	0.0	NA	0.0	0.0
Methylbutyrate inclusion	Treatment	3	-2.3	1.43	-0.7	-3.4
Mimosa	Control	2	0.0	0.00	0.0	0.0
Mimosa	Treatment	3	-2.5	3.23	0.8	-5.7
Monensin	Control	40	0.0	0.00	0.0	0.0
Monensin	Treatment	45	-14.9	23.32	27.3	-92.4
Monensin blend	Control	1	0.0	NA	0.0	0.0
Monensin blend	Treatment	3	-14.6	18.17	6.2	-27.4
Myristic acid	Control	3	0.0	0.00	0.0	0.0
Myristic acid	Treatment	4	11.8	36.35	63.0	-18.0
Myristic acid inclusion	Control	4	0.0	0.00	0.0	0.0
Myristic acid inclusion	Treatment	4	-81.6	85.88	-4.1	-156.0
Nisin	Control	2	0.0	0.00	0.0	0.0
Nisin	Treatment	2	-2.1	0.97	-1.4	-2.8
Nitrate	Control	35	0.0	0.00	0.0	0.0
Nitrate	Treatment	43	-39.5	34.65	4.0	-144.8
Nitrate and Sulfate	Control	1	0.0	NA	0.0	0.0

Nitrate and Sulfate	Treatment	1	-7.8	NA	-7.8	-7.8
Nitroethane	Control	2	0.0	0.00	0.0	0.0
Nitroethane	Treatment	5	-53.8	41.67	-28.6	-127.9
Oregano	Control	3	0.0	0.00	0.0	0.0
Oregano	Treatment	5	-127.7	120.56	-2.4	-298.0
Peppermint	Control	1	0.0	NA	0.0	0.0
Peppermint	Treatment	1	-27.5	NA	-27.5	-27.5
Polyethylene glycol	Control	4	0.0	0.00	0.0	0.0
Polyethylene glycol	Treatment	4	15.5	22.08	48.4	2.7
Propanediol	Control	1	0.0	NA	0.0	0.0
Propanediol	Treatment	1	-1.7	NA	-1.7	-1.7
Proteolytic enzyme	Control	1	0.0	NA	0.0	0.0
Proteolytic enzyme	Treatment	1	-9.3	NA	-9.3	-9.3
Quebracho	Control	2	0.0	0.00	0.0	0.0
Quebracho	Treatment	6	-15.7	19.68	3.0	-41.1
Rumen protected FA inclusion	Control	1	0.0	NA	0.0	0.0
Rumen protected FA inclusion	Treatment	1	-33.7	NA	-33.7	-33.7
Rumen protected fat	Control	1	0.0	NA	0.0	0.0
Rumen protected fat	Treatment	1	-1.4	NA	-1.4	-1.4
Saifoin maturity	Control	2	0.0	0.00	0.0	0.0
Saifoin maturity	Treatment	2	24.0	1.41	25.0	23.0
Saifoin tannins	Control	1	0.0	NA	0.0	0.0
Saifoin tannins	Treatment	5	-5.6	13.97	7.0	-23.0
Saponaria	Control	3	0.0	0.00	0.0	0.0
Saponaria	Treatment	3	-16.6	13.99	-4.5	-31.9
Sericea lespedeza tannins	Control	4	0.0	0.00	0.0	0.0
Sericea lespedeza tannins	Treatment	4	-3.2	1.01	-2.0	-4.1
Sodium bicarbonate	Control	1	0.0	NA	0.0	0.0
Sodium bicarbonate	Treatment	1	-3.3	NA	-3.3	-3.3
Sorghum tannins	Control	6	0.0	0.00	0.0	0.0
Sorghum tannins	Treatment	6	1.5	2.15	4.1	-0.9
Soybean oil inclusion	Control	2	0.0	0.00	0.0	0.0
Soybean oil inclusion	Treatment	2	-0.5	1.79	0.7	-1.8
Stearic	Control	5	0.0	0.00	0.0	0.0
Stearic	Treatment	7	-8.8	61.32	81.0	-96.0
Styzolobium tannins	Control	1	0.0	NA	0.0	0.0
Styzolobium tannins	Treatment	1	-0.1	NA	-0.1	-0.1
Sucrose	Control	1	0.0	NA	0.0	0.0
Sucrose	Treatment	1	0.0	NA	0.0	0.0
Sulfate	Control	1	0.0	NA	0.0	0.0
Sulfate	Treatment	1	-5.4	NA	-5.4	-5.4
Sulla tannins	Control	4	0.0	0.00	0.0	0.0

Sulla tannins	Treatment	4	-0.3	4.99	5.1	-6.1
Sunflower inclusion	Control	3	0.0	0.00	0.0	0.0
Sunflower inclusion	Treatment	4	-34.3	26.25	-1.7	-57.8
Sunphenon	Control	1	0.0	NA	0.0	0.0
Sunphenon	Treatment	3	-4.1	2.91	-1.8	-7.4
Tallow inclusion	Control	1	0.0	NA	0.0	0.0
Tallow inclusion	Treatment	1	-24.0	NA	-24.0	-24.0
Tea saponin	Control	6	0.0	0.00	0.0	0.0
Tea saponin	Treatment	6	-6.3	7.50	-0.7	-18.3
Triiodothyronine	Control	2	0.0	0.00	0.0	0.0
Triiodothyronine	Treatment	2	-0.7	1.94	0.7	-2.1
Valonea	Control	2	0.0	0.00	0.0	0.0
Valonea	Treatment	2	0.2	1.37	1.1	-0.8
Vitacogen	Control	2	0.0	0.00	0.0	0.0
Vitacogen	Treatment	2	5.2	9.63	12.0	-1.6
Yeast	Control	9	0.0	0.00	0.0	0.0
Yeast	Treatment	9	-9.9	16.77	7.2	-42.0
Yucca	Control	9	0.0	0.00	0.0	0.0
Yucca	Treatment	12	-2.0	4.81	6.7	-12.2

Appendix 2. Summary of manure additives investigated in this study.

Additives Type	Ingredient	Reference	Year	Species	Meta-analysis inclusion
Acidification	aluminum sulfate	Regueiro et al.	2016	pig	Yes
	calcium superphosphate	Zhang, et al.	2017	pig	No
	food industrial waste	Samer, et al.	2014	dairy	Yes
	hydrochloric acid	Petersen, et al.	2012	cattle	No
	lactic acid	Berg, et al.	2006	cattle	No
	methionine	Petersen, et al.	2012	cattle	No
	nitric acid	Berg, et al.	2006	cattle	No
	phosphogypsum	Hao, et al.	2005	cattle	No
		Luo, et al.	2013	pig	No
	sulfate	Petersen, et al.	2012	cattle	No
	sulfuric acid	Misselbrook, et al.	2016	cattle	No
		Owusu-Twum, et al	2017	cattle	No
		Wang, et al.	2014	pig	No
	wood vinegar	Wang, et al.	2018	pig	No
Adsorbent	zeolite	Wheeler, et al.	2010	dairy	No
		Wang, et al.	2018	pig	No
	clay	Chen, et al.	2018	chicken	No
Biochar	bamboo	Chen, et al.	2017	hen	No
		Liu, et al.	2017	hen	No
		He, et al.	2019	pig	No
	charcoal	Chowdhury, et al.	2014	hen	Yes
	coir	Chen, et al.	2017	hen	No
	cornstalk	Chen, et al.	2017	hen	No
	greenwaste	Agyarko-Mintah, et al.	2017	poultry	Yes
	layer manure	Chen, et al.	2017	hen	No
	poultry litter	Agyarko-Mintah, et al.	2017	poultry	Yes
	rice hull	Jia, et al.	2016	chicken	No
	rice straw	He, et al.	2019	pig	No
	woody	Chen, et al.	2017	hen	No
	N/A	Vandecasteele	2016	chicken	Yes
	N/A	Sonoki, et al	2011	cattle	No
	N/A	Mao, et al.	2018	pig	No
	N/A	Wang, et al.	2018	pig	No
	N/A	Chowdhury, et al.	2014	animal	No
Biological materials	EU200	Owusu-Twum, et al	2017	cattle	No
	Biobuster	Owusu-Twum, et al	2017	cattle	No
	Biosuper	Martinez, et al.	2003	pig	No

C/N content	sawdust	Jia, et al.	2016	chicken	No
	plastic tube pieces	Chowdhury, et al.	2014	animal	No
	woodchips	Chowdhury, et al.	2014	animal	No
	lupin residues	Chowdhury, et al.	2014	animal	No
Disinfection	sodium tetraborate decahydrate	Wheeler, et al.	2010	dairy	No
	hydrogen peroxide	Wheeler, et al.	2010	dairy	No
	oxychlorine solution	Wheeler, et al.	2010	dairy	No
Essential oil	carvacrol and pinene	Wheeler, et al.	2010	dairy	No
	eugenol	Wheeler, et al.	2010	dairy	No
	glycerol	Wheeler, et al.	2010	dairy	No
	basil	Wheeler, et al.	2010	dairy	No
	peppermint black mitchium	Wheeler, et al.	2010	dairy	No
	hyssopus oil	Wheeler, et al.	2010	dairy	No
Humate	ManureMax	Shah, et al.	2012	swine	No
Microbial digestion	aerobic/facultative microbes	Wheeler, et al.	2010	dairy	No
	mixture of chemicals and surfactants for facultative bacteria	Wheeler, et al.	2010	dairy	No
	aerobic/facultative microbes with growth factors	Wheeler, et al.	2010	dairy	No
	aerobic microorganism	Mao, et al.	2018	pig	No
	facultative microorganisms	Mao, et al.	2018	pig	No
Other chemical	Stalosan	Martinez, et al.	2003	pig	No
	NX23	Martinez, et al.	2003	pig	No
Oxidizing agent	mixture chemicals/micronutrient concentrate	Wheeler, et al.	2010	dairy	No
	mixture of chemicals in isopropyl alcohol	Wheeler, et al.	2010	dairy	No
	mixture of chemicals	Wheeler, et al.	2010	dairy	No
	complex triazine mixture	Wheeler, et al.	2010	dairy	No
	Abandoned mine drainage	Wheeler, et al.	2010	dairy	No
	dipole dibase formulation	Wheeler, et al.	2010	dairy	No
Physical agent	sand	Hao, et al.	2005	cattle	No
Straw	N/A	Yamulki	2006	cattle	Yes
	barley straw	Sommer, et al.	2000	pig	No
		Chowdhury, et al.	2014	animal	No