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Sustainability of the Dairy Industry: Emissions and Mitigation Opportunities

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Dairy cattle provide a major benefit to the world through upcycling human inedible feedstuffs into milk and associated dairy products. However, as beneficial as this process has become, it is not without potential negatives. Dairy cattle are a source of greenhouse gases through enteric and waste fermentation as well as excreting nitrogen emissions through their feces and urine. However, these negative impacts vary widely due to how and what these animals are fed. In addition, there are many promising opportunities for further reducing emissions through feed and waste additives. The present review aims to further expand on where the industry is today and the potential avenues for improvement. This area of research is still not complete and additional information is required to further improve our dairy systems impact on sustainable animal products.

Keywords: cows, sustainability, greenhouse gases, methane, ammonia, enteric emissions, waste emissions

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INTRODUCTION

Dairy production is considered a major societal asset globally due to its economic and nutritional benefits. In 2019 alone global milk production totaled 851.8 million tons in milk equivalents (Outlook, 2020). This contributes to substantial trade impacts, totaling about 76.7 million tons in 2019, as well as major per capita consumption at about 111.4 kg/year globally (Outlook, 2020). There are over 245 million dairy cows worldwide that on average produce 2,300 kg per year; although average production is less informative as there is such a major disparity between production in different countries (FAO, 2009). This vast amount of milk production has a major global benefit—for human health, society, and the economy. In countries with developing economies livestock serve many purposes including: a source of household income, a financial asset for women, a source of food security, risk management, and a direct link to human health (Herrero et al., 2013). These benefits increase substantially when viewed from a macro lens. Global dairy imports totaled over $42.2 billion in 2014 and global dairy exports expanded 175% from 2005 to 2014 (Davis and Hahn, 2016). There are many dairy commodities being produced and traded as part of these exports with whole milk powder being the highest, followed by skim milk powder, butter, and cheese (Outlook, 2020). India is the largest dairy producer globally, with 22% of global production and 52,841,810 total dairy cattle, the US is second in production, followed by China, Pakistan, and Brazil (FAO, 1997; Knips, 2005). While the US may not be the largest global milk producer, the total economic benefit of the dairy industry is substantial, totaling $628.27 billion dollars in 2018 (O'Keefe, 2018). There are currently 9,336,000 dairy cows in the U.S. that on average produce 10,610 kg of milk each year, which amassed to over 99,056,409 kg of milk in 2019 (USDA, 2020). The US dairy sector also generates over 2.9 million jobs either through direct or indirect support (O'Keefe, 2018). The top five milk producing states in the US are California, Wisconsin, Idaho, New York, and Texas, with California accounting for nearly 20% of national production (Sumner and Matthews, 2019; USDA, 2020). The dairy industry is such a major part of California's economy that in 2019 the associated impact from milk production and processing was about \$57.7 billion dollars, providing over 179,900 jobs (Sumner and Matthews, 2019). Dairy is the leading agricultural commodity produced in California, accounting for nearly 13% of the \$49.9 billion dollars in cash receipts generated for the top ranked agriculture producing state (CDFA, 2019). Not only is the dairy industry a major driver of the economy but its products serve a substantial nutritional benefit to the growing human population.

Milk and dairy products are a well-known source of calcium, vitamins, and other selected minerals as well as being a complete high-quality protein. One of the most well-documented nutritional benefits of dairy products is for bone health—particularly for its ability to prevent osteoporosis and other bone diseases. In particular the calcium in milk positively affects bone mass in children and when coupled with vitamin D, as seen in fortified milk products, will prevent bone loss and osteoporotic fractures in aging populations (Caroli et al., 2011). For low-income countries struggling with nutritional deficiencies in children, studies have shown that supplementation with dairy products causes a significant increase in vitamin B-12 plasma concentrations, improves cognition, growth and activity (Allen, 2003; Siekmann et al., 2003). In addition, maternal milk intake during pregnancy is positively associated with infant birth weight, and subsequent bone mineral content during childhood (Gil and Ortega, 2019). Other milk components, including bioactive peptides present in the whey components of milk were shown to benefit the immune system due to their antimicrobial and immunomodulatory properties (Madureira et al., 2010). Consumption of dairy products has shown an inverse relationship with cardiovascular disease in that consumption of milk and dairy is associated with a lower incidence of type-2 diabetes and improvements in glucose homeostasis (Hirahatake et al., 2014). While milk is relatively high in saturated fat it has been shown that milk intake did not increase cardiovascular risk (Visioli and Strata, 2014). Furthermore, milk intake was associated with reduced risk of childhood obesity as well as improved body composition and weight loss in adults (Thorning et al., 2016). Dairy intake was also shown to be inversely associated with incidence of cancer including colorectal, bladder, gastric, and breast cancer and was not shown to be associated with any other additional forms of cancer (Thorning et al., 2016). Although dairy production serves many benefits to overall nutrition, human health, and the economy, there has been increasing concern about the impact of dairy on the environment.

Impact of Dairies on Climate Change and Air Quality

The earth's surface has undergone massive increases in temperature, primarily in the last three decades, and the last 30 years we have seen the warmest period ever recorded (IPCC, 2014). In addition to this temperature increase, there have been other major changes to the climate including trends of increasing ocean temperature, rising sea level, as well as a major increase in greenhouse gas emissions (IPCC, 2014). Another phenomenon that has occurred over the last few centuries is an increase in ocean uptake of CO_2, causing ocean acidification and a decrease in surface water pH, as well as a rapid decrease in glaciers and ice sheets around the globe. These major changes in the climate are primarily due to anthropogenic (human caused) emissions of GHGs that have steadily increased since the beginning of the industrial revolution in the 1750s (Place and Mitloehner, 2010). Atmospheric concentrations of CO_2, CH_4, and N_2O are also the highest they have been in at least the last 800,000 years, with about 78% of these CO_2 emissions resulting from industrial processes and the combustion of fossil fuels (IPCC, 2014). Several studies have indicated that the production of livestock, including the stages of growing, transport, processing, and consumption have a relatively large impact on climate change (de Vries and de Boer, 2010; Milani et al., 2011). Dairy cattle in particular were shown to impact the environment through their potential negative contributions to air, water, and land (Naranjo et al., 2020).

In regards to the environment, the US Dairy industry has seen substantial improvements over the years. In particular it has seen a great increase in milk production primarily due to dramatic increases in milk production per cow, increase in average cow numbers per farm, as well as an overall decrease in total animal numbers (Wolf, 2003; Barkema et al., 2015). Some other major changes over the last 50 years include a shift to a primarily Holstein dairy herd (90%), an increased heifer growth rate, decreased age at first calving, and an increase in the use of artificial insemination (Capper et al., 2009). Nutrition of dairy animals has also allowed for a substantial improvement in production via use of total mixed-rations balanced for nutrient and energy requirements accounting for each animals age and stage of lactation (National Research Council, 2001). Genetic selection has also been a major driver in increased productivity, longevity, and efficiency of dairy cows, further reducing the environmental impact per unit of milk production (Pryce and Haile-Mariam, 2020). These improvements in nutrition and genetics, in conjunction with improvements to herd management, accomplished primarily through increasing density on dairy farms, have resulted in a fourfold increase in milk yield from the mid-1940s until 2007 (Von Keyserlingk et al., 2013). This efficiency of milk production has continued to improve to 2014 where 1 kg of energy and protein corrected milk (ECM) for California emitted between 1.12 and 1.16 kg of CO_2 equivalents (CO_2e) in 2014 compared with 2.11 kg of CO_2e in 1964, resulting in a 45% reduction in CO_2e (Naranjo et al., 2020). The dairy industry has continued to still further these improvements. Dairy production systems in 2017 compared with 2007 have reduced their inputs by 25.2% for animal numbers, 17.3% for total feed, 20.8% for land, and 30.5% for water of one million metric ton of energy-corrected milk, furthering the exceptional productivity gains and environmental progress of the industry (Capper and Cady, 2020). Even with these major advancements made over the last century, dairy systems still impact the environment through: GHG emissions from enteric fermentation, manure management, and feed production, water use for feed production and milk processing, water quality with contaminants including nitrogen (N) and phosphorous (P) from manure, as well as the requirement for land used in feed production (Naranjo et al., 2020). In addition to the direct impacts of cattle, such as N and P as a result of dairy production systems, there are also environmental impacts associated with dairy processing and subsequent production (Milani et al., 2011).

Manure Emissions From Dairy Cattle

Dairy manure has the potential to negatively impact the environment. Nitrogen not retained by the animal or secreted in milk will be excreted in the urine and feces of the animal (Hristov et al., 2019). Urine is more susceptible to losses of N to the environment from the animal waste as compared with fecal N (Dijkstra et al., 2013, 2018a). Dairy waste is a significant source of N and P that when land applied in excess of crop requirements can cause contamination of surface water (Knowlton and Cobb, 2006). This excess N and P in water causes a rapid bloom in the growth of algal populations that consume dissolved oxygen in water, termed eutrophication, which reduces the available dissolved oxygen required for growth of aquatic animal life (Knowlton and Cobb, 2006). Excess N can also contaminate ground water through leaching. This poses a problem for human and animal health as consumed nitrate from drinking water is converted to nitrite in the digestive tract, which replaces oxygen in hemoglobin and leads to cyanosis (oxygen starvation) (Knowlton and Cobb, 2006).

Air quality also affects human and animal health as well as the environment, and dairy cattle have been known to contribute to poor air quality. One such compound that affects air quality produced by dairy cattle is NH_3. Ammonia is produced when N in urea from the animal's urine reacts with urease present in feces (Place and Mitloehner, 2010). Ammonia production from dairy waste is dependent on a variety of factors including: urea content in urine, pH, and temperature, as well as the enzymatic activity of urease (Muck, 1982; Sun et al., 2008). In addition to NH_3 losses from fresh waste, volatilization can occur during waste application to soil as a fertilizer, as well as during the long term housing and storage of manure (Bussink and Oenema, 1998). Total losses of NH_3 can be between 0.82 and 250 g NH_3/cow/day, with the total loss dependent on the amount and composition of animal waste as well as the environment and management conditions of the manure storage (Bussink and Oenema, 1998; Hristov et al., 2011). Dairy waste management strategies greatly influence air emissions of NH_3. The greatest NH_3 emissions occur after field application, followed by the manure management strategies, for example, separated liquid and solids, aerated, straw covered, untreated, then anaerobic digested (Amon et al., 2006).

Nitrogen in waste can also contribute to GHG production through the formation and volatilization of nitrous oxide (N_2O). Nitrous oxide is created during incomplete microbial denitrification process where nitrate is converted to N gas with the potential to create N_2O, an extremely volatile byproduct (Place and Mitloehner, 2010). Land applied dairy manure on cropland as well as the long term storage of manure in lagoons can contribute to emissions of N_2O (Velthof et al., 1998; Place and Mitloehner, 2010). The N_2O emissions during storage depend on the N and carbon content of the manure (Amon et al., 2006). Nitrous oxide production and subsequent volatilization is also dependent on environment and management. Higher temperatures as well as surface coverings contribute to increasing emissions, whereas anaerobic conditions, such as those found in lagoon systems, have lower N_2O emissions (Dustan, 2002). The process of long term storage of manure seems to also contribute a larger proportion of N_2O emissions compared with land application with aerated, straw covered, digested, separated, and untreated manure contributing decreasing amounts of N_2O emissions (Amon et al., 2006).

Another substantial GHG produced by dairy cattle waste is methane (CH_4). The amount of CH_4 emitted by dairy waste is dependent on the amount of carbon, hydrogen, and oxygen present in the waste, making manure storage, diet, and bedding major contributors to total CH_4 production (Place and Mitloehner, 2010). A smaller proportion of CH_4 is also produced in the hindgut of the animal via post ruminal digestion and fermentation (Ellis et al., 2008). This CH_4 is mostly absorbed from the hindgut (89%) and eventually eructated by the animal or excreted with the manure (11%) (Murray et al., 1976; Immig, 1996; de la Fuente et al., 2019). Manure CH_4 emissions are substantially higher from long term storage compared with field application (Amon et al., 2006). These emissions are highest from straw covered manure and emissions decrease with untreated manure, followed by separation, aeration, and digested manure management methods (Amon et al., 2006).

Dairy waste can also produce volatile organic compounds (VOC). Volatile organic compounds are a class of chemicals that when reacted with oxides of N and sunlight contribute to ozone formation (Place and Mitloehner, 2010). There were 73 detectable VOCs from slurry wastewater lagoons with the most common VOCs being methanol, acetone, propanal, and dimethylsulfide (Filipy et al., 2006; Shaw et al., 2007). As with other waste emissions, VOCs from dairy waste increase with ambient air temperature with summer months having the highest rates of VOC emissions (Filipy et al., 2006). The largest contribution of VOCs on dairy systems come from fermented feedstuffs (i.e., silage) (Place and Mitloehner, 2010).

Effect of Nutrition on Emissions From Dairy Cattle

Dairy cattle enteric emissions have been shown to contain a variety of gases. For example dairy cattle emit CO_2 as a byproduct of aerobic cellular respiration, which is the GHG with the greatest contribution to climate change (Place and Mitloehner, 2010). However, this gas is not considered a net contributor to the rise in GHGs due to the CO_2 having been previously recycled from the atmosphere by fixation during photosynthesis in plants, which are then consumed by the cattle (Steinfeld et al., 2006). Dairy cattle can also produce N_2O from enteric emissions as a result of the NO_3 reduction process that takes place by the microbes in the rumen (Kaspar and Tiedje, 1981). Due to the small production of enteric N_2O, these emissions are not always considered in dairy emission analyses (Casey and Holden, 2005).

The most significant enteric emission compound from dairy cattle is CH_4. Methane acts as a hydrogen sink in the rumen and is an end product of CO_2 reduction by methanogenic archaea (Janssen and Kirs, 2008). Methanogens serve an important role in rumen health by removing this hydrogen that can be toxic to some bacterial communities and also causes the disease state rumen acidosis (Beauchemin et al., 2009). In addition to being a potent GHG, CH_4 also accounts for a 2–12% loss of potential energy available to the animal that could otherwise be used for maintenance and productive purposes as growth gestation, or lactation (Moe and Tyrrell, 1979).

Dairy cattle diets have a significant impact on enteric emissions, mostly CH_4. As there is large variability in the ingredient and chemical composition of diets fed to dairy cattle, nutrition and feeding strategies have the greatest potential for reducing CH_4 emissions, with potential reported reductions between 2.5 and 15% (Knapp et al., 2014). The amount of CH_4 produced is dependent on many factors including intake and chemical composition of the carbohydrate, retention time of feed in the rumen, rate of fermentation of different feedstuffs, as well as the rate of methanogenesis (Beauchemin et al., 2009). Altering feed digestibility and chemical composition cause a shift in the proportions of volatile fatty acids (VFA) with the predominant VFAs being propionate, butyrate, and acetate (Knapp et al., 2014). This shift in VFA proportion is important because propionate also acts as a hydrogen sink so shifting from acetate and butyrate formation to propionate will consume reducing equivalents and help preserve the pH balance in the rumen (Hungate, 2013). An overall reduction in CH_4 emissions or a shift in VFAs can be accomplished through a variety of altered feeding strategies. More energy dense or more digestible feedstuffs result in additional energy available to the animal and generate less CH_4 from fermentation (Knapp et al., 2014). An increase in starch proportion of the diet, such as through an increase in concentrate levels, also results in a more rapid fermentation of these feedstuffs and therefore decreased CH_4 production (Moe and Tyrrell, 1979; Johnson and Johnson, 1995). Feeding higher starch diets requires increased grain production, which can cause additional consumption of fossil fuel and fertilizers that results in an increase in N_2O and CO_2; however, this system is usually offset by the substantial decrease in overall in CH_4 emissions (Johnson et al., 2002; Lovett et al., 2006). Feeding of cereal forages can also favor propionate production and reduce CH_4 emissions due to the higher starch concentration (Beauchemin et al., 2009). Higher concentrations of legumes, such as alfalfa, when compared with grass forage based diets can also lead to an overall decrease in CH_4 emissions (McCaughey et al., 1999). Age of harvest of forage also has a significant impact on emissions, with advancing maturity resulting in more lignified and less fermentable substrate contributing to increasing emissions associated with higher ruminal acetate (Pinares-Patiño et al., 2003). In addition to alterations in forage or concentrate composition and ratio, supplementation of lipids to dairy cattle diets can also mitigate enteric emissions (Hristov et al., 2013b). Replacing concentrates with lipids results in a decrease in fermentable substrate by the microbes in the rumen and can also decrease total protozoa and methanogen populations (Ivan et al., 2004). An inclusion of high-oil by-products, such as distillers grains or oilseed meals, can result in decreased CH_4 emissions (Hristov et al., 2013b). Research on ensiled feeds in relation to enteric emissions is generally lacking, although it is anticipated that corn silage will mitigate emissions due to its higher starch content (Gerber et al., 2013). Furthermore, when directly comparing grass-versus corn silage, a higher inclusion of corn silage seems to mitigate enteric CH_4 emissions (Mills et al., 2008; Doreau et al., 2012). There are many potential methods to mitigate enteric emissions through alterations to nutrition strategy and composition.

Manure emissions are also significantly impacted by various dairy cattle feeding strategies. One of the main issues with altering feeding strategies to reduce enteric emissions is that fermentable substrate in the manure can increase, as has been seen with increasing the concentrate to forage ratio in the diet (Hindrichsen et al., 2006; Beauchemin et al., 2009). This response has also been seen with the supplementation of certain fatty acids (Kreuzer and Hindrichsen, 2006). To alleviate this issue, feeding concentrate with higher lignified fiber has been shown to mitigate both enteric and manure-derived emissions (Kreuzer and Hindrichsen, 2006; Aguerre et al., 2012). These changes to concentrate ratio do not have an impact on N containing manure emissions, as would be expected (Hindrichsen et al., 2006; Aguerre et al., 2012). The greatest impact of diet on waste emissions can be seen when feeding low crude protein (CP) diets to dairy animals, which results in decreased excreted N and subsequent NH_3 volatilization (Cardenas et al., 2007; Lee et al., 2012; Edouard et al., 2019). Comparing fresh grass with prepared hay at the same CP content, feeding hay causes a higher overall N and C/N ratio excreted but waste from grass fed animals tends to volatilize more NH_3 emissions (Külling et al., 2003). Corn silage inclusion in diets has also caused changes to manure emission profiles. For example when comparing corn silage versus grass silage, corn silage tended to reduce urinary N excretion (Mills et al., 2008). When adding corn silage to alfalfa silage based diets there is also an improvement in N efficiency leading to a decrease in N losses in urine and subsequent decreases in available NH_3 and N_2O volatilization (Gerber et al., 2013). Higher sugar forages also reduce N excretions, which also have the potential to limit the N available to be volatilized as gaseous emissions (Miller et al., 2001; Parsons et al., 2012; Gerber et al., 2013). Overall a variety of feeding strategies can be employed to help mitigate emissions from enteric and waste sources of dairy animals.

Mitigation Strategies for Dairy Cow Enteric Gas Emissions

In addition to changes to the diet ingredient composition, there are also additives to diets that may mitigate enteric emissions. While there are various types of strategies to alter enteric sourced emissions this section will focus primarily on methods to alter CH_4. One promising strategies for CH_4 reduction is via feed supplementation of the methanogenic inhibitor, 3-Nitrooxypropanol (3-NOP). 3-Nitrooxypropanol is a structural analog to methyl-coenzyme M, which acts on methyl-coenzyme M reductase (MCR), a nickel enzyme involved in the final reduction stages of methanogenesis (Duin et al., 2016). In the rumen system 3-NOP was shown to mimic methyl-coenzyme M and target the active site of MCR, thus inhibiting the enzymes activity and subsequently causing a decrease in CH_4 production (Duin et al., 2016). Research demonstrated that feeding 3-NOP to cattle decreased enteric CH_4 emissions up to 95% *in vitro* (Martínez-Fernández et al., 2014) and 84% *in vivo* (Vyas et al., 2016). 3-NOP was tested *in vivo* in multiple ruminant models including sheep (Martínez-Fernández et al., 2014), beef cattle (Romero-Perez et al., 2015; Vyas et al., 2016), as well as Holstein dairy cattle (Reynolds et al., 2014; Hristov et al., 2015; Lopes et al., 2016; Haisan et al., 2017). Reynolds et al. (2014) fed 3-NOP at a rate of either 500 or 2,500 mg/d via rumen fistula before each feeding and using respiration calorimetry found a reduction of 6.6 and 9.8% in CH_4 emissions, respectively. They also found a decrease in dry matter intake (DMI) and an increase in milk protein at the higher dose, without other changes in production parameters. Haisan et al. (2017) also fed 2,500 mg/d and using the SF6 systems measured a reduction in emissions from 17.8 to 7.18 g/kg of DMI without adverse effects to milk or DMI. Hristov et al., 2015 fed 3-NOP at a rate of 40, 60, or 80 mg/kg of DMI and measured reductions via a GreenFeed system of 25, 31, and 32%, respectively. They also found no changes to DMI or milk production with an increase in protein yield following supplementation. Similarly, Lopes et al. (2016) also fed 60 mg/kg of DMI and found a 31% decrease in emissions with an increase in milk fat concentration. Dijkstra et al. (2018b) evaluated the overall efficacy of 3-NOP in research trials and determined that greater 3-NOP dose results in a greater reduction of CH_4 emissions. These trials also used different diets, which did not seem to effect the impact of 3-NOP on emissions. However, this molecule has yet to be evaluated for its efficacy among different dairy breeds and the potential side effects of its use have not fully elucidated. Additionally, it has yet to be determined whether 3-NOP has any unintended consequences of carryover to the excreta of supplemented animals.

Nitrates offer great promise for their potential to mitigate CH_4 and have been well studied for their use in beef cattle diets with more recent literature focusing on the potential for use in dairy cattle. Nitrate in the diet serves as a non-protein N source that acts as an electron receptor resulting in effective and consistent reduction of enteric emissions. However, nitrate has the potential to induce methemoglobinaemia and is a known carcinogen (Lee and Beauchemin, 2014). Nitrate toxicity can generally be avoided when the rumen ecosystem is allowed time to adapt (Hristov et al., 2013b). Even with the potential for toxicity, the benefits of 16–50% reduction in CH_4 emissions continue to drive research feeding nitrates (Leng and Preston, 2010). Van Zijderveld et al. fed nitrate at a rate of 21 g/kg DMI and measured a persistent reduction in CH_4 of 16% via use of open-circuit indirect calorimetry chambers (Van Zijderveld et al., 2011). They did not measure changes in milk yield or DMI for the supplemented animals. Similar research conducted by other authors found a reduction in emissions of CH_4/d from 363 g for control animals to 263 g for nitrate supplemented animals also at 21 g/kg DMI (Klop et al., 2016). They also measured a reduction in milk protein concentration as well as DMI for nitrate-supplemented cows. When high levels of nitrate (20 g/kg DMI) were supplemented in a similar study design they found a 31% reduction in CH_4 along with a decrease in DMI during nitrate feeding (Lund et al., 2014). Another study found a 28% decrease in methanogenesis after feeding nitrate at 2.3% of DM to nonlactating cows, however they also found a significant decrease in feed intake from the supplemented animals (Guyader et al., 2015). Another trial reported a 10% decrease in DMI, coupled with a 17% decrease in CH_4 where dairy cattle diets were supplemented with nitrate at 1.5% of DMI (Meller et al., 2019). A meta-analysis found a persistent reduction in CH_4 emissions in both *in vitro* and *in vivo* studies (Lee and Beauchemin, 2014). Similar to 3-NOP, TMR composition did not seem to have a major effect on nitrate supplementation as these studies all saw a significant decrease in emissions with vastly different diets.

Plant biological compounds have also been explored for their potential to reduce emissions. Condensed tannins are secondary phenolic compounds that generally discourage consumption by herbivories and also concentrate N in the plant (Waghorn, 2008). When consumed by dairy cattle these tannins bind protein in the rumen, which reduces the degradation of protein and enhances protein flow to the intestines (Beauchemin et al., 2009). Tannin source appeared to make a major difference in subsequent mitigation of CH_4 emissions from dairy cattle. For example, the *Hedysarum coronarium* species supplemented at 27 g /kg DMI resulted in lower CH_4 emissions by dairy cattle (Woodward et al., 2002). Whereas, *Schinopsis quebracho-colorado* supplemented at 0, 1, or 2% of dietary DM did not have any effect on enteric emissions or dry matter intake of beef cattle (Beauchemin et al., 2007). Additional studies looking at *Lotus pedunculatus* (fed at 10% of dry matter) and *Medicago sativa* (fed at 0.1% of dry matter) tannin supplementation found decreased CH_4 emissions from both strains of condensed tannins, although DMI was not measured, which were attributed to reducing hydrogen production and direct inhibition on methanogenic archaea (Tavendale et al., 2005). A meta-analysis identified a general anti-methanogenic effect of tannins across different sources and that the variation in methane reduction seen in previous studies may have been due to the low tannin levels used in those trials (Jayanegara et al., 2012). They also found that dietary tannins tended to increase DMI but decrease total tract digestibility, apparent CP digestibility, and neutral detergent fiber digestibility. As with previous feed supplementation, these trials did not quantify emission changes to waste sources. Additional research into tannins in various diets as well as its effect on milk production and manure CH_4 emissions needs to be explored.

In addition to tannins, secondary plant compounds called essential oils have been explored for their antimicrobial properties. Essential oils are naturally occurring volatile components in plants that provide the plant specific color and flavor characteristics (Benchaar et al., 2008). Essential oils reduced CH_4 production through inhibiting growth and energy metabolism of selected bacteria and archaea including methanogens (Benchaar et al., 2008). Over 250 essential oils have been identified and contain mixtures of terpenoids, a variety of low molecular weight aliphatic hydrocarbons, alcohols, acids, aldehydes, acrylic esters, N, sulfur, coumarins, and homologs of phenylpropanoids (Beauchemin et al., 2009). These essential oils underwent *in vitro* screening for their potential to reduce rumen CH_4 emissions and while 35 were found to be effective only six were found to have significant decreases in emissions without disrupting digestibility (Bodas et al., 2008). It is difficult to directly compare essential oils because of the number of different compounds as well as the difference in study design and species studied. In addition, few essential oils have been thoroughly evaluated *in vivo*. Benchaar and Greathead (2011) performed additional *in vitro* testing and found decreased CH_4 production following supplementation with oregano, rhubarb, thyme, cinnamon, horse radish, frangula, and garlic. Tekippe et al. (2011) fed oregano leaf at a rate of 500 g/d to lactating dairy cattle and measured rumen CH_4 production 8 h after feeding. They found a decrease in total CH_4 yield but did not see adverse effects on DMI or milk yield with the added benefit of increased milk fat content. In a follow up study by Hristov et al. (2013a) they fed lactating dairy cows 250, 500, and 750 g of oregano leaf per day and found a linear reduction in methane per unit of DMI coupled with a linear decrease in DMI but no differences in any milk production parameters. In addition to particular isolates of essential oils, there are also commercial essential oil blends being marketed for their potential to reduce enteric CH_4. One essential oil blend is Agolin SA created in Bière, Switzerland that is comprised of coriander oil, geranyl acetate, and eugenol. Agolin was tested *in vitro* and found a significant initial decrease in rumen $CH_{4,}$ but the effect did not persist over time (Klop et al., 2017b). Another Agolin *in vitro* trial found similar results where there was an initial reduction in methane, but the effect was not constant over the total 72 h incubation period (Castro-Montoya et al., 2015). These authors also conducted feeding trials with the Agolin essential oil product. Castro-Montoya et al. (2015) found a trend in reduction of daily emissions relative to intake and (Klop et al., 2017a) found initial decrease in CH_4/DMI only for the first 2 weeks of feeding Agolin, after which Agolin did not impact CH_4. In addition, Klop et al. (2017a) reported a decrease in DMI over the second half of the supplementation period. Hart et al. (2019) also supplemented lactating dairy cattle with Agolin essential oils and measured a reduction in CH_4 emissions per pen. Changes to DMI, milk production, or fat composition after feeding of essential oils have also been reported following Agolin supplementation. For example, Santos et al., 2010 reported numerically lower DMI with an increase in milk fat production, a 0.03 kg/day increase in fat production, from Agolin supplemented cows, whereas Elcoso et al. (2019) saw an increase in ECM supplemented animals without differences in DMI. However, for Santos et al. (2010) the Agolin treatment was applied to the pen and not the individual animal. Elcoso et al. (2019) also estimated rumen CH_4 production from fermented rumen fluid and found supplemented animals to be lower, but there was an interaction between the time and treatment. Hart et al. (2019) also found a greater milk yield and ECM for Agolin supplemented animals. Clearly the large discrepancy in responses across research studies for Agolin emphasizes the need for additional research to determine if the essential oil product has application at the farm level to reduce enteric CH_4 emissions.

Mitigation Strategies for Dairy Cow Manure Gas Emissions

While there are many ways in which to alter manure emissions depending on the desired outcome this literature search will focus on methods to alter CH_4 emissions specifically, of which there are quite a few promising strategies. One manure amendment strategy includes the use of biochar. Biochar is a general term applied to products produced by thermal decomposition from a variety of biomass substrates for agricultural applications including the added benefit of optimizing the process of composting (Godlewska et al., 2017). Biochar was shown to have a multitude of benefits including improving the overall process of composting, improving N conservation, facilitating nutrient transformation, and favoring oxygen supply (Vandecasteele et al., 2016; Chen et al., 2018; Mao et al., 2018). Other studies demonstrated that biochar improved soil physicochemical properties, benefited nutrient conservation as well as boosted crop production (Li et al., 2015; Mao et al., 2017; Wu et al., 2017). While the benefits of biochar as amendments to poultry and pig manure have been well-documented (Agyarko-Mintah et al., 2017; Chen et al., 2017; He et al., 2019), its use in dairy cattle manure management has been less thoroughly studied. Jindo et al. (2012) added biochar to cattle manure to measure microbial communities, causing a significant increase in the C/N ratio from the additional of high carbon biochar materials, but they did not measure emissions from these systems. Duan et al. (2019) applied wood or wheat straw biochar with and without bacterial supplementation to cattle manure compost. While this study did not measure CH_4 emissions specifically, they found that biochar in addition to bacterial amendments enhanced the compost overall and that *Bacteroidales*, *Flavobacteriales*, and *Bacilli* were the communities with the highest abundance in the samples. Awasthi et al. (2020) also tested biochar with and without a bacterial inoculum applied to fresh cattle manure in a reactor and found treatments with the inclusion of biochar produced substantially less CH_4 as compared with the control. Overall, the impact and mechanism of action of biochar on CH_4 emissions from dairy waste specifically deserves further study.

Bacterial inoculums, as well as the supplementation of bacterial produced enzymes, have been well-researched in the literature for their potential to alter CH_4 emissions. Bacteria are involved in many of the breakdown processes that occur in manure management systems including reactions of hydrolysis, acidogenesis, acetogenesis, and methanogenesis, the latter of which has the potential to increase methane production (Juodeikiene et al., 2017). While increased CH_4 may seem in conflict with the present literature review, this manure management strategy can be applied to systems where CH_4 can be captured and transformed into biofuel or other renewable resources. One such example is through anaerobic digestion in which organic material is degraded by microbes in the absence of oxygen (Rodriguez Chiang, 2011). A variety of bacterial communities have been researched for their potential to change

CH_4 emissions from various substrates. Juodeikiene et al. studied *Lactobacillus delbrüeckii*, and found an increase in methane of 76% from dairy wastewater from milk processing, as compared with 38% without the addition of bacteria (Juodeikiene et al., 2017). Xu et al. pretreated corn straw with *Bacillus subtilis*, and increased CH_4 production 17.35% above the untreated control (Xu et al., 2018). He et al. also supplemented microalgal biomass with *Bacillus licheniformis*, and bacterial supplementation increased CH_4 production from 9.2 to 22.7% (He et al., 2016). Commercial products have also been marketed for their application in manure management systems, including BiOWiSH products. BiOWiSH products contain a proprietary mixture of *Bacillus* and *Lactobacillus*, including *Bacillus subtilis, Bacillus amyloliquefaciens, Bacillus licheniformis, Bacillus pumilus, Pediococcus acidilactici, Pediococcus pentosaceus, Lactobacillus plantarum, Bacillus meagerium, Bacillus coagulans,* and *Paenibacillus polymyxia*, a product covered by a patent outlined by Carpenter et al. (2014). The process to create BiOWiSH involves individually fermenting each organism, followed by harvesting and then drying of each organism. Finally, the dried organism ground to produce a powder with a final moisture content <5% and a final bacterial concentration between 10^5 and 10^{11} colony forming units (CFU) per gram of dried product. While these products have not been evaluated for their effect on CH_4 specifically, these products claim to digest sludge, and reduce biological oxygen demand, total suspended solids, total Kjeldahl N, and odor from manure lagoons. The BiOWiSH product has been applied to dairy waste systems and showed promise for manure mitigation including: a reduction in total suspended solids and a degradation and removal of *N* (Lee, 2012; Pal, 2012; Holland, 2017). However, BiOWiSH has not been studied with respect to its effects on CH_4 emissions from dairy wastewater systems.

Gypsum based products have been applied to dairy waste systems for manure amendments. One of the more common forms of gypsum used for manure amendment is flue gas desulphurization gypsum that is a by-product of wet gas desulphurization from coal-fired power stations (Febrisiantosa et al., 2018). This gypsum has a low heavy metal content and contains high concentrations of S, Si, and Ca that are essential minerals nutrients required by plants (Guo et al., 2016). Gypsum has been fairly well-characterized for its effects on N containing compounds. Tubail et al. found gypsum supplemented dairy manure lost significantly less N as compared with the control dairy manure without supplementation (Tubail et al., 2008). Li et al. applied gypsum to pig manure compost and found significant reductions in NH_3 and enhanced mineral and total N contents (Li et al., 2018). Hao et al. applied gypsum to beef cattle manure and found a significant reduction in CH_4 emissions from the medium and high doses of gypsum as compared with the control (Hao et al., 2005). Yang et al. studied kitchen waste compost and found the addition of gypsum to dramatically reduce CH_4 emissions by 85.8% (Yang et al., 2015). While these study designs don't quite have the same application as is intended in this literature review, the potential for use of gypsum as a manure amendment is promising. There are also commercial additives being marketed for their potential to mitigate CH_4 emissions, including SOP Srl, a company that makes the SOP Lagoon products. SOP Lagoon consists of calcium sulfate dihydrate (agricultural gypsum) that is processed with the company's proprietary technology. The product's claim is to improve liquid manure management through inhibiting the production and release of GHGs (e.g., CH_4 and N_2O) and criteria pollutants (e.g., NH_3) while also reducing the odor intensity from liquid manure. Borgonovo et al. first tested the gypsum-based commercial additive, "SOP LAGOON," on fresh dairy manure and found the additive to be effective in reducing direct NH_3 and GHG emissions, including a significant mitigation of CH_4 emissions (Borgonovo et al., 2019). Recent literature by Peterson et al. applied SOP Lagoon to liquid stored dairy cattle manure over a 2 week period and found similar results including significant reductions in NH_3 emissions (22.7% for the supplemented systems as compared with an unsupplemented control) (Peterson et al., 2020). With the strong literature documenting the potential for gypsum to decrease CH_4 emissions, this seems like a viable manure amendment strategy.

In addition to the previously described additives, a variety of additional organic substrates have been applied as amendments in diverse applications. These additional amendments include lime and coal fly ash (Fang et al., 1999; Wong et al., 2009), zeolite (Awasthi et al., 2016; Chan et al., 2016), bentonite (Wang et al., 2016), clay (Chen et al., 2018), and medical stone (Awasthi et al., 2017; Wang et al., 2017), among others. These amendments require further research to evaluate their potential use in dairy manure specifically as well as the resulting CH_4 emissions after their application.

CONCLUSIONS

There is an increasing amount of literature and research data concerning strategies to further reduce livestock's impact on the environment. However, there is no one method of environmental sustainability in these systems and even still there are many unanswered questions. Future research needs to better quantify full reduction potential and elucidate the mechanism of actions of these compounds including 3-NOP, tannins, essential oils, bacterial inoculums, and biochar, among others. Furthermore, slight alterations to dairy cattle diets can cause major changes in both enteric and waste emissions. Research on mitigating the environmental impact of dairy cattle will allow dairy producers to contribute to a more sustainable dairy production system.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

CP wrote the manuscript draft. All authors contributed to the article and approved the submitted version.

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