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Article

Pesticides Used on Beef Cattle Feed Yards Are Aerially Transported into the Environment Via Particulate Matter

Eric M. Peterson, Frank B. Green, and Philip N. Smith*

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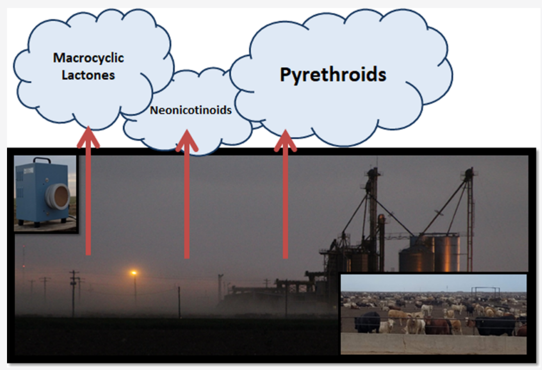
ABSTRACT: Considering the recent discovery of veterinary pharmaceutical aerial transport from industrial cattle feeding operations via particulate matter, the objective of this study is to determine the extent to which insecticides are also transported into the environment by total suspended particulates emanating from beef cattle feed yards. Of 16 different pesticides quantified in particulate matter samples collected from beef cattle feed yards, permethrin was detected most frequently at >67% of particulate matter samples and at a mean concentration of 1211.7 ± 781.0 (SE) ng/m^3. Imidacloprid was detected at a mean concentration of 62.8 ± 38.2 (SE) ng/m^3 or equivalent to published concentrations in dust from treated seed planting activities. When insecticide concentrations observed in this study are projected to all United States of America feed yards, the resulting particulate matter (669,000 kg) could contain enough insecticides (active ingredient mass basis) to kill over a billion honeybees daily. Furthermore, a novel transport pathway for macrocyclic lactone entry into the environment was identified. These data raise concern that nontarget organisms may be exposed to potentially toxic levels of pesticides from beef cattle feed yards.



■ INTRODUCTION

Global meat consumption and thus production have risen dramatically over the last 50 years.[1,2] From 1961 until 2014, beef production increased from nearly 25 million metric tons to over 61 million metric tons globally.[1] To meet the growing demands for beef, conventional small farms, often with free range cattle, are increasingly replaced by large scale confined animal feeding operations (feed yards).[3] While only 2% of global beef cattle (28.92 million head) are housed on feed yards, they account for 7% of the total beef production largely due to increased efficiency.[3,4]

The United States of America (USA) is the largest producer of beef in the world, generating 12 million metric tons annually.[1,5] The USA also contains most cattle confined to industrial feeding operations accounting for >40% of all cattle housed on feed yards globally.[3,4,6] Within the USA, the High Plains region (including Texas, Kansas, Oklahoma, Nebraska, and Colorado) has the highest density of industrial cattle feeding operations in the world with 9.1 million cattle (77% of all cattle on feed yards in the USA).[6] While feed yards are well established in the USA, Canada, and Australia, they are becoming more prevalent in Mexico, China, Paraguay, and Brazil.[3] For example, from 1990 to 2010, Brazil (the world's largest exporter of beef) saw a fourfold increase in beef produced on feed yards (0.8 million cattle in 1990 increasing to 4 million cattle in 2010).[7]

Feed yards house thousands of cattle in relatively small areas, approximately 20 sq. meters per head.[8,8] To maintain cattle health and maximize growth among high densities of animals, many countries (USA, Australia, South Africa, Japan, Mexico, New Zealand, and Chile) rely heavily on veterinary pharmaceuticals.[9−13] In these countries, more than 90% of all feed yards treat cattle with antibiotics,[12] 85% use β−agonists,[14] and over 80% use synthetic anabolic steroids.[12] Antibiotics, β−agonists, and melengestrol acetate (progesterone-like steroid) are administered to cattle via feed, whereas the primary route of administration for other growth-promoting steroids is slow release implant.[9,12] Following parenteral or enteral administration, cattle excrete a portion of metabolized and unmetabolized veterinary pharmaceuticals in urine and feces (>159 million kg dry mass of manure/day globally[15]), which can then be transported intentionally or unintentionally beyond feed yard boundaries.[16−18] In recent years, it has become clear that antibiotics, steroids, and other growth promoters consumed by, or administered to, beef cattle are transported via windblown particulate matter (PM) from the beef cattle feed yards on the High Plains of the USA.[16−19] However, no studies have examined the aerial transport of

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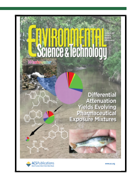

insecticides that are administered to cattle, applied topically to cattle or applied to feed yard surfaces (Table S4), despite the fact that they are used in substantially greater concentrations/volumes/masses than growth promoters and are used world-wide.[11,20,21]

The United States Department of Agriculture (USDA) estimated that insect pests cost the cattle industry $2.2 billion (USD) annually.[22] Stable flies (*Stomoxys calcitrans*), house flies (*Musca domestica*), and blow flies (*Calliphoridae spp.*) are considered to be the most important pest species to manage on feed yards throughout the world.[12,21,23,24] Taylor et al.[25] estimated that stable flies alone cost feed yards in the USA $226 million (USD) annually due to reduced cattle weight gain. A survey conducted among Australian feed yard owners revealed that 83% consider flies as major pests and 78% use chemical measures to control fly populations in an effort to reduce cattle production losses.[24] In addition to flies, other common feed yard pests include cattle grubs, scabies/mites, cattle lice, and ticks. In the USA, 99.8% of all feed yards apply insecticides directly to cattle and feed yard surfaces to combat these pests.[12] Insecticides used on feed yards include macrocyclic lactones, pyrethroids, organophosphates, neonicotinoids, and carbamates, which are administered via multiple routes (Table S4).[12,20,26] The two most commonly used insecticide classes in USA feed yards are macrocyclic lactones (88.6%) followed by pyrethroids (24%).[12] In addition to efficacious control of pests, these compounds are also highly toxic to nontarget invertebrates.[27−31] If transported beyond feed yard boundaries, both chemical classes may cause direct mortality or exacerbate stress among nontarget insects simultaneously exposed to other agrochemicals.

Aerial and spray boom pesticide application methods used in row crop agriculture can result in unintentional pesticide exposure among nontarget organisms; thus, they are regulated by product label restrictions and applicator controls.[32,33] In contrast, risks posed by aerial transport of pesticides and other agrochemicals emanating from beef cattle feed yards have not been adequately considered and are much less regulated.[34] Therefore, in an effort to better understand the risks posed by feed yard-derived pesticides, the objective of this study is to determine the extent to which insecticides used on beef cattle feed yards are transported via airborne PM to surrounding environments.

■ MATERIALS AND METHODS

Particulate Matter Collection. PM sample collection occurred in the panhandle of Texas (USA) from March until October 2019. No samples were collected in September due to frequent rainfall. Feed yards (n = 6; feed yards A−F) were identified and selected based on vehicular accessibility, and reference sites (n = 3; reference sites A−C) were situated more than 6.9 km from any feed yard to decrease the potential for cross contamination.[35] Each month, PM was collected from a minimum of five feed yards (except April, n = 4 due to weather) and two reference sites in the last 2 weeks of each month. There was a minimum of 3 weeks separating each PM collection event. Collection of PM occurred on days when there had been no precipitation during the previous 48 h and when the soil water content was less than 0.2 VWC (classified as dry to some moisture; West Texas Mesonet; The Campbell Scientific 615 Water Content Reflectometer[36]).

All PM collection events occurred between 1 h before sunset and 1 h past dusk to target optimal PM generation periods. Four Hi-Q CF-902 digital portable high-volume air samplers (HI-Q Environmental Products, San Diego, CA) were positioned 2−3 m above the ground downwind of feed yards (<0.5 km from feed yard pen and < 2 km from row crop boundary) or in excess of 6.9 km downwind from the nearest feed yard for reference sites (>1.2 km from row crop; Table S2). The total suspended PM was collected for 30 min onto preweighed (±0.1 mg) 10 cm glass fiber filters (CF-902; HI-Q Environmental Products, San Diego, CA). During PM collection, an additional filter was exposed to ambient air to serve as a sampling blank. After PM collection, the filters were placed in airtight tin containers, sealed in plastic bags, and placed on ice until transported back to the laboratory where filters were stored at −20 °C until analysis. PM was not collected upwind of feed yard locations due to sampling time constraints and because previous studies have documented infrequent detection of target analytes.[16,17,19] Distances from feed yards and row crop boundaries were determined using satellite imagery (± 1.0 m) from coordinates recorded at all PM collection sites.

Extraction of Pesticides from PM on Filters. Following PM collection, the filters were reweighed to determine the PM mass (± 0.1 mg). Pesticide (pyrethroids, neonicotinoids, macrocyclic lactones, organophosphates, synergists, phenylpyrazole, and fungicides; Table S3) extractions occurred less than 8 weeks from the PM collection date and included method blanks and laboratory matrix spikes for quality control (Table S3). Filters were placed in a 50 mL centrifuge tube and spiked with an internal standard tris (1-chloro-2-propyl) phosphate (TCPP). Next, 45 mL of methylene chloride/acetone (1:1) was added and sonicated for 1 h with heat (40 °C). Following sonication, the supernatant was poured off and filtered through a 0.45 μm polytetrafluoroethylene (PTFE) filter into a glass container. Next, 40 mL of acetonitrile/water (1:1) was added, and tubes were placed on an orbital shaker for 18 h at 350 rpm (Model MaxQ 4000, Thermo Scientific, Waltham, MA). After shaking, 50 mL tubes with sample filters were centrifuged for 10 min at 3100 rpm to facilitate decanting of the supernatant into a new 50 mL centrifuge tube with 4 g of magnesium sulfate and 1 g of sodium chloride. Tubes were then vortexed for 1 min, centrifuged for 10 min at 3100 rpm, and added to the supernatant extracted via sonication. Combined supernatants were then evaporated to dryness under nitrogen, reconstituted into 1 mL of acetonitrile, and filtered through a 0.2 μm PTFE filter. Extracts were then analyzed by triple quadrupole liquid chromatography tandem mass spectrometry with electrospray ionization (Thermo TSQ Quantum Access Max, Thermo Scientific, Waltham, MA) as described by Peterson et al.[37]

Statistics. Statistical analyses were performed using R (version 3.5.2; R Core Team, 2019) with RStudio for Windows (version 1.1.422; RStudio Team, 2016), using the "jmv" package (Friedman tests).[38] Nondetects or detections below the limit of quantitation (Table S3) were assigned a value of zero for statistical analysis. For all statistical comparisons, α was set at 0.05. Assumptions of normality were evaluated with the Shapiro−Wilk test and homoscedasticity with the Bartlett's test. PM concentrations were determined to be normally distributed and analyzed using the Pearson product−moment correlation analysis for the relationship between the PM concentration and the feed yard sampling site total pen area (Table S1). The nonparametric repeated measures analysis of variance (Friedman test) was

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used to determine the differences in PM concentration by sites with sampling events designated as the blocks for analysis.

Pesticide concentration data were found to violate the assumptions of normality, homoscedasticity, independence, and linearity even after multiple transformation adjustments. Thus, nonparametric tests were employed on the non-transformed pesticide concentration data set. Differences in the pesticide concentration between reference PM collection sites and feed yard PM collection sites were analyzed using the Friedman test, blocked by sampling events (Table S5).[38] Similarly, Friedman tests were used to examine the effects of site (reference vs feed yard), temperature, dew point, and relative humidity blocked by sampling events. Correlations between different pesticide concentrations were assessed with Spearman's rho. PM and pesticide concentrations described throughout this study are presented as mean ± standard error.

RESULTS AND DISCUSSION

PM Characterization in Air. The mean concentration of total PM collected per feed yard site (13.9 ± 1.9 mg/m^3) was similar to other sampling events conducted near feed yards in the USA and Australia.[17−19,42,43] The maximum concentration of PM in air during a single sampling event was 43.5 mg/m^3, which occurred in July. The highest monthly mean PM concentrations were collected in April (20.9 ± 5.5 mg/m^3) and July (20.6 ± 6.8 mg/m^3), whereas March produced the lowest mean PM concentration (4.5 ± 1.3 mg/m^3). The relatively lower concentration of PM collected in March was likely attributed to decreased temperatures and shorter day lengths than those in the summer months preventing drying of pen material and production of dust. Surprisingly, PM concentrations were not correlated with feed yard area (r = 0.133, p = 0.431), which may be due to differences in cattle activity levels, PM management practices, cattle densities, among other factors between feed yards. There were no differences in mean PM concentrations among different feed yard sampling locations ($\chi^2_{(6)}$ = 12.500, p = 0.052).

Pesticide Characterization in PM. All PM samples collected downwind of beef cattle feed yards contained at least one pesticide analyte (23 possible; Table S3) with boscalid, permethrin, and piperonyl butoxide detected at every feed yard sampling site. The highest numbers of detections (26 and 25 detections) occurred in June and July, whereas the fewest (16 and 13) were detected in March and April, respectively.

Throughout the duration of PM collection (all months combined), 9.5 ± 0.5 unique analytes per feed yard were identified above the limit of quantitation (LOQ data in Table S3). In comparison, the reference site PM contained 1.3 ± 0.4 unique detects per PM sampling event. Six analytes were detected relatively infrequently at reference sites; imidacloprid (17% of PM samples), azoxystrobin (8%), boscalid (33%), piperonyl butoxide (33%), bifenthrin (25%), and permethrin (17%). Additionally, the mean pesticide concentrations in PM samples collected at reference locations were, on average, 34 times lower than those at feed yard locations (Table S5). While infrequent and at relatively low concentrations, reference sites still contained at least one agrochemical in a majority of PM samples (75%). The most likely explanation for these detections is the long-range transport of agrochemicals through air currents and atmospheric transfer from agricultural fields.[44] Large particle sizes in PM fall out of suspension relatively quickly, whereas PM_{10} and $PM_{2.5}$ particle fractions may travel greater distances before settling out.[16,18,44] The generation and transport of PM are enhanced by the semiarid environment of the High Plains and frequent winds. Considering that the trans-Atlantic transport of PM from Africa to Florida is possible,[45] it is not unreasonable to assume that trace amounts of agrochemicals were detected in reference samples multiple kilometers from the source (e.g., row crops). Meteorological parameters (relative humidity, wind speed, temperature, and dew point) did not impact pesticide concentrations (temperature:$\chi^2_{(6)}$ = 11.946, p = 0.063; dew point: $\chi^2_{(5)}$ = 10.000, p = 0.075; relative humidity: $\chi^2_{(6)}$ = 12.000, p = 0.062). Dicrotophos, paraoxon-ethyl, malathion, fipronil, diazinon, and doramectin were not detected in PM collected from any location.

Pyrethroid Occurrence. Permethrin was the most frequently detected analyte in PM from feed yards (67.6%) and also at the highest mean concentration (1211.7 ± 781.0 ng/m^3; Table 1). Pyrethroids like permethrin kill insects by disrupting sodium channels resulting in depolarization of nerve cells, thereby causing paralysis and death.[46] Due to its high toxicity among nontarget organisms (honeybee contact LD_{50} = 0.008 μg/bee[28]) and aquatic organisms (fry channel catfish LC_{50} = 0.62 μg/L[47,48]), permethrin is designated as a restricted-use pesticide for wide area applications and crops.[47] However, due to its efficiency in killing insects, licensed applicators use pyrethroids for adult mosquito management and application to corn, soybeans, alfalfa, and fruit crops for controlling biting and chewing pests.[47,49] Schleier and Peterson (2010)[50] quantified permethrin 1 h after municipal mosquito control spray operations at a maximum concentration of 397 ng/m^3 in PM. In comparison, the highest permethrin concentration detected near beef cattle feed yards was 70 times greater (28,920.8 ng/m^3, Table 1). Of note though is that mosquito spraying efforts are typically separated by multiple days, sometimes weeks,[51] whereas

Table 1. Frequency and Mean Concentrations of Analytes Detected above the Limit of Quantitation in Particulate Matter (ng/g) and in Air at Feed Yard Locations (ng/m^3)

chemical	total detects	% > LOQ[a]	mean ± SE (ng/g)	mean ± SE (ng/m^3)	maximum (ng/m^3)
piperonyl butoxide[b]	27	73.0	31.8 ± 22.6	152.0 ± 42.9	1065.0
bifenthrin[c]	20	54.1	2.2 ± 1.3	8.1 ± 2.6	93.0
permethrin[c]	25	67.6	192.1 ± 117.3	1211.7 ± 781.0	28,920.8
imidacloprid[d]	10	27.0	19.9 ± 13.7	62.8 ± 38.2	1125.3
clothianidin[d]	5	13.5	0.5 ± 0.3	2.4 ± 1.0	23.3
thiamethoxam[d]	1	2.7	<0.1	1.1	41.2
abamectin[e]	1	2.7	<0.1	0.1	2.3
eprinomectin[e]	1	2.7	<0.1	<0.1	1.7
ivermectin[e]	18	48.7	1.1 ± 0.3	11.9 ± 4.2	146.2
moxidectin[e]	5	13.5	0.7 ± 0.5	5.0 ± 2.7	69.7
temephos[f]	1	2.7	0.1	2.2	81.6
azoxystrobin[f]	6	16.2	4.4 ± 4.3	7.7 ± 6.04	223.5
boscalid[f]	17	46.0	6.4 ± 2.8	73.0 ± 18.2	378.2
myclobutanil[f]	3	8.1	0.1 ± 0.1	0.8 ± 0.5	17.1
propiconazole[f]	1	2.7	0.1	0.8	28.0
pyraclostrobin[f]	1	2.7	<0.1	0.1	3.4

[a]limit of quantitation. [b]synergist. [c]pyrethroid. [d]neonicotinoid. [e]macrocyclic lactone. [f]fungicide.

permethrin concentrations detected in this study may be aerially disseminated from feed yards on a daily basis.

Though we cannot definitively exclude municipal mosquito control as a potential contributor to permethrin concentrations quantified in our PM samples, it is unlikely since beef cattle feed yard collection sites were in excess of 5 km from any residential or municipal area. Given the high permethrin detection frequencies across all feed yard sites and sampling events including those that occurred outside of the peak mosquito season (August[52,53]), mosquito control can likely be discounted as a significant contributing source.

In addition to permethrin, bifenthrin (also a pyrethroid) was commonly detected in feed yard PM samples in this study (54%; Table 1). Bifenthrin was detected in over 50% of feed yard PM samples, but at lower concentrations than permethrin (<100 ng/m^3). Bifenthrin is more toxic to honeybees than permethrin (Table 2) and requires less frequent application/spraying.[54,55] Of particular interest is that 60% of PM samples containing bifenthrin also contained permethrin. Since bifenthrin and permethrin have similar mechanisms of action, toxic effects among target and nontarget organisms are assumed to be additive.[56] In total, 32.4% of all PM samples collected near feed yards contained both permethrin and bifenthrin, indicating potential for additive toxicity. Additionally, 76% of all PM samples collected in this study contain pyrethroid concentrations higher than the honeybee contact LD_{50} concentrations,[57,58] raising concern that PM emanating from feed yards may be toxic to nontarget invertebrates.

Table 2. Projected Total Pesticide Emissions per Day across all Feed Yards in the USA Calculated Using the Total Suspended Particulate (TSP) Emission Factor of 57 g/animal/day from Bonifacio Et al.'s Study[39] and Mean Pesticide Concentrations Observed in the Current Study[a]

chemical	total pesticide emission (ng/day)	honeybee contact LD_{50} (ng/bee)[b]	honeybee death equivalencies
permethrin	128.5 x 10^9	63[c]	1,00,000,000
bifenthrin	1.5 x 10^9	15[c]	49,000,000
imidacloprid	13.3 x 10^9	61[c]	109,000,000
piperonyl butoxide	21.3 x 10^9	170,000[d]	63,000
boscalid	4.3 x 10^9	200,000[e]	11,000

[a]Total daily pesticide emission divided by honeybee contact LD_{50} value divided by two to account for median lethality yield estimates of the daily number of honeybee death equivalencies for each insecticide. [b]LD_{50} concentrations from Pesticide Manual[41] and ECOTOX database.[40] [c]highly toxic to honeybees. [d]slightly toxic. [e]not acutely toxic.

Permethrin is commonly the active ingredient in fogging applications and topical sprays used on cattle, whereas bifenthrin is used more widely in residual sprays (spraying surfaces frequented by flies[59]). Bifenthrin is also sprayed to combat cutworms, grubs, armyworms, etc. in crop production.[60] Although the majority of PM-associated bifenthrin is likely emanating from feed yards, we cannot completely discount the possibility that local application of bifenthrin to crops is a contributing source.

Piperonyl butoxide is a pyrethroid synergist frequently added to pyrethroid formulations, and it was positively correlated with permethrin concentrations (ρ = 0.548, p < 0.005) detected in the current study (present in 60% of PM containing bifenthrin). Piperonyl butoxide enhances pyrethroid toxicity in exposed insects by inhibiting cytochrome P450 activity involved in pyrethroid detoxification.[61,62] Synergism between piperonyl butoxide and pyrethroids suggests that the reported LD_{50} concentrations for pyrethroids (active ingredient only) are a significant underestimate of the risk associated with total pyrethroid concentrations detected in PM.[62]

Macrocyclic Lactone Occurrence. Interestingly, ivermectin was detected in 49% of feed yard PM samples. To our knowledge, this is the first study to quantify macrocyclic lactones associated with, and entering the environment, via PM. Moxidectin (14%) was detected less frequently than ivermectin, and eprinomectin and abamectin were only detected once each (Table 1). The macrocyclic lactones (ivermectin, doramectin, eprinomectin, abamectin, and moxidectin) are anthelmintics routinely given to cattle to control internal and external parasites such as lungworms, horn flies, cattle grubs, lice, and round worms.[12] Since most macrocyclic lactones are applied via a pour-on formulation or injection (intramuscular or subcutaneous[63]), these endectocides were not expected to be transported via PM. However, as described in the works of Blackwell et al.[18] and McEachran et al.,[16] the unique environments existing within the feed yards that promote daily suspension of elevated PM concentrations also likely facilitate macrocyclic lactone aerosolization and transport beyond feed yard boundaries. While there has been extensive research into endectocide toxicity to nontarget aquatic and soil-dwelling organisms,[30] the results of this study broaden the scope of potential nontarget organisms. Lumaret et al.[30] concluded that macrocyclic lactones are highly toxic to invertebrates, especially those with larval instars, which include a majority of native pollinators. The honeybee contact LD_{50} value for abamectin is 0.03 μg/bee, and Guseman et al.[27] used ivermectin as a positive control for toxicity assessments with honeybees (no LD_{50} data available). Furthermore, Peterson et al.[64] determined that even the least toxic macrocyclic lactone, moxidectin,[65] was extremely toxic to the painted lady butterfly larva (*Vanessa cardui*; oral LD_{50} 2.1 ng/g). Additionally, ivermectin concentrations detected in PM increased during May, which coincides with the first generation of monarch butterfly larvae (*Danaus plexippus*) emerging and maturing in the High Plains before continuing their migration north.[66,67]

Neonicotinoid Occurrence. Neonicotinoids (imidacloprid, clothianidin, and thiamethoxam) were all detected in relatively low frequencies (< 27% of all feed yard PM samples), but at potentially toxic concentrations. While neonicotinoids are not overtly toxic to mammals, they are toxic to aquatic organisms and are highly toxic to pollinators due to their mechanism of action (agonistic binding to insect nicotinic acetylcholine receptors[68,69]). Dust created during sowing of seeds treated with neonicotinoids is sufficient to kill honeybees.[70−72] Tapparo et al.[70] and Marzaro et al.[71] quantified neonicotinoid concentrations in air during maize planting at 800−13,100 ng/m^3 and at a mean concentration of 100 ng/m^3 along field margins, whereas Forero et al.[72] determined mean neonicotinoid concentrations of 2.43 ng/m^3 330−700 m from field boundaries. Neonicotinoid concentrations reported in this study (<LOQ—1125 ng/m^3, Table 1) were similar to concentrations detected during planting along field margins, even though PM was collected at a mean distance of 1 km from row crop boundaries and outside of planting periods (Table S2).[73] Furthermore, only two PM samples collected at reference locations contained a neonicotinoid (imidacloprid),

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and at relatively low concentrations (2.95 ± 2.1 ng/m^3). Limited occurrence of neonicotinoids at reference locations, and relatively high mean concentrations of neonicotinoids in PM from feed yards (located 1046 ± 73 m from row crops), suggests that neonicotinoids quantified in feed yard PM receive relatively little input from local agricultural crops.

Neonicotinoid concentrations detected in PM along the feed yard boundaries were significantly higher (>10 times) than the honeybee LD_{50} values associated with each compound (18 ng/bee for imidacloprid, 22 ng/bee for clothianidin, and 30 ng/bee for thiamethoxam[70,72,74]). Some countries have completely banned the use of neonicotinoids, and many regulatory agencies have provided guidelines to decrease the environmental release of neonicotinoids during planting.[70] However, there is no guidance for neonicotinoid emissions from beef cattle feed yards, where neonicotinoids are routinely used to kill flies and other pests.[21]

Fungicide Occurrence. Although not commonly used on feed yards, each fungicide analyte included in this study was detected in feed yard PM samples, with boscalid being the most frequently detected fungicide (46%) and at the highest concentration of any fungicide (378.2 ng/m^3). Boscalid is not registered for use on animals but is used extensively on crops consumed by cattle such as hay and corn.[75] In addition to PM originating from cattle foraging and/or feeding within the feed yard, another potential source of fungicides detected in this study was PM from local cotton fields to which fungicides had been applied. However, infrequent detection of fungicides at reference locations (azoxystrobin 8% vs feed yard 16% and boscalid 25% vs feed yard 46% of PM samples) suggests that PM from local crop fields is not the major source of fungicides detected at feed yards. Fungicide concentrations detected in PM at all locations were well below toxic levels for nontarget organisms;[74,76−79] however, fungicides like pyraclostrobin act synergistically with some insecticides.[74]

Environmental Relevance and Bee Death Equivalencies in the USA. Based on the mean pesticide concentrations quantified in this study (Table 2), total pesticide masses entering the environment daily from feed yards in the USA were estimated based on the median total suspended particulate matter (TSP) concentrations for daily PM emissions from beef cattle feed yards (57 g/animal/day) as reported by Bonifacio et al.[39] The median daily emission concentrations reported by Bonifacio et al. (57 g/animal/day) are relatively conservative, with other studies determining the median daily TSP at 81 (g/animal/day)[80] and 127 (g/animal/day),[81] respectively.

Assuming that there are 11.74 million cattle confined to feeding operations throughout the USA,[6] an estimated 669,123 kg of TSP would be generated every day. Multiplying the total mass of TSP produced daily by mean concentrations of pesticides quantified in the present study indicates that more than 150 grams of insecticide active ingredients would be released into the environment (Table 2). Since TSP collected in this study was during peak PM emissions, it may represent higher concentrations of pesticides in PM compared to other times of the day. Still, estimates of the total mass of insecticides emitted per cattle feed yards on a daily basis are 10 times higher than the mass of steroids released into the environment as reported by Blackwell et al.[16]

Using estimates of the total pesticide mass released from feed yards on a daily basis, we calculated the theoretical honeybee death equivalencies using published honeybee contact LD_{50} values and dividing by two to account for median lethality. The honeybee death equivalency estimates provide context for consideration of risks associated with airborne agrochemicals emanating from beef cattle feed yards. Recent studies have determined that humidity is a major contributor to insecticide toxicity among honeybees.[70,71,82] Elevated humidity levels allow for increased uptake through bee tegument.[70,82] Even though the High Plains is a semiarid environment, relative humidity levels begin to increase at sunset and continue to increase throughout the night (average humidity level is 74% during early mornings).[83] With peak PM emissions occurring at dusk[39] followed by increasing humidity levels throughout the nighttime hours,[83] the bioavailability of agrochemicals in PM would also likely increase.

Feed yards throughout the USA could therefore release enough permethrin via aerosolized PM to kill 1 billion honeybees each day. On a local scale, an average-sized feed yard (39,220 head[84]) emits sufficient permethrin (via TSP) to kill 6800 bees each day (> 10% of a healthy honeybee hive/day). Of note though is that honeybee death equivalency estimates do not account for bioavailability of pesticides, or additive or synergistic toxicity, and may be underestimates (or overestimates) of risk to honeybees, native bees, and other nontarget invertebrates.

While honeybee toxicity thresholds were used as a metric for other pollinators in this study, it does not account for the large variability in sensitivity among different pollinator species. To address the sensitivity concerns, the European Food Safety Authority (EFSA) has proposed using a risk assessment factor (safety factor) of 10 when applying LD_{50} concentrations of honeybees to native bees.[85] Table 2 is therefore likely a conservative estimate of total bee deaths, which is potentially an order of magnitude lower than potential bee deaths near the feed yards. These data reveal the likelihood that high quantities of pesticides are entering the environment from beef cattle feed yards. Consequently, there is a need to more fully characterize risk to affected pollinator populations. Moreover, these data highlight a need to consider regulatory limits on feed yard insecticide emissions.

Global Implications. Many feed yards established in the last 50 years have been in areas that receive relatively little rainfall, similar to the High Plains.[18,86] These regions (Plains of USA, Mexico, South America, and Northern Australia), as described in McEachran et al.'s study,[16] are prone to droughts, exacerbating PM generation and pesticide emission from feed yards. Interestingly though, dust control is a major environmental challenge even among feed yards located in tropical and subtropical regions (e.g., southern Brazil[2,87]). Therefore, aerial dissemination of insecticides into local environments, through PM, is likely to occur from feed yards worldwide regardless of climatic conditions.

Global beef production is estimated to increase by 17% in developing countries and by 8% in developed countries over the next decade.[5] In order to meet these demands, feed yards are likely to increase in number and in size, resulting in more areas being affected by fugitive PM from feed yards containing pesticides. Over the next decade, the largest percent of beef production will occur in areas (developing countries) where pesticide usage is not heavily regulated.[88,89] This is of concern because insect pollinators are declining worldwide as a result of habitat loss in addition to the extensive use of agrochemicals.[90] Expansion of feed yards and aims to increase production through the use of pesticides could come at the expense of

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insect pollinators. Assuming that insecticides from feed yards in other countries enter the environment via PM at comparable levels to those observed in this study, the total losses of bees and other similarly sensitive nontarget pollinators could exceed 2.9 billion organisms daily.[3,39]

Without pollinators, it is estimated that humans could lose over $200 billion (USD) annually in pollination services globally ($215 billion USD in 2005[91]). An example of the trade-off between beef production and pollinator production can be seen in Brazil, where beef production accounts for 9 million tons of meat,[1] while pollinated crops provide upward of 51 million tons of food.[92] It is evident that insect pollinators have a greater relative impact on food production and security than beef cattle feed yards. However, without proper management, agrochemical exposure from beef cattle feed yards may result in a dramatic loss of insect pollinators, which could decrease environmental quality, decrease crop production, and decrease agricultural revenue in areas surrounding the feed yards.[91,93−95]

Further research is needed to determine the extent (and distance) of aerial agrochemical transport from beef cattle feeding operations and the degree to which these pesticides may persist in the environment at concentrations capable of causing harm to nontarget organisms. Still, this study highlights the importance and need to more comprehensively consider pesticide management beyond outside the context of commercial spraying and row crop agriculture to protect environments from heretofore uncontrolled agrochemical dissemination from beef cattle feed yards.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c03603.

> Feed yard sampling site pen area (Table S1); particulate matter collection parameters and meteorological conditions for all sampling events (Table S2); particulate matter extraction recoveries and limit of quantitation (Table S3); pesticides used on cattle and pests on feed yard facilities (Table S4); and Friedman results depicting differences in pesticide concentrations quantified on PM between feed yard and reference locations (Table S5) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Philip N. Smith − *Department of Environmental Toxicology, Texas Tech University, Lubbock, Texas 79409, United States;* orcid.org/0000-0002-7557-7563; Phone: (806)885-4567; Email: phil.smith@ttu.edu

Authors

Eric M. Peterson − *Department of Environmental Toxicology, Texas Tech University, Lubbock, Texas 79409, United States;* orcid.org/0000-0003-4606-597X

Frank B. Green − *Department of Environmental Toxicology, Texas Tech University, Lubbock, Texas 79409, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.0c03603

Notes

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