Technical Notes & Preliminary Communications

Core Ideas

Gaseous emissions from windrow and static stockpile of cattle manure were measured.

The inverse-dispersion technique combined with the OP-FTIR techniques was used.

Windrow reduced water content by 50% while little change was found in stockpile manure.

Windrow fluxes of NH_3 , N_2O , and CO_2 were two to five times greater than from the stockpile.

Emissions of CH_4 from the stockpile were two times greater than from the windrow.

Gas Emissions during Cattle Manure Composting and Stockpiling

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Supplemental material is available online for this article.

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Abbreviations: $\text{CO}_2\text{-e}$, carbon dioxide equivalents; DM, dry manure; EF, emission factor; GHG, greenhouse gas; IPCC, Intergovernmental Panel on Climate Change; OP-FTIR, open-path Fourier transform infrared.

ABSTRACT

Manure composting is a common management practice for cattle feedlots, but gaseous emissions from composting are poorly understood. The objective of this study was to quantify ammonia (NH_3), nitrous oxide (N_2O), carbon dioxide (CO_2), and methane (CH_4) emissions from windrow composting (turning) and static stockpiling (nonturning) of manure at a commercial feedlot in Australia. An inverse-dispersion technique using an open-path Fourier transform infrared (OP-FTIR) spectrometer gas sensor was deployed to measure emissions of NH_3 , N_2O , CO_2 , and CH_4 over a 165-d study period, and 29 and 15% of the total data intervals were actually used to calculate the fluxes for the windrow and stockpile, respectively. The nitrogen (N) lost as NH_3 and N_2O emissions represented 26.4 and 3.8% of the initial N in windrow, and 5.3 and 0.8% of that in the stockpile, respectively. The carbon (C) lost as CO_2 and CH_4 emissions represented 44 and 0.3% of the initial C in windrow, and 54.8 and 0.7% of that in the stockpile, respectively. Total greenhouse gas (GHG) emissions from the manure windrow were 2.7 times higher than those of the stockpiled manure. This work highlights the value that could be accrued if one could reduce emissions of $\text{NH}_3\text{-N}$ and $\text{N}_2\text{O-N}$ from composting, which would retain manure N content while reducing GHG emissions.

Over the last two decades, Australia's beef feedlot industry has expanded to meet the demand for red meat in domestic and international markets. This has resulted in a substantial quantity of cattle manure concentrated in feedlot pens, thereby creating an opportunity for the manure to be managed as a valuable crop fertilizer (Chalk et al., 2013).

This is the author manuscript accepted for publication and has undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the [Version of Record](#). Please cite this article as [doi: 10.1002/jeq2.20029](https://doi.org/10.1002/jeq2.20029).

There are two common options for managing manure for fertilizer. Direct spreading of fresh manure to agricultural land is a common practice but can result in nitrogenous emissions and odor (Gibbs et al., 2002) and can introduce pathogens and weed seeds to soils (Sharpley et al., 1994; Stentiford, 1996). Alternatively, composting is a well-established method of managing manure prior to its field application. The composting process increases nutrient density and reduces pathogen and bacterial abundance (Stentiford, 1996; Pardo et al., 2015) and odor. Composting also reduces the water content of the manure, allowing easier handling and reducing the cost of transport. A negative aspect of composting is that it can increase the potential for direct and indirect greenhouse gas (GHG) emissions from the manure, including emissions of methane (CH_4), ammonia (NH_3), and nitrous oxide (N_2O) (Hao et al., 2001).

In Australia, the majority of large feedlots stockpile manure for <12 mo before spreading. In 2010, only 18 large feedlots (out of 75) used windrow composting, where the manure is turned regularly over periods of ~6 mo (MLA, 2017). Although these turning events result in short-term spikes in gas emissions (Bai et al., 2015), gaseous losses over the lifetime of manure stockpiling and composting are poorly understood.

The objective of this study was to measure gas emissions from the two contrasting manure management practices commonly used in Australian feedlots: stockpiling and windrow composting. We applied a micrometeorological approach to concurrently quantify the losses of NH_3 , N_2O , carbon dioxide (CO_2), and CH_4 . We calculated the losses of nitrogen (N) and carbon (C) due to gas emissions and evaluated GHG emissions (CO_2 equivalents, $\text{CO}_2\text{-e}$) from the two manure treatment options.

MATERIALS AND METHODS

Feedlot Information

The study was conducted at a commercial feedlot (36.28° S, 143.33° E; 131.7 m asl) located 220 km northwest of Melbourne, VIC, from 2 June to 12 November 2016. The terrain surrounding the feedlot was generally flat, and characterized by dry bare soil in summer and grain crops in winter. The main cattle breeds in the feedlot were Angus and Angus cross breeds (1–1.5 yr of age). The average live weight, cattle number, and feeding information are detailed in the Supplemental Tables S1 and S2.

Compost Manure Treatments and Sampling

A compost windrow and a stockpile were created on 2 June 2016 using manure collected from a single feedlot pen and transported to the experimental site just west of the feedlot (fallow land). The initial manure mass was 185.1 and 37.7 t for the windrow and stockpile, respectively. The windrow and stockpile had a size and shape typical of manure management at a feedlot. The windrow and stockpile were placed beside one another to enable concurrent emission measurements from each (Figure 1). The windrow was laid out in a north–south direction (52 m long $\times$ 5.1 m wide $\times$ 1.4 m high), and the static stockpile (10.2 m in diameter and 1.4 m high) was located 55.8 m to the north of manure windrow. From windrow formation to the end-compost product as fertilizer, the windrow was turned 21 times using a compost turner (operated by feedlot staff), with each turning event taking ~15 min. Depending on the weather and storage area, the typical windrow turning time was about seven to eight times over an average 6-mo composting period. In our study, the windrow was turned more frequently to remove the water content due to the wet weather condition. The

dates of turning are listed in the Supplemental Table S3. The static stockpile was not turned during the study.

Sampling of the manure was done during windrow turning events, with samples taken from four separate sections of the windrow and stockpile. Subsamples were oven dried and prepared for physical (pH and water content) and chemical properties analysis (total carbon [total C], total nitrogen [total N], and ammonium-nitrogen [$\text{NH}_4^+\text{-N}$]). Manure temperature was measured continuously over the course of the study. Manure sampling details are provided in the supplemental material.

Gas Emission Measurements

An open-path Fourier transform infrared spectrometer (OP-FTIR) (Matrix-M IRCube, Bruker Optics) was deployed to measure line-averaged concentrations of atmospheric CH_4 , CO_2 , NH_3 , and N_2O . A motorized mounting system (University of Wollongong) allowed the spectrometer to measure gas concentrations on three paths, each 80 m in length (Figure 1). For our measurement target of westerly winds, the three-path configuration gave the gas concentrations upwind of the manure (background) and downwind of both the compost row and the stockpile (while avoiding gas “contamination” from the large feedlot to the east of the site). During southeasterly winds, this configuration also allowed upwind and downwind measurements from the compost row (but not the stockpile). During the measurements, a 2.5-min aiming sequence for each OP-FTIR path was used, including 1.5 min of spectrum collection, 20 s of spectrum analysis and data logging, and 40 s of aiming movement. For each 15-min emission analysis interval, two measurements were collected for each path. Prior to the study, background concentrations of CH_4 , CO_2 , NH_3 , and N_2O were measured for 2 d to determine the measurement precisions: $<2 \text{ nmol mol}^{-1}$ for CH_4 , $0.4 \text{ nmol mol}^{-1}$ for NH_3 , $<0.3 \text{ nmol mol}^{-1}$ for N_2O , and $1 \text{ } \mu\text{mol mol}^{-1}$ for CO_2 at a 100-m path length. Concentration measurements were taken continually for 4 wk after windrow and stockpile formation, then over 3 wk during the next month, and thereafter for 2 wk per month. Measurements ended on 12 November, 165 d after compost and stockpile formation.

An inverse-dispersion model (WindTrax, Thunder Beach Scientific) was used to calculate gas emission rates. The principle of this technique has been reported by Flesch et al. (2004), and is based on the downwind enhancement of concentration above background levels. In calculating emissions, WindTrax assumes that gas is emitted from a ground-level area source, assumed to be the surface area (m^2) of the manure windrow and the stockpile, measured by GPS, in this study. The windrow and stockpile are not surface area sources, but more complex three-dimensional sources. We assume that the gas concentration is measured sufficiently far downwind of the manure (minimum downwind fetch to the OP-FTIR path is 20 m, >10 pile heights) so that the calculated emissions are insensitive to this error (a principle discussed by Flesch et al. (2005)).

The wind statistics needed for the emission calculations were provided by a three-dimensional sonic anemometer (CSAT-3, Campbell Scientific) adjacent to the experimental site, positioned 2.8 m above the ground. These statistics included the friction velocity (u^*), Obukhov stability length (L), turbulent velocity fluctuations in the three directional components, implied surface roughness (z_0), and wind direction. Following Flesch et al. (2014), we did not calculate emissions during error-prone atmospheric conditions: surface roughness $z_0 > 0.25 \text{ m}$, friction velocity $u^* < 0.05 \text{ m s}^{-1}$, and the percentage of particle touchdowns on the source area $< 75\%$. We also ignored wind directions from 0 to 120° , as these did not allow an emission calculation given the measurement configuration. The wind

statistics and gas concentration data were merged to create a 15-min interval time series using SAS software (version 9.4).

Statistical Modeling of Daily and Cumulative Gas Emission Trends

Statistical modeling of daily and cumulative emission trends for each gas and each manure treatment was done following three steps:

1. We calculated daily gas emissions on a per dry manure basis ($\text{g d}^{-1} \text{kg}^{-1}$ dry manure [DM]) from the 15-min emission dataset.
2. Mean daily fluxes were estimated using generalized additive models (Hastie and Tibshirani, 1990) fitted to the daily time series data using the default thin plates regression splines of the “gam” function in the “mgcv” package (Wood, 2006) in R 3.5.1 (R Core Team, 2018). Generalized additive models do not assume a predefined trend in daily gas emission, making them useful to model nonlinear trends with periods of increased and decreased gas emission over time.
3. To propagate uncertainties in daily emissions, Monte Carlo methods were used to sample 10,000 vectors of parameter values from the variance–covariance matrix of parameter estimates (Wood, 2006), simulating 10,000 daily emission curves consistent with our data. From these curves, we computed the mean and 95% confidence intervals of daily gas emissions.

RESULTS AND DISCUSSION

Gas Fluxes

With rainstorms and breaks in the OP–FTIR deployment schedule, there was a total of 57 emission measurement days during the study, or 5117 15-min intervals. Because our emission calculations were restricted to a limited range of wind directions ($120\text{--}360^\circ$) and favorable wind conditions, only a portion of these data provided suitable measurements: there was a total of 1482 and 758 suitable data intervals for the windrow and stockpile, respectively (29 and 15% of the total data intervals). When calculating cumulative emissions over our study, emissions for 108 out of the 165 d were interpolated using the statistical models described in previous section.

NH₃ Emissions

Emissions of NH₃ from the windrow were initially $0.07 \text{ g d}^{-1} \text{kg}^{-1}$ DM. Overall, the emissions gradually decreased and remained at $<0.006 \text{ g d}^{-1} \text{kg}^{-1}$ DM until the end of study (Figure 2). We did observe an emission peak of $0.19 \text{ g d}^{-1} \text{kg}^{-1}$ DM near Day 50 due to turning on Days 36 and 43. This delayed emission peak was attributed to decreasing water content, high initial N substrate levels, and increasing pH (Supplemental Figure S1). In contrast, NH₃ emissions from the stockpile showed less temporal variation, ranging from $0.002\text{--}0.05 \text{ g d}^{-1} \text{kg}^{-1}$ DM over the study (Figure 2). The windrow and the stockpile had similar emissions over the first 20 d, which suggested that the initial moving of manure to the experimental site was equivalent to turning (in terms of NH₃ emissions).

At a finer temporal scale, we observed diurnal variability in NH₃ emissions from the manure windrow: higher emissions between 1200 and 1600 h, and lower emissions between 2100 and 2400 h (Supplemental Figure S4). This pattern can be explained by the positive correlation between hourly fluxes and wind speed ($r = .49$, $P < .001$), and between hourly fluxes and ambient temperature ($r = .23$, $P = .001$) (Supplemental Figure S5). In a typical afternoon, higher winds and higher air temperatures resulted in higher NH₃ emissions. We also observed high NH₃ emission rates in the early morning on Days 2 and 16, likely due to the strong westerly wind at that period. The diurnal flux pattern was apparently in the first few weeks after windrow formation, and not detected near the end of the study (Supplemental

Figure S4). There was a diurnal emission pattern from the stockpile within the first week (higher around 1300 h and lower at nighttime), but not later in the study.

N₂O Emissions

Nitrous oxide emissions from the windrow were initially low but increased after formation (Figure 2). The greatest N₂O emissions from the windrow were seen on Day 94 (at almost 0.04 g d⁻¹ kg⁻¹ DM), which was likely due to rainfall (6.8 mm) on Day 92, inducing anaerobic conditions and promoting N₂O emissions. Emissions from the stockpile were initially low (near zero) and then increased to near 0.01 g d⁻¹ kg⁻¹ DM by Day 130.

The increase in N₂O emission over time in both treatments was opposite of that for NH₃ (decreasing with time), likely due to N₂O production through nitrification occurring mainly from late composting processes, when organic N is reduced to NH₃ before nitrification (Figure 2). The greater N₂O emissions from the windrow, compared with the stockpile, were probably due to (i) greater nitrification coinciding with a reduction in NH₃ emissions (Chadwick et al., 2011) that favored nitrifying bacteria (He et al., 2001; Pattey et al., 2005; Chadwick et al., 2011; Cayuela et al., 2012; Biala et al., 2016), and (ii) that N₂O production from denitrification process should be counted, since the undetectable NO₃⁻ (data not shown here) meant that NO₃⁻ was denitrified immediately after it was produced by nitrification process. Greater N₂O emissions from compost were also reported in Parkinson et al. (2004). Lower nitrification–denitrification processes resulted in lower N₂O emission from stockpile, which was also reported in other studies (Pattey et al., 2005; Chadwick et al., 2011; Cayuela et al., 2012). The N₂O emissions from the both treatments were comparable after Day 90, when there were similar manure temperatures and similar NH₄⁺-N contents (Supplemental Figure S1), reflecting a reduced effect of turning (aerobic) on N₂O emissions when the composting process moved toward maturing phase, with lower N content in manure.

CO₂ Emissions

The CO₂ emissions from the manure windrow initially decreased with time. Initial emission rates were near 6.0 g d⁻¹ kg⁻¹ DM and fell to near 1.0 g d⁻¹ kg⁻¹ DM on Day 9, but by Day 40, the CO₂ emissions had strongly increased, reaching a (study) peak emission rate of 6.48 g d⁻¹ kg⁻¹ DM on Day 48. Thereafter, emissions slowly decreased until the end of the study. The dynamic of CO₂ emission over the time was related to the degradation of labile material in the manure by microorganisms, which has been reported by Sommer and Møller (2000), Hellmann et al. (1997), Hobson et al. (2005), and Sánchez-Monedero et al. (2010). We also observed a positive correlation between CO₂ and NH₃ emissions from the windrow over the course of the study ($r = .46$, $P < .001$) (Supplemental Figure S5). This could be explained by the fact that both CO₂ and NH₃ production were correlated to wind speed and ambient temperature, which was also reported by Leytem et al. (2011).

From the manure stockpile we observed that CO₂ emissions were initially near 5.0 g d⁻¹ kg⁻¹ DM but fell to nearly zero by Day 49. Emissions then increased to reach a peak of 12.0 g d⁻¹ kg⁻¹ DM on Day 135, followed by a decrease until the end of the study. The increasing manure temperature associated with greater activity of microorganisms could explain the emission peak on Day 135 (Supplemental Figure S1). A positive correlation between CO₂ and N₂O emissions in the stockpile ($r = .61$, $P < .001$) (Supplemental Figure S5) was likely attributed to prevalent heterotrophic microbial activity and denitrification producing N₂O (Shi et al., 2017).

CH₄ Emissions

The CH₄ emissions from the manure windrow generally decreased over the study period. The highest emission rate of $\sim 0.02 \text{ g d}^{-1} \text{ kg}^{-1} \text{ DM}$ was measured on Day 18 after a turning event. This decrease in CH₄ emission was probably associated with the decreasing activity of anaerobic microorganisms in aerated conditions when the manure was turned more frequently (Lopez-Real and Baptista, 1996). In contrast, CH₄ emissions from the stockpiled manure generally increased over time. The emission rate was near $0.01 \text{ g d}^{-1} \text{ kg}^{-1} \text{ DM}$ on Day 1, which fell to near zero on Day 9. Thereafter, CH₄ emissions increased to a peak of $0.05 \text{ g d}^{-1} \text{ kg}^{-1} \text{ DM}$ on Day 135. While the emission rate decreased after the peak, it remained generally high ($0.02 \text{ g d}^{-1} \text{ kg}^{-1} \text{ DM}$) until the end of experiment. The high temperature and water content of the stockpiled manure would have facilitated this elevated CH₄ emissions pattern (Sommer and Möller, 2000).

We did not observe an obvious diurnal variation in CH₄ emissions from the windrow, but we did from the stockpile, which had higher emissions at night and lower emissions during the day. We found a positive correlation between hourly CH₄ and CO₂ emissions for the windrow ($r = .49$, $P < .001$) and the stockpile ($r = .37$, $P < .001$) (Supplemental Figure S5). For the windrow, this was likely due to the processes of CH₄ and CO₂ production from CH₄ oxidation, which can be regulated by a thermophilic methane oxidizer at a suitable temperature (Jäckel et al., 2005). For the stockpile, this likely corresponded to anaerobic bacteria activities. This correlation was also reported in a pig slurry study by Petersen et al. (2009).

Cumulative Gas Emissions

Cumulative emissions of NH₃, N₂O, CO₂, and CH₄ over the 165-d experimental period are given in Table 1. Overall losses of N as NH₃-N and N₂O-N were 26.4 and 3.8% and 5.3 and 0.8% of the initial N in the composted and stockpiled manure, respectively. Our finding of greater NH₃-N losses from the compost relative to the stockpile is consistent with other studies (Amon et al., 1999, 2001). The higher N₂O-N losses we observed from the compost are in contrast with studies of Amon et al. (1999, 2001) but support that of Hao and Larney (2017). This likely reflects differences in aeration that promote nitrification over denitrification and generate more N₂O from the windrow than the stockpile. The 26.4% N lost as NH₃-N from our windrow was somewhat higher than the values reported by Pardo et al. (2015), who reported that NH₃-N losses from composting were 10–25% of the initial N. The 3.8% loss of N₂O-N from our compost was much higher than the emission factor (EF) of 1% of the total N suggested by the Intergovernmental Panel on Climate Change (IPCC) (de Klein et al., 2006). The NH₃-N losses from the stockpile were higher than the 3% losses reported by Amon et al. (1999) for a manure pile. The N₂O-N losses from the stockpile (0.8%) were slightly higher than the 0.5% EF suggested by the IPCC (de Klein et al., 2006) for stored manure.

The C lost as CO₂-C and CH₄-C was 44.0 and 0.3% of total C in the windrow, respectively, and 27.3 and 0.7% of total C in the stockpile, respectively. Reduced CH₄ emissions, but increased CO₂ emissions, during the short-duration turning process were also reported by Amon et al. (1999) and Pardo et al. (2015). The C lost as CO₂-C from our study agreed with the 28–58% (compost) and 14–30% (stockpile) loss reported in Hao and Larney (2017). The 0.3% C lost as CH₄-C was comparable with the 0.41% reported by Amon et al. (2001) for compost dairy manure and the 1.6% reported by Hao and Larney (2017) from compost feedlot manure, but the 0.7% loss from our stockpile was much lower than the 3.92% measured by Amon et al. (2001) and the 7% measured by Hao and Larney (2017). The

C lost as CH₄-C in our study was lower than the EF guidelines of 1–2% suggested by the IPCC (1997).

Taking a global warming potential of 28 for CH₄ and 265 for N₂O (a 100-yr lifetime) (Hartmann et al., 2013) and assuming that 1% of manure NH₃-N emissions will be deposited on the landscape and reemitted as N₂O (de Klein et al., 2006), the total GHG emissions (in CO₂-e, including contributions of CH₄ and N₂O) over the 165-d study are estimated to be 392.1 and 143.4 g kg⁻¹ DM) for the composted and stockpiled manure, respectively. In terms of GHG emissions (CO₂-e), the N₂O component was much larger than CH₄ in the compost (92%), whereas N₂O and CH₄ had nearly equal contributions in the stockpile (49–51%).

Conclusions

We compared gaseous emissions from two typical manure handling options at cattle feedlots, composting, and static stockpile storage and found that composting inhibits CH₄ emissions but promotes NH₃ and N₂O emissions. This result is consistent with other studies of cattle manure. Adding the contributions from these gas components showed that windrow composting effectively doubled the GHG emissions compared with stockpiling. This work highlights the value that could be accrued if one could reduce emissions of NH₃-N and N₂O-N from composting—for instance by optimizing the frequency of turning event and the duration of storage period, or adding amendments, which may help retain the N values in the end-compost product while reducing GHG emissions.

SUPPLEMENTAL MATERIAL

The supplemental material includes the method of manure sampling and the results of manure physical and chemical properties. The associated figures, tables, and references are also included. Furthermore, we describe the manure temperature sensors that were used to measure manure temperature during the compost study.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

ACKNOWLEDGMENTS

We thank Meat and Livestock Australia (MLA) for funding, and the soil research group for great field support. We also thank our group members for valuable discussions during the preparation of the manuscript. Special thanks to Xiaoxian Li and Robert Impraim for collecting and analyzing the manure samples.

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FIGURE 1. Map showing the locations of open-path Fourier transform infrared (OP-FTIR, three paths in red line), manure windrow (compost row, blue), and the static stockpile (orange). Westerly winds are the target for measurement.

FIGURE 2. Daily mean fluxes of NH_3 , N_2O , CO_2 , and CH_4 measured from the manure windrow and stockpile over the 165-d measurement period (symbols) and the fitted statistic trends (lines). The shaded area represents 95% confidence intervals. The cumulative losses of NH_3 , N_2O , CO_2 , and CH_4 are also plotted. Black marks on the horizontal axis indicate windrow turning events.

TABLE 1. Daily mean and cumulative (165-d) emissions of NH₃, N₂O, CO₂, and CH₄ from the manure windrow and stockpile. Total N, C, and greenhouse gas (GHG) emissions (CO₂ equivalents [CO₂-e]) are estimated.

Emission	Manure windrow	Stockpile
Daily mean emission (mg d ⁻¹ kg ⁻¹ dry manure)		
NH ₃	34.9 (4.3) [†]	14.7 (1.4)
N ₂ O	1.3 (0.9)	0.3 (0.5)
CO ₂	3196.5 (204.4)	1390.0 (528.3)
CH ₄	9.6 (0.6)	159.0 (1.5)
Cumulative emissions (g kg ⁻¹ dry manure)		
NH ₃	6.7 (0.7)	1.4 (0.2)
N ₂ O	1.3 (0.1)	0.3 (0.1)
CO ₂	451.5 (33.7)	280.3 (87.2)
CH ₄	1.2 (0.1)	2.6 (0.2)
N loss (%)		
NH ₃ -N	26.4	5.3
N ₂ O-N	3.8	0.8
C loss (%)		
CO ₂ -C	44.0	27.3
CH ₄ -C	0.3	0.7
GHG emissions (CO ₂ -e) (g kg ⁻¹ dry manure) [‡]		
N ₂ O§	359.6	70.9
CH ₄	32.5	72.5
Total	392.1	143.4
% Total GHG emissions		
N ₂ O	91.7	49.4
CH ₄	8.3	50.6

[†] Average (SE).

[‡] Assuming a global warming potential (GWP) of 28 for CH₄, and 265 for N₂O.

§ Calculation assumes that 1% of the emitted NH₃-N is deposited and reemitted as N₂O-N.

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Title:

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Date:

2020-01-01

Citation:

Bai, M., Flesch, T., Trouve, R., Coates, T., Butterly, C., Bhatta, B., Hill, J. & Chen, D. (2020). Gas emissions during cattle manure composting and stockpiling. JOURNAL OF ENVIRONMENTAL QUALITY, 49 (1), pp.228-235. <https://doi.org/10.1002/jeq2.20029>.

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