Ammonia and greenhouse gas emissions from slurry storage - A review

Thomas Kupper[a,*], Christoph Häni[a], Albrecht Neftel[b], Chris Kincaid[c], Marcel Bühler[a,d,e], Barbara Amon[f,g], Andrew VanderZaag[c]

[a] Bern University of Applied Sciences School of Agricultural, Forest and Food Sciences, Laenggasse 85, 3052 Zollikofen, Switzerland
[b] Neftel Research Expertise, 3033, Wohlen b Bern, Switzerland
[c] Agriculture and Agri-Food Canada, 960 Carling Avenue, Ottawa, Ontario, K1A0C6, Canada
[d] Oeschger Centre for Climate Change Research, University of Bern, Hochschulstrasse 4, 3012 Bern, Switzerland
[e] Institute of Geography, University of Bern, Hallerstrasse 12, 3012 Bern, Switzerland
[f] Leibniz Institute for Agricultural Engineering and Bioeconomy (ATB), Max-Eyth-Allee 100, 14469 Potsdam, Germany
[g] University of Zielona Góra, Faculty of Civil Engineering, Architecture and Environmental Engineering, ul. Licealna 9, 65–762 Zielona Góra, Poland

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ABSTRACT

Storage of slurry is an important emission source for ammonia (NH_3), nitrous oxide (N_2O), methane (CH_4), carbon dioxide (CO_2) and hydrogen sulfide (H_2S) from livestock production. Therefore, this study collected published emission data from stored cattle and pig slurry to determine baseline emission values and emission changes due to slurry treatment and coverage of stores. Emission data were collected from 120 papers yielding 711 records of measurements conducted at farm-, pilot- and laboratory-scale. The emission data reported in a multitude of units were standardized and compiled in a database. Descriptive statistics of the data from untreated slurry stored uncovered revealed a large variability in emissions for all gases. To determine baseline emissions, average values based on a weighting of the emission data according to the season and the duration of the emission measurements were constructed using the data from farm-scale and pilot-scale studies. Baseline emissions for cattle and pig slurry stored uncovered were calculated. When possible, it was further distinguished between storage in tanks without slurry treatment and storage in lagoons which implies solid-liquid separation and biological treatment. The baseline emissions on an area or volume basis are: for NH_3: 0.12 g m^{-2} h^{-1} and 0.15 g m^{-2} h^{-1} for cattle and pig slurry stored in lagoons, and 0.08 g m^{-2} h^{-1} and 0.24 g m^{-2} h^{-1} for cattle and pig slurry stored in tanks; for N_2O: 0.0003 g m^{-2} h^{-1} for cattle slurry stored in lagoons, and 0.002 g m^{-2} h^{-1} for both slurry types stored in tanks; for CH_4: 0.95 g m^{-3} h^{-1} and 3.5 g m^{-3} h^{-1} for cattle and pig slurry stored in lagoons, and 0.58 g m^{-3} h^{-1} and 0.68 g m^{-3} h^{-1} for cattle and pig slurry stored in tanks; for CO_2: 6.6 g m^{-2} h^{-1} and 0.3 g m^{-2} h^{-1} for cattle and pig slurry stored in lagoons, and 8.0 g m^{-2} h^{-1} for both slurry types stored in tanks; for H_2S: 0.04 g m^{-2} h^{-1} and 0.01 g m^{-2} h^{-1} for cattle and pig slurry stored in lagoons. Related to total ammoniacal nitrogen (TAN), baseline emissions for tanks are 16% and 15% of TAN for cattle and pig slurry, respectively. Emissions of N_2O and CH_4 relative to nitrogen (N) and volatile solids (VS) are 0.13% of N and 0.10% of N and 2.9% of VS and 4.7% of VS for cattle and pig slurry, respectively. Total greenhouse gas emissions from slurry stores are dominated by CH_4. The records on slurry treatment using acidification show a reduction of NH_3 and CH_4 emissions during storage while an increase occurs for N_2O and a minor change for CO_2 as compared to untreated slurry. Solid-liquid separation causes higher losses for NH_3 and a reduction in CH_4, N_2O and CO_2 emissions. Anaerobically digested slurry shows higher emissions during storage for NH_3 while losses tend to be lower for CH_4 and little changes occur for N_2O and CO_2 compared to untreated slurry. All cover types are found to be efficient for emission mitigation of NH_3 from stores. The N_2O emissions increase in many cases due to coverage. Lower CH_4 emissions occur for impermeable covers as compared to uncovered slurry storage while for permeable covers the effect is unclear or emissions tend to increase. Limited and inconsistent data regarding emission changes with covering stores are available for CO_2 and H_2S. The compiled data provide a basis for improving emission inventories and highlight the need for further research to reduce uncertainty and fill data gaps regarding emissions from slurry storage.

* Corresponding author.
  E-mail addresses: thomas.kupper@bfh.ch, thomaskupper@sunrise.ch (T. Kupper).

1. Introduction

Livestock production systems around the world generate slurry—a mixture of feces and urine from housed livestock, mixed with bedding material and cleaning water (Pain and Menzi, 2011). Storage of slurry is required to enable the spreading in the field at appropriate time to supply nutrients to crops. Thus, a major part of the slurry is transferred from housings to outdoor stores such as tanks (at or above ground level) or earthen lagoons. Stores have variable forms and dimensions (e.g. up to several hectares for lagoons) according to the required storage volume. They have been identified as important emission sources for ammonia (NH_3), hydrogen sulfide (H_2S) and greenhouse gases (GHGs) including nitrous oxide (N_2O), methane (CH_4) and carbon dioxide (CO_2) from livestock production. Slurry stores are complex systems which influence emissions in many ways (Sommer et al., 2006; VanderZaag et al., 2008; Sommer et al., 2013).

A thorough description on principal mechanisms influencing the release of NH_3, GHGs and H_2S from slurry stores can be obtained from several studies (Olesen and Sommer, 1993; Ni, 1999; Sommer et al., 2006; VanderZaag et al., 2008; Sommer et al., 2013). Some important basic principles are summarized here. Slurry stores have a defined area where the gas exchange with the atmosphere takes place. It is a diffusive process and is quantified by emission rate values with the unit mass per area and time. Dissolved species of the gases are produced through microbial breakdown of nitrogen or organic compounds in the bulk slurry. Depending on prevailing chemical equilibria (e.g. NH_3/NH_4^+ which shifts to NH_4^+ at a low pH-value) and absence of microbial consumption, the gases move towards the emitting surface driven by diffusion (i.e. movement due to concentration gradients) and convection where parcels of air or liquid induce a movement of the compounds in the slurry (Sommer et al., 2013). At the slurry-air interface, the compounds pass gas- and liquid-phase resistances and diffuse into the air where they are transported to the atmosphere by convection. Transport within the liquid phase is temperature dependent and the gas-phase transfer is dependent on both temperature and turbulence (VanderZaag et al., 2015). Depending on the dry matter content of the slurry or more precisely, the amount of particles in the slurry which is influenced by the slurry type, animal species, animal diets, the thickness of the slurry bulk layer in the stores and meteorological conditions (Smith et al., 2007), a natural crust at the slurry surface can develop. It constitutes a barrier to the gas molecules between the liquid and the air. NH_3 and CH_4 may be consumed due to microbial activity in the crust leading to an emission reduction (Petersen and Ambus, 2006; Nielsen et al., 2010) while N_2O production may be enhanced (VanderZaag et al., 2009).

Ammonia has a large variety of negative environmental impacts which encompass the quality of air, soil and water, ecosystems and biodiversity. Moreover, it contributes to the formation of particulate matter which impairs human health (Sutton et al., 2011). N_2O and CH_4 are strong GHGs (Myhre et al., 2013). H_2S is often related to odor nuisances and can be lethal to animals and humans at high exposure levels (Sommer et al., 2013). NH_3 and GHG emissions have been regulated by the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (UNECE, 1999) and by the Kyoto protocol arising from the UN Framework Convention on climatic change (UN, 1997), respectively. Member countries of these protocols are obliged to calculate and report their national emissions annually, to track changes and compare to national emission ceilings where applicable. The methods for emission reporting are defined in EEA (2016) for NH_3 and in IPCC (2006) for N_2O and CH_4.

EEA (2016); IPCC (2006) and UNECE (2014) provide emission factors for slurry storage or numbers for emission reduction related to mitigation techniques which are used for emission reporting in emission inventories. However, a considerable number of recent studies on emissions from slurry storage provide updated information. The present review paper aims therefore to collect the data on NH_3, GHGs (CH_4, N_2O, CO_2) and H_2S emissions from these recent but also from previous studies and to provide a comprehensive overview on emissions from cattle or pig slurry stored uncovered and emission changes due to slurry treatment and coverage of slurry stores. This information can be used for the purpose of guide values, e.g. for the evaluation of emission data, and for improving emission inventories (greater accuracy, reduced uncertainty), e.g. for the determination of baseline emissions or emission reductions due to slurry treatment or coverage of slurry stores. The compiled data is entirely provided in the Supplementary data 2 for tracking the present or conducting future analyses.

2. Material and methods

2.1. Data search and data selection

A literature research was carried out with Web of Science [5.3] using the following search terms: "storage", "slurry", "emission"; "lagoon", "slurry", "emission". These searches were done on January 10, 2018 and yielded 601 papers in total. In a first screening, 290 papers were eliminated because they did not encompass livestock slurry. The remaining 311 articles were retained. In addition, 58 papers were found in the reference list of the screened articles. Therefore, in total, 369 articles were retained for further screening according to the following criteria:

(i) The investigated slurry was produced in an animal operation and consisted of urine and feces excreted from the animals onto a floor of a barn, a hardstanding or a milking parlor. The slurry might contain solids like bedding material or feed residues and be diluted with water. The investigated slurry was untreated or submitted to a treatment such as solid-liquid separation, anaerobic digestion, addition of an acid (acidification), additives or co-substrates. The treatment occurred under real-world conditions or after slurry sampling in the laboratory. Studies based on synthetic slurry, e.g. urine and feces collected separately from animals and subsequently combined in the laboratory, were excluded since fresh animal excretions substantially differ in chemical composition from stored slurry (Table 6). Moreover, urine and feces deposited onto a floor can rapidly undergo processes leading to gaseous losses. Hence, synthetic slurries might induce different emission levels as compared to slurries submitted to real-world conditions.

(ii) The untreated or treated slurry was transferred from the animal operation to a storage tank or a lagoon outside of animal housings and then submitted to measurements under real-world conditions or the slurry as characterized under point (i) was collected from a floor, an underfloor pit or an outside store and subsequently transferred to an experimental vessel where emissions were measured at pilot- or laboratory-scale. Studies encompassing e.g. emissions from a pit below an animal confinement were excluded since such facilities provide an environment which substantially differs from outside stores (e.g. exposure to outdoor climate, disturbance of the slurry surface due to continuous addition of animal excretions over almost the whole area of a pit).

(iii) The reported emission data are based on experimental determination of emission rates as defined by VanderZaag et al. (2008). Studies providing gas concentrations only were excluded.

(iv) The article provides numerical data encompassing emission data or percent differences in emissions between a slurry submitted to a treatment or slurry stored with covering and a reference system with untreated slurry or uncovered storage, respectively.

After evaluation, 120 papers complied with criteria (i) to (iv). 93 papers did not provide numerical data or comply with these criteria but included substantial information on emissions from slurry storage, e.g. basic mechanisms driving emissions. The remaining 156 papers were excluded because they were out of topic or did not provide substantial

information. An overview on the screened papers is in Supplementary data 1.

2.2. Data extraction

Data from the 120 papers were extracted. The parameters as shown in Table 1 were transformed, standardized or aggregated where necessary and then compiled in a database. Overall, 711 records were available for the analysis where one record is defined as an ensemble of entries listed in Table 1 (i.e. multiple records may be created from a single paper). Each record may differ in completeness according to the information provided in a paper.

2.3. Standardization of emission data

Emissions were reported in the papers using numerous units involving the gas molecule (i.e. NH_3, N_2O, CH_4, CO_2 and H_2S) or N, C or S included therein and various units for weight, time and surface or volume. Also, cumulative emissions were given over the entire experimental period. Overall, 36, 22, 31, 13 and 3 different ways for emission reporting were found for NH_3, N_2O, CH_4, CO_2 and H_2S, respectively. Standardization was performed in the present study to obtain comparable values over all records. For all emission rates, the unit g of molecules was used according to UNECE, (2015) and IPCC (2006). An emission on an area basis was applied for NH_3, N_2O, CO_2 and H_2S. For CH_4, the emission relative to the bulk volume was employed. Due to the availability of numerous additional records, data relative to the area were also provided for CH_4. For the area and the volume, the unit m^2

Table 1

Parameters extracted from the papers after transformation or standardization and transferred into the database. Explanations are given for parameters marked with symbols in the table footnote. The complete extracted data are provided in the Supplementary data 2.

Parameter	Explanation
Year	Date the study was published
Country	Location where the study was done
Slurry type	Cattle or pig
Slurry treatment	Untreated, solid-liquid separation, anaerobic digestion, acidification, aeration, addition of additives, dilution with water, addition of co-substrates (also denoted off-farm materials; mostly organic residues from e.g. food industry or energy crops) and combinations of treatments (e.g. solid-liquid separation and anaerobic digestion)
Slurry characteristics	Chemical analysis of the slurry: dry matter (DM), volatile solids (VS), total nitrogen (N_{tot}), ammonium (NH_4^+; TAN (total ammoniacal nitrogen) is often used instead of NH_4^+), total carbon (C), total sulfur (S) in $g\,L^{-1}$, pH
Type of study*	Farm-scale, pilot-scale, laboratory-scale
Type of store	For farm-scale studies: tank, lagoon according to Pain and Menzi (2011)**
Replicates	Number of replicates of real-world stores or experimental vessels
Store characteristics	Investigated store surface (m^2), depth (m), and volume (m^3); agitation of slurry (number of agitation events); other producer events or meteorological conditions; slurry temperature (°C)
Experimental conditions	Duration of storage of investigated slurry (days); duration of the study (days); number of measurement periods and total duration of the measurement (hours); season of measurements: cold, temperate, warm; for the determination of the season, the meteorological winter, spring or fall and summer were considered
Meteorological conditions	Air temperature during measurements (°C); air speed over the emitting surface during measurements ($m\,s^{-1}$); rainfall (cumulative amount during measurements in mm)
Measurement methods applied	Measurement method for the gases: dispersion modeling based on a backward Lagrangian stochastic (bLS) dispersion model or UK-ADMS atmospheric dispersion model, flux chamber method, flux gradient method, micrometeorological mass balance method (e.g. integrated horizontal flux, IHF; vertical radial plume mapping, VRPM), sampling at exhaust chimney, tracer gas method, method not further defined; instrument used for the concentration measurements of the gases
Cover type	Storage uncovered or covered; For covered storage: cover type according to VanderZaag et al. (2015): impermeable structural covers: lid (wood or concrete), tent covering; impermeable floating covers: plastic film; permeable synthetic floating covers: plastic fabrics, expanded clay, other materials such as expanded polystyrene, plastic tiles; permeable natural floating covers: peat, straw, vegetable oil, other organic materials (wood chips, sawdust etc.), other cover types such as storage bag
Occurrence of a natural crust at the store's surface	Formation of natural crust: yes or no, crust thickness (cm), time for natural crust formation (days)***
Measurement data****	NH_3 ($g\,NH_3\,m^{-2}\,h^{-1}$, $g\,NH_3\,m^{-3}\,h^{-1}$, $g\,NH_3\,AU^{-1}\,h^{-1}$), NH_3-N in % TAN and in % N, N_2O ($g\,N_2O\,m^{-2}\,h^{-1}$, $g\,N_2O\,m^{-3}\,h^{-1}$, $g\,N_2O\,AU^{-1}\,h^{-1}$), N_2O-N in % TAN and % N, CH_4 ($g\,CH_4\,m^{-2}\,h^{-1}$, $g\,CH_4\,m^{-3}\,h^{-1}$, $g\,CH_4\,AU^{-1}\,h^{-1}$), CH_4-C % VS, CO_2 ($g\,CO_2\,m^{-2}\,h^{-1}$, $g\,CO_2\,m^{-3}\,h^{-1}$, $g\,CO_2\,AU^{-1}\,h^{-1}$), CO_2-C in % VS, CO_2eq ($g\,CO_2eq\,m^{-2}\,h^{-1}$, $g\,CO_2eq\,m^{-3}\,h^{-1}$, $g\,CO_2eq\,AU^{-1}\,h^{-1}$), H_2S ($g\,H_2S\,m^{-2}\,h^{-1}$, $g\,H_2S\,m^{-3}\,h^{-1}$, $g\,H_2S\,AU^{-1}\,h^{-1}$); Difference between untreated and treated slurry or between slurry stored uncovered and stored covered in percent for NH_3, N_2O, CH_4, CO_2, CO_2eq, H_2S

* Type of study: Farm-scale: measurements carried out at real-world storage facilities at a farm site. This information could be obtained from the description of the experimental setup given in the papers. Pilot-scale and laboratory-scale: measurements conducted under controlled conditions in experimental vessels. Due to a lack of definition for these study types, a discrimination according to the following characteristics was employed: Pilot-scale: volume of slurry investigated: ≥ 500 L with experimental vessels situated outdoors, with or without a shelter and submitted to ambient meteorological conditions. Laboratory-scale: volume of slurry investigated: < 500 L. Most of the studies defined as laboratory-scale studies were conducted indoors in a temperature-controlled room. Three studies deviated from the conditions regarding study situation or temperature control and for four studies, this information was not available (Supplementary data 2). Despite these gaps in information, the studies were retained.

** A tank is a large, normally open-top, in most cases circular vessel made from pre-fabricated vitreous enameled steel, concrete or wood panels charged from a reception pit and emptied using a pump. It is a facility constructed at or below ground level and may extend above ground with a depth of several meters. Earthen storage basins not designed for biological treatment of slurry are considered as stores equivalent to tanks. Like earthen storage basins, a lagoon is a large rectangular or square shaped structure with sloping earth bank walls and may be lined with water impermeable material. Lagoons are designed for both storage and biological treatment (Pain and Menzi, 2011). They are not emptied below a specific depth necessary for slurry treatment except for maintenance (Hamilton et al., 2001).

*** We did not consider a natural crust as a mitigation technique equivalent to covering of slurry stores. The significance of crusting and considerations regarding distinction between crusting and storage covering are specifically addressed in Section 4.2.4.

**** For units: see Section 2.3. Acronyms: AU: animal unit = animal with a live weight of 500 kg; CO_2eq: carbon dioxide equivalent. CO_2eq is a standardized unit for different greenhouse gases. The numbers reported rest on data provided by the authors of the papers which were mostly based on IPCC (2007); TAN: total ammoniacal nitrogen; N: nitrogen; VS: volatile solids.

and m^3 was used, respectively. For all gases, the time unit hour was applied (reasons are given in section 4.1). Where useful for inventories, the time unit year was additionally provided for emissions. In this paper, the emission data standardized as explained above are denoted emission on an area or volume basis.

Since emission inventories do usually not apply emissions on an area or volume basis but emission factors which express emissions as a proportion of a compound present in the slurry store, data were additionally scaled as follows: percent of TAN for ammonia (EEA, 2016), percent of N for N_2O (IPCC, 2006) and percent of VS for CH_4 (IPCC, 2006) and CO_2. To be consistent with the notion "emission on an area or volume basis" regarding terminology, we used the term flow-based emission. Flow-based emissions were either taken from the papers or determined based on the emission rate, the N, TAN or VS content of the slurry, the volume of the store and the duration of the experiment. Dividing the cumulative emission which was derived from the emission rate and the duration of the study by the amount of the compounds present in the store at the beginning of the experiment (derived from the slurry content of N, TAN or VS and the slurry volume) yielded the flow-based emission. It was only calculated if no slurry addition or discharge occurred during the experiment.

2.4. Data analysis

2.4.1. Descriptive statistics of the emission data

In a first step, descriptive statistics (number of records, minimum, 1 st quantile, median, average, 3rd quantile, maximum, standard deviation) were calculated over all records encompassing slurry stored uncovered. There were eight categories for data reporting resulting from the combination of two slurry types (cattle and pig) with four study types (farm-scale lagoon comprising solid-liquid separation and biological treatment of slurry; farm-scale tank, pilot-scale and laboratory-scale which include untreated slurry).

2.4.2. Baseline emissions
2.4.2.1. Definition.
We define the term baseline emission as the average emission occurring with slurry storage according to the reference technology without emission control similar to VanderZaag et al. (2015). This implies uncovered storage in the following types of store: i) tanks or earthen stores without slurry treatment; ii) lagoons with solid-liquid separation and biological treatment occurring during storage (Hamilton et al., 2001). The baseline emission is considered as representative for average emissions over the whole course of a year. According to EEA (2016), baseline emissions are given separately for cattle and pig slurry. We further distinguished between storage in tanks (or earthen stores) and lagoons. Baseline emissions were calculated from uncovered slurry stores regardless of the occurrence of a natural crust because its formation can be only partially controlled and thus varies widely between stores (Smith et al., 2007). Moreover, there was insufficient information about the presence of crusts in the data impeding a distinction between crusted and non-crusted store's surfaces.

2.4.2.2. Determination.
Baseline emissions were calculated using farm-scale and pilot-scale studies published in peer-review papers. For the calculation of representative emissions, important influencing factors should be considered such as the meteorological conditions (mainly air temperature, wind speed, precipitation) and operations at storage facilities (Sommer et al., 2013). Among these factors, we were able to include air temperature since the season used for emission measurements which can be used as surrogate for the temperature was available for more than 90% of the records. Records were dropped where conditions prevailed which are not representative for slurry storage in practice over a longer period, e.g. if daily agitation of slurry occurred. More detailed information on meteorological conditions and operations at storage facilities was not available and could not be

included in the evaluation of emissions (e.g. only approx. 60% of records provided numerical air temperature data). Information on wind speed, precipitation and crust formation was available for less than half of the records.

We hypothesized that for generating emission data which are representative over the whole course of a year, emission values generated during the cold, the warm and the temperate season (spring, fall) should be equally covered. To achieve this, a weighting of the emission data for season was done. Values were aggregated according to the categorization "Season code" ("c": cold season = winter, "t": temperate season = spring or fall, "w": warm season = summer, "c,t": cold and temperate season, "c,w": cold and warm season, "t,w": temperate and warm season, "c,t,w": cold, temperate and warm season), "Slurry type" (cattle, pig), "Type of study" (farm-scale, pilot-scale) and "Type of store" (for farm-scale studies: lagoon, tank). For some papers, emission values for each individual season "c", "t", and "w" were provided and also the average value over the year, i.e. the "c,t,w" value. In these cases, the "c,t,w" value is denoted as redundant in the database (Supplementary data 2). It was used for the further calculations and not the values of the individual seasons. The aggregated values were averaged afterwards in the following manner:

i) Study duration varied considerably, i.e. individual experiments ranged from less than one day up to several months. The individual records were thus weighted according to measurement durations of records within each "Season code" category. The individual records were aggregated to four classes of measurement durations: a) ≥ 1 month, b) ≥ 1 week to < 1 month, c) ≥ 1 day to < 1 week, d) < 1 day. Weighting was done based on the square-root of the median of the measurement duration for each class to avoid over-emphasis of long-term measurements. The median values of the measurement duration for the 4 classes a, b, c and d were 146.5 days, 16.9 days, 4.5 days and 0.34 days, respectively. This implied the following respective weights 12.1, 4.1, 2.1 and 0.6. Therefore, a record based on a measurement of more than one month received a weighting 20.8 times higher than a record based on a measurement over less than a day.

ii) Average values for each season "c", "t" and "w" were calculated from all available values within one category (based on "Slurry type", "Type of study" and "Type of store"). Averaging was done in a way that values spanning over more than one season were attributed to the respective seasons, i.e. a value for "c,t" was attributed half to "c" and half to "t", a value of "c,t,w" was counted one fourth to seasons "c" and "w" and one half to season "t". For example, to average a "c" value based on a 2 weeks measurement (c_{2weeks}), a "c" value based on a 2 days measurement (c_{2days}) and a "c,w" value that based on a 2 months measurement ($cw_{2months}$) led to the following average "c" value: "c_{avg}" = (c_{2weeks}*$weight_{2weeks}$ + c_{2days}*$weight_{2days}$ + $cw_{2months}$*0.5*$weight_{2months}$)/($weight_{2weeks}$ + $weight_{2days}$ + 0.5*$weight_{2months}$).

iii) These average values were further averaged to annual emission rates "c,t,w" by weighting the value for season code "t" twice as high as the seasons "c" and "w" (i.e. "c,t,w_{avg}" = ¼*"c" + ½*"t" + ¼*"w") since the temperate season code "t" includes two seasons (spring and fall). These final averaged values are listed in column "Avg" in Tables 8, 9 and Supplementary data 4.

Numbers for baseline emissions are reported as average emission values if at least one record for each of the season "c", "t" and "w" was available. Included can be a record from an individual season (i.e. "c", "t" or "w"), or any kind of seasons combination (i.e. "c,t" "c,w" "t,w" or "c,t,w"). The lower and upper 95% confidence bounds (l95, u95) for baseline emissions were determined using bias-corrected and accelerated bootstrap intervals (Efron, 1987) if at least three individual records for each of the season "c", "t" and "w" were available. Again, this can be in the form of an individual season or any kind of seasons

combination as for the calculation of the average. The bootstrapping was done as non-parametric bootstrapping with sampling stratified by season. To test whether there are significant ($p < 0.05$) differences in these baseline emission values, 95% confidence intervals were obtained from bootstrapping the differences between each combination of values. If a confidence interval of a difference did not include 0, the difference was marked as statistically significant.

The data resulting from this procedure related to emissions on an area or volume basis were aggregated according to the slurry type (cattle and pig) and the study types farm-scale tank and pilot-scale and for the two study types combined which were denoted as baseline emissions tank. The baseline emissions for lagoons are based on measurements carried out at farm-scale for lagoons. Baseline emissions expressed as flow-based emissions were given separately for cattle and pig slurry for tanks only due to a lack of appropriate data for lagoons. The calculation procedure is additionally illustrated based on an example in the Supplementary data 9.

2.4.3. Emissions and emission changes due to slurry treatment and covering of slurry stores

We determined the emission changes due to slurry treatment techniques and covering of slurry stores using records with a treatment or a cover and a reference system (uncovered storage with untreated slurry) to compare the emissions on an area or volume basis from both. Due to the limited number of available records, the restriction to peer review papers and exclusion of laboratory-scale studies was not applied. For storage covering, all records with less than 20 cm of slurry depth were excluded from the data analysis since it is likely that such conditions differ too much from the real-world and even more evident if the thickness of the cover material is similar to that of the bulk slurry layer. Studies where slurry depth was not provided were excluded.

Although a natural crust is often listed as abatement measure together with slurry store covers (Bittman et al., 2014) we did not consider it as a mitigation technique equivalent to covering of slurry stores. In contrast to coverings such as impermeable floating covers, it is not applicable for all stores since it does not form at each slurry type. Crusting was neither considered for the analysis on emission changes due to slurry treatment and covering of slurry stores because of insufficient information about the presence of crusts in the experimental data. The significance of crusting is specifically addressed in Section 4.2.4.

The numbers from different studies were aggregated without a weighting for season or measurement duration due to the limited number of records. We tested whether the differences between treatments or covers and the reference system (untreated slurry or uncovered storage) were significantly different from zero by a two-sided t-test.

3. Results

3.1. Characterization of the database

3.1.1. General characteristics

The literature review yielded a total of 711 records. Among them, 13% were from before 2000. The period between 2000 and 2010 contributed 43%, and 44% were published after 2010 (Table 2). US and CA generated 28% and 19%, respectively, of the records while 11 European countries provided 48%. Two countries from Asia and Oceania contributed 3% and 2% of the records. Ammonia was studied in 38% of the records, while 59% were on GHGs, and 3% on H_2S. Among GHGs, CH_4 was most often investigated with a share of 30% of all records. 47% of the records included one gas and 53% several gases.

Table 3 shows the types of studies. A share of 46% of the records are based on studies conducted at farm-scale. Pilot-scale studies contributed 31% and laboratory-scale 23% of the records. Records from pilot-scale studies are similarly represented over all three periods

before 2000, between 2000 and 2010 and after 2010. In contrast, data from farm-scale studies and conducted in the laboratory occur more frequently from 2000 onwards.

An overview of the investigated slurry types is shown in Table 4. Cattle and pig slurry each account for about 50% of the investigated slurries. Cattle slurry mostly originated from dairy cows while for pig slurry fattening pigs and breeding pigs or a mixture of both was studied. Other types of slurry were included in measurements as well, but these occur much less. The proportion of untreated slurries is 65% and 87% for cattle slurry and pig slurry, respectively. Solid-liquid separation occurs for 16% (cattle slurry) and 3.7% (pig slurry) of the records. Anaerobic digestion of unseparated slurry applies for 7.2% (cattle slurry) and 3.7% (pig slurry) of the records while for anaerobically digested and separated slurry, the numbers are 8.1% for cattle slurry and 0.6% for pig slurry, respectively. Other treatments encompass acidification, aeration, supplementation with additives or dilution of slurry, but these treatments occur less.

Approximately 140 records compare the emissions between covered and uncovered storage. More than 80% of these data are from pilot and laboratory studies. Straw covers and other natural materials such as wood chips or maize stalks were most often investigated (51 in total). Also, cover types such as a lid, plastic film and fabrics were frequently addressed resulting in approximately 15 records for each.

Measurement methods employed in the experiments are shown in Table 5. Roughly, two thirds of all measurements were carried out using a flux chamber method. While this is almost the only option for pilot- and laboratory-scale studies, this system was also used for approximately 30% of the measurements conducted at farm sites. Methods like dispersion modeling or micrometeorological mass balance method make up about 60% of the records from farm-scale studies. Other methods e.g. using a tracer gas were rarely applied.

Slurry analyses are shown in Table 6. Not all studies provided analytical data of the slurry (e.g., only 84% of NH_3 studies presented TAN values). While most laboratory studies analyzed TAN, only 67% of the studies carried out at farm sites reported this parameter. Pilot-scale studies lie in between with 92% of records reporting TAN data. The availability of analytical data is similar for other parameters (e.g. DM) as for TAN but with somewhat lower numbers. The composition of the mixture of urine and feces as excreted by animals published by ASAE (2005) and Richner et al. (2017) is added at the bottom of Table 6. They provide numbers for cattle on DM, VS and TAN in the range of 80 to 90 g L^{-1}, 53 to 70 g L^{-1} and 1.4 to 2.1 g L^{-1}, respectively. For pigs, the values for DM, VS and TAN are in the range of 50 to 90 g L^{-1}, 36 g L^{-1} and 3.4 to 5.0 g L^{-1}. The slurry analyses given in the records show substantially lower numbers for DM and VS contents for untreated slurries which is most likely due to dilution with water from farm operation and rainfall at the farms (Table 6). Studies at farm-scale based on tanks, at pilot-scale and at laboratory-scale exhibit DM contents which are in a similar range within cattle and pig slurry. Numbers for DM are lower for pig slurries compared to cattle slurry except for laboratory scale studies. Pig slurry exhibits higher N_{tot} and TAN contents than cattle slurry. Within farm-scale studies, the numbers for all analytes strongly differ between slurry from tanks and from lagoons. Values for DM, VS, N_{tot} and TAN are lower for lagoons by a factor of approximately two to eight as compared to slurry stored in tanks. Slurries from lagoons compare better with slurries after solid-liquid separation (Table 6) than with untreated slurries.

3.1.2. Descriptive statistics of emission data from cattle and pig slurry stored uncovered

Descriptive statistics are shown in Table 7 for NH_3, N_2O, CH_4, CO_2 and H_2S over all records encompassing untreated cattle and pig slurry stored uncovered from studies conducted at farm-, pilot- and laboratory-scale (farm-scale studies with lagoons include biologically treated and separated slurry; see section 2.4.2.1). Data from measurements conducted during warm, temperate and cold seasons are unevenly

Table 2

Number of records listed by country and year of publication and share of total records by country.

Country	Before 2000		2000 to 2010		After 2010		total			Share of total
	Cattle	Pig	Cattle	Pig	Cattle	Pig	Cattle	Pig	Cattle and pig	
AT	0	0	15	6	0	0	15	6	21	3%
AU	0	0	0	0	0	4	0	4	4	1%
CA	1	1	47	10	72	5	120	16	136	19%
CN	0	0	0	0	0	17	0	17	17	2%
DE	8	9	4	2	0	0	12	11	23	3%
DK	20	17	0	0	6*	14	22	35	57	8%
ES	0	0	0	0	1	0	1	0	1	0.1%
FR	0	0	2	33	0	6	2	39	41	6%
IT	0	0	23	32	12	12	35	44	79	11%
JP	0	0	0	0	3	0	3	0	3	0.4%
LT	0	0	0	0	0	21	0	21	21	3%
NL	13	11	4	4	0	0	17	15	32	5%
NZ	6	0	1	1	2	0	9	1	10	1%
PT	0	0	4	0	4	8	8	8	16	2%
SE	0	0	1	0	6	3	7	3	10	1%
UK	1	3	13	2	6	13	20	18	38	5%
US	1	3	13	86	74	25	88	114	202	28%
Total	50	44	127	176	182	132	359	352	711	100%
Share of total	13%		43%		44%		100%			

* Cattle slurry with addition of other types of manure and feedstock materials.

Table 3

Number of records classified by type of study (farm-scale, pilot-scale, laboratory-scale) and time periods of publication and in percent of the total.

Type of study	Before 2000	2000 - 2010	After 2010	Total	Share of study types
Farm-scale	27	157	141	325	46%
Pilot-scale	54	75	90	219	31%
Laboratory-scale	13	77	77	167	23%
Total	94	309	308	711	100%
Publication of study types over time (in percent of total)					
Farm-scale	8%	48%	43%	100%	
Pilot-scale	25%	34%	41%	100%	
Laboratory-scale	8%	46%	46%	100%	

Table 4

Overview on investigated slurry types stored uncovered or covered: number of records listed by slurry treatments, slurry types and share of the total records in percent.

Slurry treatment	Cattle n	Pig	Other*	Cattle Percent of total	Pig	Other*
Untreated	233	302	–	65%	87%	–
Solid-liquid separation	57	13	–	16%	3.7%	–
Anaerobic digestion	26	13	–	7.2%	3.7%	–
Anaerobic digestion, solid-liquid separation	29	2	4	8.1%	0.6%	100%
Acidification	5	3	–	1.4%	0.9%	–
Acidification, anaerobic digestion	–	2	–	–	0.6%	–
Acidification, anaerobic digestion, solid-liquid separation	–	1	–	–	0.3%	–
Acidification, solid-liquid separation	–	1	–	–	0.3%	–
Dilution	5	2	–	1.4%	0.6%	–
Addition of additives	3	3	–	0.8%	0.9%	–
Aeration	1	4	–	0.3%	1.1%	–
Aerobic treatment	–	2	–	–	0.6%	–
Total	359	348	4	100%	100%	100%

* Cattle slurry with addition of other types of manure and feedstock materials.

Table 5

Number of records classified by the measuring method and by the type of study.

Measuring method	Farm-scale	Pilot-scale	Laboratory-scale	Total	
Dispersion modeling based on bLS* or ADMS**	107	2		109	15%
Dispersion modeling based on bLS* and VRPM***	8			8	1.1%
Flux chamber method	98	213	167	478	67%
Flux gradient method	4			4	0.8%
Micrometeorological mass balance method	92			92	13%
Sampling at exhaust chimney	4			4	0.6%
Tracer gas method	7			7	1.0%
Method not defined	5	4		9	1.3%
Total	325	219	167	711	100%

* backward Lagrangian stochastic (bLS) dispersion model.
** UK-ADMS atmospheric dispersion model (Hill et al., 2008).
*** Vertical Radial Plume Mapping.

distributed over all records (Supplementary data 3). The minimum and maximum emission values differ by one to several orders of magnitude for all gases. The average often exceeds the median by a factor of two or more which is most pronounced for N_2O. This indicates a distribution of data being right skewed by high values. The variability of data and the occurrence of high maximum values is most pronounced for laboratory-scale studies. Striking high values exceeding the median by at least one order of magnitude for NH_3, CH_4 and CO_2 are reported in the laboratory-scale study of Guarino et al. (2006). For N_2O, high values were found from three studies conducted at farm- and pilot-scale (Clemens et al., 2006; Amon et al., 2007; Leytem et al., 2011) (Supplementary data 2,8,11). For H_2S, one figure from a laboratory-scale study stands out which exceeds all other values by two orders of magnitude (Hobbs et al., 1999).

Table 6

Number of records of a slurry type (cattle, pig), type of study (f: farm-scale; p: pilot-scale; l: laboratory-scale), type of store for farm-scale studies, and slurry treatment (untreated, sol-liq sep: solid-liquid separation) in the database. Number of records (n) with analytical data on DM, VS, N_{tot}, TAN and average contents of DM, VS, N_{tot}, TAN in g L^{-1} for untreated slurry.

Slurry type	Type of study*	Type of store	Slurry treatment**	Total number of records	DM	VS	N_{tot}	TAN	DM	VS	N_{tot}	TAN
				n	n				g L^{-1}			
Cattle	f	lagoon	untreated	73	19	7	13	14	17	3.7	1.2	0.2
Cattle	f	tank	untreated	39	21	9	19	25	67	48	3.1	1.5
Cattle	p		untreated	106	97	36	93	97	62	53	3.2	1.6
Cattle	l		untreated	35	31	24	29	31	57	43	3.0	1.3
Pig	f	lagoon	untreated	109	19	23	50	76	9.7	4.5	0.8	0.6
Pig	f	tank	untreated	55	35	9	33	35	42	37	3.3	1.9
Pig	p		untreated	63	56	30	56	54	50	33	4.6	3.2
Pig	l		untreated	68	68	43	64	64	59	56	4.7	2.9
Cattle	f tank, p, l		sol-liq sep	23	19	10	17	17	39	29	2.4	1.2
Pig	f tank, p, l		sol-liq sep	14	10	5	8	12	29	23	3.8	2.3
Cattle	Contents of mixture of urine and feces obtained from ASAE (2005)								80	53	3.0	1.4
Pig									61-90	n.a.	4.7-7.0	3.4-5.0
Cattle	Contents of mixture of urine and feces obtained from Richner et al. (2017)								90	70	3.9	2.1
Pig									50	36	6.5	4.6

n.a.: not available.

* f: farm-scale; p: pilot-scale; l: laboratory-scale.

** sol-liq sep: solid-liquid separation.

Table 7

Emissions from cattle and pig slurry stored uncovered in tanks at farm-scale, pilot-scale and laboratory-scale without slurry treatment and in lagoons with solid-liquid separation and biological treatment; descriptive statistics for NH_3, N_2O, CH_4, CO_2 and H_2S in g m^{-2} h^{-1} or g m^{-3} h^{-1}. n: number of records; Min: minimum; 1 st Qu: first quartile; 3 st Qu: third quartile; Max: maximum; Std: standard deviation. Additional information is provided in Supplementary data 3.

Slurry type	Study type		n	Min	1 st Qu	Median	Average	3rd Qu	Max	Std
				NH_3 g m^{-2} h^{-1}						
Cattle	Farm-scale	lagoon	35	< 0.01	0.10	0.13	0.21	0.36	0.11	
Cattle	Farm-scale	tank	20	0.02	0.04	0.06	0.13	0.16	0.68	0.15
Cattle	Pilot-scale		53	< 0.01	0.03	0.07	0.09	0.11	0.44	0.08
Cattle	Laboratory-scale		19	< 0.01	0.02	0.04	0.26	0.33	1.4	0.43
Pig	Farm-scale	lagoon	74	< 0.01	0.04	0.08	0.15	0.18	0.68	0.18
Pig	Farm-scale	tank	23	0.03	0.06	0.10	0.22	0.27	1.0	0.26
Pig	Pilot-scale		22	0.01	0.06	0.20	0.24	0.26	0.92	0.23
Pig	Laboratory-scale		20	< 0.01	0.03	0.23	0.69	0.71	4.5	1.16
				N_2O g m^{-2} h^{-1}						
Cattle	Farm-scale	lagoon	13	< 0.001	< 0.001	< 0.001	0.002	0.001	0.02	0.006
Cattle	Farm-scale	tank	3	< 0.001	0.001	0.002	0.002	0.003	0.003	0.002
Cattle	Pilot-scale		46	< 0.001	< 0.001	0.001	0.003	0.004	0.04	0.007
Cattle	Laboratory-scale		6	< 0.001	< 0.001	< 0.001	0.005	0.001	0.03	0.01
Pig	Farm-scale	lagoon	6	< 0.001	< 0.001	< 0.001	0.003	0.002	0.01	0.005
Pig	Farm-scale	tank	5	Not detected						
Pig	Pilot-scale		17	< 0.001	< 0.001	< 0.001	0.01	0.001	0.06	0.02
Pig	Laboratory-scale		4	< 0.001	< 0.001	< 0.001	0.003	0.003	0.01	0.006
				CH_4 g m^{-3} h^{-1}						
Cattle	Farm-scale	lagoon	3	0.27	0.29	0.30	0.77	1.0	1.7	0.83
Cattle	Farm-scale	tank	7	< 0.01	0.26	0.75	0.83	1.3	1.9	0.71
Cattle	Pilot-scale		46	0.01	0.07	0.42	0.56	0.75	3.6	0.69
Cattle	Laboratory-scale		15	< 0.01	0.15	0.64	10	16	51	16
Pig	Farm-scale	lagoon	2	< 0.01	0.88	1.8	1.8	2.6	3.5	2.5
Pig	Farm-scale	tank	10	0.02	0.25	0.55	1.6	3.1	5.0	1.8
Pig	Pilot-scale		21	0.01	0.13	0.18	0.77	1.0	3.4	1.1
Pig	Laboratory-scale		18	0.02	1.3	2.9	7.4	6.6	33	10
				CO_2 g m^{-2} h^{-1}						
Cattle	Farm-scale	lagoon	18	0.27	1.9	2.3	4.7	5.3	27	6.4
Cattle	Farm-scale	tank	3	11	11	11	16	18	25	8.1
Cattle	Pilot-scale		15	0.17	2.8	4.3	5.6	6.3	21	5.2
Cattle	Laboratory-scale		14	0.45	2.4	8.0	86	189	332	120
Pig	Farm-scale	lagoon	7	< 0.01	< 0.01	0.03	0.89	0.74	4.7	1.8
Pig	Farm-scale	tank	1	5.7	5.7	5.7	5.7	5.7	5.7	–
Pig	Pilot-scale		7	3.2	3.6	4.4	6.6	9.0	13	4.1
Pig	Laboratory-scale		14	1.0	6.3	9.1	52	80	217	75
				H_2S g m^{-2} h^{-1}						
Cattle	Farm-scale	lagoon	3	0.02	0.04	0.06	0.05	0.07	0.07	0.03
Cattle	Laboratory-scale		3	< 0.01	< 0.01	0.01	0.01	0.01	0.02	0.01
Pig	Farm-scale	lagoon	14	< 0.01	< 0.01	< 0.01	0.01	0.03	0.08	0.02
Pig	Laboratory-scale		6	< 0.01	< 0.01	< 0.01	0.47	0.02	2.8	1.1

Table 8

Emissions on an area or volume basis from cattle and pig slurry stored uncovered in tanks at pilot-scale and at farm-scale without slurry treatment and in lagoons with solid-liquid separation and biological treatment. Baseline emissions for storage in tanks and lagoons given in g m^{-2} h^{-1} / kg m^{-2} y^{-1} for NH$_3$, N$_2$O and CO$_2$ and in g CH$_4$ m^{-3} h^{-1} / kg CH$_4$ m^{-3} y^{-1}. n: number of records after aggregation; Avg: average; l95, u95: lower and upper 95% confidence bounds; cells denoted with "-": value is not available; #: values denoted with different letters are significantly different ($p < 0.05$). Detailed information is provided in Supplementary data 4.

Slurry type	Study type/baseline emissions		n	Avg	l95	u95	#	Avg yearly amount
				NH$_3$ g m^{-2} h^{-1}				NH$_3$ kg m^{-2} y^{-1}
Cattle	Pilot-scale studies		34	0.08	0.07	0.09	a	–
Cattle	Farm-scale studies	tank	11	0.09	0.05	0.13	ab	–
Cattle	**Baseline emissions***	**lagoon**	**28**	**0.12**	**0.10**	**0.15**	**bc**	**1.1**
Cattle	**Baseline emissions****	**tank**	**45**	**0.08**	**0.07**	**0.09**	**a**	**0.67**
Pig	Pilot-scale studies		15	0.24	0.15	0.38	def	–
Pig	Farm-scale studies	tank	8	0.23	0.13	0.37	cdef	–
Pig	**Baseline emissions***	**lagoon**	**40**	**0.15**	**0.12**	**0.19**	**ce**	**1.3**
Pig	**Baseline emissions****	**tank**	**23**	**0.24**	**0.17**	**0.34**	**f**	**2.1**
				N$_2$O g m^{-2} h^{-1}				N$_2$O kg m^{-2} y^{-1}
Cattle	Pilot-scale studies		33	0.002	0.001	0.002	a	–
Cattle	Farm-scale studies	tank	–	–	–	–	–	–
Cattle	**Baseline emissions***	**lagoon**	**11**	**< 0.001**	**–**	**–**	**–**	**< 0.01**
Cattle	**Baseline emissions****	**tank**	**35**	**0.002**	**0.001**	**0.002**	**a**	**0.02**
Pig	Pilot-scale studies		12	0.002	< 0.001	0.005	a	–
Pig	Farm-scale studies	tank	2	< 0.001	–	–	–	–
Pig	**Baseline emissions***	**lagoon**	**–**	**–**	**–**	**–**	**–**	**–**
Pig	**Baseline emissions****	**tank**	**14**	**0.002**	**< 0.001**	**0.005**	**a**	**0.01**
				CH$_4$ g m^{-3} h^{-1}				CH$_4$ kg m^{-3} y^{-1}
Cattle	Pilot-scale studies		35	0.49	0.38	0.70	a	–
Cattle	Farm-scale studies	tank	6	1.2	0.88	1.5	b	–
Cattle	**Baseline emissions***	**lagoon**	**3**	**0.95**	**0.40**	**1.5**	**ab**	**8.3**
Cattle	**Baseline emissions****	**tank**	**41**	**0.58**	**0.46**	**0.76**	**a**	**5.1**
Pig	Pilot-scale studies		16	0.67	0.38	1.1	a	–
Pig	Farm-scale studies	tank	3	0.76	–	–	–	–
Pig	**Baseline emissions***	**lagoon**	**1**	**3.5**	**–**	**–**	**–**	**31**
Pig	**Baseline emissions****	**tank**	**19**	**0.68**	**0.41**	**1.1**	**a**	**6.0**
				CO$_2$ g m^{-2} h^{-1}				CO$_2$ kg m^{-2} y^{-1}
Cattle	Pilot-scale studies		6	7.0	–	–	–	–
Cattle	Farm-scale studies	tank	–	–	–	–	–	–
Cattle	**Baseline emissions***	**lagoon**	**14**	**6.6**	**2.6**	**17**	**–**	**58**
Cattle	**Baseline emissions****	**tank**	**8**	**8.0**	**–**	**–**	**–**	**70**
Pig	Pilot-scale studies		4	8.8	–	–	–	–
Pig	Farm-scale studies	tank	1	5.7	–	–	–	–
Pig	**Baseline emissions***	**lagoon**	**3**	**0.30**	**–**	**–**	**–**	**2.7**
Pig	**Baseline emissions****	**tank**	**5**	**8.0**	**–**	**–**	**–**	**70**

Cells denoted with "-": value is not available.

  * Baseline emissions lagoon are entirely based on values from farm-scale studies lagoon.

  ** Based on the average from studies at farm-scale tank and pilot-scale.

3.2. Baseline emissions

3.2.1. Emissions on an area or volume basis

Table 8 shows emissions on an area or volume basis from cattle and pig slurry stored uncovered in tanks at farm-scale and at pilot-scale without slurry treatment and in lagoons with solid-liquid separation and biological treatment for NH$_3$, N$_2$O, CH$_4$ and CO$_2$. Average NH$_3$ emissions from farm-scale studies conducted at lagoons are higher than those from tanks for cattle slurry but lower for pig slurry. Pilot-scale studies exhibit similar emissions as farm-scale studies conducted at tanks, but they differ when compared to measurements from lagoons. The range between the lower and upper 95% confidence bounds is relatively small for cattle slurry but large for pig slurry with the greatest range for farm-scale studies from tanks (0.13 to 0.37 g NH$_3$ m^{-2} h^{-1}). The baseline emission for lagoons is 0.12 g NH$_3$ m^{-2} h^{-1} and 0.15 g NH$_3$ m^{-2} h^{-1} for cattle and pig slurry, and for tanks 0.08 g NH$_3$ m^{-2} h^{-1} and 0.24 g NH$_3$ m^{-2} h^{-1} for cattle and pig slurry, respectively. Baseline emissions given as a yearly average emitted amount for lagoons are 1.1 kg NH$_3$ m^{-2} y^{-1} and 1.3 kg NH$_3$ m^{-2} y^{-1} for cattle and pig slurry, and for tanks 0.67 kg NH$_3$ m^{-2} y^{-1} and 2.1 kg NH$_3$ m^{-2} y^{-1} for cattle and pig slurry, respectively. The differences between baseline emissions for cattle slurry and pig slurry, and the difference between lagoons and tanks are both statistically significant ($p < 0.05$).

Values for N$_2$O emissions mostly originate from pilot-scale studies. The data from the three studies which exhibit high values mentioned in section 3.1.2 were excluded for the calculation of baseline emissions. The N$_2$O losses shown in Table 8 are very low and often close to the limit of detection. Negative fluxes are reported e.g. in VanderZaag et al. (2009) or values lower than the limit of detection in Misselbrook et al. (2016). Pig slurry exhibits a large range between the lower and upper 95% confidence bounds (< 0.001-0.005 g N$_2$O m^{-2} h^{-1}). Baseline emissions are 0.002 g N$_2$O m^{-2} h^{-1} for cattle and pig slurry stored in tanks. Storage in lagoons for cattle slurry is 0.0003 g N$_2$O m^{-2} h^{-1} while for pig slurry no baseline value is available. Statistically significant differences were not found for N$_2$O.

Farm-scale studies exhibit higher CH$_4$ emissions than pilot-scale studies (Table 8). For both study types, pig slurry has a higher emission level as compared to cattle slurry. The baseline emission values for lagoons are 0.95 g CH$_4$ m^{-3} h^{-1} (cattle slurry) and 3.5 g CH$_4$ m^{-3} h^{-1} (pig slurry), and for tanks 0.58 g CH$_4$ m^{-3} h^{-1} (cattle slurry) and 0.68 g CH$_4$ m^{-3} h^{-1} (pig slurry), respectively. The baseline emission for lagoon storage of pig slurry is based on one record only. But its distinctly higher emission level as compared to the baseline for tank storage and relative to the baseline emissions of cattle slurry stored in lagoons and tanks is confirmed by the area based CH$_4$ emissions where the data basis is much larger and statistically significant differences ($p < 0.05$) were found (Supplementary data 4).

For CO$_2$, the number of observations is relatively small. Some studies exhibit high values for cattle slurry which are greater than 20 g CO$_2$ m^{-2} h^{-1} (Leytem et al., 2011; Minato et al., 2013; Misselbrook

Table 9

Flow based baseline emissions for tanks from untreated cattle and pig slurry stored uncovered for NH_3 given in percent of total ammoniacal nitrogen (TAN), N_2O in percent of nitrogen (N), CH_4 and CO_2 in percent of volatile solids (VS). The average (Avg) and the lower and upper 95% confidence bounds (l95, u95) are shown. The numbers are mainly based on pilot-scale studies. Cells denoted with "-": value is not available; #: values denoted with different letters are significantly different ($p < 0.05$). Detailed information is provided in Supplementary data 4.

	n	Avg	l95	u95	#
		NH_3% TAN			
Cattle	31	16%	14%	19%	a
Pig	17	15%	9.2%	23%	a
		N_2O% N			
Cattle	16	0.13%	0.08%	0.18%	a
Pig	8	0.10%	0.01%	0.18%	a
		CH_4% VS			
Cattle	27	2.9%	2.3%	3.7%	a
Pig	14	4.7%	2.1%	10%	a
		CO_2% VS			
Cattle	4	11%	–	–	–
Pig	3	9.2%	–	–	–

Cells denoted with "-": value is not available.

et al., 2016). The baseline emissions for lagoon storage are 6.6 g CO_2 m^{-2} h^{-1} and 0.30 g CO_2 m^{-2} h^{-1} for cattle and pig slurry, respectively, and for tank storage, 8.0 g CO_2 m^{-2} h^{-1} for both slurry types. Data on H_2S emission are sparse and a calculation of baseline emissions is only feasible for lagoon storage which are 0.04 g H_2S m^{-2} h^{-1} for cattle slurry and 0.01 g H_2S m^{-2} h^{-1} for pig slurry (Supplementary data 4).

3.2.2. Flow-based emissions

Flow-based emissions, i.e. emissions given in percent of TAN, N or VS present in the store are shown for NH_3, N_2O, CH_4 and CO_2 in Table 9. Almost all data originate from pilot-scale studies (Supplementary data 4) which can be used for baseline emissions for tanks but not for lagoons. Baseline emission values for NH_3 are 16% of TAN for cattle slurry and 15% of TAN for pig slurry, respectively. N_2O emissions are 0.13% of N for cattle slurry and 0.10% of N for pig slurry. Baseline emissions for CH_4 are 2.9% of VS for cattle slurry and 4.7% of VS for pig slurry. Emissions for CO_2 reach 11% of VS and 9.2% of VS for cattle and pig slurry, respectively, but the data basis is limited. The ranges between the lower and upper 95% confidence bounds are large in most cases and are partially skewed to high values, especially for N_2O and CH_4 from pig slurry. There were no statistically significant differences.

3.3. Emission changes due to slurry treatments

Acidification clearly reduces NH_3 emissions by ca. 70% during

storage compared to untreated cattle and pig slurry (Table 10). The effect is even higher for CH_4 (61%–96%) but lower for CO_2. For NH_3 and CH_4, the differences are statistically significant ($p < 0.05$). An emission reduction also occurs for digested slurries and slurries after solid-liquid separation combined with acidification for all gases except for N_2O (Supplementary data 6). In contrast, the emissions are enhanced for N_2O emissions compared to untreated cattle and pig slurry although limited data are available and the differences not statistically significant. Data on H_2S emissions are sparse. Fangueiro et al. (2015) state in their review that H_2S emissions were either unaffected or decreased following acidification.

The number of studies on emission changes due to anaerobic digestion is limited. Where more than one observation is available, both an increase and a decrease in emissions occur for storage after anaerobic digestion as compared to untreated slurry (Supplementary data 6). NH_3 and N_2O exhibit on average greater emissions from anaerobically digested slurry. Most studies comparing anaerobically digested and untreated slurry exhibit lower emissions of CH_4 for the former. An emission increase is observed for N_2O and CO_2 for pig slurry, although this is based on only one observation for both gases. Statistically significant differences do not occur for anaerobic digestion.

Average NH_3 emissions during storage from the liquid fraction are significantly ($p < 0.05$) higher as compared to untreated cattle slurry (Table 10). But for pig slurry, only a slight effect of solid-liquid separation on NH_3 release can be observed which is statistically insignificant. CH_4 and CO_2 exhibit lower emissions from the liquid fraction as compared to untreated slurry with a statistically significant difference for CH_4. A statistically significant reduction ($p < 0.05$) in N_2O emissions occurs for cattle slurry. But the release of N_2O is greater for pig slurry compared to untreated slurry where the difference is statistically not significant.

Five studies examined the effect of slurry dilution with water and found an average reduction of all investigated gases in the range of approximately 30–50%. Statistically significant effects occurred for cattle slurry for NH_3 and N_2O. Maximum abatement effects of 88% and 86% were found for N_2O and CH_4, respectively (Supplementary data 6).

3.4. Emission changes due to covering of slurry stores

The average NH_3 emission percent reduction due to covers ranges between approximately 50% up to ca. 90% for most cover types (Table 11). However, the variability of values is large. Minimum values can be around 15% and maximums higher than 95% (Supplementary data 7). The emission mitigation does not systematically differ between cattle and pig slurry on a percentage basis. Emission reductions lie in a similar range for structural covers, impermeable floating covers, permeable floating covers and the other cover materials. The differences are statistically significant ($p < 0.05$) for the following covers and

Table 10

Percentage emission change (i.e. % change of emissions on an area or volume basis) during storage due to acidification, anaerobic digestion, solid-liquid separation and dilution of cattle and pig slurry for NH_3, N_2O, CH_4 and CO_2 relative to untreated slurry. Positive figures indicate a decline, negative numbers an increase in emissions. n: number of records, Avg: average emission change; Std: standard deviation; cells denoted with "-": value is not available. Detailed information is provided in Supplementary data 6.

		NH_3			N_2O			CH_4			CO_2		
		n	Avg	Std	n	Avg	Std	n	Avg	Std	n	Avg	Std
Acidification	Cattle	5	71%*	17%	1	−4%	–	5	61%*	36%	5	7%	23%
	Pig	3	77%*	22%	1	−39%	–	3	96%*	3%	1	67%	–
Anaerobic digestion	Cattle	3	−59%	64%	3	−16%	29%	5	−2%	129%	1	53%	–
	Pig	1	45%	–	1	−363%	–	1	99%	–	1	−22%	–
Solid-liquid separation	Cattle	12	−23%*	21%	6	43%*	36%	10	32%*	27%	7	18%	24%
	Pig	7	−1%	18%	1	−258%	–	7	39%*	39%	5	13%	12%
Dilution	Cattle	5	48%*	29%	5	57%*	38%	5	39%	33%	–	–	–
	Pig	–	–	–	–	–	–	2	47%	15%	2	30%	11%

Cells denoted with "-": value is not available.

* Numbers with an asterisk indicate a statistically significant difference ($p < 0.05$) between the treated and the untreated slurry.

Table 11

Percentage emission change (i.e. % change of emissions on an area or volume basis) from storage of cattle and pig slurry due to different types of covers relative to uncovered storage for NH_3, N_2O, CH_4, CO_2 and H_2S. Positive figures indicate a decline, negative numbers an increase in emissions. n: number of records, Avg: average emission change; Std: standard deviation; cells denoted with "-": value is not available. Detailed information is provided in Supplementary data 7.

		Slurry type	NH_3			N_2O			CH_4			CO_2			H_2S		
			n	Avg	Std	n	Avg	Std	n	Avg	Std	n	Avg	Std	n	Avg	Std
Impermeable structural covers	Lid (wood or concrete)	Cattle	6	73%*	29%	2	−4%	23%	2	15%	2%	–	–	–	–	–	–
		Pig	7	64%*	35%	4	31%	56%	4	45%*	17%	–	–	–	–	–	–
	Tent covering	Cattle	2	77%	9%	–	–	–	–	–	–	–	–	–	–	–	–
		Pig	2	89%	7%	–	–	–	–	–	–	–	–	–	–	–	–
Impermeable synthetic floating covers	Plastic film	Cattle	4	66%*	22%	–	–	–	–	–	–	–	–	–	–	–	–
		Pig	6	88%*	18%	2	100%	0%	2	62%	54%	–	–	–	–	–	–
Permeable synthetic floating covers	Plastic fabrics	Cattle	1	89%	–	1	68%	–	1	−2%	–	1	15%	–	–	–	–
		Pig	5	39%*	15%	–	–	–	3	−17%	18%	–	–	–	4	50%*	20%
	Expanded clay	Cattle	4	59%	39%	–	–	–	2	11%	7%	2	0.1%	1%	–	–	–
		Pig	12	74%*	20%	1	−8%	–	6	8%	17%	5	29%*	8%	–	–	–
	Expanded polystyrene	Cattle	2	79%	2%	–	–	–	–	–	–	–	–	–	–	–	–
		Pig	4	64%*	32%	–	–	–	2	−26%	41%	2	26%	35%	–	–	–
	Plastic tiles	Cattle	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
		P	2	88%	11%	1	−7%	–	1	25%	–	–	–	–	–	–	–
Permeable natural floating covers	Peat	Cattle	2	90%	13%	–	–	–	–	–	–	–	–	–	–	–	–
		Pig	3	59%	31%	–	–	–	1	−33%	–	1	−31%	–	–	–	–
	Straw cover	Cattle	8	71%*	19%	2	−79%	30%	4	3%	30%	4	−6%	10%	–	–	–
		Pig	8	73%*	22%	–	–	–	7	0.2%	36%	2	13%	9%	–	–	–
	Other organic material#	Cattle	4	51%*	32%	–	–	–	4	−13%	37%	4	−46%	71%	–	–	–
		Pig	4	45%	44%	–	–	–	4	−9%	37%	4	20%	17%	–	–	–
	Vegetable oil	Cattle	4	71%*	16%	–	–	–	2	39%	6%	2	27%	9%	–	–	–
		Pig	4	94%*	10%	–	–	–	2	11%	2%	–	–	–	–	–	–

Cells denoted with "-": value is not available; # materials like maize stalks or wood chips; cells denoted with "-": value is not available.

* Numbers with an asterisk indicate a statistically significant difference ($p < 0.05$) between storage with a cover and uncovered storage.

both slurry types: lid, plastic film, straw cover, vegetable oil; other organic materials for cattle slurry; plastic fabrics, expanded clay, expanded polystyrene for pig slurry.

For N_2O, an increase in emissions is observed in many cases. But reduced emissions occur as well (Supplementary data 7). However, the number of records providing emission changes from slurry storage due to store covers is sparse and the effects are statistically insignificant. CH_4 emissions being lower by approximately 10% to 60% occur for impermeable covers (lid and plastic film), plastic tiles and vegetable oil compared to uncovered storage (Table 11). For plastic fabrics, expanded polystyrene and peat, the emissions are higher by 2% to 33%. The other cover types (expanded clay, straw and organic materials such as corn stalks or wood chips) show both increases and reductions in CH_4 emission (Supplementary data 7). On average, CH_4 emissions from slurry stores covered with permeable materials moderately differ in emission levels as compared to uncovered storage. The differences in CH_4 emissions are statistically not significant ($p < 0.05$) except for pig slurry covered with a lid. Stores covered with plastic fabrics, expanded clay and expanded polystyrene emit less CO_2 while higher emissions are observed for peat and straw covers than for the uncovered controls, but the differences are statistically not significant. Plastic fabrics induce a significant ($p < 0.05$) emission reduction for pig slurry by 50% for H_2S. Data on both CO_2 and H_2S emissions are sparse.

4. Discussion

4.1. Variability in emissions

The high variability of emission levels as shown by descriptive statistics (Table 7) may be due to different meteorological conditions, disturbance of the slurry surface induced by operations at the stores and slurry characteristics. The enhanced variability in laboratory-scale studies compared to the other study types is striking. In laboratory-scale studies, the environment is expected to be largely uniform since the experiments were mostly conducted in a temperature-controlled room with ambient temperatures lying in a narrow range and the slurry being undisturbed. As most of the laboratory-scale studies aimed at a

comparison of different techniques or systems, the representativeness of the resulting emission rates for real-world conditions was not the primary focus and discrepancies between different approaches are very likely present. A thorough evaluation of potential biases of the laboratory studies is not possible due to missing information on the measuring systems and is beyond the scope of this paper (see Liu et al. (2020) and the related discussion). Also, for other study types, the occurrence of methodological biases cannot be ruled out which may lead to implausible results. Detection of striking values might be hampered due to the multitude in units used in the papers. Therefore, a standardization as used here and providing guide values are important issues. For this, a favorable option is the unit g m^{-2} h^{-1} or g m^{-3} h^{-1} of a molecule. It is equally suitable to illustrate an emission pattern within one day, also in combination with important influencing factors such as temperature or wind speed which can change over short time periods and to compare them with e.g. average emissions over one year. If a yearly amount of a gas release is required, data can be obtained from Table 8. Alternatively, the unit mol m^{-2} h^{-1} could be used to facilitate the comparability between different molecules, even if to date, it is generally not used in the context of emission inventories.

4.2. Important factors influencing emissions

The relevance of important influencing factors on emissions from slurry stores is discussed in this section in order to support interpretation and understanding of the data used to determine baseline emissions and emission changes due to slurry treatment and coverage of slurry stores. It should be noted that a part of these influencing factors could not be included in the data processing such as the weighting or the statistical analysis of emission data due to insufficient information in the records. This data limitation applied for operations at stores, the meteorological parameters rain and wind speed and the natural crust.

4.2.1. Type of slurry

Records from the same study where both cattle and pig slurry have been investigated using the same approach were compared. Eight studies (De Bode, 1991; Sommer et al., 1993; Husted, 1994; Kaharabata

et al., 1998; Balsari et al., 2007; Dinuccio et al., 2008; Mosquera et al., 2010; Misselbrook et al., 2016; Baral et al., 2018) with a total of 14, 2, 8 and 3 pairs of records on NH_3, N_2O, CH_4 and CO_2 emissions, respectively, were available. For NH_3, 85% of data pairs, exhibited higher emissions for pig slurry than for cattle slurry. Similar for N_2O, CH_4 and CO_2, pig slurry exceeds emissions of cattle slurry in most cases. These findings agree with the data reported in Table 8 (except for CO_2) and with data previously published by Sommer et al. (2006) and VanderZaag et al. (2015).

4.2.2. Operations at stores

Operations at the storage tank, such as agitation, filling and removing of slurry are necessarily related to real-world storage systems. Their effects are usually reflected in farm-scale measurements using non-intrusive methods but rarely included in pilot studies or farm-scale studies using chamber systems. A series of studies specifically investigated such processes (see Supplementary data 10). They showed consistent results and provided evidence that disturbance of the manure surface due to slurry agitation, filling and discharging of the stores induces large episodic emissions for NH_3, CH_4, CO_2 but not for N_2O. While emissions of CH_4 and CO_2 rapidly decline after cessation of the operations and can even drop to levels below the previously undisturbed stores, increased emission levels persist for NH_3. Due to the relatively short time duration of agitation over the year and the subsequent drop below average levels for CH_4, this operation per se does not substantially contribute to annual NH_3 and GHG emissions from slurry (VanderZaag et al., 2009). A more detailed overview on emissions during and following operations at stores is given in the Supplementary data 10.

4.2.3. Meteorological conditions

Increasing air temperature and wind speed enhance the emissions since they directly affect diffusion and convection of gases near the emitting surfaces (Sommer et al., 2013). The relationship between the temperature as represented by the season of measurements and the emission level could be demonstrated in the present study (Supplementary data 5). It must be considered however, that the air temperature is a simplistic surrogate for the slurry temperature which is a determinant factor for GHG emissions. Rennie et al. (2018) demonstrated that slurry store design and operations (i.e. filling level, agitation) influence the slurry temperature and the emission level of gases such as CH_4. In 25 studies, slurry temperatures during different seasons are available. Slurry temperatures increase as expected in the order cold < temperate < warm for 94% of the cases. The effects of temperature and wind speed are not discussed further in the present study because this topic has been previously covered by e.g. Ni (1999) or Sommer et al. (2006). In contrast, emission changes related to the influence of rain events and thawing of the slurry surface are summarized here since they have been less frequently addressed in the literature. Petersen et al. (2013) found lower NH_3 emission from uncovered storage of pig slurry with precipitation than from the treatment without rain although the differences are not statistically significant. It was shown that ammonia emissions can decline towards zero during rain events after slurry spreading (Hafner et al., 2019) due to sorption of NH_3 onto wet surfaces. Moreover, the TAN-concentration at the emitting surface may decrease with precipitation due to dilution or transport of TAN from a crusted slurry surface into the bulk liquid. Overall, it can thus be assumed that NH_3 emissions from slurry storage during rain events are low. In contrast, an increase in emissions of CH_4 has been observed. Balde et al. (2016b) reported average emissions of 1.8 g CH_4 m^{-2} h^{-1} for digested slurry while peak emissions during rain events reached 10 g CH_4 m^{-2} h^{-1}. This was likely due to bursting of bubbles at or near the surface. Elevated emissions were also observed by Balde et al. (2016a) from storage of the liquid fraction of cattle slurry which confirms earlier findings from Kaharabata et al. (1998) and Minato et al. (2013) on slurry stored in open tanks or lagoons. Kaharabata et al.

(1998) suggested that the emission increase is due to more disturbance at the slurry surface induced by rain and thus enhancement of the CH_4 exchange through the liquid surface area and of incidental outburst of gas bubbles (ebullition). Petersen et al. (2013) found a drop of N_2O emissions to zero as a result of rewetting of the crust after rainfall inducing a shift towards anaerobic conditions. Grant and Boehm (2015) did not find a relationship between H_2S emissions and rain events.

VanderZaag et al. (2011) observed important bubble flux events in late winter/early spring that coincided with surface thawing which were probably due to a release of previously produced CH_4 that was trapped under the frozen slurry surface. In the study of VanderZaag et al. (2010a) which encompassed winter and spring, N_2O release was only recorded during spring thaw. A moderate increase in CH_4 emissions was observed during the same period at slurry temperatures above 0 °C while NH_3 and CO_2 flows were unaffected by spring thaw. Elevated CH_4 emissions due to thawing of the manure store were also reported by Leytem et al. (2017).

4.2.4. Natural crust

There is agreement that crusting impacts the gas release in many ways: enhanced resistance to mass transfer (Olesen and Sommer, 1993), oxidation of NH_3 (Nielsen et al., 2010) and CH_4 (Petersen et al., 2005) and formation of N_2O related to nitrification and denitrification occurring in liquid–air interfaces near air-filled pores present in crusts (Petersen and Miller, 2006). Several studies investigated the effect of a natural crust on the emission level (Sommer et al., 1993; Misselbrook et al., 2005; Aguerre et al., 2012; Wood et al., 2012). All studies showed that a natural crust provided an efficient barrier leading to an emission reduction for NH_3. Baldé et al. (2018) confirmed these findings by measurements conducted under farm conditions at tanks and earthen basins containing slurries with differing ability to form natural crusts (i.e. raw cattle slurry, the liquid fraction of cattle slurry produced by solid-liquid separation, digested slurry with and without solid-liquid separation). They confirmed that slurry stored with a thick surface had lower NH_3 losses. Grant and Boehm (2015) found that crusting of a lagoon surface containing dairy cow slurry reduced NH_3 but not H_2S emissions. Nielsen et al. (2010) showed that NH_3-oxidizing bacteria may contribute to a significant reduction of NH_3 emissions if a natural crust is present on a slurry store. Grant and Boehm (2018) found emissions from a tank containing pig slurry to be greater by 10% when the surface was covered with a crust than without crusting (difference not statistically significant). They explained their findings by the higher TAN content of the crusted slurry surface as compared to a non-crusted one. Sommer et al. (2000) and Husted (1994) found higher emissions of CH_4 from slurry without than from slurry with a natural surface crust. Wood et al. (2012) investigated the emissions from dairy slurry with varying DM contents and thus natural crusts with different thicknesses and coverage of the storage surfaces. They were not able to relate the CH_4 fluxes to the presence of a natural crust. N_2O production was found to be enhanced after build-up of a natural crust (VanderZaag et al., 2009).

In the literature (e.g. Vanderzaag et al., 2015), a natural crust is often classified as abatement measure for NH_3 similar as slurry store covers. However, crust formation can only be controlled to a limited extent. Crusts are of variable thickness, coverage of the store and durability. Their effectiveness for emission abatement has therefore be considered as inconsistent (Vanderzaag et al., 2015). Crusts only develop for slurry types with a high content of fibrous material (Bittman et al., 2014). This applies mainly for cattle slurry (Smith et al., 2007) and less for pig slurry (Sommer et al., 2006). Crusting is likely to occur at a slurry DM content of more than 20 g L^{-1} (Sommer et al., 2006; Wood et al., 2012) which mostly does not apply for slurry stored in lagoons (Table 6). Consequently, they have much less ability to form a natural crust as shown by e.g. Balde et al. (2018).

We therefore did not consider crusting as an emission mitigation technique equivalent to slurry store covers but rather as a parameter

influencing emissions from stored slurry and thus excluded it from the analysis of emission changes due to covering of stores. But we stress that if a natural crust is present, it is likely to significantly contribute to an emission reduction and, therefore, should be preserved by e.g. reducing slurry agitation and addition of manure below the surface.

The limited information in the data impedes our ability to clearly distinguish between crusted and non-crusted stores. This may be relevant for (i) the calculation of baseline emissions and (ii) emissions changes due to slurry treatments and covering of stores: (i) baseline emissions determined here may include stores with variable occurrence of a natural crust. For lagoons, information on crusting was available for 45% (cattle slurry) and 19% (pig slurry) of the records, respectively. Among these, 62% of the lagoons containing cattle slurry were fully or partly covered by a crust during the emission measurements. For pig slurry, this applies for 21% only. Among records used for the determination of baseline emissions for tanks, 78% and 50% included information regarding crusting for cattle and pig slurry, respectively. Of these, 83% (cattle slurry) and 48% (pig slurry) had a fully or partly crusted surface. This complies with findings that crusting occurs less on lagoon surfaces and stores containing pig slurry. The proportion of crust occurrence for cattle slurry stored in tanks is in line with earlier findings (Smith et al., 2007). We thus suggest that the baseline emissions determined here are based on studies which appropriately reflect the range of store surface crusts occurring at farms. (ii) A natural crust may occur in combination with a storage cover and thereby be enhanced (Chadwick et al., 2011) since the slurry surface is less exposed to wind turbulence. In experiments comparing uncovered and covered storage, it is thus difficult to stringently distinguish between the effect of covering and of crusting. Moreover, this information is not always available: only 60% of the records used to determine the emission change due to covering included information on crusting. From these, about half had crusted surfaces and the other half not. This might partly explain the variability of emission changes due to covering found here. These considerations should be taken into account for the discussion in the Sections 4.4 and 4.6.

4.3. Study types to be included for baseline emissions

Data should only be included for the calculation of baseline emissions if they can be considered as representative or typical for gas flows occurring at farm conditions. In principle, this applies for farm-scale studies. Pilot-scale studies imply some aspects of farm-scale studies due to measurements conducted in outdoor facilities and a slurry volume in the order of several cubic meters. But there are concerns extrapolating data from pilot-scale studies to real-world systems. VanderZaag et al. (2009, 2010a, 2010b) who performed pilot-scale studies state that although measured fluxes were reported, emission trends and treatment differences or temporal trends were the focus of their analysis. Moreover, almost all pilot-scale studies are based on flux chambers. VanderZaag et al. (2010b) argued that steady-state chambers alter the enclosed environment and concluded that absolute fluxes measured might deviate from emissions that would occur without chambers. Nevertheless, several studies conducted in pilot-scale facilities similar to that of VanderZaag et al. (2009, 2010a, 2010b) quantified emissions of NH_3 and GHGs and derived emission factors for slurry storage (e.g. Amon et al., 2006, 2007; Rodhe et al., 2012). Petersen et al. (2009) presented a pilot-scale facility and suggested to use the obtained results for better documentation of emission data for GHG and ammonia inventories. Pilot-scale studies have occasionally been conducted with simulation of real-world conditions by including mixing of the slurry or filling of tanks during the experiment (VanderZaag et al., 2009; Rodhe et al., 2012).

Emission peaks for CH_4 were observed in several studies (VanderZaag et al., 2011; Balde et al., 2016a) due to ebullition. They may remain unrecorded (Rodhe et al., 2012) unless the gas measurements are continuous with a high temporal resolution. This

shortcoming may apply for pilot-scale studies where e.g. a flow chamber is used which is moved between several experimental tanks (e.g. Amon et al., 2006). Intermittent gas sampling can hamper measurements at a farm-scale as well. Grant et al. (2015) assumed differences in emission levels between two locations due to under-sampling of ebullition events given the short measurement periods. Sampling large storage areas using chambers might be hampered if the sampled surface areas are not representative for the entire store. Balde et al. (2016b) found average emissions of CH_4 measured at an earthen storage containing liquid digestate with a floating chamber which were about four-fold greater than measured at the same time with a non-intrusive bLS technique. The authors explained this by the limited area covered by the chamber and by disturbances induced by the chamber causing bubble formation and bursting thereby increasing emissions.

To summarize, it can be hypothesized that farm-scale measurements using non-intrusive methods are a preferential option. Still, data from such studies are limited at present time. Therefore, inclusion of records from pilot-scale and farm-scale studies appears to be the best opportunity for the determination of baseline emissions. This approach provides a larger data basis as if only farm-scale studies were included. Moreover, Table 8 shows that emissions from pilot-scale studies comply with farm-scale studies tank for NH_3 but less for CH_4. On the other hand, we excluded laboratory-scale studies for the determination of baseline emissions. They are mostly not designed for generating emission rates. Their experimental conditions strongly deviate from an environment that occurs under practical conditions. The enhanced variability found in emissions level from laboratory-scale studies (section 3.1.2) points to severe methodological shortcomings which might bias baseline values.

4.4. Baseline emissions

4.4.1. Emissions on an area or volume basis

NH_3 emissions from pig slurry are higher as expected due to its higher TAN content and its lower ability to form a natural crust compared to cattle slurry. Sommer et al. (2006) and VanderZaag et al. (2015) suggested lower emissions on an area basis from pig slurry stored in lagoons than from storage in tanks. This complies with the results of this study (Table 8). Lagoons are the prevailing system for slurry storage in the US (Sorensen et al., 2013). They usually have a greater surface area than tanks which would imply more exposure to the ambient air turbulence suggesting a higher emission potential. Slurries from lagoons have on average a lower dry matter and TAN content as compared to tanks. This might be due to a stronger dilution with water: e.g. five out of six lagoons investigated by Leytem et al. (2017) collected parlor wash water and not slurry from a pit of a livestock housing. We assume that solid-liquid-separation was applied at the farms studied which have lagoons although this was not always clearly defined in the papers. This is supported by the low contents in DM and TAN in slurry from lagoons as shown in Table 6. The lower solids content would enhance the emission potential due to less ability for formation of a natural crust at the slurry surface (Wood et al., 2012). But the lower TAN content induces the opposite effect on NH_3 emissions (Sommer et al., 2006). The overall impact of these effects combined on the emission level is difficult to assess. The present data suggest higher emissions from lagoons than from tanks containing cattle slurry but the opposite for pig slurry.

The baseline emissions (lagoons: 0.12 g NH_3 m^{-2} h^{-1} and 0.15 g NH_3 m^{-2} h^{-1} for cattle and pig slurry, tanks: 0.08 g NH_3 m^{-2} h^{-1} and 0.24 g NH_3 m^{-2} h^{-1} for cattle and pig slurry, respectively), are mostly lower than numbers given by VanderZaag et al. (2015), Sommer et al. (2006) and Bittman et al. (2014). VanderZaag et al. (2015) suggested emissions for crusted and non-crusted cattle slurry of 0.11 and 0.19 g NH_3 m^{-2} h^{-1}, respectively, from tanks or lagoons. For pig slurry stored in a lagoon, they give 0.12 g NH_3 m^{-2} h^{-1}, and stored in a tank, 0.40 g NH_3 m^{-2} h^{-1}. Sommer et al. (2006) provided similar values. Bittman

et al. (2014) gave baseline emissions between 0.19 and 0.40 g NH_3 m^{-2} h^{-1}. They attributed the lower value to slurry which is frozen in the store for several months, and the higher value applies to warm countries. For N_2O, most studies exhibit emissions clearly below 0.01 g N_2O m^{-2} h^{-1} (Supplementary data 8). In contrast, three papers reach values from 0.02 to 0.06 g N_2O m^{-2} h^{-1} and N_2O losses ranging between 25% and 160% of the NH_3 emissions determined concomitantly (Clemens et al., 2006; Amon et al., 2007; Leytem et al., 2011). Unless at very low levels of NH_3 emissions, flows of both NH_3 and N_2O in the same order of magnitude do not occur in other records and have not been reported in the livestock sector (e.g. EEA, 2016). Therefore, the data from the three studies were excluded for the calculation of baseline emissions. If they were kept, the baseline emissions for N_2O would be higher by a factor of two and three for cattle and pig slurry stored in tanks, respectively. Chadwick et al. (2011) stated in their review that N_2O emission from slurry stores without a surface cover are negligible which supports the baseline emissions shown in Table 8.

For CH_4, higher emissions occur for farm-scale studies than for pilot-scale studies (Table 8; statistically significant differences for emissions on an area basis; $p < 0.05$; Supplementary data 4). This could be due to the temperature dependency of methanogenesis (Elsgaard et al., 2016). Pilot-scale studies exhibit lower slurry volumes as compared to farm-scale stores which suggests faster cooling of the slurry and therefore a lower methane conversion rate. Another reason could be the batch-filling of vessels used for pilot-scale studies which differs from continuous filling and incomplete removal of slurry at farm-scale stores. Under such conditions, aged slurry may act as inoculum which was shown to enhance emissions of CH_4 (Wood et al., 2014). Overall, the lower emission level for CH_4 measured at pilot-scale included for the determination of baseline emissions tank could lead to an underestimation thereof.

The review of Owen and Silver (2015) reported CH_4 emission data from lagoons and tanks of dairy systems being 2.3 and 2.7 g CH_4 m^{-2} h^{-1}, respectively. This is higher than data from farm-scale studies reported here which are 1.2 and 1.3 g CH_4 m^{-2} h^{-1} for cattle slurry stored in lagoons and tanks, respectively (Supplementary data 4). However, the data basis of Owen and Silver (2015) is smaller and measurements carried out in the warm season tend to be overrepresented. The higher CH_4 emissions from pig slurry as compared to cattle slurry are expected due to the higher methane production potential of pig slurry (Triolo et al., 2011). Both cattle and pig slurry exhibit lower losses from tanks than from lagoons. Moreover, lagoon storage produces a solid fraction which includes a large proportion of the slurry VS generating additional emissions. According to VanderZaag et al. (2010b), a CO_2-C:CH_4-C ratio of 50:50 is expected from stores. Looking at records which include emission data of both CH_4 and CO_2, a large variation occurs. The average CO_2-C:CH_4-C ratio is approximately 65:35 which also differs from the CH_4 to CO_2 relationship expected from anaerobic digestion of livestock slurries (ca. 55%–70% CH_4 content of dry biogas; Triolo et al., 2011). This could be due to a tendency for greater CO_2-C:CH_4-C ratios in pilot-scale studies which increases the average CO_2-C:CH_4-C ratio of all records included. The greater ratios were also linked to studies with low CH_4 fluxes. This is likely because pilot-scale studies had less ability to provide appropriate conditions for CH_4 production as mentioned above. On the other hand, CO_2 seems to be emitted more consistently in all studies.

As the aim of all studies considered for the calculation of baseline values in the present paper was the determination of emission rates we think that the baseline emissions are robust and reflect the current state of knowledge. But the confidence intervals shown in Table 8 may be substantial. This suggests an inherent variability in the systems which can be due to differing conditions regarding meteorological conditions, operations at stores and occurrence of a natural crust. Baseline values must be considered as average numbers. In a specific situation and e.g. for representative regional values, deviations from the presented baseline values can occur. Moreover, methodological biases cannot be

ruled out and different experimental approaches might entail systematic differences in results (e.g. possibly CH_4 emissions from pilot-scale studies). Such effects have been observed for experimental data on NH_3 emissions from slurry application (Hafner et al., 2018).

4.4.2. Flow-based emissions

The determined baseline emission values for NH_3 of 16% of TAN and 15% of TAN for cattle and pig slurry, respectively, which are mostly based on pilot-scale studies exhibit similar values as the emission factors of EEA (2016) which give 20% of TAN and 14% of TAN as Tier 2 default values for cattle and pig slurry. Data from farm-scale studies have comparable numbers for storage in tanks for cattle slurry: 16% of TAN (Baldé et al., 2018) and 13% of TAN (McGinn et al., 2008) but lower emissions for pig slurry (Dinuccio et al., 2012) with 2% and 5% of TAN. Flow-based emissions are not available for lagoons. IPCC (2006) and EEA (2016) suggest an N_2O emission factor being zero for slurry storage without a natural crust. For a crusted store, IPCC (2006) and EEA (2016) give EFs of 0.5% of N and 1% of TAN entering the store, respectively. These values are higher than the values determined in this study which are 0.13% of N and 0.10% of N for cattle and pig slurry, respectively (Table 9). The eight highest values for flow-based N_2O emissions originate from records that include slurry stores with a crust which supports the occurrence of N_2O emissions with crusted store surfaces. This complies with Sommer et al. (2000) who suggest that N_2O is produced in drying of natural crusts where aerobic and anaerobic zones exist. Drying enhances convection of liquid upward through the cover, where dissolved ammonium can be oxidized by nitrifying bacteria in an aerobic environment and under such conditions, molecules produced from nitrification can be denitrified. During ammonium oxidation and denitrification, N_2O is released as an intermediate or final product. At limited oxygen availability, formation of N_2O is enhanced.

For CH_4, a direct comparison between the suggested baseline emissions with default emission factors used in models for emission inventories is not possible. A simplified application of the approach of Mangino et al. (2001) and IPCC (2006) using the methane conversion factor (MCF) for slurry in a cool climate with an annual average temperature of 14 °C results in a CH_4 emission of ca. 4.0% of VS and 7.5% of VS for cattle slurry and pig slurry, respectively. These figures are somewhat higher than the baseline emissions suggested (Table 9) which are 2.9% of VS and 4.7% of VS for cattle and pig slurry, respectively. It should be noted that emission values for CH_4 derived from such model approaches can strongly deviate from measured values as shown by several studies (e.g. Kariyapperuma et al., 2018).

For the determination of flow-based emissions, analytical data of the slurry, the flow volume of slurry into storage and its residence time in the store must be known. Determining these three parameters is not straightforward which might explain the high degree of absence regarding flow-based emissions in farm-scale studies. This particularly applies for lagoons where extended slurry residence times, accumulation of solids over long time periods and repeated recycling of liquids used for flushing or recharging pits of livestock housings represent additional challenges. Generally, lagoons have a greater surface to volume ratio and longer slurry residence times as compared to tanks. These two factors will lead to higher flow-based emissions for lagoons as compared to tanks if identical emissions on an area basis are assumed for both storage systems.

For inventory purposes, emission factors could be calculated using the baseline emissions on an area or volume basis and an assumption of the surface or volume of the storage system, the average values for the residence time of the slurry in the store and the slurry contents of TAN, N or VS. These values are specific for different countries and production systems. A further assessment thereof is outside the scope of this paper. The calculation of flow-based emissions (and emission factors) is subject to additional uncertainties as compared to emissions on an area basis due to the requirement of further parameters.

4.5. Emission changes due to slurry treatments

The pH value has a strong effect on gaseous emissions from slurry stores (Sommer et al., 2013). This is appropriately reflected by the data on emission changes due to slurry acidification through addition of inorganic acids shown in Table 10. The variability in the achieved reduction is likely related to the degree of acidification and the different pH values in slurry (Dai and Blanes-Vidal, 2013). The emission reductions found for NH_3 and CH_4 are in line with the review of Fangueiro et al. (2015). Similarly, Petersen et al. (2012) observed significant reduction effects for NH_3 and CH_4 due to acidification. The data point at an increase in N_2O emission but this is based on limited data. Bastami et al. (2016) concluded that self-acidification of slurry induced by addition of substrates rich in carbon may be a promising alternative to slurry acidification using concentrated acids for abatement of CH_4 emissions. Additives other than acids to reduce gaseous emissions or odor nuisance from manure storage have been investigated in some studies (Martinez et al., 2003; Sun et al., 2014; Owusu-Twum et al., 2017). A clear emission reduction due to other additives did not occur. Similarly, Van der Stelt et al. (2007); Wheeler et al. (2011) and Holly and Larson (2017a) found little evidence that manure additives other than acids have a clear influence on the release of ammonia and GHGs. Still, individual investigations have shown an emission reduction potential for certain additives (Bastami et al., 2016).

The number of studies allowing a direct comparison of emissions from storage of untreated slurry and anaerobically digested slurry is limited since biogas plants are mostly fed with manure and off-farm organic feedstock material which hampers a direct comparison with unamended untreated slurry. The increase of NH_3 emissions due to anaerobic digestion complies with studies which include anaerobic digestion with addition of organic feedstock material. Baldé et al. (2018) measured NH_3 emissions from two stores at different farms containing untreated livestock slurry and liquid digestate obtained from livestock slurry and organic feedstock materials under farm conditions. Emissions from the untreated slurry were lower. Koirala et al. (2013) suggested that anaerobic digestion of dairy slurry significantly increased the NH_3 volatilization potential. The most important factor was the enhanced ammonium dissociation. Anaerobic digestion seems to reduce CH_4 emissions during slurry storage. Maldaner et al. (2018) found lower CH_4 losses from the liquid fraction of anaerobically digested slurry amended with organic feedstock material compared to unamended raw slurry from the same farm before the installation of anaerobic digestion. This is likely due to the reduction of the VS load after digestion but also a consequence of solids removal with solid-liquid separation of the major part of the digestate. Furthermore, Maldaner et al. (2018) suggested that VS remaining in the digestate was less degradable which leads to a reduced CH_4 production. VanderZaag et al. (2018) showed the CH_4 emission potential (B_0) from digestate was 35% lower than the B_0 of untreated manure. In contrast, Sommer et al. (2000) and Rodhe et al. (2015) measured higher emissions from anaerobically digested slurry as compared to untreated cattle slurry. They explained this by the presence of a larger and more active microbial community in digested slurry. However, most storage tanks are never completely empty. Residual aged slurry may act as inoculum and enhance the production of CH_4 (Sommer et al., 2007; Ngwabie et al., 2016). Although, the microbial population in aged slurry may be less efficient for methane production as compared to microbes present in anaerobically digested slurry, the higher amount of degradable organic carbon available in untreated slurry might compensate this. It can therefore be hypothesized that untreated slurries as occurring in real-world stores imply a higher potential for CH_4 emissions than anaerobically digested slurry.

Solid-liquid separation reduces the solids content of the slurry and thus the potential to develop a natural crust. This enhances NH_3 emissions during storage which complies with the increasing emissions of cattle slurry due to solid-liquid separation. Pig slurry exhibits a lower ability to form a natural crust which could explain why almost no effect of solid-liquid separation on NH_3 emissions can be observed (Table 10). Baldé et al. (2018) investigated the NH_3 emissions from two stores situated at different dairy farms containing untreated slurry and separated liquids under farm conditions. They measured higher emissions from the liquid fraction than from the untreated slurry and reported similar findings for the separated liquids from digestate derived from livestock slurry and organic feedstock materials. In contrast, Hjorth et al. (2009) found significantly higher NH_3 emissions from raw and digested slurries than from the corresponding liquid fractions. They explained their findings by the higher ammonium and N contents of the unseparated raw and pre-digested slurries compared with the liquid fractions, which increased the potential for NH_3 volatilization. The lower N_2O storage emissions of the liquid fraction for cattle slurry as compared to raw slurry is in line with the conclusions of the review paper published by Chadwick et al. (2011). The reduced emissions of CH_4 due to solid-liquid separation results from the reduction of the total solids content in the slurry which can be considered as a surrogate for the available VS pool. This leads to a lower amount of organic matter which can be degraded to CH_4 and CO_2 (Wood et al., 2012). However, yearly average CH_4 emissions of 1.4 g CH m^{-3} h^{-1} and 2.2 g CH m^{-3} h^{-1} for the first and second year of measurements were reported for dairy slurry from a farm-scale study conducted at a tank (Balde et al., 2016a). These numbers exceed the baseline emission and emissions from farm-scale studies from tanks for cattle slurry given in Table 8. Balde et al. (2016a) explained the elevated emission levels by the high biodegradability of the liquid fraction and the limited crust development. VanderZaag et al. (2018) showed that the speed of CH_4 production was much higher for the separated liquid fraction, compared to untreated slurry. Grant et al. (2015) found CH_4 emissions on an area basis from the liquid fraction of cattle slurry stored in a lagoon which are similar to the baseline emission for cattle slurry (Supplementary data 4). The discrepancy between the emission changes given in Table 10 and the high emissions found in these two studies is difficult to explain.

Dilution of slurry with water changes its DM content. DM of slurry can be considered as an indicator for the N/TAN- and VS-content which influences the potential production of NH_3, N_2O, CH_4 and CO_2, respectively (Wood et al., 2012). However, DM affects the formation of a natural crust as well (section 4.2.4). Overall, dilution leads to a reduction of all investigated gases which complies with the findings of Ni et al. (2010) for NH_3 and CO_2 and of Habetwold et al. (2017) for CH_4. But this conclusion is based on pilot-scale studies where the slurry volume is identical for the diluted and untreated slurry. Under practical conditions, addition of water leads to a higher amount of slurry. If the area of manure stores is thereby increased due to requiring larger storage capacities, the reduction might be overcompensated due to a rise in emitting surface (Ni et al., 2010).

Aeration of slurry is a technique which is used to remove excess N from slurries. It induces nitrification and denitrification that converts TAN in the slurry to nitrite/nitrate with the aim of a complete denitrification to N_2. If the process is not properly controlled aeration can produce substantial amounts of NH_3 and N_2O (Loyon et al., 2007). The effect of aeration was investigated in several laboratory- and pilot-scale studies. Amon et al. (2006) found a strong increase of NH_3 emissions by up to a factor of five. Molodovskaya et al. (2008) reported NH_3 emissions of up to 50% of total slurry N. Losses were increased at greater aeration rates. Many studies found a strong increase in emissions for N_2O with aeration (Beline et al., 1999; Beline and Martinez, 2002; Amon et al., 2006; Loyon et al., 2007). Low emissions for both NH_3 and N_2O were achieved from low flow phased oxic/anoxic treatment (Molodovskaya et al., 2008). A reduction of CH_4 by ca. 50% to almost 100% emissions was observed by Martinez et al. (2003) and Amon et al. (2006) if slurry aeration was applied. Concomitantly, CO_2 emissions were reduced (Martinez et al., 2003).

An increase in emissions of a specific gas during slurry storage due to a treatment technique does not necessarily indicate a conflict related

to emission mitigation. Enhanced losses during storage can be reduced by e.g. storage covering and might be overcompensated by reduced emissions during subsequent field application. The overarching goal of manure management is the reduction of gaseous losses between the excretion by livestock and uptake by arable and fodder crops. Therefore, the discussion on effects of slurry treatments on emissions from slurry storage must consider the context of good management practices along the whole manure management chain (Sajeev et al., 2018).

4.6. Emission changes due to coverage of slurry stores

Almost all types of covers induce a substantial emission reduction for NH₃ which complies with the review of VanderZaag et al. (2008). Emission reductions lie in a similar range for all categories of covers and for both cattle and pig slurry. This contrasts to Bittman et al. (2014) who give distinct values for the different cover types and lower values for "Low technology" floating covers such as permeable natural floating covers. VanderZaag et al. (2015) give an emission reduction of 80% for impermeable structural covers and for impermeable synthetic floating covers which is in the range of the values given in Table 11. A larger layer thickness of natural floating covers leads to a higher emission reduction (Guarino et al., 2006; VanderZaag et al., 2009). This is probably due to a more efficient barrier for the gas transport between the slurry and the ambient air (VanderZaag et al., 2009). Other cover types not included in the data analysis according to section 2.4.3 also efficiently reduce NH₃ emissions. Organic materials such as steam-treated wood or biochar were shown to exhibit similar effects as floating covers consisting of straw or peat (Holly and Larson, 2017b). Minerals like perlite or zeolite were also found to be efficient in NH₃ emission reduction (Hörnig et al., 1999; Portejoie et al., 2003).

The increase in emissions observed for N₂O in many cases agrees with the previously published literature (VanderZaag et al., 2008; Chadwick et al., 2011). Petersen et al. (2013) observed lower N₂O flows with a straw cover exposed to precipitation as compared to straw covers where wetting by precipitation was excluded. Sommer et al. (2000) suggested that N₂O is only produced in periods with drying surface layers. Storage covers can influence the formation of a natural crust (Chadwick et al., 2011). A natural surface crust on slurry can provide sites with aerobic conditions where nitrification occurs which produces N₂O (Sommer et al., 2000). Therefore, the variability in emission changes due to slurry storage covering could be driven by differing moisture contents of the manure surfaces and differing formation of a natural crust due to covering. However, the number of records related to emission changes for N₂O due to store covers is sparse and these findings can be uncertain.

The observed increase in CH₄ emissions for plastic fabrics, expanded polystyrene and peat complies with the review of VanderZaag et al. (2008). Straw covers provide additional carbon but might reduce ebullition and increase aerobic microbial activity at the upper storage layer (VanderZaag et al., 2009). This induces contrasting effects on CH₄ net emissions. VanderZaag et al. (2009) suggested that the reduction of CH₄ emissions due to a straw cover is related to areas in a crust where microbial breakdown of CH₄ might occur. It can be assumed that the enhanced CH₄ consumption overcompensates the increased potential for CH₄ production due to the additional carbon supply with straw. An opposite effect can occur if straw is incorporated into the bulk slurry during storage due to e.g. agitation. Petersen et al. (2013) found that an elevated CH₄ concentration in the gas phase above the slurry surface is required for a significant stimulation of methane oxidation. This would support the preponderant emission reduction found for impermeable covers. For other cover types, the CH₄ concentration above the emitting surface might have been inconsistent in the experiments which could explain the contrasting emission changes for CH₄ due to storage covering. Similar to NH₃, a larger layer thickness of straw covers leads to a higher emission reduction for CH₄ although the differences of the

emissions are low (Guarino et al., 2006; VanderZaag et al., 2009).

The contribution of the different gases to the total of GHG emissions is largest for CH₄ with a proportion of ca. 80% (VanderZaag et al., 2009; Petersen et al., 2013). Therefore, the changes of GHG equivalents due to effects of covers is moderate with a slight trend towards lower total GHG emissions.

VanderZaag et al. (2010b) have shown that a permeable synthetic floating cover was more efficient regarding emission reduction of NH₃ and CH₄ when slurry is agitated as compared to undisturbed slurry. VanderZaag et al. (2009) found that agitation increased NH₃ losses from straw covered tanks less than from the uncovered reference. CH₄ emissions from covered and control tanks were similarly changed.

Although found efficient in emission reduction, low cost floating covers such as straw covers are probably not efficient for emission mitigation in practice since they may be destroyed when the slurry surface is disturbed due to strong winds or operations at a store. Therefore, we consider impermeable structural covers or synthetic floating covers as most reliable for emission mitigation.

4.7. Recommendations for further research

The emission data provided in records from different studies range over several orders of magnitude even for the same slurry type, the same type of study and identical seasons of measurement. This may be partly due to varying conditions related to manure management and meteorological conditions occurring during the measurements. In addition, different study designs and measuring methods are likely to contribute to the variability in emissions. An important issue in future research should thus focus to identify and quantify potential experimental biases. This aspect requires the simultaneous use of independent approaches to determine emissions.

Farm-scale studies using non-intrusive methods such as micrometeorological mass balance (Wagner-Riddle et al., 2006) or dispersion modeling (Flesch et al., 2009) are likely to be a preferential option. Such approaches avoid interactions with emitting processes and determined emission rates best reflect the emissions occurring under conditions at farm-scale. They have the ability to cover large area sources (Gao et al., 2008) and can thus integrate the large inhomogeneity of emissions over space and time. For dispersion modeling, the limiting factor is the requirement of a simple topography allowing for representative turbulence measurements. Most of the micrometeorological methods require a minimum wind speed. Many sensors, e.g. for NH₃, have a minimum detection limit which may be higher than gas concentrations occurring under conditions with low emissions (Balde et al., 2019). Consequently, farm-scale studies using non-intrusive methods have a risk to overestimate the true average emissions (Baldé et al., 2018). This risk can be minimized by using recently developed sensors such as DOAS systems (Volten et al., 2012; Bell et al., 2017). Moreover, it would be important to quantify such potential biases by an assessment of gap filling procedures used for missing data due to e.g. non-detection at low concentration levels as done by Voglmeier et al. (2018). For reliable results from farm-scale studies, extended measurement periods are required covering all seasons of a year. Moreover, recent research has demonstrated that the history of the storage may play an important role for CH₄ emissions (Kariyapperuma et al., 2018) pointing at the necessity of measurement campaigns over several years for an adequate determination of representative emission rates. This implies a large effort in labor and costs. In addition, thorough recording of the operations at the storage facilities (agitation, filling, discharging of the stores by using e.g. a webcam, continuous measuring of the slurry volume stored), of crusting at the stores surface (thickness, structure, coverage of the surface), of slurry temperature at several depths, of meteorological conditions as well as slurry sampling and analyses are required. A few studies comply with these requirements (e.g. Baldé et al., 2018; Kariyapperuma et al., 2018). Still, collection of such data can be challenging or even hardly

feasible (e.g. representative slurry sampling at large lagoons). Also, operations at stores can largely differ between individual farms and consequently, it is difficult to select an experimental site at farm-scale which is appropriate to generate baseline emissions. Therefore, several measurement campaigns that consider the variety of different conditions occurring at slurry stores are required.

Pilot-scale studies are indispensable for studying principal mechanisms and influencing factors driving emissions or to evaluate the effectiveness of emission mitigation techniques. Facilities allowing for continuous measurements e.g. as presented by Petersen et al. (2009) are probably the best option. Further advantages of pilot-scale studies are the possibility to conduct experiments in replicates and a better control of the experimental conditions. There is also a potential to generate bases for modeling which could be used to complement data from farm-scale studies. Further progress for the quantification of emissions from slurry storage could be achieved by an analysis of individual measurement intervals from several experiments and model construction on this basis as e.g. done for slurry application by Hafner et al. (2019). The measurement intervals should include the relevant information regarding influencing factors. For this, we recommend that the researchers provide the emission data along with parameters as given in the Supplementary data 2. For indistinct parameters such as crusting, we suggest the elaboration of a standardized procedure to achieve a definition which reliably reflects its influence on the emission level.

5. Conclusions

The present article provides a comprehensive overview on published emission data from slurry storage which serves as a basis to determine guide values and baseline emissions for NH_3, GHGs and H_2S. Standardization of the emission data is an important issue in the present study due to the use of a large variety of units in the studies. Accompanying parameters (e.g. data on slurry analyses) were only partly available in the papers and could thus not be used for a more advanced data analysis. However, the season of the experimental period which served as a surrogate for the temperature was provided in most studies. Descriptive statistics of the emission data revealed a large variability for all gases. Data generated during warm, temperate and cold seasons are unevenly distributed over all records. Therefore, the calculation of an average annual value completed with a confidence range based on a weighting of the emission data according to the season and measurements duration was done. The baseline emissions on an area or volume basis determined for cattle and pig slurry stored in lagoons and tanks (Table 8) are mostly lower than existing reference values. NH_3 baseline emissions for tanks related to TAN are 16% of TAN (range: 14%–19% of TAN) and 15% TAN (range: 9.2%–23% of TAN) for cattle slurry and for pig slurry, respectively, and thus similar to emission factors used in emission inventory models. The flow-based baseline emissions for N_2O and CH_4 are lower than current emission factors. Total GHG emissions from slurry stores based on the global warming potential using a 100-year time horizon are dominated by CH_4.

Techniques for slurry treatment exhibit contrasting effects on emission levels during storage. Acidification was found to be efficient in reducing the emissions of NH_3 and CH_4 but less for CO_2 while the release of N_2O was enhanced in few studies. Solid-liquid separation causes higher losses for NH_3 and a reduction in CH_4, N_2O and CO_2 emissions. Anaerobic digestion promoted NH_3 emissions in most studies. In contrast, emission changes during slurry storage were less explicit for CH_4, although there is evidence toward an emission reduction. The effect of anaerobic digestion on N_2O and CO_2 emissions is unclear. It is essential to consider the context of good management practices along the whole manure management chain when the effect of slurry treatments on emissions from slurry storage is assessed.

All storage cover types reduce emissions of NH_3 while the effect is small for CH_4 and CO_2 with a trend toward a reduction. Permeable covers increase emissions of N_2O. Total GHG emissions tend to be lower

with coverage of slurry stores. Overall, coverage of slurry is efficient to abate NH_3 emissions involving a minimum risk of pollution swapping.

The present study provides a robust data basis for the determination of baseline emissions except for flow-based baseline emissions for lagoons which could not be calculated. The emission data in the records from different studies may vary over several orders of magnitude even for the same slurry type, the same type of study and identical seasons of measurement. For future research, appropriate study designs are required to generate baseline emissions appropriate to improve emission inventories. For this, farm-scale studies using non-intrusive methods are likely to be a preferential option. Pilot-scale studies are important to complement results from farm-scale studies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.agee.2020.106963.

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