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Effect of natural and regulatory conditions on the environmental impacts of pig slurry acidification across different regions in Europe: A life cycle assessment

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ABSTRACT

Animal manure handling is an essential but challenging part of Europe's intensive agriculture. To safeguard the environment and facilitate sustainable livestock production, lower-impact manure management technologies need to be identified, evaluated and implemented. Slurry acidification has been developed to address some of the environmental challenges faced by pig farmers, such as methane and ammonia emissions, that contribute to environmental impacts such as climate change, terrestrial acidification and air fine particulate matter formation. However, the efficiency of this technology has been found to depend on local environmental and regulatory conditions. The current study compared two slurry treatment options (no treatment versus slurry acidification in storage tanks) under the climatic, agronomic and legislative conditions found in Denmark, the Netherlands and Spain using Life Cycle Assessment (LCA). Data for the LCA model was collected from various sources and emissions following field application and crop yields were simulated over a 100-year period using the Daisy agricultural model. To address the uncertainty in LCA modelling, parameter analyses and Monte Carlo simulations with 1000 iterations were conducted followed by pairwise statistical tests. The results indicated that slurry acidification reduces the impact in all countries in impact categories mostly related to direct emissions from agriculture, such as methane and ammonia. For impact categories related to the provision of materials and energy to the farm, acidification increased the impacts in some cases. Additional requirements for lime application to counteract potential soil acidification did not result in significant changes in the performance of slurry acidification. The sulphur in the applied acid as an alternative to mineral S fertiliser can in some cases reduce the environmental impact of slurry acidification, but should not be advertised as doing so *per se*. Introducing stricter P application limits would seem to be the preferred option compared with slurry acidification in the Netherlands, while the opposite appeared true for Denmark.

1. Introduction

The agricultural sector is a significant source of ammonia (NH_3) and methane (CH_4) emissions and a substantial contributor to air pollution through fine particulate matter and human-induced climate change. Agriculture is responsible for 10% of greenhouse gases (GHG) and 90% of NH_3 emissions in the EU-28 (European Commission, 2017; Eurostat, 2012). Of these, 15% of GHG and 80% of NH_3 emissions are attributable to manure management on livestock farms. In addition, losses of reactive nitrogen (N) during slurry handling lead to inefficient nutrient recycling rates and environmental issues on a local, regional and global scale (Fangueiro et al., 2015; Pilar et al., 2015; Sommer et al., 2013).

Some of these emissions can be mitigated by new technologies that decrease emissions during manure storage and field application. Slurry acidification is a technology primarily designed to reduce emissions of ammonia along the manure management chain (Fangueiro et al., 2015; Hou et al., 2015). It is commercially available (Technological Readiness Level: 9) and has been declared by the European Commission to be a Best Available Technique (European Comission, 2015; Jensen, 2019). The technology takes advantage of the pH-dependent physical state of NH_3. At high pH levels, the chemical equilibrium between ammonium (NH_4^+) and NH_3 is shifted towards NH_3, which is susceptible to volatilisation (Lagerwerf et al., 2019). By lowering the pH of animal slurry, the dissociation of NH_4^+ decreases and NH_3 emissions are reduced

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accordingly. Decreasing slurry pH from 7.5 to 5.5 reduces the acid-base equilibrium concentration of $NH_{3(aq)}$ from 1.8% to 0.02%, which decreases the NH_3 volatilisation potential due to the equilibrium of NH_3 in the liquid and gaseous phase (ten Hoeve et al., 2016a). The result of the technology is that NH_3 emissions are reduced while the ammonium-N content of the slurry increases. Furthermore, a low slurry pH slows down microbial degradation of organic matter and decreases CH_4 emission during storage (Petersen et al., 2012, 2014).

However, the implementation of an additional technology often results in a trade-off between the benefits they bring locally and the environmental impacts they might have elsewhere in the chain. For slurry acidification, the trade-offs stem from the fact that production and provision of acid results in material and energy consumption. In addition, as acidification retains more nitrogen in the slurry because of reduced NH_3 volatilisation, this could result in increased nitrate (NO_3^-) leaching after the slurry is applied on agricultural land.

Life cycle assessment (LCA) is a standardised and well-established method to assess the potential environmental impacts resulting from products or services over the course of their life cycle (Finnveden et al., 2009; ISO, 2006). In the past, LCAs have been used to evaluate and compare the environmental performances of various slurry and manure management strategies, such as anaerobic digestion (Croxatto Vega et al., 2014; De Vries et al., 2012; Lopez-Ridaura et al., 2009), transport versus treatment (Lopez-Ridaura et al., 2009; Ten Hoeve et al., 2014), direct slurry application versus mineral fertiliser production (De Vries et al., 2012; Makara et al., 2019) and acidification (Miranda et al., 2021; Pexas et al., 2020; ten Hoeve et al., 2016a, 2016b). Miranda et al. (2021) conducted an LCA on liquid fraction cattle manure acidification supported by laboratory data on emission rates during storage. They found acidification to be environmentally superior to other treatment options (biochar amendment) but with greater impacts than no treatment given the additional needs of materials and energy, pointing to the requirement to include not only local achievements in emission reductions but also broader infrastructural changes. Implications for changes in yield, fertiliser needs and emission patterns along with specific regional features were not included in the analysis. Pexas et al. (2020) studied the environmental impact of Danish pig production and the impacts that different manure management strategies may have on its performance. In-house acidification was found to reduce impacts in some categories, but to increase it in others. Sensitivity testing suggested robust performance and that slurry acidification 'could potentially be used in a variety of systems across Europe' (Pexas et al., 2020). ten Hoeve et al. (2016a) studied the environmental impacts of slurry acidification in the animal house and during field application combined with slurry separation under different N and P application regulations. They concluded that in-house acidification results in better environmental performance than field application, and that stricter P application limits are also beneficial (ten Hoeve et al., 2016a). However, in-house acidification involves the retrofitting of animal houses to allow for recirculation of acidified slurry and thus requires substantial investment. Unless required to do so by environmental regulations or permits, farmers are unlikely to be willing or able to make such investments. While changes in crop yield and associated replacements were included in their study, agricultural modelling and Monte Carlo-supported uncertainty analysis were not. Since slurry acidification alters the chemical composition of slurry, retaining more N, nitrate leaching is an important aspect to consider. Nitrate leaching intensity is influenced by soil texture, precipitation and crops and their rooting system (Sørensen and Jensen, 2013), which all vary across Europe. These differences and interdependencies have only been reflected to a limited extent in previous LCA studies.

To the authors' knowledge, no LCA has assessed the outdoor storage acidification of pig slurry, which might constitute a compromise between in-house and field acidification and offer a reasonable balance between achievable emission reductions and financial burdens. In addition, all LCA studies have been conducted to represent either Danish or laboratory conditions, which means that there is a lack of knowledge about how environmental and regulatory conditions across Europe would affect the performance of the acidification technology.

The aim of the present study was to assess the benefits and drawbacks of implementing slurry storage acidification in three European regions characterised by intense animal production that have different climatic, soil and regulatory conditions. The European regions were represented by case-study areas in Denmark, the southern Netherlands and Catalonia in Spain.

2. Material and methods

This study was conducted in accordance with LCA principles to provide decision support on whether the implementation of slurry acidification as an emission mitigation technology is sensible in areas of high pig density in Denmark (DK), Limburg (NL) and Catalonia (ES).

2.1. Scenarios

To analyse the effect of slurry acidification in the three different regions, six standard scenarios were defined:

- slurry handling without acidification in Denmark: DK_UA
- slurry acidification implemented in Denmark: DK_SA
- slurry handling without acidification in Limburg in the Netherlands: NL_UA
- slurry acidification implemented in Limburg in the Netherlands: NL_SA
- slurry handling without acidification in Catalonia in Spain: ES_UA
- slurry acidification implemented in Catalonia in Spain: ES_SA.

2.2. Functional unit

The functional unit was the *handling of 1000* kg *of slurry-N entering outdoor storage* in each respective region.

It should be noted that since the slurry entering storage differs between the three regions (especially differences in dry matter and nutrient content – see Table S1), the functional unit essentially means that the reference flow is different in the different regions. Therefore the functional unit allowed for the comparison of scenarios with and without acidification in each of the three regions. However, any comparison between countries is only useful to a limited extent.

2.3. Regionalised LCA

To conduct regionalised LCAs, geographical aspects should be considered in all four stages of an LCA study (namely goal and scope definition, inventory analysis, impact assessment and interpretation) (Loiseau et al., 2018; Wowra et al., 2021). This study followed these recommendations and site-specific systems were defined in the three countries. Distinct inventories were created based on agricultural modelling calibrated to each of the three sites. The site-specific effects of the technology were compared with generic site effects, but were only differentiated between farm-level and anywhere else in the world. No site-specific or country-specific characterisation factors were used to account for different degrees of sensitivity in the ecosystems potentially affected.

2.3.1. Geographical scope and selection of representative European regions of intense pig production

The three regions were chosen based on their relevance to Europe's intensive pork production and their trade, regional distinctiveness, degree of technological establishment and urgency of reducing agricultural emissions in order to comply with international emission reduction targets.

In Denmark, slurry acidification has been established as a tried-and-tested and accepted ammonia emission abatement technology by livestock farmers as well as policy-makers (Bull, 2016; Fangueiro et al., 2015; Jacobsen, 2017; Kupper, 2017; UBA, 2019). Pig production is an important part of Denmark's agricultural sector, representing a share of more than 25% of total agricultural economic output in 2020 and exceeding the economic output of all other animal products (such as milk or cattle meat) combined (EU, 2020). In 2018, the Danish agricultural sector was responsible for 95% of the country's ammonia and 23% of the country's greenhouse gas emissions, mainly originating from livestock production (EU, 2020; European Environment Agency, 2021). Given its high livestock density, Denmark was chosen as a representative EU pig production region.

Pork is among the most important animal products in the Netherlands in terms of economic return (EU, 2020). The Dutch agricultural sector has become one of the most efficient and productive in the EU, however emissions of NH_3 and surplus nitrogen per hectare of agricultural land are also among the highest (van Grinsven et al., 2019). Adding elevated N emissions from other sectors such as construction and road traffic over many years, the Netherlands has reached what is often called a 'nitrogen crisis', forcing the country to take drastic action (Stokstad, 2019). Combined with societal and regulatory pressure on a reduction in GHG emissions, Dutch farmers will be forced to decrease livestock density and/or implement further emission abatement technologies. Limburg is one of the provinces with the highest density of pigs in the country and was therefore used as a study region in the current analysis.

In 2019, Spanish pig farms had the single largest share of pigs in the EU-28, amounting to 22% of total EU pig livestock numbers (EU, 2020). In 2017, Spain reported the highest NH_3 exceedance rates (47%) of all EU member states and is currently expected to be unable to meet agreed targets for NH_3 and fine particulate matter emissions by 2030 (European Environment Agency, 2019). Catalonia is the largest region with intensive pig production in Spain and was therefore chosen as the third region for analysis in this study.

The three countries were chosen because of the different circumstances encountered by farms in these countries. These include slurry characteristics, storage and application requirements, and field and environmental conditions. Slurry characteristics differ in dry matter content and thus nutrient concentration due to different management strategies in the animal houses. In Spain, pigs need to be supplied with ample amounts of drinking and cooling water to increase their resistance to heat, resulting in rather diluted slurry. In the Netherlands, water usage is reduced to a minimum because the slurry often has to be transported long distances, making Dutch slurry more viscous. Danish slurry characteristics lie between the two; while pigs are provided with water *ad libitum*, water needs for cooling are lower than in Spain. Slurry storage conditions vary in accordance with national regulations. In the Netherlands storage tanks have to be sealed with an impermeable cover, while in Denmark a permeable cover suffices in most cases and in Spain there are currently no cover requirements. Further differences exist in the ways the slurry is field applied. Under national regulations, farms in Denmark can use a trailing hose application when the slurry is either acidified or applied to a winter crop, and in the Netherlands when the slurry is applied to a winter crop, otherwise the default application method in all countries is open slot injection. Apart from regulations on the modes of slurry application, differences arise from the rates of slurry application allowed as country-specific legislation restricts the rate at which slurry N and P can be applied on fields and thus the area required to dispose of a given amount of slurry. In Denmark and the Netherlands, P legislation is the limiting factor for slurry application, while in Spain N is the limiting factor. A lack of agricultural area forces Dutch farmers to export some of their slurry to fields in neighbouring countries to comply with application standards for N and phosphorus (P), while Danish farmers usually have access to sufficient land and produce the majority of feed crops themselves (Willems et al., 2016). Denmark and the Netherlands both have a temperate, oceanic climate (Köppen Cfb), while Catalonia has a Mediterranean climate with hot summers (Köppen Csa). The pH status of soils in the study region of southern Netherlands is comparable to that in Denmark at values of 5.5 or lower. Catalonian soils have a rather high pH, ranging from 6.5 to 7.5 (Joint Research Centre, 2010). Combined, these countries represent a good variety of conditions in Europe's pig production and cover a wide range of potential effects of slurry acidification.

2.4. Life cycle inventory

The LCA provides a 'storage-to-field' perspective. A process diagram of the life cycle inventory model is shown in Fig. 1.

Data for the foreground processes were determined using agricultural emission factors and models allowing for estimates specific to the climatic, soil, agronomic and regulatory conditions of the modelled regions. The actual production of pigs, including feed and housing as well as storage tank or agricultural machinery construction and demolition, were excluded from the study since no or only insignificant differences were assumed between the management strategies studied (i.e. acidification against no acidification).

2.4.1. Outdoor slurry storage

The studied system starts with a country-specific reference flow, i.e. an amount of pig slurry corresponding to the functional unit of 1000 kg of slurry-N entering outdoor storage. This reference flow was derived from the nitrogen content of the pig slurry and of a composition corresponding to country average values (see Supplementary information (SI) Table S1).

In the slurry acidification (SA) scenario, some slurry is pumped out of the storage tank into a tank container mounted on a tractor trailer where it is mixed with sulphuric acid (H_2SO_4) and then returned to the outdoor

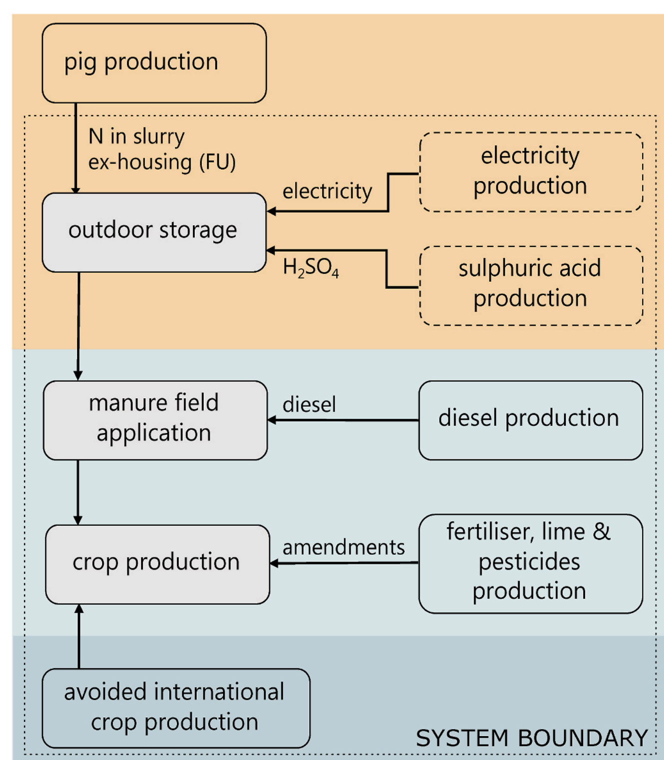

Fig. 1. Structure of the analysed product system in the acidification scenario. Boxes indicate the main activities associated with slurry acidification (grey boxes indicate on-farm activities). Arrows indicate flows of products. FU: functional unit. Background colours - > orange: processes associated with slurry storage; light blue: processes associated with regional crop production; dark blue: international crop production.

storage. The process is repeated until the slurry in the outdoor storage reaches a target pH of about 5.5 (Hjorth et al., 2015). Given a storage period of several months, re-acidification is assumed to take place between four and six times (Fangueiro et al., 2015). Assumptions regarding emissions during storage for the unacidified (UA) and SA scenarios can be seen in SI Table S2.

2.4.2. Field application & agricultural modelling

Following outdoor storage of four to six months, the slurry is field applied in accordance with local legislation (see below). Environmental emissions in the field were estimated using agricultural models. Ammonia emissions during field application were calculated using the ALFAM2 model (Hafner et al., 2018), taking into consideration application rates and methods, slurry composition and environmental conditions such as wind speed, air temperature and precipitation. Average weather conditions in the month of application in the past 30 years in the three regions were used as input parameters to the ALFAM2 model. Estimated fractions of the applied total ammoniacal nitrogen that is volatilised were 9.4% for Denmark, 13.3% for the Netherlands and 9.6% for Spain in the UA scenarios. In the SA scenarios, those fractions were reduced to 2.9% in DK, 3.7% in NL and 2.5% in ES (see SI, Table S7). Calculated NH_3 emissions, slurry compositions and environmental data formed the input to the Daisy agro-ecosystem model (Abrahamsen and Hansen, 2000). This model was used to estimate carbon (C) and N-related emissions (such as nitrous oxide emissions and nitrate leaching to freshwater ecosystems) as well as crop yields including biomass (dry matter), and carbon and nitrogen content in the harvested products and residues returned to the soil. This was done in representative scenarios for each study region where crop rotations realistic for a pig farm in the area were simulated with historical weather data (approximately 30 years) and respective soil properties. The chosen crop rotations for DK were spring barley – winter rapeseed – 2 x winter wheat – spring barley – winter barley, for NL maize – potato – sugar beet – winter wheat and for ES maize monoculture (SI, Table S18). The 30-year historical weather data were reused several times during the 100-year simulation. To reduce the coincidence of the same crop and weather during the extended simulation, a weather sequence was generated for the latter 70 years by randomising the year sequence of the historical data, which in the meantime creates a larger sample space with different combinations of crops and weather, allowing a better estimation of uncertainty. To simulate the short-term turnover of slurries in the soil, the model was calibrated on the specific C and N mineralisation patterns of untreated and acidified slurry based on CO_2 and soil N evolutions from laboratory incubation experiments (Fangueiro et al., 2009, 2010; Kirchmann and Lundvall, 1993). Since slurry acidification lowers nitrogen emissions, more N is retained in the slurry and becomes field applied. In these simulations, increased N concentrations were taken into account and mineral N fertilisation revised downwards where applicable. An overview of the Daisy simulation results is given in the SI in Table S21. In previous studies, Daisy has proven to be a valuable inventory model for LCA studies on agricultural systems and management alternatives (Andrade et al., 2021; ten Hoeve et al., 2016b; Yoshida et al., 2018).

2.4.3. Crop production

In order to construct processes for the production of different crops in the rotations, the Daisy modelling results were taken as a point of departure, including fertiliser-associated emissions of nitrate (to ground and surface waters) and nitrous oxide (greenhouse gas) and crop yields. To complement crop production processes with all the remaining emissions and materials needed, existing ecoinvent processes were selected for each respective crop. Emissions and material inputs, such as pesticides, were then added or modified after being scaled to the area of land used. This was based on the assumption that agrochemicals are applied on a per hectare basis and not in relation to how much the farmer expects to harvest. For a full description of altered flows, please refer to the SI (changed_ecoinvent.xlsx).

The modelled system provides two functions (referred to as multi-functionality): handling pig slurry and producing crops. Since crop yields are a 'by-product' of slurry handling and because they differ between scenarios (UA vs. SA) despite the amount of slurry-N applied being equal (FU: handling 1000 kg slurry-N), the system needs to be balanced in terms of available crops on the global market, otherwise the systems would lack comparability. In consequential LCA modelling, multi-functionality is solved by system expansion rather than allocation, and it was assumed that the crops produced on the studied imaginary farm replace an equal quantity of crops produced elsewhere. In this way, the difference between globally marketed crops (either coming from the study farm or from elsewhere) between scenarios (UA vs. SA) equals zero and fair comparability is ensured. In so doing, the way ($< > =$) in which the environmental impact (EI) of the system changes relative to the production elsewhere when introducing slurry acidification can be evaluated:

Equation 1: Simplified mathematical representation of presented LCA study

$$EI_{crop,\ UA} - EI_{crop,\ g} \ < > = EI_{crop,\ SA} - EI_{crop,\ g}$$

where EI_{crop} is the environmental impact of a given crop at a given quantity, UA, SA and g are production with unacidified or acidified slurry or elsewhere (global production) respectively.

Following the overall trend of the world's growing population and increasing crop production, crop production here is assumed to alleviate the increase in demand on the global market instead of causing an actual decrease in demand elsewhere. Following consequential LCA theory on the identification of marginal suppliers, providing goods to a growing market may lead to a delay in the implementation of new production technology in the fastest growing market (Hauschild and Rosenbaum, 2018). Here, market segments equal countries, and the countries recording the strongest growth in crop production were identified. It was assumed that they would respond to the global increase in demand if the studied farm did not. In accordance with the ecoinvent database, the long-term marginal crop was identified (see SI). Like-for-like replacement was considered, i.e. potatoes replace potatoes and are not substitutable by another crop in terms of nutritional value or other dietary factors. A global market for crop trade was assumed and marginal suppliers not limited to European regions or the EU were identified.

2.5. Variation analysis

Several critical assumptions were made. To test how alternative assumptions would affect the results, a variation analysis was conducted in which some of the assumptions were changed. This included assumptions about how sulphur fertilisation strategies could be affected by the addition of sulphuric acid to slurry, how the addition of acid could affect the need for liming, and how stricter P regulations could alter the environmental performance of slurry handling. The parameters tested in the variation analysis to examine how alternative assumptions affect results are described below.

2.5.1. Sulphur fertilisation (S)

In the unacidified slurry UA scenarios without addition of sulphuric acid, it was assumed that farmers do not fertilise with sulphur (S), thus sulphur added in the form of H_2SO_4 in the corresponding acidification SA scenario does not lead to a decreased application of S fertiliser. However, flue gas desulphurisation in the energy sector leads to a decrease in atmospheric S deposition, which in turn increases the need for S fertiliser in agricultural soils across Europe (Eriksen et al., 1995; Feinberg et al., 2021; Haneklaus et al., 2000; Zhao et al., 2002). This trend is expected to continue and S deficits will become more widespread and intense over time. As of today, crops with a higher S demand, such as oilseed rape, have been reported to be S deficient in countries such as France, Germany and Denmark (Zhao et al., 2002). In reality, most farmers in Europe will regularly be applying synthetic fertilisers containing S. Adding S to slurry in the form of H_2SO_4 can fully replace the S in synthetic fertilisers as the quantities suffice for all crops and it is equally plant available (Eriksen et al., 2008; UBA, 2019). In the S fertilisation scenario and on the assumption that S fertilisation is or will become common, the H_2SO_4 added to slurry to acidify it fully replaces the synthetic S fertiliser needs. Accordingly, a new UA scenario with S fertilisation (UA_S) was introduced in which S fertiliser is applied to maintain the S balance of the soil. No potential crop effects of S deficits where not fertilised with S were modelled: crops were assumed to meet optimal conditions either because S fertilisation is not necessary or because it is provided.

2.5.2. Soil liming (L)

The effect on soil pH resulting from the application of acidified slurry depends not only on the slurry itself, but also on the buffer capacity of the soil (Jensen et al., 2018). In the standard scenarios, H_2SO_4 was not assumed to affect soil pH substantially; however, on soil with a low buffer capacity, additional liming might be necessary to keep pH levels within a range optimal for plant growth. The Danish agricultural advisory and research organisation (SEGES) recommends applying on average 75 kg of agricultural lime (75% $CaCO_3$) per hectare per year on typical Danish soils if the manure is acidified with 1 L of H_2SO_4 per tonne of slurry applied at a rate of 30 t/ha (SEGES, 2014). In the LCA, soil liming involves natural lime (mainly $CaCO_3$) extraction, the application of lime to the field using agricultural machinery, and CO_2 emissions in the field as a consequence of $CaCO_3$ decomposition. In NL and DK, soils have a rather low natural pH (4.5–5.5), whereas in Catalonia calcareous soil with a high pH prevails (6.5–7.5) (Joint Research Centre, 2010). To account for this in the LCA and in accordance with SEGES' recommendations, scenarios were modelled with additional liming to fields in DK and NL in the SA scenarios to counteract potential soil acidification (SA_L).

2.5.3. P application limits (P)

Up to 2025, the P application limit in DK is 34.5 kg P ha^{-1} y^{-1} (standard scenario in DK) (Environmental Protection Agency, 2017), but thereafter will be reduced to 30–31 kg P ha^{-1} (Environmental Protection Agency, 2017). Therefore a low P scenario was modelled (variation scenario in DK, DK_P). In Limburg in the southern NL, the P application limit directly depends on the P status of the soil, which in some parts was in the range of medium high to high (Pw 51 -> 100) between 2000 and 2004 (Reijneveld et al., 2010). A statistical analysis of soil samples between 2005 and 2015 showed no significant change in the Pw value (Brolsma et al., 2017). Thus, application rates of 60 kg P_2O_5 ha^{-1} y^{-1}, i. e. 26.2 kg P ha^{-1} y^{-1}, were assumed to be in accordance with national legislation (standard scenario in NL). However, a study in 2016 and 2020 on P-AL values identified areas with the highest country-wide values (65 - > 75 P_2O_5/100 g) in Limburg (Agro, 2020). Thus, in the variation analysis, a reduced application rate of 40 kg P_2O_5 ha^{-1}, i.e. 17.5 kg P ha^{-1} was tested (variation scenario in NL, NL_P).

Spain has no legislation on P application in the studied region and the EU Nitrates Directive sets the limit for slurry application at 170 kg N ha^{-1} y^{-1} (Amery and Schoumans, 2014; EC, 1991). Due to the increased N content in acidified slurry at application, area requirements increase. An alternative application scenario in ES related to P was not modelled.

2.6. Life cycle modelling and uncertainty analysis

All analyses were performed with the LCA open-source software openLCA v.1.10.2 (https://openlca.org). The impacts of each system were determined in accordance with the ReCiPe 2016 midpoint methodology (Huijbregts et al., 2017) and included the following impact categories:

- fine particulate matter formation
- fossil and mineral resource scarcity
- freshwater and marine eutrophication
- freshwater and terrestrial ecotoxicity
- global warming
- human (non-)carcinogenic toxicity
- terrestrial acidification.

All background processes were modelled using the consequential ecoinvent database 3.6 (Wernet et al., 2016).

To account for the uncertainty of the results and at the same time determine the environmentally superior alternative, error propagation and subsequent statistical testing were used (Henriksson et al., 2015; Pexas et al., 2020; Pizzol, 2019; Ross and Cheah, 2017). Instead of reporting single values for each indicator, as has been done in many LCA studies, the aim here was to obtain and compare uncertainty distributions in order to facilitate more meaningful and robust conclusions about the two alternatives (Heijungs and Lenzen, 2014).

To estimate the uncertainty of the conclusions, propagation of errors in the parameter values was performed via Monte Carlo simulations (MCS) (1000 runs). This requires uncertainty estimates and distributions for all model parameters associated with appreciable uncertainty. Uncertainty distributions were already estimated for the ecoinvent processes. Uncertainty related to the foreground system, such as emissions during land application and attainable yield, were obtained through Daisy modelling. The respective datasets were tested for their distribution and the mean and standard deviation were determined in accordance with the type of distribution. Most datasets followed a lognormal distribution, and geometric mean and standard deviation were used for the MCS. Following the MCS, the distribution calculated for each impact category by the means of the 1000 runs was tested for normality using the Shapiro-Wilk test ($p < 0.05$). To determine whether slurry-handling scenarios are likely to differ from each other, the nonparametric pairwise Wilcoxon Rank Sum test ($\alpha = 0.05$) or the Student's t-test was applied, depending on whether normality was rejected or not. In a second round, MCS was run on the difference between two management scenarios (e.g. ΔDK) to determine the range of their standard deviation. To do so, the process without acidification was subtracted from its respective counterpart in the acidification scenarios and simulations were run on its results (for instance DK_SA – DK_UA = ΔDK). Given recent criticism of this practice (Heijungs, 2021; von Brömssen and Röös, 2020), it should be noted that the results of inferential statistics on Monte Carlo simulations are unlikely to reveal the ultimate truth, just as no LCA in itself will ever be able to reveal the true impact of human action. If the number of iterations is set too high, a significant difference might be indicated while in fact the gap between two alternatives is very small. However, inferential statistics are very unlikely to underestimate differences, may point to negligible discrepancies and can serve as an objective screening tool. Apart from the statistical analysis, a data quality assessment was conducted to identify and highlight model uncertainty (see SI Table S22).

The statistical analysis of the LCA results and data visualisation were conducted using Python and respective packages (Hunter, 2007; van Rossum and Drake, 2009; Virtanen et al., 2020) as well as inkscape (https://inkscape.org/). More detailed descriptions of the assumptions and methods as well as the complete inventory can be found in the SI.

3. Results & discussion

3.1. Effect of slurry acidification

The effect of slurry acidification is best illustrated by examining the difference between impacts in the scenarios with and without acidification in the studied regions. These differences can be found in Table 1. In the SA scenarios, slurry handling becomes more complex and the system is expanded by additional processes, such as sulphuric acid provision and energy consumption for slurry mixing. The altered

Table 1

Change in environmental impact when switching from one slurry management strategy (A) to another (B) (A – B). The strategy listed first forms the reference, the strategy listed second the new strategy. UA: unacidified slurry; SA: slurry acidification; _P: stricter P application legislation. Empty cells denote that there is no significant difference between the two strategies following analysis of the Monte Carlo simulations. Functional unit: 1000 kg slurry N entering outdoor storage.

	Denmark			The Netherlands			Spain
	UA - SA	UA - UA_P	UA - SA_P	UA - SA	UA - UA_P	UA - SA_P	UA - SA
Fine particulate matter formation (kg PM2.5 eq) [FPMFP]	−25.9	5.1	−32.2	−16.9	−87.0	−104.2	−3.5
Global warming (kg CO_2 eq) [GWP]	−6443.7	5250.8	−1829.7	−6670.4	6472.5	147.8	−7092.0
Terrestrial acidification (kg SO_2 eq) [TAP]	−230.2	−46.1	−282.5	−152.8	−604.4	−761.	−47.1
Freshwater eutrophication (kg P eq) [FEuP]	–	–	–	–	−2.1	−1.9	0.4
Marine eutrophication (kg N eq) [MEuP]	–	−28.4	−29.9	–	−83.1	−81.7	1.2
Freshwater ecotoxicity (kg 1.4-DCB eq) [FEcoP]	62.8	362.6	397.8	–	–	–	–
Terrestrial ecotoxicity (kg 1.4-DCB) [TEcoP]	3724.9	17,888.5	18,049.4	–	37,778.2	41,213.0	6228.9
Human carcinogenic toxicity (kg 1.4-DCB eq) [HCTP]	28.1	76.1	98.4	24.8	34.5	62.5	40.7
Human non-carcinogenic toxicity (kg 1.4-DCB eq)	2676.6	7240.6	9616.8	–	–	–	–
Mineral resource scarcity (kg Cu eq) [MRSP]	6.8	6.2	10.5	5.6	−90.9	−85.2	10.3
Fossil resource scarcity (kg oil eq) [FRSP]	148.0	331.0	209.2	125.5	−1711.0	−1579.3	204.9

chemical properties of the slurry change the susceptibility to leaching and emission of its components: while ammonia and methane emissions to the air decrease, nitrate leaching to groundwater increases. Since it was assumed that a farmer adjusts mineral N fertiliser application to the concentration of N in the slurry, no great differences in yield were observed in the Daisy simulation results.

In all three study regions, slurry acidification indicated both beneficial and harmful environmental effects (Table 1). A decrease in the environmental impact potential was observed for all countries in the categories *global warming potential* (GWP), *terrestrial acidification potential* (TAP) and *fine particulate matter formation potential* (FPMFP). These reductions are achieved mainly because of lower CO_2, CH_4 and NH_3 emission during storage and field application of acidified slurry. The magnitude of the decrease in GWP was identified as being similar between the three countries although the percentage changes were very different. In DK, reductions of 6443 kg CO_2 eq (−18%), in NL of 6670 kg CO_2 eq (−66%), and in ES reductions of 7092 kg CO_2 eq (−18%) per FU were noted when switching to acidification of slurry. The decrease in greenhouse gases is primarily due to emission savings during slurry storage. The strongest decrease in TAP was registered in Denmark, with emissions about twenty times lower than without acidification – a reduction corresponding to 230 kg SO_2 eq per FU. The Dutch system achieved reductions of about 153 kg SO_2 eq (−16%) per FU when switching from UA to SA. Slurry acidification showed similar effects on NH_3 emission reductions during storage (about −50% SO_2 eq) and field application (about −70% SO_2 eq) in both countries, but these improvements must be viewed in relation to the crops assumed to be substituted. The crops produced elsewhere, which are replaced by Danish production, perform better in environmental terms than the crops replaced by Dutch production. Small improvements in Danish production therefore carry more relative weight than similar improvements to the Dutch cropping system. The same was true for FPMFP, which also responds to changes in NH_3 emissions.

Other categories suggested increases in the environmental impacts as a result of an implementation of slurry acidification, namely *fossil & mineral resource scarcity potential* (FRSP, MRSP), *terrestrial ecotoxicity potential* (TEcoP) and *human carcinogenic toxicity potential* (HCTP). In the UA scenarios of DK and NL, on-farm crop production was found to be less impactful in terms of fossil and mineral resource depletion than international production of the crops they replace. The introduction of slurry acidification decreased DK and NL crop production performance in the above-mentioned categories and reduced this advantage. Spanish maize production was found to be more fossil and mineral resource intensive than the replaced international crop production, and slurry acidification was found to further widen this gap.

Furthermore, slurry acidification indicated adverse effects on *freshwater ecotoxicity potential* (FEcoP) and *human non-carcinogenic toxicity potential* in DK and *freshwater & marine eutrophication potential* (FEuP, MEuP) in ES. The increase in DK of FEcoP and HNCTP, by 83 kg 1.4-DCB (+12%) and 2677 kg 1.4-DCB (+13%) per FU respectively, can be explained by the production of sulphuric acid and diesel burned in agricultural machinery to perform the mixing as well as the increase in rape seed production, which replaces less harmful international rape seed production. The increase in FEuP, MEuP and TEcoP in the ES case also resulted from sulphuric acid production and the use of agricultural machinery. Furthermore, higher N concentrations in the slurry resulted in larger area requirements and consequently higher demands for fertiliser, pesticides and other activities related to soil cultivation and harvesting per FU. Slightly increased yields and thus higher crop replacement rates are insufficient to counteract these effects. Negligible impacts on *human non-carcinogenic toxicity potential* in the NL case can be associated with the needs of acid and energy during storage leading to an additional 1814 kg 1.4 DCB (+1%) in the SA scenario.

A study of liquid cow manure acidification compared with no treatment concluded that there is an increase in GWP, acidification, eutrophication and HNCTP (Miranda et al., 2021). These findings contradict the results of the present study, which may be due to the different manure characteristics and the exclusion of changes in field emissions, fertiliser needs and crop substitutions. A past LCA study comparing in-house and field acidification in Denmark indicated a strong impact improvement on terrestrial eutrophication potential, with decreases in impact potential of 31% for field and 72% for in-house acidification. The same study suggested that the former decreases overall GHG emissions, while the latter causes a 60% increase (ten Hoeve et al., 2016a). This is because the environmental costs of producing sulphuric acid and lime needed in the acidification scenarios can only be counteracted when the acidification takes place early in the management chain (in-house) and higher overall slurry emission savings are achieved. The present study suggested that storage acidification is still sufficient to achieve reductions in GWP. In the above-mentioned study, both acidification treatments showed little effect on freshwater and marine eutrophication potential and caused an increase in fossil resource depletion (ten Hoeve et al., 2016a, 2016b). In the present study, slurry acidification also had a negligible impact on FEuP and MEuP in DK and NL, with changes in impact below 1.5%, and all country scenarios suggested an increase in FRSP and MRSP. The same study suggested little difference in terms of ecotoxicity between treatment scenarios (ten Hoeve et al., 2016b), which is only in partial agreement with the present findings since increased impacts in the *freshwater* and *terrestrial ecotoxicity potential* in all countries were identified due to the introduction of slurry acidification. Another study on in-house acidification under Danish conditions concluded that acidification results in reductions in acidification and eutrophication and increases in emissions related to GWP and non-renewable energy and resource consumption potential (Pexas et al., 2020). The same conclusions were drawn here except for GWP, which may be because this study did not take into account changes in CO_2 and CH_4 emissions during storage and field application.

3.2. Variation analyses

Fig. 2 provides an overview of the change in impacts caused by switching to acidification under different assumptions and stricter P application limits with or without acidification.

3.2.1. Sulphur fertilisation (S)

As can be seen in part in Fig. 2, whether sulphuric acid addition to slurry replaces S fertilisation or not has little or no impact on the environmental performance of the studied system. It indicated small effects in ES and negligible effects in DK and NL. If acidification were previously found to be beneficial, these benefits would now be more pronounced, while if slurry acidification were found to have negative effects, these effects would be cushioned. The differences arise from the S demand of the crops in each rotation and thus the rates at which S is applied to a field. Maize in the Spanish case has a rather high demand in

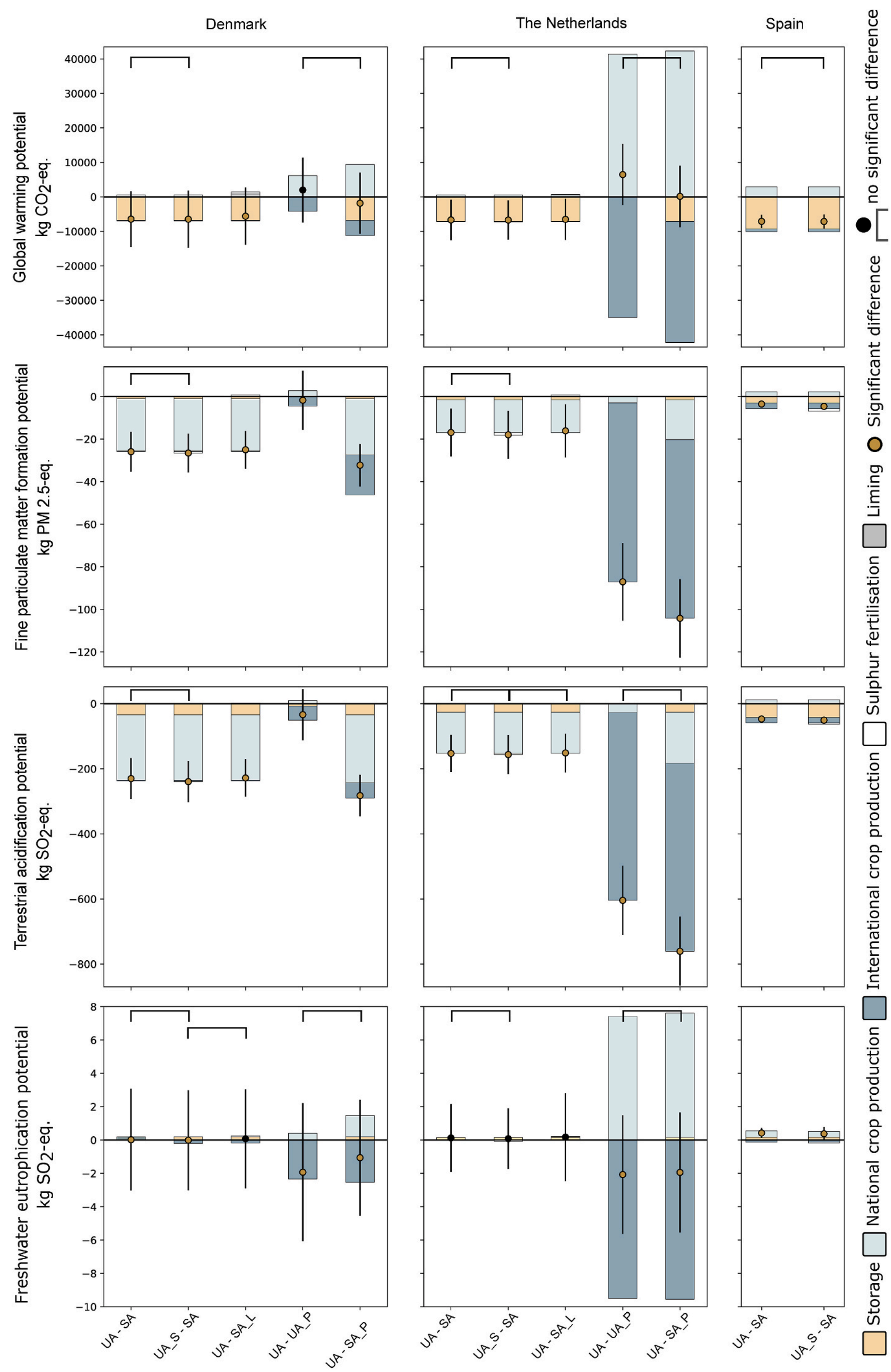

Fig. 2. Change in environmental impact by shifting from UA → SA (no acidification to acidification), UA_S → SA (no acidification assuming S fertilisation to acidification), UA → SA_L (no acidification to acidification assuming lime application), UA → UA_P (no acidification to stricter P application limits without acidification) and UA → SA_P (no acidification to stricter P application limits with acidification). Standard deviations (SD) derived from Monte Carlo simulations.

S at 35 kg $ha^{-1}yr^{-1}$, whereas the average S demand of the Danish crop rotation is around 18 kg $ha^{-1}yr^{-1}$ and that of the Dutch rotation is 20 kg $ha^{-1}yr^{-1}$. The higher the demand in S of the crops in the rotation, the lower the additional impact from acidification as a result of sulphuric acid production. To a certain extent it could be said that the S is only taking a detour through the slurry storage tank before being field applied and thus serves two purposes: to lower slurry pH and to fertilise crops. However, while S fertilisation is generally recommended in DK, it is not common practice throughout Europe. For instance, in Spain S fertilisation is rather uncommon (IRTA, personal communication), which again makes it difficult to speak of a true diversion. The promotion of sulphur provision to crops as an additional benefit of slurry acidification should thus be limited to crop rotations of high S demand under conditions (soil, climate) deficient in S, where sulphuric acid will partly or fully substitute mineral S fertilisation. However, if farmers are fertilising with S, they could be incentivised to implement slurry acidification instead. In the Dutch case, adding H_2SO_4 to slurry could be seen as a valorisation measure and boost its acceptance by recipient farms.

Going beyond Europe's borders, slurry acidification could be a valuable treatment option in China where maize is the most common energy feed and open-air tanks are the most common way to store manure (Liu et al., 2021). The S in the slurry could find optimal 'secondary' usage as a fertiliser and emission savings during storage are likely to be considerable.

3.2.2. Soil liming (L)

Modelling the need for additional soil liming to compensate for the acidifying effect of sulphuric acid had little or no effect on the performance of acidification relative to no acidification (Fig. 2). It either slightly decreased the beneficial effects of acidification or intensified its already adverse effects. Liming decreased the beneficial impacts in both DK and NL on FPMFP, GWP and TAP, but increased the harmful effects in FRSP and FEuP. Given that liming did not impact the relative comparison between UA and SA, the pH status of the soil need not be part of the decision-making process.

Another study comparing in-house slurry acidification including liming against a baseline without further treatment under Danish conditions found reductions in terms of *acidification potential* (kg SO_2^- eq) and *eutrophication potential* (kg PO_4^- eq), but large increases in the categories *non-renewable resource use potential* (kg Sb eq), GWP and *non-renewable energy use potential* (MJ) (using CML-IA 3.05 methodology) (Pexas et al., 2020). This is in line with the present study's findings except for the impacts on GWP, which was still reduced in the present case despite the introduction of liming into the system because emission savings from slurry acidification were greater than the emissions related to the provision and application of additional lime. However, as mentioned above, the difference in interpretation of GWP might be due to different system boundaries and the exclusion or inclusion of carbon-related emissions during storage and field application. No differentiation was made by ten Hoeve et al. (2016b) between the impacts of addition of lime and sulphur, but that study found that the achieved reductions still sufficed when acidification took place early in the management chain, i.e. in-house. The present study also suggested that outdoor acidification, like in-house acidification, resulted in savings great enough to justify the additional needs for liming in the respective categories.

3.2.3. P application limits (P)

Under stricter P application limits, the area needed to apply a given amount of slurry increases. In the present case, the area increased from 6 ha to 6.7 ha (+13%) in DK (with P application limits tightened from 34.5 to 30–31 kg P ha^{-1}) and from 9.2 ha to 13.7 ha (+50%) in NL (from a limit falling from 26.2 to 17.5 kg P ha^{-1}) per FU of 1000 kg slurry-N entering the outdoor storage. This has both beneficial and adverse impacts on the environment, depending on the category examined. Under stricter P application rules, the slurry has to be transported longer distances and the farmer is forced to apply slurry-N well below recommended rates, leading to higher application rates of mineral N fertiliser, which is of high resource demand in its production. However, applying the slurry to a larger area results in higher yields in respect of the FU. Storage emissions are not impacted because the same amount of slurry is handled.

In both countries, stricter P application limits have beneficial effects on MEuP and TAP. The decrease in MEuP, by 28 kg N eq in DK (13%) and 83 kg N eq in NL (47%), can be explained by an increased application rate of mineral N fertiliser relative to slurry N application (Tables S8 and S9), which decreases the overall susceptibility to nitrate leaching. A larger area of application results in higher yields and higher replacement rates of crops produced elsewhere, often with higher N leaching rates per kg crop. *Terrestrial acidification potential* also decreases as a result of higher replacement rates of foreign crop production that is more intense in its acidifying potential. Stricter P application limits further positively impact FPMFP, FEuP (−23%) and *fossil* and *mineral resource scarcity potential* (~-30%) in NL, but negatively affect GWP, HCTP and TEcoP in both countries. The increase in GWP, by 5251 kg CO_2 eq in DK (15%) and 6473 kg CO_2 eq in NL (64%), is among other things caused by an increased need for transportation and agricultural machinery operation due to the increased area. The increase in DK of HCTP and TEcoP (both by more than 40%) is caused by the greater demand for mineral N fertiliser because the same amount of slurry-N is now applied on a larger area and thus at lower concentrations. At this point, it is crucial to bear in mind that as this study is looking at a FU of a certain quantity of slurry-N, the larger area has to be seen in relation to this FU and not in total terms. The field to which the surplus slurry is now applied has almost certainly been fertilised with mineral N fertiliser before, such that more mineral N fertiliser is not in fact being used – it is only 'more' in relation to the slurry handled in the model.

When combining stricter P application rates with acidification, the effects were found to be either reduced or intensified. In those categories where SA showed favourable consequences under less strict P application limits, the beneficial effects of stricter legislation were intensified and adverse effects alleviated. Slurry acidification can improve the performance of slurry handling under stricter P application rules in the categories FMPFP and TAP in DK and NL. GWP did not seem to be affected. In other categories, acidification led to either even greater increases in emissions (such as TEcoP) or decreases in its beneficial impact (for example FRSP).

Previously, tougher P application limits have been found to have favourable consequences for both the reference and acidification scenario (ten Hoeve et al., 2016a). This is not entirely in accordance with the present findings, since adverse impacts were found in various categories in the DK cases for both the UA as well as the SA scenarios. For NL cases, however, the findings are more in accordance with those of ten Hoeve et al. (2016a) as improvements were found in almost all (7 out of 10) categories in both scenarios.

All in all, stricter P application rates seemed to be more favourable under Dutch conditions than under Danish ones, with improvements in 67% and 28% of the impact categories respectively. From a global environmental perspective, reducing the amount of P that can be applied seems to be a more promising strategy for the Netherlands than slurry acidification, with improvements in only 33% of the examined impact categories. When looking at impact categories mostly related to agriculture, such as *fine particulate matter formation potential*, *global warming potential* and *terrestrial acidification potential*, slurry acidification seems to be better under Danish conditions than if stricter P laws were introduced in the country. However, these savings come at a cost and the provisioning of materials and energy play a crucial role in other less related impact categories. This is an example of the difficulties related to finding one-fits-all solutions and illustrates how important it is to look at specific circumstances when implementing technologies elsewhere, even if they have been proven to work well under some conditions.

3.2.4. *On-farm versus off-farm effects*

Fig. 3 shows the percentage change as a consequence of the introduction of acidification in selected environmental impacts divided into on-farm and off-farm impacts. On-farm relates to emissions on the premises of the fictive case study farm and includes emissions from outdoor storage and during and after field application of one and the same slurry. Off-farm emissions comprise all emissions taking place away from the farm, such as during the production of fertiliser used on the farm or during crop production elsewhere in the world.

In all three countries, FPMFP and GWP decrease on-farm but increase off-farm as a consequence of the implementation of slurry acidification. This is because acidification causes reductions in CH_4, CO_2 and NH_3 emissions on-farm, but requires additional inputs of material and energy that are produced off-farm. However, as overall impacts improve in these two categories, these off-farm increments do not exceed on-farm improvements (e.g. change in FPMFP in DK: on-farm −70%, off-farm +10%; change in GWP in NL: on-farm −13%, off-farm +1%). *Terrestrial acidification potential* follows a similar pattern in DK and NL, where acidification causes on-farm reductions of 70% and off-farm increments of 1–2%. For the ES case, TAP decreases on-farm and off-farm because the decrease in demand elsewhere in the world results in sufficient savings to compensate for additional material requirements. *Marine eutrophication potential* increased locally in DK and NL, but was reduced off-farm since slurry-N leads to more leaching than mineral N in the Daisy simulations. In DK and NL, field-applied slurry-N increases in the acidification scenario, while needs for mineral N fertiliser produced elsewhere decrease. *Freshwater ecotoxicity potential* declined as a consequence of acidification on-farm and off-farm in all countries, but to differing degrees, with an on-site rise of below 0.1% in DK and NL but an off-site rise of 10% and 2% respectively. In some categories, an increase in impact was observed locally as well as globally, such as for FEcoP, HNCTP and MEcoP in DK, HCTP and TEcoP in DK and NL, and FEuP in ES.

Of course, the location of GHG emissions does not play a role in terms of global climate change as emissions of GHGs are equally impactful no matter where they are emitted. However, due to the obligation of countries to account for their GHG emissions under the Paris Agreement and agreed national reduction targets, countries are incentivised to reduce national emissions before reducing global emissions. There is a tendency for slurry acidification to be used as a technology to shift impacts from one country to another, but overall the tendency is that the benefits of the acidification technology are much greater locally than the impacts inflicted on other places.

FPMFP is a problem for the agricultural sector, especially in the Netherlands, and its reduction is a matter of great concern. The increase in FPMFP elsewhere in the world against local reductions could potentially be justified if the affected areas are impacted less by the additional loads. This question could be answered by applying regionalised characterisation factors in future studies.

The local increase in MEuP in DK and NL could be problematic since the whole territory of each country has been declared a Nitrate Vulnerable Zone (Hou et al., 2018). The Baltic Sea around Denmark, for instance, suffers from eutrophication, with potential further deterioration under future climate conditions (Skogen et al., 2014).

Taking such local effects into account would require site-specific characterisation factors to be built into the impact assessment, whereas the ReCiPe methodology currently used is representative of Europe on average. Additional research could also be conducted on the impact of the provision of off-farm supplies, such as different energy sources or alternative production methods for fertilisers and pesticides to those provided by ecoinvent.

4. Conclusions

This study compared the acidification of pig slurry with no treatment in three intensive pig production regions of Denmark, Limburg in the Netherlands and Catalonia in Spain in terms of their environmental performance.

The results suggested that slurry acidification reduces the environmental impact of slurry management in those categories mostly related to agriculture, such as terrestrial acidification and global warming potential, and has the potential to contribute to enhanced nutrient recycling overall. Given its lower investment costs and easier implementation into existing systems compared with in-house acidification, storage acidification could be a viable technology for reducing GHG and fine particulate matter emissions in slurry management.

However, this study also showed that slurry acidification can have harmful impacts on those categories related more to the provision of energy (e.g. fossil resource depletion) or manufacturing (e.g. human toxicity). To justify slurry acidification as a cleaner production technology on all levels, energy and material sources should be examined and carefully selected. The study showed that the performance of the acidification technology depends on the context it is applied under. Therefore, it is important to bear in mind that regulatory and environmental conditions can have an impact on the performance of clean technologies when transferring them to new contexts.

Whether or not slurry acidification replaces S fertilisation and whether additional liming becomes necessary has little impact on the environmental performance of slurry acidification against the baseline. However, in terms of cost and ease of implementation, the replacement of S fertilisation with outdoor storage slurry acidification could be a policy change worthy of further investigation. When comparing slurry acidification against stricter P application rates, stricter P application limits in the Netherlands would appear more favourable than the introduction of acidification. In future studies, it should be investigated how slurry acidification can be combined with other slurry treatment technologies like slurry separation to achieve better environmental performance. In addition, it should be investigated how different

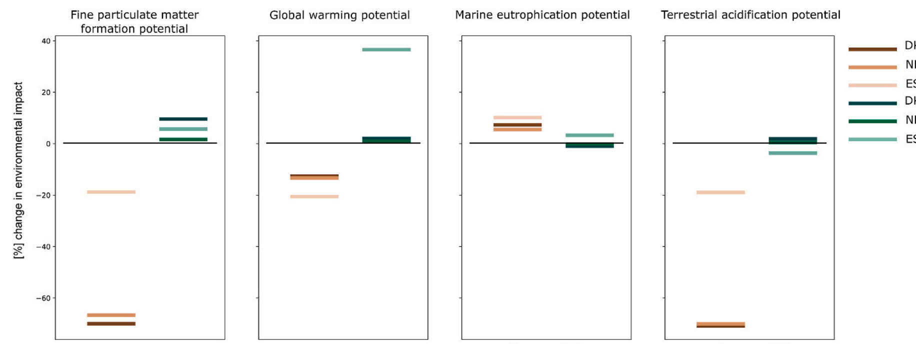

Fig. 3. Changes in environmental impact as a consequence of using slurry acidification in the three study regions of Denmark (DK), Limburg in the Netherlands (NL) and Catalonia in Spain, subdivided into farm-site effects (brown) and off-site effects (blue).

acidification strategies (in the animal house, in the storage tank or during field application) and alternatives to sulphuric acid can reduce environmental impacts in categories where the effect of the acidification is less favourable.

This study could be used to support the decision-making of different stakeholders involved in the agricultural sector and interested in closing nutrient loops and improving the environmental performance of pig production and manure handling. It could also be used as a starting and reference point for slurry acidification studies under circumstances and assumptions other than the ones presented here.

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CRediT authorship contribution statement

Miriam Beyers: Conceptualization, Data curation, Formal analysis, Methodology, Visualization, Writing – original draft, Writing – review & editing, LCA modelling. **Yun-Feng Duan:** Methodology, Resources, Writing – review & editing, Daisy modelling. **Lars Stoumann Jensen:** Conceptualization, Funding acquisition, Methodology, Project administration, Supervision, Writing – review & editing. **Sander Bruun:** Conceptualization, Data curation, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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