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Evaluation of the Sustainability of a Prototype for Atmospheric Ammonia Capture from Swine Farms Using Gas-Permeable Membrane Technology

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Abstract: Ammonia (NH_3) emissions from animal wastes are directly related to serious environmental problems and can be reduced by using gas-permeable membranes (GPMs) in animal housings, but not many studies have been conducted on the environmental impact of the entire system. Thus, the aim of this work was to analyze the environmental impacts caused by the implementation of GPM technology in a 920-animal swine farm with a closed cycle (i.e., birth, breeding, transition, and fattening take place on the same site), using life-cycle analysis (LCA). Two scenarios were studied: a reference scenario in which there was no NH_3 reduction from the air captured in the sheds and a treatment scenario that used the GPM technology. The LCA results were evaluated by using the ReCiPe 2016 Midpoint (H) V1.04 method, showing that using the GPM technology had beneficial environmental impacts. Terrestrial acidification (TA) showed a reduction of 14.68 kg SO_2 eq compared with the reference scenario, whilst human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HnCT), and land use (LU) showed reductions of 1.52 kg 1,4-DCB, 66.26 kg 1,4-DCB, and 44.55 m^2a crop eq, respectively.

Keywords: ammonia reduction; air scrubbing; emissions in livestock farming

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1. Introduction

Over the years, the European Union has been implementing different strategies to mitigate and reduce total greenhouse gas emissions [1–3]. Spain ranks fifth among EU countries for emissions generated by the agricultural sector [4]. Most of the agricultural emissions are of nitrogen compounds, such as ammonia (NH_3), and come from pastures as a consequence of the application of animal manures. Ammonia is a compound that reacts with acids to promote the formation of fine particulate matter (FPM), causing soil acidification, eutrophication, and environmental degradation, and although it is not a greenhouse gas (GHG), it has serious effects on air quality and terrestrial ecosystems [4]. The main atmospheric effects related to nitrogen oxides (NO_x) and NH_3 emissions include elevated ozone concentrations that increase the greenhouse effect [5]. Due to the reactivity of ammonia with a variety of atmospheric acids, such as nitric acid (HNO_3), hydrochloric acid (HCl), and sulfuric acid (H_2SO_4), particulate matter (aerodynamic diameter of around

2.5 μm) is formed, which not only impairs atmospheric visibility, but also has an adverse effect on human health [5,6]. Additionally, the deposition of nitrogen compounds contributes to the acidification and eutrophication of ecosystems [5].

At ambient temperature, ammonia is a colorless gas with an intense smell and high toxicity in the form of ammonium (NH_4^+), and it is very soluble. Both forms are kept in a dynamic balance; however, the balance can be affected by changes in different variables [7], such as temperature [8] and pH [9]. The conversion of ammonium to ammonia and its volatilization increases with the temperature and pH level of the medium [10].

As a consequence of the great variety of emission sources from the agricultural and livestock sectors, different mitigation strategies have to be taken into account to reduce impacts according to different systems and their own characteristics [11,12]. Most of the environmental impacts generated in swine production systems occur during crop and feed production, manure management, and the facilities and operational activities required for animal farming [13]. More than 70% of the nitrogen (N) consumed by pigs in their diet is excreted in manures, mainly in the form of urea in their feces and urine (>75% of total N excretion); in the case of excrement, N is bound to feed proteins. Managing excretions results in certain amounts of soluble N being lost by runoff into surface waters, leaching to underground waters as nitrate, or conversion into volatilized N_2; however, the largest amount of N is finally released to the atmosphere as ammonia [14]. As a result of this, and taking into account the involvement of agriculture in the European emissions of ammonia (i.e., approximately 94%) [15], different sustainable practices have been put in place to reduce the impact on livestock and farming operations. In the case of gaseous emissions that pollute waters, eutrophying gases, mainly NO_x and NH_3, are the main environmental pollutants that cause an excess of nutrients in aquatic ecosystems and adverse effects [16]. Approximately 80% of the manure produced in livestock farming is treated, of which 22% is leached and 15% is volatilized; 30% of the manure applied directly to pastures is leached and 20% is volatilized [17].

Decreasing ammonia formation and increasing nitrogen use efficiency are among the best options proposed to decrease ammonia emissions [6].

Different strategies to capture ammonia from the air have been evaluated. Some examples include the use of a catalyst to extract ammonia, with the aim of purifying substances and compounds [18], and the use of air purifiers [19,20] that can reduce NH_3 emissions and also strong smells and dust [21]. These gas-capture devices are frequently used in some European countries to achieve the level set by maximum emission indexes [22]. Adding acids is one of the most usual forms of ammonia capture; acid capture devices have been demonstrated to have high efficiency, with up to 100% reduction of NH_3 and also resulting in the production of solutions with a high content of nitrogen (N) and sulfur (S), which can be used as fertilizers [20]. A new innovative technique that is being implemented is the use of a gas-permeable membrane (GPM), in which the NH_3 in the air is discharged from the production shed through a microporous hydrophobic membrane through diffusion [23] with a solution rich in sulfuric acid (H_2SO_4) that captures and recovers the ammonia in the form of ammonia sulfate ($(NH_4)_2SO_4$). This solution is used as fertilizer in agricultural soils [22] and is thus considered a valuable product [24], facilitating the circular economy through the transformation and exploitation of air in animal shed compounds.

Life-cycle analysis (LCA) is a methodology that is used to evaluate the environmental impact of processes and/or products in an integral way [25]. One of the main advantages of LCA is the integral perspective of processes and the inclusion of environmental issues, which allows a detailed comparison that includes different products, processes, resources, and emissions that take place in different locations and at different times. LCA has been used in the pig-production industry to evaluate feed production [26], animal breeding systems [27–31], and waste management [13,28]. LCA is considered an effective tool to analyze the potential environmental impact through the identification of emissions in production chains [29] due to the detailed inventory and process analysis.

This research work follows up on a European Union-funded project to develop an environmental and economically viable solution that will allow the reduction of emissions from manures in livestock anaerobic digestion processes, with the overall objective of minimizing atmospheric emissions and improving the circular economy in rural regions where pig production is established. The result of this development was prototype ammonia-capture equipment based on GPM technology. The aim of the current work is to evaluate the environmental impacts of the ammonia-capture prototype using LCA. The application of the LCA methodology in the first development stages of this innovative technology will allow the identification of hotspots within the process for future improvements of the device. In order to evaluate the sustainability of the prototype, two scenarios were considered: a treatment scenario (TS) that considers the use of the prototype and the production of $(NH_4)_2SO_4$ in situ, and a "reference scenario" (RS) without any processes to reduce the emissions of ammonia in the air.

The evaluation of the sustainability of GPM technology in the management of ammonia emissions in swine farms represents an innovative analysis that aims to establish a baseline for the large-scale implementation of this alternative. It takes into consideration the benefits of obtaining a liquid nutrient that can be directly spread into the soil as fertilizer and also a reduction in atmospheric ammonia. For this reason, this work focuses on a scenario in which GPM technology was implemented in a prototype to capture and treat the air from swine housing. The aim of using LCA is to determine the processes that have the greatest relevance in the impacts generated during the operation of the prototype. The environmental performance of the prototype was compared with a reference scenario in which any air treatment alternative was available. The data inventory used in this LCA study came from primary sources and from real-time measurements of the processes that were considered. Thus, a sustainability evaluation was completed of a prototype developed by Inderen S.L., an engineering company dedicated to the promotion and development of environmentally sensitive technology. The prototype was designed for the capture of NH_3 in the atmosphere of farms and the subsequent recuperation of N as an ammonia sulfate solution ($(NH_4)_2SO_4$) using GPM technology. The main objective of this work is to present an environmental evaluation of the implementation of ammonia-capture equipment with GPM. The results presented correspond to the data obtained from the use of the prototype in a pig farm in Spain.

2. Materials and Methods

LCA methodology with ISO 14040 and 14044 standards was used to perform a sustainability assessment regarding the implementation of the ammonia-capture prototype using GPM technology [25–30]. A reference scenario (RS) was established to show the differences with the air treatment. This study had an attributional approach, which attributed the impacts to defined stages in the systems and was modeled with SimaPro® software (Version 9.1.1.1 PhD, The Netherlands) following the impact analysis method ReCiPe 2016 Midpoint (H) V1.04, middle point level with a hierarchic perspective.

Primary data were collected directly from the farm, and Ecoinvent 3—Allocation at Point of Substitution—System [31] and ELCD [32] databases were the sources of secondary data described in Section 2.2.

2.1. Goal and Scope of LCA

The goal of the LCA was the analysis of the implementation of an ammonia-capture prototype with data obtained from a farm located in Guardo (Palencia, Spain) to identify the benefits obtained by using the prototype in the farm; the rest of the activities associated with the production of animals in the farm were excluded from this study for both scenarios, as they had the same conditions and, thus, the same environmental loads. The shed had an area of approximately 1840 m^2, with a 6808 m^3 volume and a total of 920 sows in a closed cycle; the capture prototype was implemented in the shed as a pilot plant. The analysis period was one complete year, commencing in August 2020. The detailed protocol and methodology of the operation were described by Soto-Herranz et al. [24], who performed an analysis of the operation of the prototype. The different stages considered in both scenarios were the purchase of mineral fertilizer (RS), transport of the fertilizer to the point of application near the farm (RS), prototype operation for ammonia capture and extraction of the subproduct, which is then used as fertilizer (TS), and fertilizer transport to distribution area (RS, TS). The full description of each scenario is presented in Section 2.2.1.

The functional unit was defined as the 'capture of 1 kg of ammonia from the air in the swine shed'. The same functional unit was selected in a study with the objective of ammonia capture in the air [22].

2.2. Inventory Data and Assumptions

Primary data used in both scenarios were taken from an operating pilot plant located on the farm. Gas emissions to the atmosphere were calculated from manure production in the sheds. Other data were taken from Ecoinvent 3.6 [31] and ELCD [32] databases. Different methodologies from different authors were selected to establish the proportion of emitted gases according to the functional unit to be studied.

Emissions estimates in the sheds.

According to the literature, during the storage of manure in sheds, the most important gaseous emissions are those of ammonia (NH_3), methane (CH_4), nitrous oxide (N_2O) [28], and nitrogen oxides (NOx) [33]. The emissions generated in the sheds were estimated where the ammonia capture equipment was installed. Enteric fermentation was not included, as the emissions generated did not have significant importance compared with the emissions generated from the storage of manure. The amount of NH_3-N generated by the farm was calculated to be 5277.9 kg/year. The calculation was performed using the manure characterization data obtained in the lab (See Table 1, Equation (1)) considering the concentration of total ammoniacal nitrogen (TAN) (i.e., 2.8 kg/m^3) in the manure produced by the farm every year (17,136 m^3/year) according to the data provided by the farm staff. The sheds were naturally ventilated. The amounts of greenhouse gases produced were calculated based on occupancy and shed operation data.

Table 1. Chemical characterization of raw manure based on experimental data. Source: Own work from data generated by González-García et al. [23].

Parameter	Value	Units
pH	7.6	-
Total Solids (TS)	41.7	kg/m^3
Volatile solids (VS)	31.3	kg/m^3
Total ammonia nitrogen (TAN)	2.8	kg/m^3
Total Kjeldahl nitrogen (TKN)	3.6	kg/m^3

Ammonia NH_3 emissions from the storage of manure in sheds

The sows mating shed has a system of a combined deep septic tank with nutritional management techniques and pressure washing. (MTD 30.a4). (UE) 2017/302 [34] used the Tier 2 method proposed by the EMEP/EEA (2019) [35] Inventory Guide:

$$NH_3 - N = EF_{NH_3} \times TAN \quad (1)$$

where:

$NH_3 - N$: Ammonia produced in swine housing, kg $NH_3 - N$;

EF_{NH_3} : Emission factor, 0.11 $\frac{kg NH_3 - N}{kg TAN}$ (Table 3.10; Chapter 3B) [35].

The amount of NH_3 is obtained by multiplying by the factor 17/14,

$$NH_3 = NH_3 - N \times \frac{17}{14} \quad (2)$$

Methane CH_4 emissions from the storage of manure in sheds

CH_4 emissions were estimated following the IPCC Guidelines [36]:

$$CH_4 = VS \times B_0 \times 0.67 \times MCF \quad (3)$$

where:

CH_4 : Methaneemissions, $kgCH_4$;

VS : Excreted volatile solids, kg;

B_0 : Maximum methane − producing capacity for swine manure, m^3CH_4 /kg VS;

0.67 : Methane density, $m^3CH_4/kgCH_4$;

MCF : Methane conversion factor.

Nitrogen N emissions from the storage of manure in the sheds

A concentration of 6446.82 mg N/l was obtained from the manure sample in the shed, equivalent to 119.79 g N/(place·day).

Thus,

$$N = 0.11979 \times N^{\circ}\, of\ places \times 365 \quad (4)$$

where:

N : Nitrogen produced in swine housing, kg N.

Dinitrogen (N_2) emissions from the storage of manure in sheds.

The N_2 emissions were estimated using the Tier 2 methodology proposed by the EMEP/EEA [35]:

$$N_2 = EF_{N_2} \times TAN \quad (5)$$

where:

N_2 : N_2 emissions, kg N_2;

EF_{N_2} : Emission factor, $0.003 \frac{kgN_2}{kgTAN}$ (Table 3.10; Chapter 3B);

TAN : Total ammonia nitrogen, kg TAN.

Based on the total emission data, the NH_3-N emissions captured per kg were calculated. Furthermore, the prototype operation inputs were considered during the process of ammonia capture. Table 2 shows the emissions in the sheds.

Table 2. Emissions generated in swine farms annually.

Type of Waste/Emissions	Quantity	Units	Data Source
N	40225.5	kg/year	Experimental data
CH_4	4851.3	kg/year	IPCC (2019)
NH_3-N	5277.8	kg/year	EMEP/EEA Inventory Guidebook (2019)
NH_3	6408.9	kg/year	EMEP/EEA Inventory Guidebook (2019)
N_2	143.9	kg/year	EMEP/EEA Inventory Guidebook (2019)

2.2.1. Description and System Limits for the Reference Scenario

In the 'reference scenario' of this study, the current practices that are considered in animal housing consist of emissions released to the atmosphere without any treatment, mainly caused by the storage of manure. The main stages considered were the storage of manure in the sheds and the production of mineral fertilizers, which not only involves expensive manufacturing processes, but also represents a considerable environmental burden. Transport to the farm was also considered, but transport to application points in the field or the application itself were not taken into account because the application of fertilizer would take place in the farm itself where the prototype is located; thus, the environmental burden in the two scenarios would be the same (Figure 1).

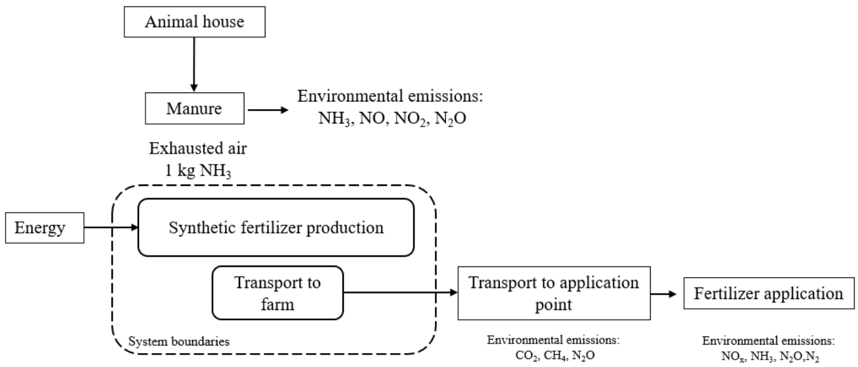

Figure 1. System boundaries for the reference scenario.

2.2.2. Description of Systems and System Boundaries for Treatment Scenario

In this scenario, the following steps were taken into account: storage of manure in the sheds and the operation of the prototype with GPM technology. The resources and processes used in the analysis of the operation of the prototype and the environmental impacts of its operation were the production of sulfuric acid (H_2SO_4), the manufacture of the expanded polytetrafluoroethylene tubular membrane (e-PTFE), the energy used for the operation of the prototype, and the water consumed in the process (Figure 2).

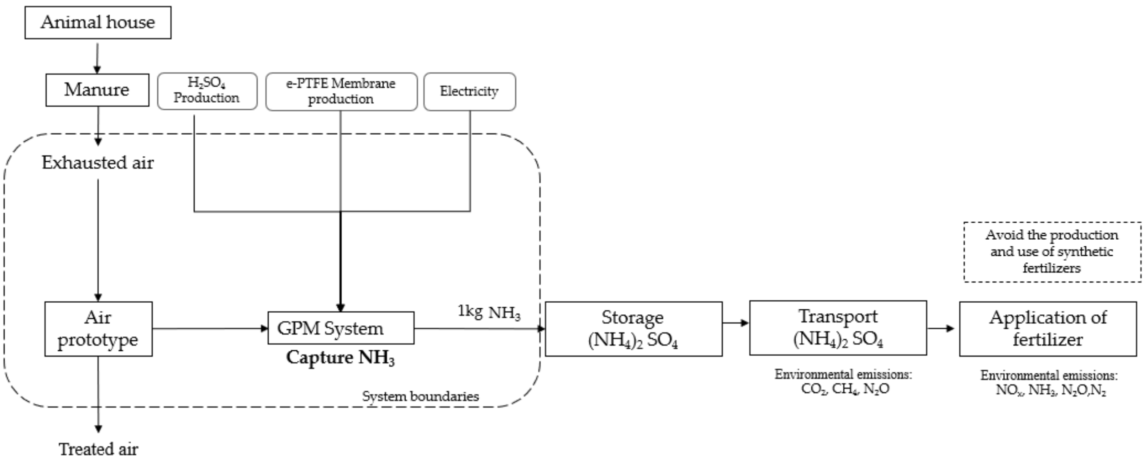

Figure 2. System boundaries for the treatment scenario.

Air Prototype Operation

The prototype (Figure 3) with GPM technology has an air reservoir in which the grids that support the tubular membrane of e-PTFE are located; sulfuric acid circulates continuously through the interior of the membranes. The air collected from the pig farms system passes through the membranes, allowing the air to cross the microporous membrane and the ammonia present in the air to be captured in a H_2SO_4 solution with an initial 1N concentration. The resulting product is ammonium sulfate $(NH_4)_2SO_4$, which is stored in a reservoir. The prototype has a pump to drive sulfuric acid through the membrane and a fan to force the circulation of air containing volatile nitrogen compounds through the membrane. The prototype was designed and manufactured by an engineering company, Inderen S.L., located in Valencia, Spain.

The ammonia GPM capture process is shown in Figure 4. Air containing NH_3 passes through the porous membrane, where the acid is circulating. NH_3 is combined with acid-free protons and forms non-volatile ammonium ions (NH_4^+); in this case, ammonium sulfate is obtained by using sulfuric acid [37], which is used to capture ammonia due to its lower cost among inorganic acids and also because sulfur is a valuable nutrient.

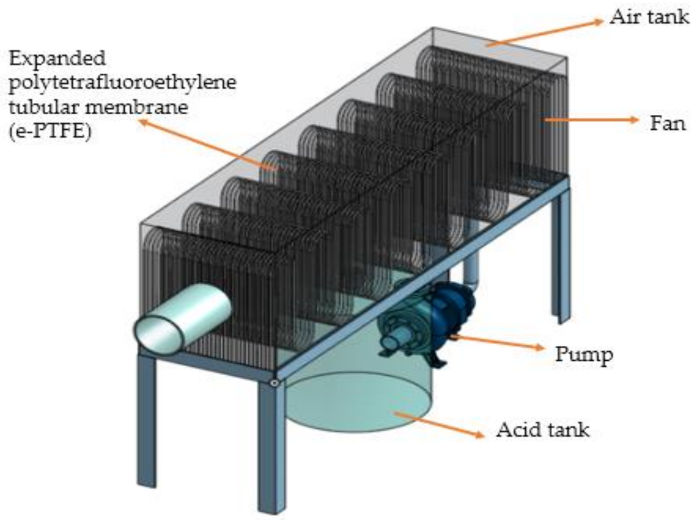

Figure 3. Ammonia capture prototype.

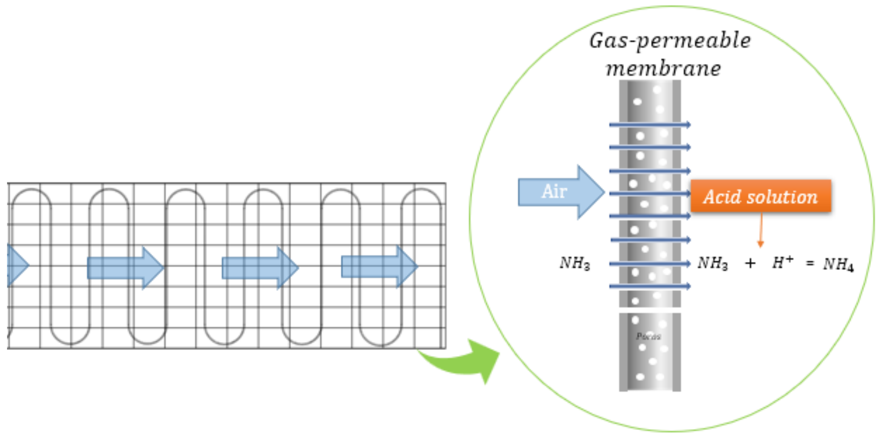

Figure 4. Operation of the NH_3-capture process.

It was determined that for one year of operation of the prototype, 15 m of membrane and 156 l of 1 N sulfuric acid were required. The prototype had an initial volume of 150 l of sulfuric acid, and two corrections of 3 l each were added in order to maintain the concentration as close to 1 N as possible. The annual trapping rate was 28.2 g N/l ammonium sulfate.

2.2.3. Transport

Transport from the place where the mineral fertilizer was purchased to the application point close to the farm (400 km average) was considered for the reference scenario. Transport from the farm to the fertilizer-spreading point was not included in the same environmental burden results for both scenarios.

2.2.4. Environmental Impact Analysis

The evaluation of the environmental impacts was carried out with the life-cycle impact assessment method ReCiPe 2016 V1.04 [38] at the midpoint level, which has, as the main objective, the transformation of inventory data into a limited number of scores for different impact categories (global scope), assuming a hierarchist perspective. For the evaluation, the method considers the following impact indicators: global warming (GW), stratospheric ozone depletion (SOD), ionizing radiation (IR), ozone formation—human health (OFH), fine particulate matter formation (FPM), ozone formation—terrestrial ecosystems (OFT), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME), terrestrial ecotoxicity (TEC), freshwater ecotoxicity (FEC), marine ecotoxicity (MEC), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HnCT), land use (LU), mineral resource scarcity (MRS), fossil resource scarcity (FRS), and water consumption (WC).

In relation to the assessment of the impact of the prototype for the capture of ammonia in the air, the categories with the greatest involvement were GW, OFH, FPM, OFT, TA, TEC, FEC, MEC, HCT, HnCT, LU, and WC. These midpoint impact categories were selected for the following reasons: GW is associated with global warming, while OFH and OFT are associated with health problems in people, atmospheric pollution, and ecosystem impacts. Additionally, FPM involves the formation of fine particles of 2.5 microns in size that pollute the environment. TA is strongly associated with the processes considered in the analysis due to the environmental loads that are created by the emissions of nitrogen oxides and sulfur to the atmosphere and soil, as well as the macronutrients present in fertilizers, such as nitrogen, that can cause eutrophication in soils and waters (TEC, FEC, and MEC). HCT and HnCT are related to toxicity to human health. LU can be affected by changes in land use due to activities developed to carry out the processes analyzed, and WC was included due to the need for this resource to be able to operate the prototype adequately. Additionally, the analyses carried out by other authors using LCA methodology for the treatment or recovery of ammonia in liquids using GPM technology [23] and in the air of pig farms [22] were considered. The impact categories that were not considered had a minimal score in the global impact.

3. Results

Figure 5 shows the results for the reference scenario where the two processes analyzed were the purchase of ammonium sulfate as a fertilizer and transport to the point near the farm. It is evident that the production of ammonium sulfate had a greater effect on the impact categories compared with the transport phase. The indicators with the greatest impact were those associated with the atmospheric environment, such as ozone formation—human health (OFH), ozone formation—terrestrial ecosystems (OFT), terrestrial acidification (TA), terrestrial ecotoxicity (TEC), freshwater ecotoxicity (FEC), marine ecotoxicity (MEC), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HnCT), land use (LU), and water consumption (WC).

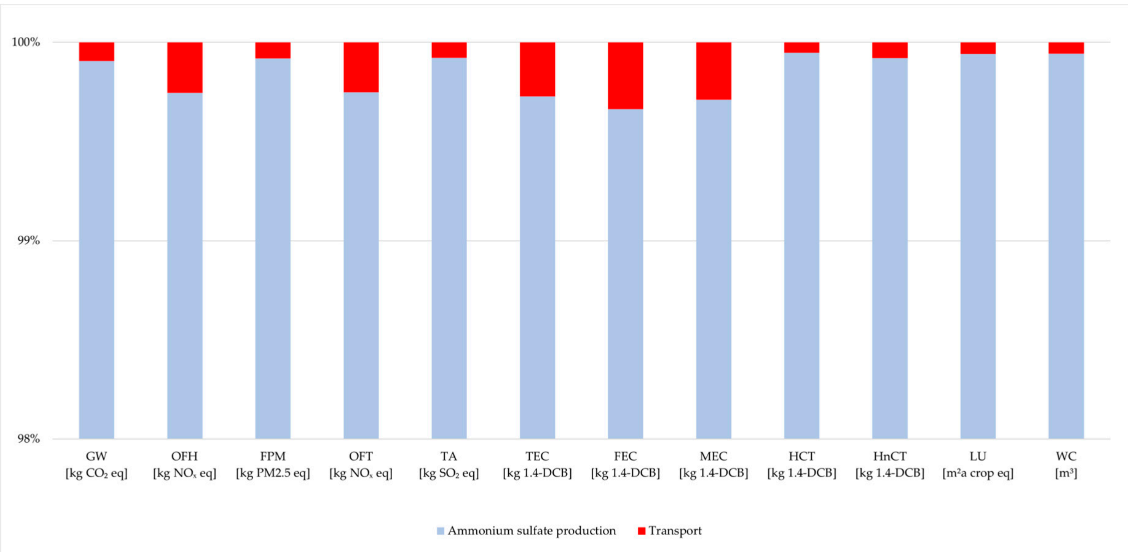

Figure 5. Normalization of impacts for the reference scenario source: Own work with SimaPro data. Global warming (GW), ozone formation—human health (OFH), fine particulate matter formation (FPM), ozone formation—terrestrial ecosystems (OFT), terrestrial acidification (TA), terrestrial ecotoxicity (TEC), freshwater ecotoxicity (FEC), marine ecotoxicity (MEC), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HnCT), land use (LU), and water consumption (WC).

The data were normalized to establish a common scale that would allow their influence on the impacts generated to be determined. Values for the indicators calculated by the ReCiPe method that had the greatest impact were analyzed and are shown in Table 3.

Table 3. Categorization of impacts for the reference scenario.

Id	Impact Category	Unit	Total	Ammonium Sulfate Production	Transport
GW	Global warming	kg CO_2 eq	124.03	123.92	0.12
OFH	Ozone formation—human health	kg NO_x eq	0.24	0.24	0.00
FPM	Fine particulate matter formation	kg PM2.5 eq	0.21	0.21	0.00
OFT	Ozone formation—terrestrial ecosystems	kg NO_x eq	0.25	0.25	0.00
TA	Terrestrial acidification	kg SO_2 eq	0.49	0.49	0.00
TEC	Terrestrial ecotoxicity	kg 1.4-DCB	810.57	808.36	2.21
FEC	Freshwater ecotoxicity	kg 1.4-DCB	0.10	0.10	0.00
MEC	Marine ecotoxicity	kg 1.4-DCB	0.57	0.57	0.00
HCT	Human carcinogenic toxicity	kg 1.4-DCB	1.29	1.29	0.00
HnCT	Human non-carcinogenic toxicity	kg 1.4-DCB	49.80	49.76	0,04
LU	Land use	m^2a crop eq	24.27	24.25	0.01
WC	Water consumption	m^3	0.39	0.39	0.00

Figure 6 shows the results obtained for the treatment scenario with the prototype for all impact categories. The LCA results show that electricity was the process with the greatest load in most of the impact categories, followed by sulfuric acid production and the material used for the manufacture of the e-PTFE membrane as a result of its high resistance to degradation. Negative values refer to the improvements in the impact categories, which were the result of the production of ammonia sulfate from ammonia that was concentrated from the air in the sheds.

Table 4 shows the values for the results obtained for the treatment scenario.

SimaPro Comparison Scenarios

Figure 7 shows the indicators for the impact categories comparing both scenarios. The results show that the impact categories related to toxic contaminants had a higher impact in the reference scenario, which was mainly associated with the industrial processes of ammonia sulfate production. The global net impact of the GPM technology in the air of pig sheds can have a positive impact; however, further work is required to improve some of the processes and prevent the impacts from moving across.

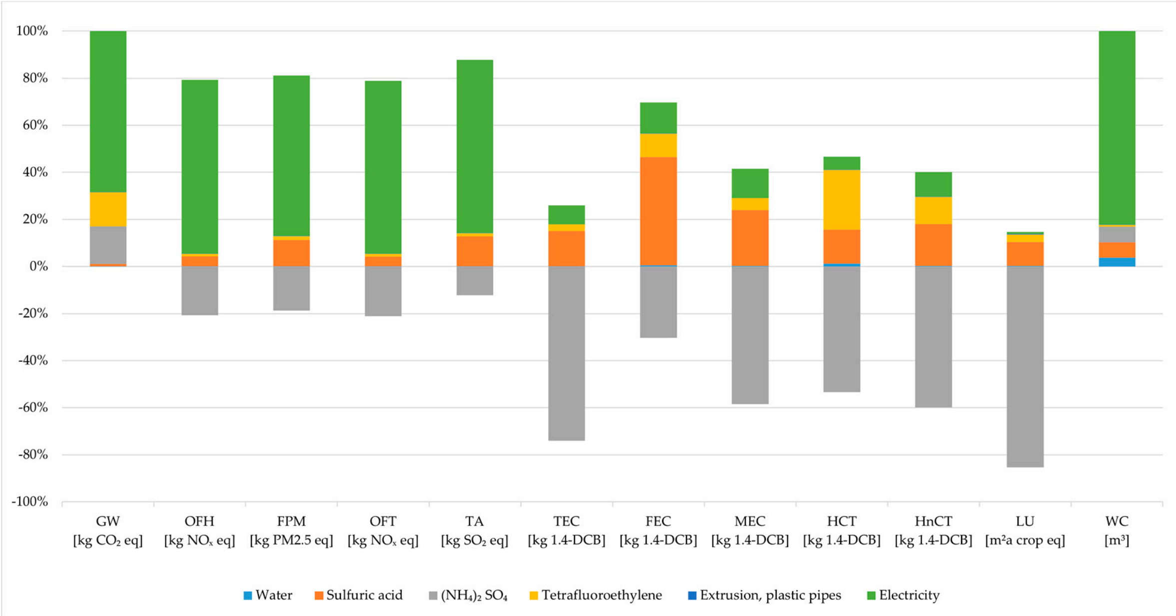

Figure 6. Normalization of impacts for the treatment scenario. Source: Own elaboration with SimaPro data. Global warming (GW), ozone formation—human health (OFH), fine particulate matter formation (FPM), ozone formation—terrestrial ecosystems (OFT), terrestrial acidification (TA), terrestrial ecotoxicity (TEC), freshwater ecotoxicity (FEC), marine ecotoxicity (MEC), human carcinogenic toxicity (HCT), human non-carcinogenic toxicity (HnCT), land use (LU), and water consumption (WC).

Table 4. Categorization of the impacts of the treatment.

Id	Unit	Total	Water	Sulfuric Acid	$(NH_4)_2$ SO_4	Tetrafluoroethylene	Extrusion, Plastic Pipes	Electricity
GW	kg CO_2 eq	553.50	0.49	5.52	88.42	80.17	0.30	378.59
OFH	kg NO_x eq	0.58	0.00	0.04	−0.20	0.01	0.00	0.73
FPM	kg PM2.5 eq	0.44	0.00	0.08	−0.13	0.01	0.00	0.48
OFT	kg NO_x eq	0.58	0.00	0.04	−0.21	0.01	0.00	0.74
TA	kg SO_2 eq	1.51	0.00	0.25	−0.25	0.03	0.00	1.47
TEC	kg 1.4-DCB	−462.50	1.09	144.42	−712.04	26.95	0.36	76.72
FEC	kg 1.4-DCB	0.05	0.00	0.06	−0.04	0.01	0.00	0.02
MEC	kg 1.4-DCB	−0.12	0.00	0.17	−0.42	0.04	0.00	0.09
HCT	kg 1.4-DCB	−0.14	0.02	0.29	−1.07	0.50	0.00	0.11
HnCT	kg 1.4-DCB	−13.36	0.13	12.06	−40.36	7.70	0.05	7.05
LU	m^2a crop eq	−18.89	0.07	2.72	−22.81	0.84	0.06	0.23
WC	m^3	11.45	0.44	0.73	0.77	0.09	0.01	9.42

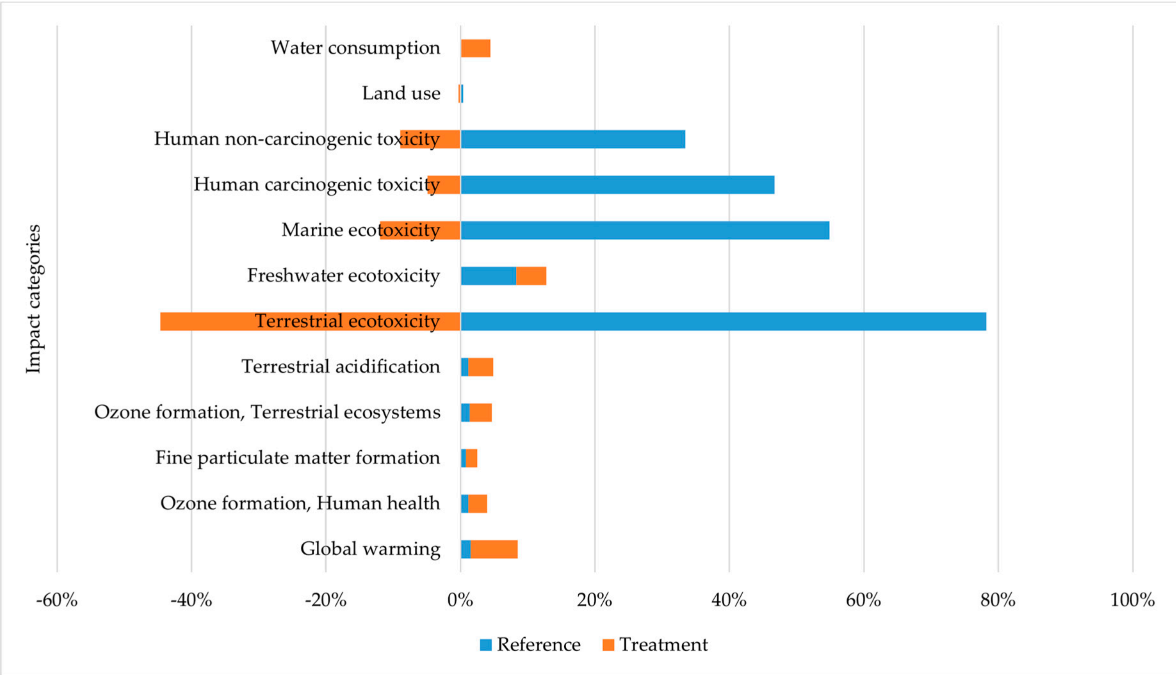

Figure 7. Categorization of impacts for both scenarios: Comparing "Reference with Treatment"; ReCiPe 2016 Midpoint (H) V1.04/World (2010) H/Characterization /Excluding infrastructure processes/Excluding long-term emissions. Source: Own elaboration with SimaPro data.

4. Discussion

The objective of the LCA was to evaluate the implementation of a prototype with GPM technology to capture ammonia. For this reason, the limits of the system were delimited in the operation of the prototype in the pig sheds. Considering that the prototype produced ammonium sulfate that could be used as fertilizer, the synthetic fertilizer equivalent to ammonium sulfate was established as a reference scenario and was transferred to the same point of application.

The results of the analysis showed that using the prototype for ammonia capture improved the environmental impact of the system compared with the reference scenario for most of the impact categories. The main reason for this was that there was a reduction in the categories related to toxicity (i.e., HnCT, HCT, MEC, FEC, TEC, and LU), which was mainly caused by the manufacture of the synthetic fertilizer. The reason behind this result is that, during the manufacture of these types of compounds, large amounts of NO_x, NH_3, N_2O, SO_2, and other emissions are released as a consequence of the techniques used for production [39].

The approach of using attributional LCA was appropriate because it allowed the unitary processes included in the analysis that are more relevant to the impact categories to be established, which would allow specific improvements in the development of the technology to be determined.

In this work, in order to reduce the uncertainties of the calculations, real information was used during the period of the prototype's operation; additionally, it was conducted using estimations from other authors [35,36], as presented in Section 2. This information provides an overview of the behavior of emissions in swine sheds, which was also supported by information from databases, such as electricity production in Spain [32], as well as the production of sulfuric acid and fertilizer (ammonium sulfate) [31]. González-García et al. [23] used an ammonia capture prototype with GPM technology submerged in manure, similar to that used in this study. In their treatment scenario, they obtained a value of 97 kg CO_2 eq, which shows a decrease in this impact category compared with the baseline scenario (i.e., 113 kg CO_2 eq). On the contrary, in the present work, the value obtained for the treatment scenario was 553.50 kg CO_2 eq, which showed an increase in the impact compared with the reference scenario (i.e., 124.03 kg CO_2 eq). This different effect was the result of the use of an additional engine for the prototype to propel air through the membranes, resulting in higher energy consumption, which caused 68% of the impact. This additional engine was required for the operation of the ventilator that extracted air from the sheds. In the study of air scrubbers for ammonia abatement, De Vries and Melse [22] showed that electricity consumption represents a higher contribution of GW during the operation of devices similar to that used in this study. The trend of this indicator was due to the phases that were considered within the electricity production process, as well as its different sources in Spain [32]; Melse and Ogink [20] stated that the installation of a capture device increases electricity consumption by 57 W per 1000 m^3/h of installed ventilation capacity; however, manufacture companies tend to decrease energetic use. Regarding the membranes, the manufacture of the e-PTFE caused an impact of 14%. The use of engines, electricity consumption, and the production of raw materials for the ammonia capture equipment (Figure 6) were the most relevant factors when evaluating the environmental performance of the system in the short term (i.e., 1 year); however, these impacts could be decreased when considering a longer period, as the lifetime of the components is considerably high; then, energy consumption will be the main source of impacts.

Focusing on efficiency, the captured TAN by the prototype measured for a day was approximately 18 g/d. Soto-Herranz et al. [24] obtained a value of 0.022 g/d in a study in labs using a GPM membrane with swine manure. The increase in the farm was linked to the surface of the membrane used in the labs (i.e., 0.01634 m^2) as compared with the surface of the current prototype (i.e., 7.7 m^2), as well as the availability of the emitted gases being higher in farms than in laboratories. On the other hand, Soto-Herranz et al. [24] reported a TAN recuperation efficiency of 73.3% in lab trials using GPM in manure gases, which corresponds to 1.31 g of recuperated NH_3. They compared these results with the operation of the same prototype in poultry houses and found that the efficiency was associated with the ventilation of the shed, as there was a higher NH_3 concentration in the pig farms with less ventilation; therefore, the performance was better. With the aim of reducing emissions, these results confirm that, for the prototype to be more sustainable during its operation, it must be assured that air is propelled through the membranes and as little as possible is released outside; however, the operation time must be constant to avoid health problems in the animals associated with toxic environments, which makes it necessary to use alternative energy sources from renewables. However, Molinuevo-Salces et al. [40] achieved average TAN values of 26.85 $g/(m^2 \cdot d)$ when working with a membrane with the same characteristics directly from manure, which means higher ammonia capture. Previous studies showed that ammonia recuperation in liquids is higher than that in gases, which could be related to the higher NH_3 concentration in manures as compared with that in air [24].

In order to improve the capture process, the contact surface of the air membrane and the flow of captured air should be increased; however, this modification will require higher energy consumption that will affect the impact indicators related to electricity consumption.

The calculated value for fine particulate formation (FPM) was 210 g PM2.5 eq/kg of NH_3-N for RS, which was close to the results obtained by González-García et al. [23] (i.e., 231 g PM2.5 eq/m^3 manure) for a conventional scenario. This indicator increased for the treatment scenario (i.e., 440 g PM2.5 eq/kg of NH_3-N) and, similar to GW, it could be mainly attributed to the operation of the prototype [22]. This result makes sense, as ammonia is a precursor of particulate matter [22].

For terrestrial acidification (TA), the net value for the RS was approximately 0.49 kg SO_2 eq/kg of NH_3-N, which was fully attributed to the mineral fertilizer. For the TS, the value obtained was 1.51 kg SO_2 eq/kg owing to the operation of the prototype, and consequently the electricity consumed (about 97.4 %) and the production of sulfuric acid (16.8 %), which was compensated by the ammonium sulfate (−16.2 %) that was obtained as a by-product. In this type of analysis, in which alternatives for the handling and management of fertilizers and slurry are proposed, acidification plays a fundamental role because the main compounds that take are involved in the processes, from raw materials to the products themselves, are NH_3, NO_x, and SO_2 [22]. Therefore, although adding chemicals might suggest a higher rate of nutrient recovery, it is important to use chemical inputs efficiently because the additional environmental impacts cannot be fully compensated [22].

In the terrestrial ecotoxicity (TEC) category, a net total of 810.57 kg 1.4-DCB was obtained, which represents excess toxic agents that are harmful to the terrestrial ecosystem, mainly caused by industrial processes to produce chemicals, such as mineral fertilizer (i.e., 99.7%), and transport (in this case with a negligible participation of 0.3%). For the TS, this indicator was calculated to be −462.5 kg 1.4-DCB, which was attributed to the ammonia capture process with the prototype, indicating improvements for this impact category as its value was considerably reduced. This coincides with the work by Verdi et al. [41], who reported that conventional agriculture had a greater impact on terrestrial ecotoxicity. In addition, for freshwater ecotoxicity (FEC), the value obtained was 0.10 kg 1.4-DCB, which, in relation to the previous category, had a significantly lower impact (i.e., 0.013 %) due to the production of ammonium sulfate (99.7%) and transport. For FEC, in the treatment scenario, the value was 0.05 kg 1.4-DCB, which, in relation to the RS, was reduced by almost 50% due to the production of ammonium sulfate. Verdi et al. [41] indicated that, for this impact category, they also obtained relevant results for terrestrial ecotoxicity in the evaluation of conventional farming systems. The marine ecotoxicity (MEC) impact was 0.57 kg 1.4-DCB, with the same participation of stages as FEC and TEC. For TS, the value of MEC was −0.12 kg 1.4-DCB, also indicating improvements in the impact category. Similar to the results obtained by Verdi et al. [41], MEC relies heavily on fertilizer production.

The production of mineral fertilizer was the main contributor to carcinogenic toxicity in humans (HCT) (i.e., percentage contribution of 99.9%), with a value of 1.29 kg 1.4-DCB/kg NH_3-N, which was related to the generation of waste harmful to human health. For non-cancerous toxicity in humans (HnCT), the value achieved was 50 kg 1.4-DCB, which, compared with the previous value, shows a greater influence due to the industrial production of fertilizer (corresponding to 99.9%). For the treatment scenario, the net value for HnCT was −0.23 kg 1.4-DCB/kg NH_3-N, which was related to an improvement in this impact category using the prototype for ammonia capture. On the other hand, HnCT also showed an improvement compared with the previous one, with a net value of −16 kg 1.4-DCB; difference with the reference scenario was 66.26 kg 1.4-DCB/kg of NH_3-N. These impact indicators are related to the toxicity indicators, which means that a reduction or improvement in the processes that contribute to these environmental loads can directly affect these related categories. Regarding land use (LU), the value of the indicator was calculated to be 24.27 m^2 a crop eq for the RS, and it is important to note that this result was mainly attributed to the mineral fertilizer production process. However, the LU for the TS resulted in a value of −18.89 m^2 a crop eq, which was attributed to the use of the prototype, showing an improvement for the impact category when compared with the reference scenario, as the charges for the production of synthetic fertilizer were eliminated. This could be attributed to the size of the prototype being smaller in relation to the whole infrastructure required for a fertilizer production plant. Water consumption (WC) during the operation stage of the ammonia capture prototype would vary depending on the environmental conditions of the site, and its use basically depends on the dissolution of sulfuric acid during the production of ammonium sulfate.

It can be concluded that, for the reference scenario, the majority of the participation of impact indicators occurred during the stage of mineral fertilizer production.

In general, the operation of the prototype had a higher impact in some categories; this was the case for the GW of the TS, with a difference of 429.46 kg CO_2 eq, which accounted for approximately 80% above the RS values. The value of the global impact for most categories was higher for the TS, contrary to the results obtained by González-García et al. [23], who used GPM in manure; the increase in the impact categories analyzed was mainly caused by the energy requirements of the system to move the air through the membranes. The total net TEC for the TS was −462.5 kg 1.4-DCB, indicating that this indicator improved considerably. As for

FEC and MEC, with results of 0.10 kg 1.4-DCB and 0.57 kg 1.4-DCB for the RS; these values differed by −0.05 kg 1.4-DCB and −0.69 kg 1.4-DCB for the TTS, showing that the impact generated was reduced. Likewise, the decreased values indicated improvements in the LU due to impacts being avoided. It can be inferred that the environmental loads of the industrial production of ammonia sulfate were shifted to the operation of the prototype, mainly due to electrical consumption and, in a lower proportion, to the manufacture of the membrane.

Similar to the results obtained by González-García et al. [23] in the analysis of the use of GPM technology in manure, there was a variation in the different impact categories, and they should be studied taking into consideration the environmental indicators that should be improved; in addition, the proper management of the operation of the prototype could significantly decrease NH_3 emissions through the recovery and redistribution of nutrients to the soil in a controlled manner.

Using the prototype showed an improvement in the following categories: HnCT, HCT, MEC, and TEC. As previously mentioned, the main problem with GW was electricity consumption; for this reason, a new EU Life project is currently being developed under the name "Green Ammonia", which aims to improve the energy efficiency of the operation of the prototype.

5. Conclusions

The purpose of this study was to use LCA methodology to analyze the environmental impact of a prototype to capture the NH_3 present in the air of swine-production sheds using GPM technology.

The evaluation was performed based on 12 environmental impact categories considering the main sources of NH_3 emissions in the livestock sector. It was concluded that using the capture prototype resulted in a higher environmental impact in some of the evaluated categories (i.e., GW, OFH, FPM, OFT, TA, and WC) compared with the reference scenario in which there was no ammonia capture; however, significant improvements were found in other impact categories (i.e., HnCT, HCT, MEC, and TEC).

Overall, the alternative that employs the prototype showed improvements due to the use of NH_3 in the production of a valuable fertilizer by-product and the consequent reduction in the consumption of synthetic products. The main impacts generated by the prototype were associated with energy consumption during its operation, which could improve with the implementation of renewable energy in the system. It is necessary to evaluate the environmental performance in comparison with other scrubbers to determine their efficiency in the process of reducing atmospheric NH_3 and transforming it into a valuable by-product. It is possible to reduce the impacts caused by the operation of the prototype by optimizing the processes and characteristics of the ammonia-capture system.

The analysis of the environmental performance of this type of technology is a key component in establishing improvements and making decisions that make it possible to take advantage of the by-products and residues obtained from agricultural activities without causing additional negative impacts on the environment. This project was funded by the European Union under the Life+ AMMONIA TRAPPING project (LI-FE15-ENV/ES/000284) "Development of membrane devices to reduce ammonia emissions generated by manure in poultry and pig farms".

At present, the LIFE projects are working together to find improvements in the energy efficiency and environmental performance of new technologies. For this prototype, further work will be focused on a phase of development and improvements based on the results obtained in this first version.

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Abbreviations

LCA	Life-Cycle Assessment
LCI	Life-Cycle Inventory
GPM	Gas-Permeable Membrane technology
RS	Reference Scenario
TS	Treatment Scenario
e-PTFE	Expanded Polytetrafluoroethylene Tubular Membrane

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