

Article

Assessing Ammonia and Greenhouse Gas Emissions from Livestock Manure Storage: Comparison of Measurements with Dynamic and Static Chambers

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Abstract: Emission quantification from the agricultural sector, and especially from livestock manure management, is relevant for assessing mitigation strategies and for inventory purposes. There are different direct techniques used to monitor emissions from quiescent surfaces. Common techniques include the closed static chamber and the open dynamic chamber. The aim of this study was to evaluate and compare different direct methods, two dynamic hoods and one static hood, for monitoring NH₃ and greenhouse gas (GHG) emissions (N₂O, CO₂, and CH₄) from different emission sources. These sources are ammonia solutions and different by-products of manure (compost, liquid fraction of digestate, and solid fraction of pig slurry). The use of dynamic hoods, despite their differences in size, operation, and applied air flux, presents comparable emission rates for all emissions and compounds assayed. These rates are always higher than those obtained using static hoods. Therefore, it can be concluded that the use of dynamic hoods is a valuable technique for refining the indirect estimation of emissions.

Keywords: ammonia emissions; greenhouse gas emissions; dynamic hood; static chamber; emission monitoring; manure management



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1. Introduction

Animal manure contains valuable nutrients for plants. Nevertheless, they can also be the cause of soil, water, and air pollution if handled incorrectly [1].

Regarding air pollution, agriculture represents the main source of anthropogenic ammonia emissions and malodorous gases, such as sulphur compounds, organic acids, phenolic compounds, and indoles [2,3]. NH₃ emissions have been increasing in the EU-28 since 2015, mainly driven by the agricultural sector. In 2020, agriculture was the largest emitter of NH₃, with 93% of total emissions in the EU-28 [4]. Gaseous ammonia in the atmosphere also leads to the formation of secondary inorganic aerosols, the main constituents of anthropogenic PM_{2.5} emissions. Fine particulate matter causes the onset of lung cancer and cardiopulmonary disease [5]. Indirectly, NH₃ emissions also contribute to N₂O secondary emissions [6]. Emissions from livestock effluents and livestock production in general are also related to greenhouse gases (GHGs), which include methane (CH₄), carbon dioxide (CO₂), and nitrous oxide (N₂O) [7].

The quantification of emissions from the agricultural sector, and especially from livestock manures, is relevant both for inventory purposes [2,8] and for assessing the reduction that can be achieved by adopting mitigation strategies [9]. Therefore, suitable methods are required to determine the emissions from manures.

There are different methods to estimate the emission rate (ER), including [10]:

1. The mass balance approach: based on the difference between the total nitrogen manure content before and after the emission period considered.
2. Indirect methods: these measure environmental concentrations and use dispersion models to calculate ER. The most common indirect methods are micrometeorological techniques, whereby gas emissions are calculated from a large footprint using tower-based instrumentation (e.g., gradient methods, eddy covariance, and the backward Lagrangian dispersion model).
3. Intergovernmental Panel on Climate Change (IPCC) approaches: range from the most generic estimations (tier 1) to more accurate ones provided that we have the required information, which is not always possible (tier 3) [11,12].
4. Direct methods: when emissions are measured using different devices, for example, wind tunnels, flux chambers (dynamic hoods), or static hoods. Samples are taken and analysed or directly measured using specific sensors.

As highlighted by other authors, the different approaches do not give comparable ER, and the discrepancies between methods can be greater than one order of magnitude [13,14].

Usually, in order to select the appropriate method to measure emissions, it is necessary to evaluate different factors, such as the typology of the emitting source, the type of gas compounds, the range of measurements, and the homogeneity of the source [13]. The direct methods are commonly applied for earth–atmosphere and water–atmosphere fluxes of various compounds, including ammonia, biogenic sulphur, nitrogen oxides, methane, and general GHG [14].

There are two main direct techniques most commonly used in the case of emissions from quiescent surfaces: the closed static chamber (non-steady-state chamber) and the open dynamic chamber (steady-state chamber). The first is a physical enclosure that creates a limited headspace above the surface from which the emissions are measured. Inside the chamber, the only present flux is that generated by the gradient concentration between the source and the headspace. This will be quantified over time in order to obtain the undisturbed flux into or out of the emission source [15]. Many studies use this type of direct measuring method [16–18]. Alternatively, in open dynamic chambers, an air flux is forced to pass through the hood, flushing out the headspace over the chamber so as to maintain the concentration at a constant value [10]. Airflow is generated by a fan. This method is also applied in many studies for measuring emission flux [19–23]. A wind tunnel is another option, but, despite being able to better imitate a natural airflow, a wind tunnel cannot simulate exact wind conditions, and the investment costs are high [23].

When estimating emissions, direct methods present limitations linked to the spatial variability of the emission source and possible disturbances induced by the chamber size, air velocity, etc. This can alter the natural gradient between the emission source and the atmosphere [21,24]. Nevertheless, the errors presented by the use of chambers can be reduced with correct chamber designs, proper data analyses, and spatial and temporal sampling regimes [13]. In addition, the advantage of using chambers is to connect a particular emission site to its measurable array of physical, chemical, and microbiological properties using the emissions of particular compounds or their reaction products [10]. Also, the investment cost of flux chambers is low, and they are generally easy to use [10]. Compared with indirect methods, e.g., the IPCC guidebook, direct methods allow us to consider specific situations for which the guides do not provide specific ER (e.g., emissions from by-product storage from a processing plant) [20].

The numerous and varied existing techniques for measuring emissions from agricultural fields show us that there is not a single consensual and widely used technique. However, it is crucial to rely on the data obtained both for research objectives and to properly define mitigation techniques and interventions aimed at reducing emissions. Previous studies aimed to compare the performance of chamber methods to measure emissions. Yang et al. compared the closed static chamber, semi-open static chamber, and open dynamic chamber methods for measuring ammonia emissions from fields and canopy in rice and wheat fields [14]. Recently, Alexander et al. determined the capture efficiency

of four chamber designs—open, open with polytetrafluoroethylene (PTFE), semi-open, and closed—for measuring ammonia emissions. However, these studies were carried out with synthetic ammonia sources. Comparisons involving ammonia and GHG in the real storage of manure by-products are lacking. The aim of this study was to: (i) evaluate and compare the performance of different direct methods (two dynamic chambers and one static chamber) for monitoring NH_3 and GHG emissions from different storages of manure by-products; and (ii) obtain specific ER for the gases and emission sources assessed.

2. Materials and Methods

2.1. Experiment Set-Up

In order to evaluate and compare the use of different chambers for measuring ammonia and GHGs, two types of experiments were carried out. The first set of tests was performed using a synthetic ammonia solution in order to assess ammonia emissions in a controlled environment without any disturbance induced by the complex characteristics of livestock manure. The second set of experiments was carried out in field conditions using hoods for measuring ammonia and GHG emissions from the storage of different by-products of livestock manure.

The experiments under controlled conditions were carried out at IRTA (Institut de Recerca i Tecnologia Agroalimentàries), located in Caldes de Montbui, Barcelona (41°36'43.704" N, 2°10'10.621" E). For these experiments, an ammonia solution was prepared by adding ammonium hydroxide, an extra pure, 25% solution in water ($\geq 25\%$ as NH_3) (Acros Organics, Janssen Pharmaceutica, Geel, Belgium), to different amounts of water in order to obtain the desired ammonia concentration.

The ammonia solution was placed inside heavy-duty PVC and polyester square containers with triple-layer sided walls enclosed in a metal frame (1.22×1.22 m; A: 1.49 m^2). Specifically, the plastic vessel was filled with 30 L of tap water mixed with a defined amount of NH_4OH , based on the desired ammonia concentration (Table in Section 3.1). In addition, a group of tests were performed on ammonia solution by adding sulphuric acid (97%), in order to reduce pH from 12 to 7.8, and sodium bicarbonate to simulate the buffer capacity of the slurry (NaHCO_3) (concentration: 0.1 mol TIC/L, where TIC is total inorganic carbon defined as: $\text{TIC} = \text{CO}_{2\text{aq}} + \text{HCO}_3^- + \text{H}_2\text{CO}_3$). Throughout all the experiments, the content of ammonia was also monitored analytically.

Trials at field conditions for estimating emissions from livestock manure by-products were performed at a biogas treatment plant located in Vilademuls (Catalonia, Spain), where cow slurry is digested together with other co-substrates. The emissions of ammonia, CO_2 , CH_4 , and N_2O were analysed at a pile where the thick phase of the digestate is composted (Compost, C) and at the storage of the digested liquid fraction (liquid fraction of digestate, LFD). Emissions were also measured at a biological treatment plant located in Vic (Catalonia, Spain), where pig slurry is processed in a nitrification–denitrification plant to remove nitrogen. In this case, NH_3 , CO_2 , CH_4 , and N_2O were measured from a stored pile of solid fraction after solid/liquid separation (solid fraction of pig slurry, SFPS).

2.2. Equipment Used

Three different hoods were used in the experiments. Two of these were dynamic hoods, and the third was a static hood. The dimensions of each chamber are shown in Table 1, and a picture of them is given in Figure 1.

Table 1. Characteristics of different hoods used in the tests (DH1: Dynamic Hood 1, DH2: Dynamic Hood 2, SH: Static Hood).

	L1 Length (m)	L2 Width (m)	H Height (m)	Emitting Surface (m ²)
DH1	0.96	0.96	0.15	0.92
DH2	1.03	0.53	0.25	0.55
SH	0.4	0.405	0.75	0.16



Figure 1. Images of the hoods used in the tests: Dynamic Hood 1 (DH1); Dynamic Hood 2 (DH2); Static Hood (SH).

The first dynamic hood (DH1) is a Lindvall hood. The air sampling was done by fitting a pipe and inserting it into the middle of the outlet hole of DH1. The flow velocity required inside the hood should be between 0.2 and 0.3 m/s [25], which corresponds to an input air flow of 0.7–0.9 m/s. To obtain this air speed inside the hood, a blower was connected to the inlet port of the Lindvall hood with a nalophane pipe. The air speed velocity was measured by means of a thermal anemometer flowrate device at the outlet port of the hood. In order to find the velocity inside the hood (v_2), a mass balance was applied (Equations (1) and (2)).

$$S_1 * v_1 = S_2 * v_2 \quad (1)$$

where S_1 is the hood surface, $L1 \times L2$ (mm²), v_1 is the air speed inside the hood, S_2 is the outlet hood surface = $\Pi(D/2)^2$ (mm²), v_2 is the air speed at the outlet of the hood, and D is the diameter of the outlet.

In this way, v_2 can be calculated as:

$$v_2 = 0.25 * \frac{S_1}{S_2} \quad (2)$$

Therefore, in order to ensure the required air speed velocity inside the hood, the measured velocity at the outlet of the hood must be around 0.8 m/s. In the case of DH1,

to estimate the ER, it is also necessary to define the hood factor w . This can be calculated according to Equation (3) and the hood dimensions.

$$w = \text{flow path section (m}^2\text{)} / \text{covered area (m}^2\text{)} \quad (3)$$

In the DH1 hood, gas samples were collected via a cross-sectional sampling tube at the outlet point of the hood.

The second dynamic hood (DH2) (Odournet ventilating hood, ODOURNET, S.L., Sant Cugat del Vallès, Spain), whose dimensions are stated in Table 1, consists of two radial fans with a constant volumetric flow. In this case, the velocity of air cannot be checked inside the hood, and the flux is set by the equipment manufacturer (30 m³/h, cf. Odurnet technical brochure), corresponding to an airflow equal to 0.005 m³/s. Also, for full-scale use of the DH2 application, a floating system was required to ensure the sealing of the hood when positioned on a liquid surface. A pressure-sealing connection on the structure enabled the PTFE pipe to be plugged into the hood for air conduction and sampling.

The static hood (SH) is a low-density polyethylene plastic enclosure (0.12 m³). It has a thickness of 150 µm and a self-regulating pressure system to avoid a vacuum effect. This hood is fitted with two holes: one to insert the temperature probe and another used as an air sampling port. When the SH was used over solid manure, it was introduced 2 cm into the heap to guarantee the sealing of the hood.

2.3. Emission Sampling and Calculation

2.3.1. Sampling Procedure

NH₃ emissions were sampled by means of an absorption method. A known volume (5 L and 1 L for the dynamic hoods and the static hood, respectively) of air outflow from the hood was bubbled into two serial glass impingers containing 10 mL of sulphuric acid (0.1 N) by means of a pump (Universal PCXR8, 1000 mL/min, SKC, Inc., Eighty Four, USA). Then, the ammonium absorbed in the sulphuric acid was measured by means of a colorimetric method (see Section 2.4). Each measure was repeated in triplicate.

Air samples for N₂O, CO₂, and CH₄ emission measurements for the DHs were collected with a syringe from the pipe connected to the hood outlet, 10 mL of which was then injected into a vacutainer tube with a butyl rubber stopper to further analyse.

For the SH, air was collected from the headspace at 0, 10, 20, 30, and 40 min for lab-trial experiments throughout each sampling period. Two more samples were taken in the field campaigns (at 50 and 60 min) to be certain that chamber headspace had reached saturation.

2.3.2. Emission Rate (ER) Calculation

The ER calculation for each hood was performed as follows:

Regarding DH1, the specific emission rate (ER) was calculated according to Equation (4).

$$ER = (C_{out} - C_{in}) * w * v \quad (4)$$

where:

ER is the emission rate expressed in terms of mg/m²/s; C_{out} is the NH₃, N₂O, CH₄, or CO₂ concentration of air outlet from each chamber in mg/m³; C_{in} is the NH₃, N₂O, CH₄, or CO₂ concentration of air inlet into each chamber in mg/m³; v is the air speed expressed in m/s; and w is the hood factor (–).

The GHG and ammonia ER in the dynamic chamber DH2 (ER) were calculated as follows (Equation (5)).

$$ER = \frac{Q * (C_{out} - C_{in})}{A} \quad (5)$$

where:

ER is the emission rate expressed in terms of mg/m²/s;
 C_{out} is the NH₃, N₂O, CH₄, or CO₂ concentration of the air outlet from each chamber in mg/m³;

C_{in} is the NH_3 , N_2O , CH_4 , or CO_2 concentration of air inlet into each chamber in mg/m^3 ;

Q is the airflow expressed in m^3/s ; and A is the emitting surface (m^2).

For the SH, in order to calculate ammonia emissions and GHG emissions flux, it is necessary to build a saturation curve and define the regression coefficient γ . γ was calculated for each experiment using simple linear regression based on the concentration of gases as a function of collection time for the five or seven samples [26,27]. The ER of the SH was calculated as follows (Equation (6)).

$$\text{ER} = \frac{\Delta \text{gas}}{\Delta t} * \frac{V}{A} \quad (6)$$

where:

ER is the emission rate expressed in terms of $\text{mg}/\text{m}^2/\text{s}$;

$\Delta \text{gas}/\Delta t$ is the angular coefficient of the curve (γ) (mg measured $\text{gas}/\text{m}^3/\text{s}$);

V is the volume of the chamber's headspace (m^3); and

A is the emitting surface (m^2).

2.4. Analytical Methods

Manure by-products, compost (C), liquid fraction of digestate (LFD), and solid fractions of pig slurry (SFPS) used as the emission source in full-scale trials were characterised for Total Kjeldahl Nitrogen (TKN) (4500-Norg C), ammonium nitrogen ($\text{NH}_4^+\text{-N}$) (4500-NH3 B C), pH (4500- H + B), electrical conductivity (EC), total solids (TS) (2540 B), volatile solids (VS) (2540 E) [28] and the chemical oxygen demand (COD) (410.4). NO_2^- , NO_3^- , SO_4^{3-} , and PO_4^{3-} were analysed using ionic chromatography following Standard Method 4110. Total phosphorus (P) was analysed using the colorimetric Standard Method 4500-P C. During all the experiments, the liquid temperature and air temperature (inside and outside of the chamber for the SH) were monitored.

Regarding the concentration of ammonia in air samples, a colorimetric method was applied (Spectroquant[®] kit, Merck KGaA, Darmstadt, Germany), analogous to the phenate method (APHA 4500-F). Then, absorbance was measured in a spectrophotometer (DR2800, Hach Lange, Loveland, USA) using a wavelength equal to 640 nm.

GHG concentrations (N_2O , CO_2 , and CH_4) were performed using a gas chromatograph coupled with a flame ionisation detector and an electron capture detector. CH_4 was determined using a THERMO TRACE 2000 (Thermo Fisher Scientific, Waltham, USA) gas chromatograph equipped with a flame ionisation detector (FID). N_2O and CO_2 emissions were determined with an Agilent 7820A (Agilent Technologies, Inc, Santa Clara, USA) GC system using an electron capture detector (ECD).

3. Results and Discussion

3.1. Ammonia Emissions from Ammonia Solution

Table 2 shows results relating to experiments with ammonia solution. Regarding physicochemical parameters, the data average refers to different measurements carried out during the same experiment for each hood. All the samples have been analysed in duplicate.

As can be seen, the higher the ammonium concentration in the emission source, the higher the ammonia ER. For example, in the DH2 chamber, the ER increased from $0.3 \pm 0.03 \text{ mg}/\text{m}^2/\text{s}$, when the ammonium concentration in the solution was $0.31 \pm 0.01 \text{ gN-NH}_4/\text{L}$ to $2.14 \pm 0.18 \text{ mg}/\text{m}^2/\text{s}$ when the ammonium concentration was $2.26 \pm 0.3 \text{ gN-NH}_4/\text{L}$ (Table 2). NH_3 emissions are strongly dependent not only on ammonia concentration at the emission source [14,21], but also on pH [29], buffer capacity [30], and the ionic strength that has an inverse relation to ammonia emission [31]. In order to better simulate pig slurry composition, a buffer was added to the synthetic ammonium solution ($0.1 \text{ mol TIC}/\text{L}$). As expected, the ER decreased to $0.35 \pm 0.07 \text{ mg}/\text{m}^2/\text{s}$ and $0.37 \pm 0.09 \text{ mg}/\text{m}^2/\text{s}$ for DH1 and DH2, respectively (Table 2).

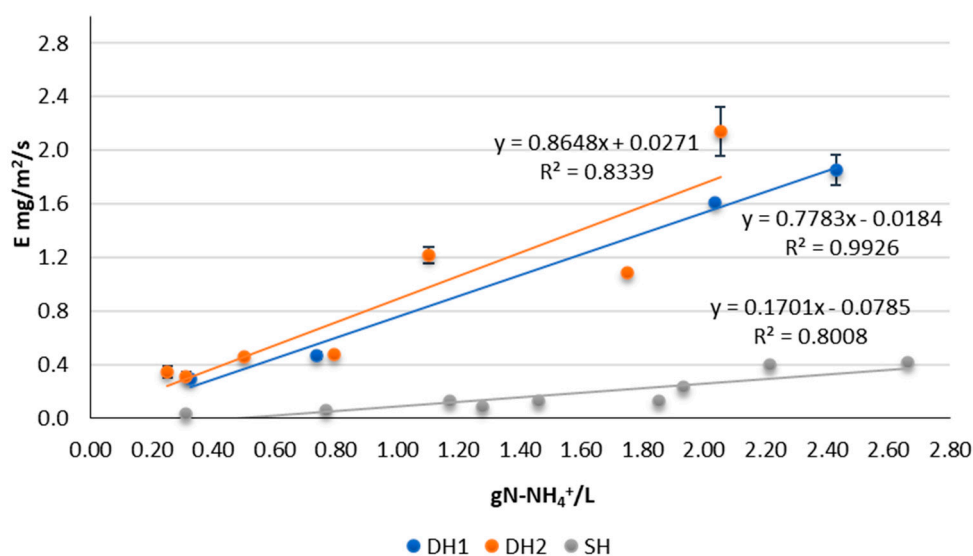
Table 2. Experiment set-up and ammonia ER for ammonia solution experiments (DH1: dynamic hood1; DH2: dynamic hood2; SH: static chamber; ER: Emission Rate; N: No; Y: Yes).

Buffer	N° Rep. DH1	N° Rep. DH2	N° Rep. SH	Environmental T (°C)	Slurry T (°C)	TAN (gN-NH ₄ ⁺ /L)	pH	ER_DH1 (mg N-NH ₃ /m ² /s)	ER_DH2 (mg N-NH ₃ /m ² /s)	ER_SH (mg N-NH ₃ /m ² /s)
N	3	3	3	10.96 ± 1.99	11.33 ± 1.39	2.26 ± 0.3	11.32 ± 0.08	1.85 ± 0.12	2.14 ± 0.18	0.35
N	3	3	1	12.37 ± 0.2	10.52 ± 0.08	1.88 ± 0.14	10.72 ± 0.12	1.6 ± 0.02	1.1 ± 0.02	0.1
N	-	-	3	13.02 ± 3.06	10.8 ± 1.43	1.26 ± 0.27	11.67 ± 0.31	-	-	0.12
N	-	3	-	13.25 ± 0.07	13.3 ± 0.14	1.1 ± 0.01	10.92 ± 0.04	-	1.2 ± 0.06	-
N	3	3	1	13.72 ± 0.37	11.72 ± 0.47	0.77 ± 0.03	10.33 ± 0.08	0.5 ± 0.03	0.5 ± 0.01	0.1
N	3	3	1	14.17 ± 0.1	12.05 ± 0.14	0.29 ± 0.06	11.1 ± 0.06	0.3 ± 0.03	0.3 ± 0.04	0
N	-	3	-	14.2 ± 1.13	13.5 ± 0.42	0.5 ± 0.01	10.56 ± 0.03	-	0.5 ± 0.004	-
N	-	3	-	17.35 ± 0.92	13.5 ± 0.28	0.31 ± 0.01	10.38 ± 0.05	-	0.3 ± 0.03	-
Y	3	3	-	9.03 ± 0.76	13.77 ± 0.83	2.36 ± 0.05	8.3 ± 0.13	0.35 ± 0.07	0.37 ± 0.09	-
Y	-	-	1	18.94 ± 3.00	15.96 ± 2.05	1.76	8.7	-	-	0.01
Y	-	-	3	13.38 ± 1.49	12.72 ± 0.94	1.44 ± 0.04	8.53 ± 0.06	-	-	0

When comparing emission factors from different hoods, the SH always gives an ER one order of magnitude less than when using a DH, in any given situation, with or without a buffer. These results are similar to those observed by Alexander et al. [24] and Yang et al. at laboratory experiments with synthetic ammonia source solutions [14]; emission factors obtained in a steady-state chamber are 50% lower. Also, when comparing the static chamber with other methods, such as the periodic integrated horizontal flux (IHF) technique [32,33] or boundary layer equation [27], the emission of gases measured was much lower.

The possible causes of disparity between hood types may be (1) the mode of transport of the dissolved gases, (2) air and water temperature, and (3) wind speed [27]. These conditions can be affected by the higher resistance to atmospheric vertical transfer in the absence of or under low headspace air movement [34]. The reduced values obtained with the SH may also be derived from a linear model applied for calculating ammonia flux (and other gas flux) that cannot accurately represent the fundamental physics of the diffused gas when transported in static chambers [35].

In order to compare the three hoods, a linear regression of the ammonia ER in the presence of the TAN concentration of the emissions source was used (Figure 2), showing a good correlation, with R² above 0.8 in all cases.

**Figure 2.** Linear regression of the ER in the presence of N concentration in the emission source for each chamber. (Note: error bars represent standard deviation.)

Emission rates obtained with the two dynamic hoods (DH1 and DH2) are of the same order of magnitude. Nevertheless, when nitrogen concentration in the emission sources increases, differences between the ER increase, attaining a difference of 33% with the highest N concentration in the emission source ($2.26 \pm 0.3 \text{ gN-NH}_4^+/\text{L}$). In contrast, the ER of the SH is substantially different (lower) from that obtained with the two DHs (Figure 2).

3.2. Ammonia and GHG Emissions in Field Conditions

3.2.1. Manure By-Products Characterization

Table 3 summarises the main characteristics of the different manure by-products used as emission sources during the field trials. The characteristics of different slurries are typical of these kinds of by-products [20]. The solid fraction of pig slurry has similar characteristics to those reported in the study by Dinuccio et al. [36].

Table 3. Characteristics of the manure by-products. LFD: liquid fraction of digestate; C: compost; and SFPS: solid fraction of pig slurry.

Parameter	Units	LFD	C	SFPS
pH	-	7.86	8.62	8.80
EC	mS/cm	20.60	4.10	2.73
ST	%	5.43	28.45	27.75
SV	%	3.68	24.91	23.96
NH_4^+	gN/kg	3.05	2.89	1.96
TKN	gN/kg	4.33	8.36	3.68
NO_2^-	mgN/kg	0.00	0.00	763.00
NO_3^-	mgN/kg	0.00	0.00	578.00
COD	mg O_2/kg	58,546	459,854	412,196
SO_4^{2-}	mg/kg	-	313.00	-
PO_4^{3-}	mg/kg	62.00	73.00	-
P_{tot}	g/kg	0.43	0.85	1.15

3.2.2. Ammonia Emissions

Table 4 shows the results of ammonia emissions taken from the storage of different manure by-products. As can be seen, the ER calculated with dynamic hoods, DH1 and DH2, is similar and one order of magnitude higher than those reported with the SH, and the ER is similar to those obtained with ammonia solution tested under controlled conditions (Table 3).

Table 4. Experiment set-up and ammonia ER for different manure by-products: compost (C), liquid fraction of digestate (LFD), and solid fraction of pig slurry (SFPS) (note: DH1 = dynamic hood1; DH2 = dynamic hood2; SH = static chamber; and ER = emission rate).

Emission Source	N° Rep.			Environmental T (°C)	Slurry T (°C)	TAN (gN-NH ₄ ⁺ /L)	pH	ER_NH ₃ (mg N-NH ₃ /m ² /s)		
	DH1	DH2	SH					DH1	DH2	SH
C	3	3	2	16.04 ± 2.27	25.56 ± 3.32	3.05	8.62	0.5 ± 0.05	0.51 ± 0.23	0.03
LFD	3	3	2	17.21 ± 2.93	14.39 ± 3.06	2.95 ± 0.04	7.86	0.12 ± 0.02	0.09 ± 0.02	0.02
SFPS	3	3	2	6.43 ± 1.59	13.70	1.96	8.8	0.22 ± 0.03	0.33 ± 0.03	0.02

The ammonia emissions resulting from digestate storage obtained using the dynamic chamber DH1 are very similar to those observed in the study by Baldé et al. (annual average of $15.5 \text{ gNH}_3/\text{m}^2/\text{d}$), even if, in that research, the technique used was the micrometeorological mass balance method, which limits its comparison [37]. Kupper et al. collected published emission data from stored cattle slurry, reporting emission values for cattle digested liquid fraction in a range of $0.07\text{--}0.13 \text{ mg N-NH}_3/\text{m}^2/\text{s}$, which is very similar to the results obtained with DH1 and DH2 [2].

Emissions results from the solid fraction of pig slurry were twice as high as those observed in the study by Dinuccio et al. [36] (60–180 mg/m²/h at 5 °C and 20–280 mg/m²/h at 25 °C). This difference can be attributed to the composition of the slurry (as the content of nitrogen was much higher, 4.05 g/kg as TAN and 12.2 g/kg as TKN). Comparing the different manures tested, the highest ammonia emissions were detected from compost and the lowest in the liquid fraction of digestate (C > SFPS > LFD).

Besides TAN content, the ammonia ER depends on environmental temperatures as well as the temperature of the emission source. In the case of compost, another factor that should be taken into account is the C/N ratio: the lower the C/N ratio, the higher the ER of ammonia [38]. In addition, the structure and porosity of the solid fraction and compost can favour ammonia volatilization, as a low apparent density allows the passage of air through the solid matrix, thereby favouring ammonia emissions. pH also plays an important role in ammonia emissions [39], as can be seen from the results: the lower the pH, the lower the ammonia ER.

3.2.3. Greenhouse Gas Emissions

The measurements of GHG emissions were taken from different manures on a farming scale.

With regard to N₂O, the ER observed was noticeably low in all tests, and the use of SH, similar to what had been observed in the ammonia ER, gave figures that were two orders of magnitude below those obtained with the DH (Table 5). As mentioned previously, the emission of N₂O is related to incomplete ammonium oxidation or to an incomplete denitrification process. In the manures considered, neither of these two processes were involved.

Table 5. GHG Emission Rate for different manure typologies: processing compost (C), liquid fraction of digestate (LFD), and solid fraction of pig slurry (SFPS) (Note: DH1 = dynamic hood1; DH2 = dynamic hood2; SH = static chamber; and ER = Emission Rate).

Emission Source	ER_N ₂ O (mg/m ² /min)			ER_CO ₂ (mg/m ² /min)			ER_CH ₄ (mg/m ² /min)		
	DH1	DH2	SH	DH1	DH2	SH	DH1	DH2	SH
C	0.2	0.2	0.0	201.2	158.7	24.1	1.1	0.5	0.5
LFD	0.2	0.2	0.0	152.8	150.3	9.2	0.4	0.4	0.3
SFPS	0.0	0.0	0.0	75.4	30.3	67.4	0.2	0.1	0.6

In the study by Petersen et al., 2013, a dynamic method was used for measuring GHG emissions [40]. They found that storage of pig slurry resulted in total GHG emissions of up to 1.3 kg CO₂ eq/m³/d during the summer. No N₂O emissions were observed from pig slurry during storage in any period of observation except in the presence of a crust, whereby N₂O emission was 80 mg/m²/h.

In this study, the maximum emission value detected for the liquid fraction of digestate was 11.6 mgN₂O/m²/h, measured with DH2. Nevertheless, this value is comparable with the ones obtained using DH1. Also, in the study by Perazzolo et al., 2015, N₂O emissions were not observed before a storage period of 32 days [39]. Values in a range of 14.1–41.7 mgN₂O/m²/h, similar to the results obtained in this study, have been reported for cattle digested liquid fraction [2]. In the literature, cases of N₂O emission from stored slurries are also reported, but, in those cases, the emissions were imputable to a crust presence [41]. In the study by Bao et al., N₂O emissions from swine manure compost were reported to be extremely low during the temperature-heating, thermophilic, and temperature-cooling phases [42].

Despite CO₂ being a greenhouse gas, in the context of manure management, its emission is considered to be biogenetic and is, therefore, not computed as a GHG. Nevertheless, CO₂ emissions were studied, and ER was calculated. As with the other gases, ER_CO₂ measured from compost and digestate liquid fraction using different hoods gave similar

results, except in the case of the SH, which was one order of magnitude lower compared to the DHs. In contrast, ER_CO₂ of the solid fraction of pig slurry presented some differences, but in the same order of magnitude for all three hoods. These results show that static chambers could be appropriate for low emission fluxes [43].

The ER_CH₄ reported was of the same order of magnitude in the three hoods. ER_CH₄ showed low values, resulting in carbon lost in the form of CH₄, which was much lower than that observed in the form of CO₂. Perazzolo et al., 2015 also reported similar results [39]. The smallest emission was observed when using DH2 for the solid fraction of pig slurry, and the highest was reported with DH1 for compost. It is remarkable that digestate (liquid fraction) presented lower ER_CH₄ than compost. This can be explained by the fact that digestate had been stored for a long period of time before measuring CH₄ emissions. This long storage period resulted in the release of all the occluded CH₄. Experiments performed with fresh digestate presented considerably higher CH₄_ER, in the range of 18.3–37.6 mgCH₄/m²/min [2].

Figure 3 shows a summary of nitrogen compounds (NH₃ and N₂O) and carbon compounds (CO₂ and CH₄) emission rates from the different manure typologies and hoods.

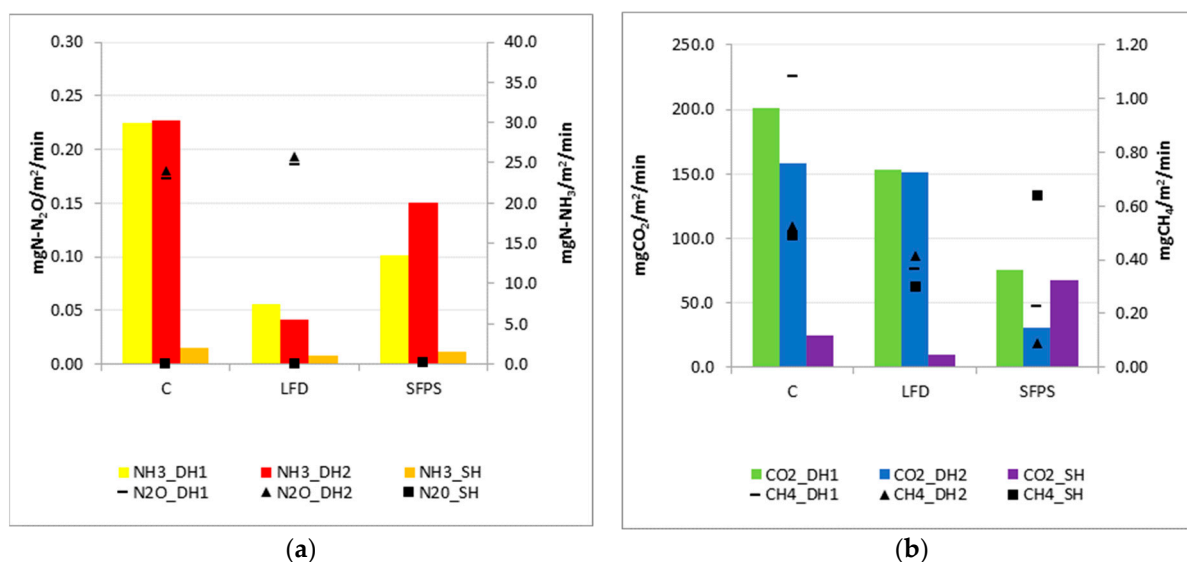


Figure 3. NH₃, N₂O (a) and CO₂, CH₄ (b) ER for all manure by-products considered. C: compost, LFD: liquid fraction of digestate; and SFPS: solid fraction of pig slurry.

As can be seen in Figure 3, NH₃, N₂O, and CO₂ measured with DHs showed similar values and were always higher than the ER measured with the SH. In contrast, ER_CH₄ did not show any clear pattern. The use of DHs imposes a constant value of wind speed that is far from natural conditions. Furthermore, this method does not consider factors such as rainfall, creating, as defined by Miola et al., 2015, an oasis effect that could overestimate the ER [34]. On the other hand, the SH gives lower values of ER as the gas concentration in the hood increases over time, limiting gas emissions according to Henry's law.

3.2.4. Implications of the Study and Future Recommendations

Measuring GHG and NH₃ fluxes in livestock manure storages is critical to understanding the processes driving air pollution and climate change in order to develop and evaluate mitigation strategies based on management practices. In addition, it is important to find a balance between high-precision measurement and the cost of the infrastructure necessary to produce reliable inventories of ammonia and GHG emissions. From the measurement data obtained, the application of mitigation strategies can be prioritised based on the emissions profile of each source.

The results obtained in this study show that dynamic chambers are the preferable option to measure ammonia and GHG emissions from manure storage due to an underestimation of the ER when using the static hoods. However, the low-cost requirements of the static chamber method make it still useful for measuring ammonia and GHG fluxes in livestock manure storages, especially for estimating differences between management techniques [44] or exploring storage dynamics over time (seasons or years). Static chambers allow measurements of small areas and numerous treatments to be obtained without major investments in infrastructure, although they obtain high coefficients of variation due to the time and spatial variation of the manure storage gas flow. Temporal extrapolation of short measurements of ammonia and GHG fluxes from static chambers is associated with some error in cumulative emissions. Likewise, the area covered by the chambers is small, so only a fraction of the total surface area of the manure piles or pits is measured. To obtain representative data, a comprehensive experimental design, following the numerous standardised guidelines available [45], is essential to extend local findings to global models and translate research results into viable mitigation strategies. For example, the time between samplings should be determined by an initial calibration, and the sampling period must be representative of the average temperature of the day to reduce bias.

The specific measurements carried out in this study are less representative of the storage emissions at an annual level since they are related to very specific conditions of temperature or age of the stored material. In addition, emissions must be assessed at different stages of storage since composting involves changes due to mixing, biological processes, and the evolution of the process temperature over time, and emissions from liquid or solid manure storage can be influenced by the aging of the substrates. In addition, similar research will also need to be conducted on different manure management practices, including mitigation strategies such as changes in livestock diet or the addition of additives to manure to reduce emissions.

To reduce measurement uncertainties, the static chamber design used in this study could also be improved. In general, the smallest chambers produce the lowest errors, and for larger chambers (above 20 L), such as the one used in this study, the use of fans appears critical to reducing measurement error, making it comparable to smaller chambers when fans are used [46].

ER can also be calculated through other approaches, such as IPCC, which is not site-specific but a regional estimation that considers different factors, such as average temperature, manure typology, storage site, etc. The direct methods assessed in this study give ER that can be used to assess real conditions and, despite generalisation, can provide correct estimations when taking into account site-specific characteristics, such as actual temperature. To sum up, if direct methods cannot be directly compared with IPCC emission factors, they can be a valuable method to compare the emissions resulting from different manure management techniques. Moreover, with adequate assessment, they can be used to modulate the general emission factors when specific techniques are used. For this purpose, further studies are required to compare the emissions obtained from long-term measurement in operative conditions with those obtained using direct methods like those used in this paper.

4. Conclusions

A comparison of ammonia and GHG emission measurements was carried out using two different technologies, both applying direct methods: dynamic hoods and static chambers. Tests were performed with different emission sources: synthetic ammonia solution, digestate, compost, and a solid fraction of pig slurry. The use of dynamic hoods, despite their differences in size, operation, and applied air flux, presents comparable emission rate results for all emission sources assayed. The static chamber has been associated with a general underestimation of emission rates. It can be concluded that the use of dynamic hoods could be a valuable technique for refining indirect estimations of emissions. Nevertheless, considering that NH_3 and GHG emissions are an issue that should be tackled urgently, a

unified criterion should be established for the measurement of these emissions, since it has been observed that different systems for capturing emissions can give rise to different results.

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