

Alma Mater Studiorum Università di Bologna
Archivio istituzionale della ricerca

Measuring ammonia concentrations by an infrared photo-acoustic multi-gas analyser in an open dairy barn:
Repetitions planning strategy

This is the final peer-reviewed author's accepted manuscript (postprint) of the following publication:

Published Version:

D'Urso P.R., Arcidiacono C., Valenti F., Janke D., Cascone G. (2023). Measuring ammonia concentrations by an infrared photo-acoustic multi-gas analyser in an open dairy barn: Repetitions planning strategy. COMPUTERS AND ELECTRONICS IN AGRICULTURE, 204, 1-8 [10.1016/j.compag.2022.107509].

Availability:

This version is available at: <https://hdl.handle.net/11585/935086> since: 2023-07-17

Published:

DOI: <http://doi.org/10.1016/j.compag.2022.107509>

Terms of use:

Some rights reserved. The terms and conditions for the reuse of this version of the manuscript are specified in the publishing policy. For all terms of use and more information see the publisher's website.

This item was downloaded from IRIS Università di Bologna (<https://cris.unibo.it/>).
When citing, please refer to the published version.

(Article begins on next page)

1 **Highlights**

- 2 - NH₃ concentrations were measured by INNOVA in a semi-open dairy barn;
- 3 - The number of repetitions for each sampling location were assessed;
- 4 - The first repetition should be discarded to reduce the variability due to the changing of SL;
- 5 - The second or the third repetitions have the lowest variability;
- 6 - Up to four repetitions are recommended to avoid measurement change.

7 **Research Paper**

8 **Measuring ammonia concentrations by an infrared photo-acoustic** 9 **multi-gas analyser in an open dairy barn: repetitions planning** 10 **strategy**

11

12 Provvienza Rita D'Urso^a, Claudia Arcidiacono^{a*}, Francesca Valenti^a, David Janke^b, Giovanni
13 Cascone^a

14 ^a University of Catania, Department of Agriculture, Food and Environment (Di3A), Building and Land
15 Engineering Section, via S. Sofia 100, 95123, Catania, Italy

16 ^b Leibniz Institute for Agricultural Engineering and Bioeconomy (ATB), Department Livestock
17 Engineering, Max-Eyth-Allee 100, Potsdam 14469, Brandenburg, Germany

18 *Corresponding author: claudia.arcidiacono@unict.it. Tel. 0039 0957147576, Fax 0039
19 0957147600/5,

20

21 **Abstract**

22 The infrared photoacoustic spectroscopy is a common gas sensing technique used for research activities to
23 perform continuous measurement of gas concentrations. Few research works have investigated how to collect
24 data and obtain reliable measurements in open and semi-open dairy houses for gas concentration estimation.

25 In the present study, measurements of ammonia (NH₃) concentrations were carried out in a semi-open free-
26 stall dairy barn by using a commercial photo-acoustic spectroscopy system (INNOVA by LumaSense
27 Technologies A/S, Denmark), widely used for scientific purposes. Several repetitions for each sampling
28 location (SLs) were investigated in in-field conditions to study the variability of the measurement in each
29 repetition. Based on acquired data, one-way analyses of variance (ANOVA) were carried out for different
30 groups of data. The achieved results showed that the measurement strategy depended on the number of
31 repetitions with effect on the variability of each measurement. In detail, the number of repetitions performed
32 had a significant influence on data collection (P<0.001). From the outcomes of the research, it is recommended
33 to: perform three repetitions, or at most, four repetitions for each SLs; exclude the first repetition from the data
34 analyses because it reduces measurement variability; consider the mean value of the second and third
35 repetitions as the NH₃ concentration value determined at each SL; up to four repetitions (that would take a
36 five-minutes acquisition) are recommended because measurement changed in time when a higher number of
37 repetitions were carried out. The measurement design should be planned based on the number of repetitions
38 and the number of SLs in order to perform more than one SL measurement cycle within an hour. Since the
39 measurement cycles are strictly related to the number of SLs in the barn, they depend on the barn typology and
40 dimensions.

41 The application of a specific measurement strategy is valuable to obtain reliable data that could be used in the
42 research activities focused on estimation of gas concentrations for monitoring air quality, improving animal
43 welfare, assessing mitigation strategies, or estimating emissions in open and semi-open free-stall dairy barns.

44 *Keywords:* ammonia, gas concentrations, repetitions, measurement strategy, open-sided dairy barn

45

46 **1. Introduction**

47 In the last decades, the scientific community has been mainly focused on the estimation of gas emissions from
48 the livestock sector due to environmental concerns. It is widely acknowledged, in fact, that agriculture is
49 responsible for the 94% of the ammonia (NH₃) production within Europe (Tullo et al. 2019). In detail, the
50 dairy sector is a relevant category that produces NH₃ emissions at farm level with negative effects on both
51 human health and ecosystems (de Vries, 2021). Since the estimation of NH₃ emissions plays a relevant role in
52 the identification of mitigation strategies, the application of precise measurement techniques and instruments
53 can improve the quality of data for both estimation purposes and monitoring of the pollutants in the barn
54 environment.

55 Among the main measurement techniques (Hassouna and Heglin, 2016), the infrared photoacoustic
56 spectroscopy is used for research activities to perform continuous concentrations measurement of one or more
57 gases. The measurement principle is based on the analysis of an air sample in a measurement chamber,
58 equipped with optical filters. First of all, the sample is sucked into the measurement chamber, then sealed by
59 valves. Secondly, an intermittent infrared source passes through a chopper and an optical filter into the chamber
60 and is irradiated onto gas sample. Based on the radiation energy absorbed by the gas in proportion to the
61 concentration, the gas sample emits acoustic waves that are detected by microphones and the signal is, finally,
62 post processed and the gas concentration can be estimated.

63 In the literature many authors (Ngwabie et al. 2011; Bijerg et al., 2012; Hassouna et al., 2013; Ngwabie et al.
64 2014; Saha et al., 2014a; Hempel et al., 2016; Schmithausen et al. 2018, D'Urso and Arcidiacono, 2021) have
65 monitored gas concentrations from different livestock housing by using INNOVA multi-gas analysers
66 (LumaSense Technologies A/S, Denmark). This device is based on the infrared photoacoustic spectroscopy
67 principle, and it can measure up to five gases (e.g., NH₃, methane, carbon dioxide, nitrous oxide) in a sampling
68 point. When the INNOVA is combined with a multiplexer, it allows acquiring gas concentrations in different
69 points. However, INNOVA is not able to measure the gases simultaneously in all the sampling points, but it
70 measures simultaneously all the gases in a specific sampling point accordingly to a continuous sequence. In
71 some studies, INNOVA was applied in both in-field conditions (Saha et al. 2014b) and a scale model building
72 (Shen et al. 2012) to acquire carbon dioxide concentrations in many sampling points in order to assess the
73 ventilation rate. Since this kind of multi-gas analysers provides precise and reliable measurements (Zhuang et
74 al., 2020), the INNOVA is also used in research studies as a reference method to assess the performance of
75 other devices (Li et al., 2015; Mendes et al., 2015; Schmithausen et al. 2016; Wang et al., 2016). Moreover,
76 this device was recently assessed in the study of von Jasmund et al. (2022) by using a gas calibration unit.

77 During the measurement with INNOVA, non-target gas can produce an interference. According to Hassouna
78 et al., (2013) non-compensated interferences between gases induce an overestimation of gas concentrations
79 and a cascade effect induces underestimation. A relevant role for obtaining reliable measurements is carried
80 out by the instrument (e.g., precision) and the design of the measurement campaign (e.g., sampling frequency,
81 number of measurement points, sampling positions, and time duration of the measurement). Moreover, the gas
82 concentrations from animal housing are influenced by the spatial and temporal variability of the measure due
83 to the environmental conditions and barn management (Saha et al., 2013, Rong et al. 2014, Mendes et al. 2017,
84 D'Urso et al., 2021a) thus it is unlikely that a uniform distribution of a gas within the building occurs.
85 According to Calvet et al. (2013), the execution of measurement repetitions in time can reduce random errors
86 that derive from unpredictable temporal variations of the measured values.

87 Based on the literature, few studies have investigated the number of repetitions to be performed by an
88 INNOVA device. In detail, the study of Brehme (2003) defined specific information about experiment design

89 based on the analyses carried out in a duck farm during 20 months of observation. The results reported that the
90 adequate number of repetitions is ten. Moreover, the author suggested performing three repetitions and
91 rejecting the first and the second values for each sampling point. This conclusion is based on the fact that
92 although excluding the first and the second repetition would produce a data loss of 66%, the third repetition
93 will have the 33% of the realistic value. However, this study showed neither error analysis nor statistical
94 evidence. Another study was carried out by Rom & Zhang (2010) in laboratory conditions; they recommended
95 to measure gas concentrations from a minimum of 12.5 to a maximum of 25 minutes on each position. The
96 authors proposed some suggestions regarding the design of the measurement set up and measurement
97 strategies, however the results were not verified in in-field conditions. In the study of Ngwabie et al. (2009),
98 similar recommendations were applied by sequentially measuring at different indoor sampling locations before
99 switching to outdoor ones, although it was not specified whether replicates for each location were performed.
100 Wu et al. (2012) measured six times at each sampling point with a sampling frequency of less than one minute.
101 The NH₃ concentration at each sampling point was determined by the average of the last three repeated values
102 in the measurement, while other gas concentrations were calculated as the mean of all the six repeated values.
103 In the study of Hassouna et al. (2013), the gas concentrations were acquired in a poultry building every two
104 minutes for 20 minutes before switching to the next sampling location. A sampling frequency of 1 minute was
105 also applied by De Vogeeler et al. (2017) in a naturally ventilated animal mock-up building. In this study, gas
106 concentrations were continuously measured in five sampling points and two measurements were carried out
107 for each sampling point before measuring the next point. In different studies, no replicates for each sampling
108 point were carried out. In detail, Zhang et al. (2005) measured gas concentrations at six sampling points with
109 a sampling frequency of 20 minutes, whereas D'Urso et al. (2021b) used a sampling frequency of 15 min for
110 each sampling point. Other studies (Saha et al., 2013; Hempel et al., 2016) measured gas concentrations at
111 twelve sampling points in a naturally ventilated dairy barn. The INNOVA required 1 minute for each sampling
112 point and, thus, the sequence of the 12 sampling points was completed in about 12 minutes. In another study
113 by Fiedler et al. (2014), gas concentrations were continuous acquired in many sampling points according to a
114 continuous sequence, yet no further information on sampling frequency or number of replicates was provided.
115 Since the measurement of gas concentrations is useful for the emission estimation, the recent VERA Test
116 Protocol (2018) provides a guideline on the measurement strategy (i.e., measuring period, measurement
117 locations, and measuring methods) for emission estimation. However, specific recommendation on the set-up
118 of number of replicates to acquire gas concentrations was not specifically provided for the measurement
119 device.

120 Based on the literature, the research studies have applied different methods to acquire gas concentrations and
121 most of them did not set the INNOVA device in order to make repetitions for each sampling point. Therefore,
122 the aim of this research study was to evaluate the optimal number of repetitions for gas concentrations
123 measured by the INNOVA device under in-field conditions. The objective was to identify the optimal number
124 through statistical analyses in order to provide specific criteria for the design of the measurement campaign.
125 In detail, several repetitions were assessed by studying the variability associated to a specific measurement.
126 Specifically, the number of repetitions needed to minimise the variability were investigated.

127

128 **2. Materials and methods**

129 *2.1. Site description and gas concentration measurements*

130 Measurements were carried out in a cubicle free-stall dairy barn located in South Italy (37°01'N, 14°32'E)
131 within the province of Ragusa. The building is about 55.50 m long and 20.80 m wide with three sides
132 completely open (Figure 1). The roof is symmetric with a central ridge vent having a 7m ridge height and a
133 4m eave height. Different areas compose the indoor environment of the barn: the farmer office, boxes for
134 calves, and cow's functional areas including the resting area with 64 head-to-head cubicles organised in three
135 pens. In each pen the cubicles are organised in two rows built of concrete kerbs and covered with sand (Figure
136 2).

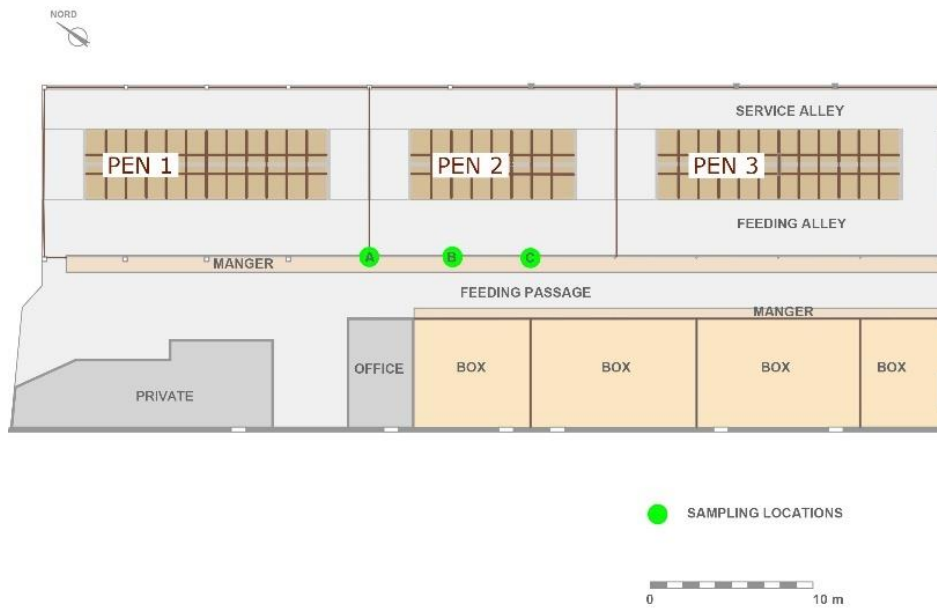


137

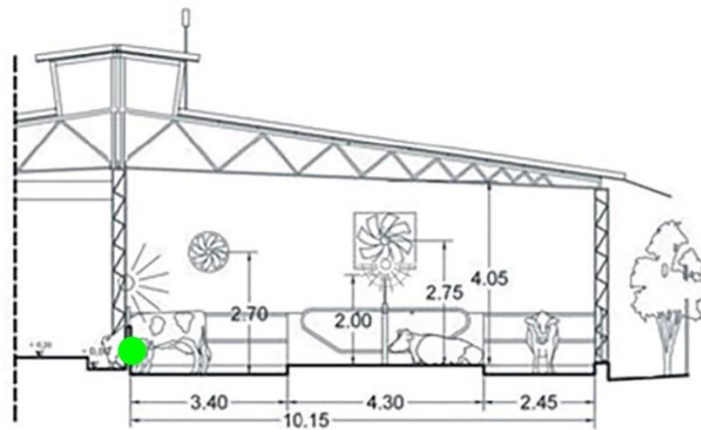
138 *Figure 1. View of the barn with the open-sided walls.*

139

140



141



142

143 *Figure 2. Plan and section of the barn with localisation of sampling locations.*

144 NH₃ concentrations were continuously measured by an INNOVA photo-acoustic analyser, which was
145 calibrated by the manufacturer before the beginning of the measurement campaign in 2018. The INNOVA
146 analyser was composed of a Multigas Monitor mod 1412i and a multipoint sampler 1409/12 (Lumasense
147 Technology A/S, Denmark) with 12 inlet channels. The sampler system is made of AISI-316 stainless steel
148 and PTFE (poly-tetrafluoroethylene) tubing to minimise adsorption of samples. An air-filter was attached to
149 the end of each sampling tube to keep the 1409 free of particles. The declared resolution of the instrument by
150 the manufacturer is equal to 1 % of the measured value and its accuracy is lower than the 3 % of the measured
151 value under standard condition with T° = 20°C and RH = 60%.
152

153 Data were acquired from 19/06/2018 to 30/06/2018 with an indoor and outdoor air temperature of 27.6 ± 5.1°C
154 and 26.5 ± 4.9°C, respectively, and indoor and outdoor air velocity of 0.73 ± 0.46 m s⁻¹ and 1.29 ± 0.68 m s⁻¹.
155 During the experiment, gas concentrations of NH₃ were continuously acquired at three sampling locations (i.e.,
156 SL_A, SL_B, and SL_C) located in the central pen of the barn (Figure 2). The SLs were installed at 20 cm
157 from the barn floor in the animal occupied zone (AOZ) according to the findings of Arcidiacono et al. (2015).
158 In detail, the localisation in the central area of the barn allows obtaining gas concentrations values that are not
159 significantly different (D'Urso et al., 2021a). Moreover, the sampling points were located at 20 cm from the
160 floor to reduce the influence of the indoor conditions related to the open envelope, as it was found in the study
161 by D'Urso et al. (2022).

162 The measurement cycle was composed by the sequence of the SL_A, SL_B, and SL_C. INNOVA device was
163 set to perform continuously ten repetitions for each SL before switching to the next SL. For each repetition,
164 the INNOVA measures the gas concentrations in a specific SL. In the present research study, the INNOVA
165 required about 1 minute 15 seconds for each repetition, about 11 minutes and 15 seconds for each SL (i.e.,
166 time required to perform ten repetitions) and less than one hour for a measurement cycle.

167

168 2.2. Processing datasets and statistical analyses

169 Data collected during the observation periods were organised into different datasets for each SLs by using
170 Excel® software.

171 The variability related to a specific repetition ε_i (%) was computed by using the following relation:

$$172 \quad \varepsilon_i(\%) = \frac{x_{GS} - x_i}{x_{GS}} * 100 \quad \text{with } i = 1, \dots, 10$$

173 where x_{GS} was the gas concentration value of repetition used as benchmark and x_i was the value of gas
174 concentrations in the i -th repetition. Different values of the benchmark were assessed in this study based on
175 specific recommendation found in the literature. In detail, based on the study of Brehme (2003), the tenth
176 repetition was considered as x_{GS} . Moreover, according to the author, the third repetition was considered as x_{GS}
177 based on the balance between the number of sampling locations when more repetitions are carried out and the
178 data loss. In this study, the first and the second repetitions were also considered as x_{GS} to quantify the error in
179 considering those repetitions. With regard to the repetition time length, Rom and Zhang (2010) recommended
180 to measure gas concentrations for at least 12.5 minutes. Since the INNOVA requires about 11 minutes and 15
181 seconds to execute ten repetitions in each SLs, the present study investigated both the tenth repetition and the
182 mean values of the ten repetitions as x_{GS} . In addition, also other repetitions or repetition means were tested as
183 benchmark to investigate the variability of the measurement.

184 Datasets of positive and negative ε_i (%) associated to the i -th repetition were organised in a database for each
185 SLs and the considered benchmark. Then, the one-way analysis of variance (ANOVA) was applied in order to
186 statistically determine whether significant differences occurred between the positive (or negative) variability
187 associated to a specific repetition in a SL. The level of significance was a P value (p) lower than 0.05. If the
188 test was significant (p < 0.05), the applied post hoc test was the Tukey test which identified differences between
189 groups of positive (or negative) variability for a specific repetition in a SL.

190
191
192
193
194
195

196
197
198
199
200
201

202
203

204
205
206
207
208

209
210
211

212
213
214
215
216

217
218
219
220
221
222
223

224
225
226
227

228

3. Results and discussion

3.1. Variability related to the repetition chosen as benchmark

The results of the one-way ANOVA applied for each benchmark under analysis showed p-values lower than 0.05 for each considered SL. Based on the post hoc analysis (Table 1), the positive (or negative) variability showed significant differences related to the i-th considered repetition. Since the results of this study were confirmed for all the SLs, only the values related to the SLB were reported in the following of the text.

In detail, when the benchmark in SLB was the first repetition, it was found that the ε_2 differs from the others, except for ε_3 . However, the negative ε_2 value (i.e., -4.73) obtained by fixing the first repetition as the benchmark was greater compared to the negative ε_2 (i.e., -3.12) when the third repetition was considered the benchmark. Therefore, the first repetition cannot be considered as a benchmark. Moreover, the study of the frequency of the variability showed an underestimation of the measurement in the 55.8% - 59.5% of the cases rather than an overestimation (40.5-44.2%) when the first repetition was considered as the benchmark.

When the benchmark was the second repetition, the ε_3 differed from the variability associated to all those repetitions from the 5th to the 10th, except for ε_1 and ε_4 for positive variability and for ε_4 for negative variability.

When the third repetition was considered as the benchmark, positive and negative ε_1 were higher than ε_2 and ε_4 . Therefore, the first repetition overestimated or underestimated the value acquired in the third repetition with a variability higher than ε_2 and ε_4 . This result highlights that it is not equivalent to consider the first or the third repetition whereas the second, third or fourth repetition are statistically equivalent. In addition, also the fifth repetition could be considered from the statistical point of view, though accepting a higher variability.

When the considered benchmark was the mean value of gas concentration of the second and the third repetitions, the ε_2 and ε_3 were significantly different from the variability in other repetitions with values lower than $\pm 2\%$, except for ε_4 (for positive variability).

In the Tukey test, carried out by considering the fifth repetition as benchmark, ε_3 was higher than ε_4 and ε_6 with a difference of about 1-1.5%. Based on the results, the fifth repetition overestimated or underestimated the value of the third repetition because the measurement value was changing over time, influenced by different factors. Therefore, considering the fifth or third repetition as benchmark is not equivalent, whereas the fourth or the sixth repetitions are statistically equivalent when the benchmark is the fifth repetition.

When the benchmark was the tenth repetition, the error ε_1 , ε_3 , and ε_5 ranged from about 7% to about 14% for both positive and negative values of variability. In detail, the ninth repetition highly overestimated or underestimated the measured values compared to the fifth, the third and the first repetitions. Therefore, taking the ninth or the fifth repetitions was not equivalent, whereas the ninth repetition was equivalent to the eighth repetition. This result confirmed that the measurement value was changing over time. In the last analysis, the mean value of the tenth repetition considered as benchmark produced the lowest variability in the fifth repetition and high variability in the first, second and third repetitions.

The outcomes of SL_B were verified in the other SL_A and SL_C. Based on the statistical analyses, there was not a relevant difference among the results in SL_B, SL_A and SL_C. The results confirmed that: the first repetition should not be considered; a high number of repetitions increases the variability due to the changing of gas concentration in time during in-field experiments.

Table 1. Results of the post-hoc Tuckey test for the variability associated to the number of repetitions ε_i (%) for SLB.

Xgs	First repetition			Second repetition		
	Rep.	Mean	Grouping	Rep.	Mean	Grouping
positive	10	11.11	A	10	9.47	A
	9	10.45	A B	9	9.00	A
	8	10.02	A B	8	8.42	A B
	7	9.15	A B	7	8.28	A B
	6	8.50	B C	6	6.72	B C

	5	7.91	B C	5	5.83	C D
	4	6.58	C D	4	4.50	D E
	3	5.20	D E	1	4.39	D E
	2	3.71	E	3	2.97	E
	2	-4.74	A	1	-3.96	A
	3	-7.08	A B	3	-4.01	A
	4	-8.71	B C	4	-6.13	A B
	5	-9.26	B C D	5	-7.10	B C
	6	-10.54	C D E	6	-8.52	C D
negative	7	-11.78	D E F	7	-9.07	C D E
	8	-12.39	E F	8	-10.36	D E F
	9	-13.45	F	9	-11.16	E F
	10	-13.79	F	10	-11.55	F

XGs	Third repetition			Mean value of the second and third repetitions		
	Rep.	Mean	Grouping	Rep.	Mean	Grouping
	10	7.98	A	10	8.73	A
	9	7.63	A B	9	8.20	A B
	8	7.01	A B	8	7.47	A B C
	1	6.33	B C	7	7.03	B C
positive	7	6.31	B C	6	5.95	C D
	6	5.31	C D	1	5.14	D E
	5	4.53	D	5	5.05	D E
	2	3.76	D E	4	3.56	E F
	4	2.82	E	2	1.94	F G
				3	1.52	G
	2	-3.12	A	2	-1.52	A
	4	-3.85	A B	3	-1.94	A
	5	-4.86	A B C	4	-4.65	B
	1	-5.74	B C D	1	-4.70	B
	6	-6.51	C D	5	-5.71	B C
negative	7	-7.61	D E	6	-7.18	C D
	8	-9.04	E F	7	-8.23	D E
	9	-9.95	F	8	-9.59	E F
	10	-10.51	F	9	-10.38	F
				10	-10.75	F

XGs	Fifth repetition			Tenth repetition			Mean of the ten repetitions		
	Rep.	Mean	Grouping	Rep.	Mean	Grouping	Rep.	Mean	Grouping
	1	7.98	A	1	11.35	A	1	7.30	A
	10	6.87	A B	2	9.77	A B	10	5.49	B
	2	6.35	B C	3	8.99	B C	2	5.41	B C
	9	6.30	B C	4	8.30	B C	9	4.86	B C D
positive	8	5.13	C D	5	7.58	C D	3	4.10	C D E
	3	4.46	D E	6	6.19	D E	8	4.06	D E
	7	4.35	D E	7	5.54	E	7	3.57	D E
	4	3.16	E F	8	4.57	E F	4	3.33	E
	6	2.76	F	9	3.02	F	5	3.18	E
							6	2.88	E
	4	-2.98	A	9	-2.90	A	5	-3.03	A
	6	-3.52	A B	8	-4.52	A B	6	-3.25	A
	3	-4.88	A B C	7	-5.76	B C	4	-3.32	A B
	7	-5.18	B C	6	-7.27	B C D	7	-3.79	A B
	2	-6.47	C D	5	-7.85	C D	3	-3.86	A B
negative	8	-6.85	C D E	4	-8.33	C D	8	-4.73	B C
	9	-7.98	D E F	3	-9.42	D E	2	-5.57	C D
	10	-8.77	E F	2	-11.52	E F	9	-5.67	C D
	1	-9.18	F	1	-14.05	F	10	-6.44	D

229

230

3.2. Measurement strategy

231

232

233

234

235

236

237

238

239

240

241

242

243

244

245

246

247

248

249

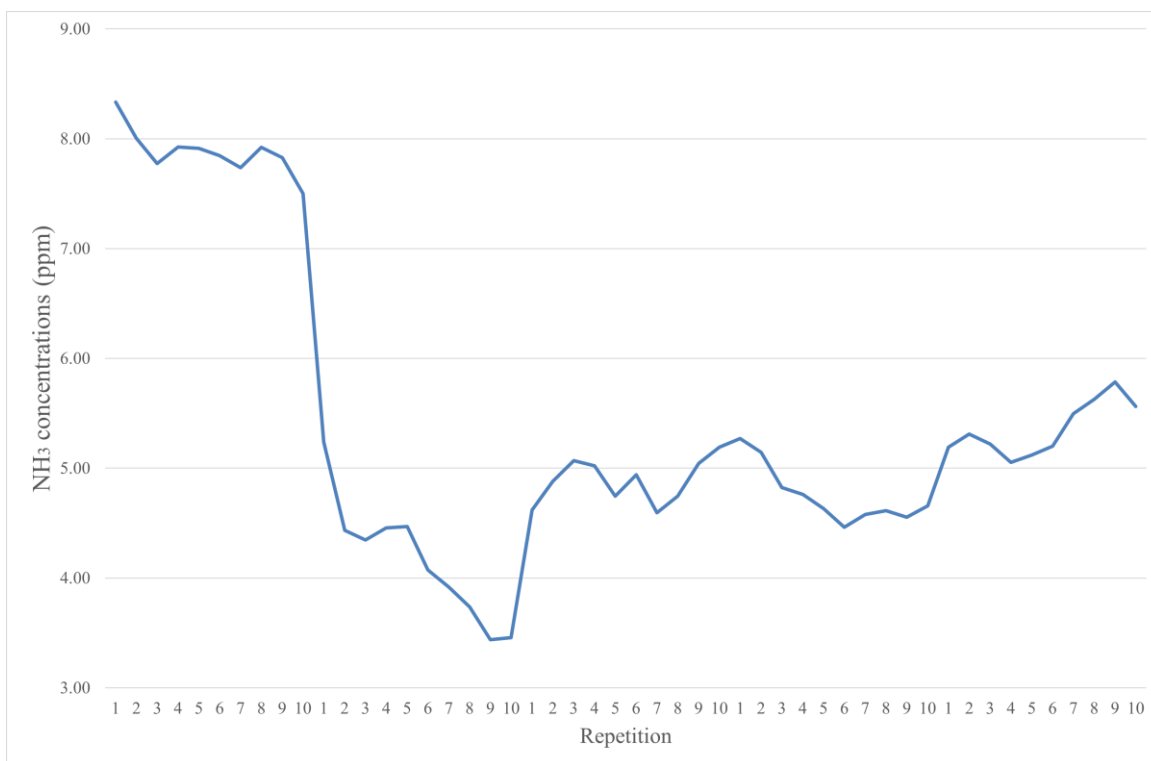
250

251

252

253

Based on the investigation carried out in these experiments, the number of repetitions for each SL had a significant influence on data collection. For this reason, the measurement design should be organised with some repetitions for each SLs. The results recommend excluding the first and the tenth repetitions because their value ε_i had the highest variation from about -14% to the 11%. Excluding the first repetition is in line with what suggested by Brehme (2003) that stated that the first and the second repetitions were less realistic than the third repetition. Moreover, this author reported that the best number of repetitions were ten. In the present work, the results did not corroborate this statement. In fact, besides the highest variability ε_i when the benchmark was the tenth repetition, the ten repetitions had the highest variability ε_{10} when the benchmark was the first, the second and the third repetitions. When the benchmark was the fifth and the mean value of all the ten repetitions, the errors ε_9 and ε_{10} had highest value and were not significantly different from ε_1 . Therefore, increasing the number of the repetitions, in order to have representative data for uniform conditions, does not ensure a higher reliability of the concentrations because the time required by the instrument to perform the measurement increases and, at the meantime, the gas concentrations could be modified in the barn environment. In fact, since each repetition required about 1 minute 15 seconds, ten repetitions required about 11 minutes for each SL. During the range of about 11 minutes, the gas concentration was not constant under in-field conditions (Figure 3). In fact, different typologies of the NH_3 trend with a different variation of the curve slope during the time period of the ten repetitions were found in the experiments. This variation could be attributed not only to the random error of the measure (Calvet et al. 2013), but also to other factors that make the analysis much complex. In detail, as it was recently found in the literature (Saha et al., 2014a, Poteko et al., 2019; Zou et al., 2020; D'Urso et al., 2021a; D'Urso and Arcidiacono, 2021; D'Urso et al., 2022), multiple factors influence the measurement of NH_3 concentrations, such as building configurations, measurement locations, size of the openings, climatic conditions, animal behaviour, barn management, and housing design.



254

255

256

Figure 3. NH_3 concentrations trend during five measurement cycles at SLB with 10 repetitions for each measurement cycle.

257 For this reason, the NH₃ concentration recorded in the fifth repetition generally overestimated or
258 underestimated the value of the second or third repetition and the number of repetitions should not be more
259 than five. Although Zhang and Rong (2010) found that measuring periods should last from 12.5 to 25 minutes
260 in laboratory conditions, the outcome of this study recommended to reduce the measuring periods under in-
261 field conditions to obtain more precise values of the gas. In the present experiment, ten repetitions for each
262 SLs required about 11 minutes but, starting from the fifth repetitions the values of gas concentrations had a
263 significantly higher variability. Therefore, in this barn typology it is recommended to reduce the number of
264 repetitions and perform three or, at least, four repetitions. In this way, this number of repetitions could increase
265 the precision of the results. In fact, precision is acknowledged as the measurement of how tightly clustered the
266 forecast dispersion is. Based on the results of this study the best solution is to perform three repetitions for
267 each SLs, then exclude the first repetition due to the high variability and, then again, compute the mean value
268 of gas concentrations recorded in the second and in the third repetitions.

269 By reducing the number of repetitions, a relevant effect is saving time to perform further measurements cycles
270 and obtain more data with a higher frequency. In fact, a measurement cycle with three SLs and ten repetitions
271 required about 34 minutes to the INNOVA and allowed achieving a complete measurement cycle every hour.
272 When the repetitions are five for each SL and the SL are three, the measurement cycle required about 15
273 minutes thus allowing the data acquisition data in four measurement cycles within an hour. A measurement
274 cycle at three SLs and with three repetitions will allow performing up to six measurement cycles during an
275 hour. This could have positive effect also in the emission estimation. In fact, in the literature (Saha et al., 2014,
276 Hempel et al. 2016, Janke et al., 2020), the authors generally estimate the emissions using the hourly mean
277 value of gas concentrations when more than one measurement is available. In other cases, when there is not a
278 value of gas concentration for each hour, it was used the interpolation method (Hassouna et al., 2013). A
279 correct planning of the number of repetitions as well as the number of SLs could increase the reliability of the
280 measurement because, besides the number of repetitions for each SLs, a higher number of measurement cycles
281 could be planned in order to obtain more values to determine the hourly mean value of gas concentrations,
282 useful for emission estimation. For this reason, the barn dimension and typology have a great influence on the
283 design of the measurement campaign. Reduced barn plan dimensions allow decreasing the number of SLs and
284 increasing repetitions with a significant improvement of the precision of collected data. Moreover, the correct
285 location of SLs in the barn plan could avoid monitoring those places in the barn that can increase the
286 uncertainty in the emission estimation (König et al., 2018; Janke et al., 2020; D'Urso et al. 2021c)

287

288 **4. Conclusions**

289 In this study, the concentration of NH₃ was measured in an open naturally ventilated dairy barn during the
290 warm period in the Mediterranean area. The measurement campaign was carried out by using the INNOVA
291 photo-acoustic analyser, and different repetitions were performed for each sampling location (SL). The results
292 of this work highlighted a measurement strategy for acquiring data of gas concentrations with INNOVA in a
293 open barn typology. Based on this research study, it is possible to conclude that increasing in the number of
294 the repetitions does not ensure a higher reliability of the gas concentrations.

295 A measurement protocol for open or semi-open dairy barn could represent a fundamental step for monitoring
296 air quality, improving animal welfare, assessing mitigation strategies, or estimating emissions. Moreover, the
297 implication of this research study could be helpful in elaborating a potential revision of the next VERA
298 protocol. An improved knowledge is needed related to the vertical position of SLs, the distance from the
299 perimeter (without wall) to the SLs, the influence of climatic factor or barn management in the uncertainty of
300 measurements. Based on the advancement in measuring gas concentrations, the following step is to evaluate
301 the impact of different positions of SLs in the monitoring of gas concentrations in open and semi-open dairy
302 barns. Moreover, the application of a measuring strategy in this barn typology to obtain emission factor has
303 not yet been assessed.

304

305 **5. Acknowledgements**

306 The authors are grateful to the farm ALPA S.S. for providing the opportunity of carrying out the tests.

307 The research study was funded by the University of Catania through the ‘Piano incentivi per la ricerca di
308 Ateneo 2020-2022-Linea 2’ project on ‘Engineering solutions for sustainable development of agricultural
309 buildings and land’ - LANDSUS (ID: 5A722192152), coordinated by Claudia Arcidiacono.

310 The INNOVA device was funded by the project “Centro per l’innovazione dei sistemi di qualità tracciabilità
311 e certificazione dell’agroalimentare”- AGRIVET (ID: G46D15000170009).

312 **6. References**

313 Arcidiacono, C., Porto, S. M. C., & Cascone, G., 2015. On ammonia concentrations in naturally ventilated
314 dairy houses located in Sicily. *Agricultural Engineering International: CIGR Journal*, 2015, 294–310.

315 Bjerg, B., Zhang, G., Madsen, J., Rom, H.B. Methane emission from naturally ventilated livestock buildings
316 can be determined from gas concentration measurements. *Environmental Monitoring and Assessment*.
317 184(10):5989-6000. DOI: 10.1007/s10661-011-2397-8

318 Brehme G., 2003. True measuring with „ Innova “. *Measuring Technology*. 58 *Landtechnik* 3, 196–197

319 Calvet, S., Gates, R. S., Zhang, G. Q., Estellés, F., Ogink, N. W. M., Pedersen, S., & Berckmans, D., 2013.
320 Measuring gas emissions from livestock buildings: A review on uncertainty analysis and error sources.
321 *Biosystems Engineering*, 116(3), 221–231. DOI: <https://doi.org/10.1016/j.biosystemseng.2012.11.004>

322 de Vries, W, 2021. Impacts of nitrogen emissions on ecosystems and human health: A mini review. *Curr. Opin.*
323 *Environ. Sci. Health*, 21 p. 100249. DOI: <https://doi.org/10.1016/j.coesh.2021.100249>

324 De Vogeeler, G., Pieters, J.G., Van Overbeke, P., Demeyer, P., 2017. Effect of sampling density on the
325 reliability of airflow rate measurements in a naturally ventilated animal mock-up building. *Energy and*
326 *Building*. 152, 313–322. DOI: <https://doi.org/10.1016/j.enbuild.2017.07.032>

327 D’Urso, P.R.; Arcidiacono, C.; Valenti, F.; Cascone, G., 2021a. Assessing Influence Factors on Daily
328 Ammonia and Greenhouse Gas Concentrations from an Open-Sided Cubicle Barn in Hot Mediterranean
329 Climate. *Animals*, 11, 1400. <https://doi.org/10.3390/ani11051400>

330 D’Urso, P.R.; Arcidiacono, C.; Cascone, G., 2021b. Environmental and Animal-Related Parameters and the
331 Emissions of Ammonia and Methane from an Open-Sided Free-Stall Barn in Hot Mediterranean Climate: A
332 Preliminary Study. *Agronomy* 2021, 11, 1772. DOI: <https://doi.org/10.3390/agronomy11091772>

333 D’Urso, P.R., Arcidiacono C., and Cascone, G., 2021c. Uncertainty in determining ammonia and methane
334 emissions at different sampling locations in an open-sided dairy barn," 2021 IEEE International Workshop on
335 Metrology for Agriculture and Forestry (MetroAgriFor), pp. 145-150, doi:
336 10.1109/MetroAgriFor52389.2021.9628493.

337 D’Urso P.R., Arcidiacono C., Cascone G, 2022. Spatial variability of ammonia concentrations in an open-
338 sided dairy barn. *Lecture Notes in Civil Engineering*, 252 LNCE, pp. 76–84

339 D’Urso, P.R. and Arcidiacono, C., 2021. Effect of the Milking Frequency on the Concentrations of Ammonia
340 and Greenhouse Gases within an Open Dairy Barn in Hot Climate Conditions. *Sustainability* 2021, 13, 9235.
341 <https://doi.org/10.3390/su13169235>

342 Fiedler, M.; Saha, C.K.; Ammon, C.; Berg, W.; Loebstin, C.; Sanftleben, P.; Amon, T., 2014. Spatial
343 Distribution of Air Flow And CO2 Concentration in a Naturally Ventilated Dairy Building. *Environmental*
344 *Engineering and Management Journal*. 13, 2193–2200.

345 Hassouna M. and Heglin. 2016. Measuring emissions from livestock farming: greenhouse gases, ammonia and
346 nitrogen oxides. Ademeand INRA, Paris, France. ISBN2-7380-1392-9. hal-01567208. Available online at:
347 <https://hal.archives-ouvertes.fr/hal-01567208>

348 Hassouna, M., Robin, P., Charpiot, A., Edouard, N., & Méda, B., 2013. Infrared photoacoustic spectroscopy
349 in animal houses: Effect of non-compensated interferences on ammonia, nitrous oxide and methane air
350 concentrations. *Biosystems Engineering*, 114(3), 318–326. DOI:
351 <https://doi.org/10.1016/j.biosystemseng.2012.12.011>

352 Hempel, S.; Saha, C.K.; Fiedler, M.; Berg, W.; Hansen, C.; Amon, B.; Amon, T., 2016. Non-linear temperature
353 dependency of ammonia and methane emissions from a naturally ventilated dairy barn. *Biosystems*
354 *Engineering.*, 145, 10–21. DOI: <https://doi.org/10.1016/j.biosystemseng.2016.02.006>

355 Janke, D., Willink, D., Ammon, C., Hempel, S., Schrade, S., Demeyer, P., Hartung, E., Amon, B., Ogink, N.,
356 Amon, T., 2020. Calculation of ventilation rates and ammonia emissions: Comparison of sampling strategies
357 for a naturally ventilated dairy barn. *Biosystems Engineering*, 198, 15–30. DOI:
358 <https://doi.org/10.1016/j.biosystemseng.2020.07.011>

359 König, M.; Hempel, S.; Janke, D.; Amon, B.; Amon, T., 2018. Variabilities in determining air exchange rates
360 in naturally ventilated dairy buildings using the CO₂ production model. *Biosystems Engineering*, 174, 249–
361 259. DOI: <https://doi.org/10.1016/j.biosystemseng.2018.07.001>

362 Li, H., Zhang, C., Xin, H., 2015. Performance of an Infrared Photoacoustic Single Gas Analyzer in Measuring
363 Ammonia from Poultry Houses. *Applied Engineering in Agriculture* 31: 471–477, DOI:
364 10.13031/aea.31.10826

365 Mendes, L.B.; Ogink, N.W.M.; Edouard, N.; Van Dooren, H.J.C.; Tinôco, I.D.F.F.; Mosquera, J., 2015. NDIR
366 Gas Sensor for Spatial Monitoring of Carbon Dioxide Concentrations in Naturally Ventilated Livestock
367 Buildings. *Sensors*, 15, 11239–11257. <https://doi.org/10.3390/s150511239>

368 Mendes, L.B.; Pieters, J.G.; Snoek, D.; Ogink, N.W.; Brusselman, E.; Demeyer, P., 2017. Reduction of
369 ammonia emissions from dairy cattle cubicle houses via improved management- or design-based strategies: A
370 modeling approach. *Science of the Total Environment*, 574, 520–531. DOI: 10.1016/j.scitotenv.2016.09.079

371 Ngwabie, N. M., Jeppsson, K. H., Nimmermark, S., Swensson, C., & Gustafsson, G., 2009. Multi-location
372 measurements of greenhouse gases and emission rates of methane and ammonia from a naturally-ventilated
373 barn for dairy cows. *Biosystems Engineering*. <https://doi.org/10.1016/j.biosystemseng.2009.02.004>

374 Ngwabie, N.M., Jeppsson, K.-H., Gustafsson, G., Nimmermark S., 2011. Effects of animal activity and air
375 temperature on methane and ammonia emissions from a naturally ventilated building for dairy cows.
376 *Atmospheric Environment*. 45: 6760–6768. DOI: <https://doi.org/10.1016/j.atmosenv.2011.08.027>

377 Ngwabie, N.M., Vanderzaag, A., Jayasundara S., Wagner-Riddle C., 2014. Measurements of emission factors
378 from a naturally ventilated commercial barn for dairy cows in a cold climate. *Biosystems Engineering*. 127,
379 103–104. DOI: <https://doi.org/10.1016/j.biosystemseng.2014.08.016>

380 Poteko, J.; Zähler, M.; Schrade, S. 2019. Effects of housing system, floor type and temperature on ammonia
381 and methane emissions from dairy farming: A meta-analysis. *Biosystem Engineering.*, 182, 16–28. DOI:
382 <https://doi.org/10.1016/j.biosystemseng.2019.03.012>

383 Rom, H. B., and Zhang, G., 2010. Time Delay for Aerial Ammonia Concentration Measurements in Livestock
384 Buildings. *Sensors*, 10, 4634–4642. DOI: 10.3390/s100504634

385 Rong, L., Liu, D., Pedersen, E. F., & Zhang, G., 2014. Effect of climate parameters on air exchange rate and
386 ammonia and methane emissions from a hybrid ventilated dairy cow building. *Energy and Buildings*.
387 <https://doi.org/10.1016/j.enbuild.2014.07.089>

388 Saha, C.K.; Ammon, C.; Berg, W.; Loebstin, C.; Fiedler, M.; Brunsch, R.; Von Bobrutski, K., 2013. The effect
389 of external wind speed and direction on sampling point concentrations, air change rate and emissions from a
390 naturally ventilated dairy building. *Biosystems Engineering*, 114, 267–278. DOI:
391 <https://doi.org/10.1016/j.biosystemseng.2012.12.002>

392 Saha, C.; Ammon, C.; Berg, W.; Fiedler, M.; Loebstin, C.; Sanftleben, P.; Brunsch, R.; Amon, T., 2014a.
393 Seasonal and diel variations of ammonia and methane emissions from a naturally ventilated dairy building and
394 the associated factors influencing emissions. *Science of the Total Environment*. 468–469, 53–62. DOI:
395 <https://doi.org/10.1016/j.scitotenv.2013.08.015>

396 Saha, C.K., Fiedler, M., Ammon, C., Berg, W., Loebstin, C., Amon, B., Amon, T., 2012. Uncertainty in
397 calculating air exchange rate of naturally ventilated dairy building based on point concentrations.
398 *Environmental engineering and management journal*. Vol. 13 n.9, pp. 2349-2355; DOI:
399 [10.30638/eemj.2014.262](https://doi.org/10.30638/eemj.2014.262)

400 Schmithausen, A.J., Trimborn, M., Büscher, W. 2016 Methodological Comparison between a Novel
401 Automatic Sampling System for Gas Chromatography versus Photoacoustic Spectroscopy for Measuring
402 Greenhouse Gas Emissions under Field Conditions. *Sensors*, 16, 1638. DOI:
403 <https://doi.org/10.3390/s16101638>

404 Schmithausen, A.J.; Schiefner, I.; Trimborn, M.; Gerlach, K.; Südekum, K.-H.; Pries, M.; Büscher, W., 2018.
405 Quantification of Methane and Ammonia Emissions in a Naturally Ventilated Barn by Using Defined Criteria
406 to Calculate Emission Rates. *Animals*, 8, 75. DOI: <https://doi.org/10.3390/ani8050075>

407 Shen, X.; Zong, C.; Zhang, G., 2012. Optimization of Sampling Positions for Measuring Ventilation Rates in
408 Naturally Ventilated Buildings Using Tracer Gas. *Sensors*, 12, 11966-11988. DOI:
409 <https://doi.org/10.3390/s120911966>

410 Tullo, E., Finzi, A., Guarino, M., 2019. Review: Environmental impact of livestock farming and Precision
411 Livestock Farming as a mitigation strategy. *Science of the Total Environment*, 650, pp. 2751-2760, DOI:
412 [10.1016/j.scitotenv.2018.10.018](https://doi.org/10.1016/j.scitotenv.2018.10.018)

413 VERA. 2018. Vera test protocol: For livestock housing and management systems.

414 von Jasmund, N., Schmithausen, A.J., Krommweh, M.S., Trimborn, M., Boeker, P., Büscher, W., 2022.
415 Assessment of ammonia sensors and photoacoustic measurement systems using a gas calibration unit.
416 *Computer and Electronics in Agriculture*. 194: 106744. DOI: <https://doi.org/10.1016/j.compag.2022.106744>

417 Wang, X.; Ndegwa, P.M.; Joo, H.; Neerackal, G.M.; Harrison, J.H.; Stöckle, C.O.; Liu, H., 2016. Reliable
418 low-cost devices for monitoring ammonia concentrations and emissions in naturally ventilated dairy barns.
419 *Environmental Pollution*, 208, 571–579. DOI: <https://doi.org/10.1016/j.envpol.2015.10.031>.

420 Wu, W., Zhang, G., Kai, P., 2012. Ammonia and methane emissions from two naturally ventilated dairy cattle
421 buildings and the influence of climatic factors on ammonia emissions. *Atmospheric Environment*. 61: 232-
422 243. DOI: <https://doi.org/10.1016/j.atmosenv.2012.07.050>

423 Zhang, G.; Strøm, J.S.; Li, B.; Rom, H.B.; Morsing S.; Dahl, P.; Wang C., 2005. Emission of Ammonia and
424 Other Contaminant Gases from Naturally Ventilated Dairy Cattle Buildings. *Biosystems Engineering*, 92 (3),
425 355–364. DOI: [10.1016/j.biosystemseng.2005.08.002](https://doi.org/10.1016/j.biosystemseng.2005.08.002)

426 Zhuang, S., Brusselman, E., Sonck, B., & Demeyer, P., 2020. Validation of Five Gas Analysers for Application
427 in Ammonia Emission Measurements at Livestock Houses According to the VERA Test Protocol. *Applied
428 sciences*, 10, 5034. DOI: <https://doi.org/10.3390/app10155034>

429 Zou, B., Shi, Z., Du, S., 2020. Gases emissions estimation and analysis by using carbon dioxide balance method
430 in natural-ventilated dairy cow barns. *International Journal of Agricultural and Biological Engineering*. Vol.
431 13 n.2, pp. 41-47. DOI: [10.25165/j.ijabe.20201302.4802](https://doi.org/10.25165/j.ijabe.20201302.4802)