

1 **Advances in sensing ammonia from agricultural sources**

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28 **Abstract**

29 Reducing ammonia emissions is one of the most difficult challenges for environmental
30 regulators around the world. About 90% of ammonia in the atmosphere comes from
31 agricultural sources, so that improving farm practices in order to reduce these emissions is a
32 priority. Airborne ammonia is the key precursor for particulate matter (PM_{2.5}) that impairs
33 human health, and ammonia can contribute to excess nitrogen that causes eutrophication in
34 water and biodiversity loss in plant ecosystems. Reductions in excess nitrogen (N) from
35 ammonia are needed so that farms use N resources more efficiently and avoid unnecessary
36 costs. To support the adoption of ammonia emission mitigation practices, new sensor
37 developments are required to identify sources, individual contributions, to evaluate the
38 effectiveness of controls, to monitor progress towards emission-reduction targets, and to
39 develop incentives for behavioural change. There is specifically a need for sensitive,
40 selective, robust and user-friendly sensors to monitor ammonia from livestock production
41 and fertiliser application. Most currently-available sensors need specialists to set up,
42 calibrate and maintain them, which creates issues with staffing and costs when monitoring
43 large areas or when there is a need for high frequency sampling. This paper reports
44 advances in monitoring airborne ammonia in agricultural areas. Selecting the right method of
45 monitoring for each agricultural activity will provide critical data to identify and implement
46 appropriate ammonia controls. Recent developments in chemo-resistive materials allow
47 electrochemical sensing at room temperature, and new spectroscopic methods are sensitive
48 enough to determine low concentrations in the order of parts per billion. However, these new
49 methods still compromise selectivity and sensitivity due to the presence of ambient dust and
50 other interferences, and are not yet suitable to be applied in agricultural monitoring. This
51 review considers how ammonia measurements are made and applied, including the need for
52 sensors that are suitable for routine monitoring by non-specialists. The review evaluates how
53 monitoring information can be used for policies and regulations to mitigate ammonia
54 emissions. The increasing concerns about ammonia emissions and the particular needs

55 from the agriculture sector are addressed, giving an overview of the state-of-the-art and an
56 outlook on future developments.

57 **Keywords:** Ammonia, Sensors, Nitrogen losses, Livestock production, Fertiliser Application.

58

59 **1. Introduction**

60 In most developed countries, the dominant source of ammonia (NH_3) emissions is the agro-
61 industry . Efforts to mitigate these emissions must therefore focus on improving those
62 agricultural activities that are important contributors - such as livestock feeding and housing,
63 excreta management, and fertilisation practices. To develop better agricultural practices it is
64 critical to know which sources contribute to these emissions, when, and by how much.
65 Ammonia measurements are primarily needed to understand emission rates, the
66 effectiveness of control measures, the impact of atmospheric processes and the outcomes
67 of exposure. Many types of sensors and devices for measuring NH_3 are commercially
68 available, and some promising new developments will be discussed in this paper. However,
69 most of the current devices are relatively specialised, costly or difficult to apply in “real world”
70 farming situations, so they are not readily used for day-to-day monitoring by non-specialist
71 practitioners. Also, there are only a few sensors that have enough sensitivity to measure
72 NH_3 emissions from fertilised soils, such as those using mid-ultraviolet absorption
73 spectroscopy , photoacoustic spectroscopy or the recent artificial nose with fluorescence
74 detection . NH_3 measurements are needed to improve agricultural efficiency, and to
75 contribute to a better quality of life – because excess concentrations of NH_3 lead to adverse
76 impacts on human health and on the health and diversity of ecosystems. Reductions in NH_3
77 emissions would support more efficient food production that uses fewer resources, and so
78 would reduce the environmental impacts of food supply chains. Availability of simple, reliable
79 and low cost measurements and procedures will help developing an improved understanding
80 of NH_3 emissions which is important for:

81 i) Agricultural producers: reduced NH_3 emission will generate savings in nitrogen
82 fertilisers . More efficient use of crude protein in animal feed reduces N excretion,

83 and these emission reductions could attract farm payments if they are linked to
84 environmental improvements.

85 ii) Researchers: to develop new and more suitable technologies to reduce emissions
86 and solve problems for the agroindustry and society.

87 iii) Wider society and the economy: to decrease the combined cost for human health
88 and biodiversity from excess NH_3 , which is estimated to be 4-30 euros per kg N
89 pollution in 2011 .

90 iv) Regulators: to target control measures effectively onto culpable sources, and to
91 evaluate source performance and the effectiveness of controls.

92 v) Policymakers and government: monitoring NH_3 emissions is required to meet
93 international emission reduction targets .

94 **2. Ammonia in the environment**

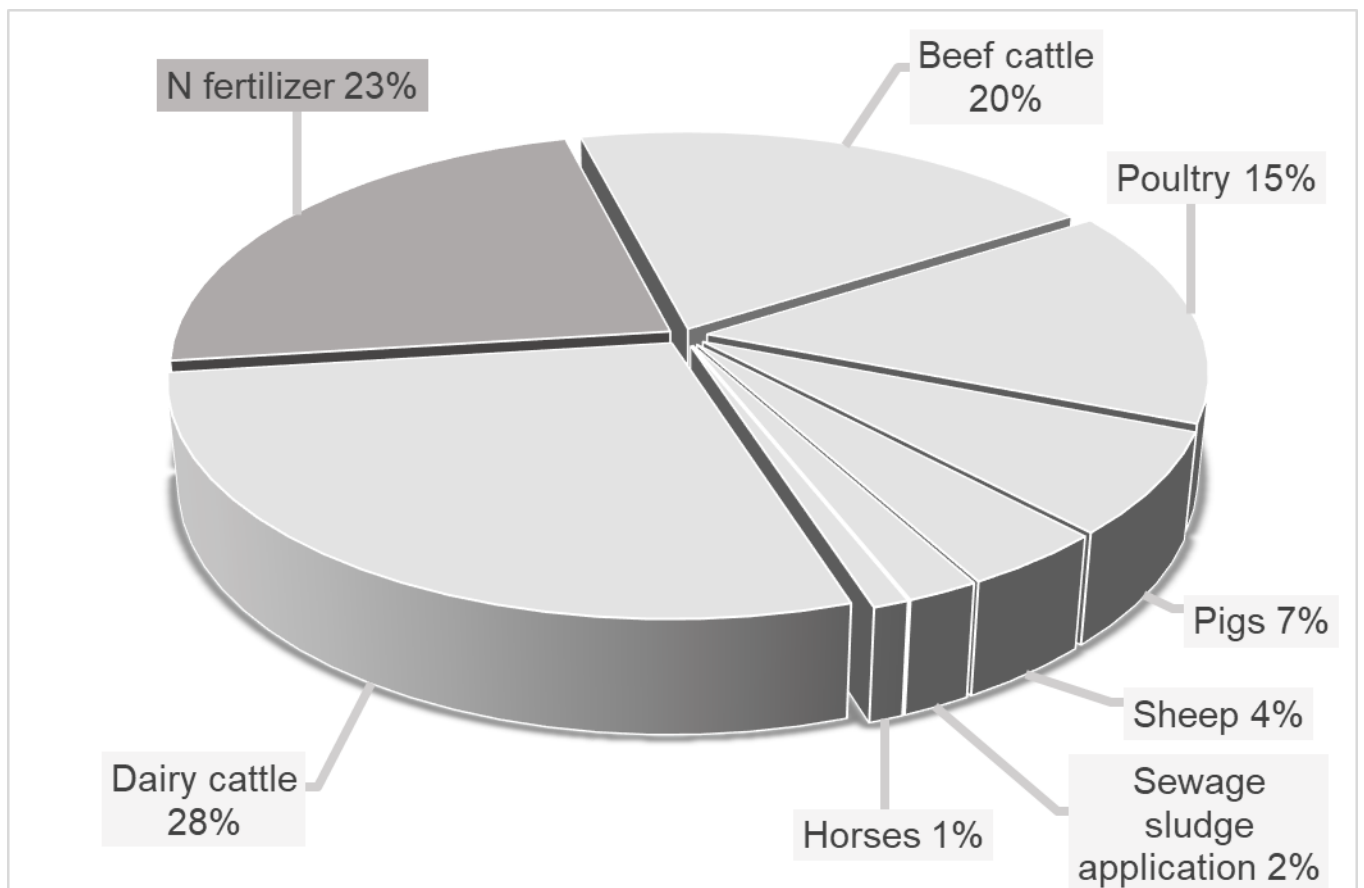
95 The distinct smell of ammonia is often noticed in rural parts of developed countries with high-
96 density livestock farming, especially near intensive pig, poultry or cattle facilities or after
97 liquid manure has been spread on the land. NH_3 is a colourless with a very perceptible
98 odour, even at very low concentrations. It is a common gas that is released in the natural
99 environment from decomposing organic material and animal waste. In the agricultural
100 industry, NH_3 can be used as a fertiliser in the pure gas form of anhydrous NH_3 , but it is also
101 widely used to make nitrogen-based fertilisers such as urea, urea-ammonium nitrate,
102 ammonium sulfate, ammonium nitrate and diammonium phosphate. NH_3 is also used as a
103 refrigerant, and in the manufacture of plastics, explosives, textiles, pesticides, dyes and
104 other chemicals. Although NH_3 is commonly present in nature and widely used in industry, it
105 is classified as an extremely hazardous substance because it is both caustic and flammable
106 . The United States Environmental Protection Agency has classified NH_3 as a hazardous
107 substance because of its toxicity to humans, and has set a maximum safe level of 25 ppm
108 for long term exposure (8 hours) and of 35 ppm for short term exposure (15 min) . For
109 livestock, exposure to 25 ppm of NH_3 can adversely affect weight gain, feed conversion and
110 mortality rates . Several institutions regulate exposure and peak instantaneous

111 concentrations, but there are no regulations for NH_3 emission rates. The 'Environmental
112 Permitting Regulations' relate to ammonia emissions , which set a limit for large livestock
113 facilities on the number of animals that can be kept near a sensitive habitat. Critical level of
114 ammonia concentration cannot be exceeded, and these concentrations are calculated based
115 on accepted emission factors for livestock for different production systems. Another relevant
116 piece of legislation is the National Emission Ceilings Directive (NECD), which sets targets for
117 maximum annual emissions from individual European countries . There is also the
118 Gothenburg Protocol of The United Nations Economic Commission for Europe (UNECE)
119 which is an international agreement to abate acidification, eutrophication and ground-level
120 ozone, that aims to reduce emissions of several pollutants including NH_3 .

121

122 Livestock excreta and fertilisers are major sources of ammonia. Large amounts of NH_3 are
123 released from excreta immediately after deposition and during decomposition. Livestock are
124 often fed high-protein feeds containing N to ensure that their nutritional requirements are
125 met, but the N that is not converted to animal protein is excreted in the urine and the faeces.
126 Once the N compounds are in the faeces or urine, enzyme reactions or microbial activity
127 quickly release NH_3 to the atmosphere . This is because the urea produced and excreted by
128 mammals is rapidly converted to NH_3 gas by the urease enzyme. In the case of poultry, it is
129 uric acid that is excreted and mostly converted to urea, and then to NH_3 . There are several
130 technologies that can be implemented to reduce NH_3 emissions by reducing the availability
131 of both precursors (urea and the urease enzyme) such as controlling the contents of
132 livestock diets or using urease inhibitors such as is N-(n-Butyl)-thiophosphoric triamide
133 (NBPT) . Another control option is to reduce urea hydrolysis by separating the urea and
134 faeces . Key methods to control ammonia emissions from animal excreta are to keep it as
135 dry as possible (i.e. by using indirect heating or heat exchangers); to keep it covered (e.g.
136 floating covers for slurry stores and lagoons); to acidify it; and to avoid adverse weather
137 conditions such as high temperatures, high humidity and adverse wind direction when
138 manure is injected into the soil. The other large source of NH_3 gas is the application of N

139 fertilisers, which represents a significant fraction (23%) of NH₃ emissions (Figure 1, data
140 from the UK Code COGAP). All fertilisers containing ammonium (NH₄⁺) are subject to volatile
141 losses and the NH₃ emissions is largely controlled by pH . Precision agriculture can help
142 adjusting fertiliser rates to reduce excess N application and hence losses by volatilization or
143 run-off.



144
145 **Figure 1:** Agricultural NH₃ emissions in the UK by livestock (light grey) and fertiliser category
146 .

147
148 Presently, NH₃ stands out as the air pollutant whose ambient concentrations are not
149 decreasing rapidly, and occasionally increase slightly. Similarly, emissions of NH₃ during the
150 last 3 decades have reduced much more slowly than those of other major air pollutants such
151 as NO_x, SO₂ and non-methane volatile organic compounds . A recent study shows that there
152 are numerous NH₃ hotspots and unknown sources unidentified around the world . Areas with
153 high beef cattle density are reported to produce an average of 50 µg N m⁻² s⁻¹ (85 g animal⁻¹

154 day⁻¹) . The Po Valley in northern Italy is considered the region in Europe with the highest
155 intensity of NH₃ emissions, which can reach maximum values close to 250 µg m⁻² s⁻¹ during
156 the season of manure application . After urea application to soils, NH₃ emissions can reach
157 an peak of about 5 µg m⁻² s⁻¹ around midday if there is no rain . In order to address the
158 environmental burden of NH₃ emissions and to design management strategies, it is
159 necessary to distinguish the main sources of agricultural NH₃, because 88-94% of NH₃ in the
160 troposphere comes from farming practices , of which 65-71% is from livestock and 16-23%
161 from fertiliser application. There are several options to mitigate NH₃ emissions and
162 increased efforts are needed to lower concentrations to safer levels. To support the
163 implementation of policies and changes to farming practices are effective, there is a need to
164 monitor emissions and ambient levels of NH₃. For that purpose, a transformative
165 improvement in monitoring technology is required to get real-world data from multiple sites
166 and activities.

167

168 **3. Impacts of ammonia emissions**

169 Ammonia in the atmosphere has become a concern for environmental and health agencies
170 for two main reasons: because of excess N deposition and because NH₃ is a precursor of
171 fine particulate matter, both of which can impair human health. NH₃ is an alkaline compound,
172 and when released into the air it is rapidly adsorbed to surfaces and significant deposition
173 (up to 20%) may occur over distances ranging from a few hundreds of meters of the source
174 to hundreds of miles . The remaining atmospheric NH₃ can rapidly react with acid gas
175 compounds (such as SO₂ and NO_x from vehicular and industrial combustion processes) to
176 form very small secondary aerosol particles . This fine particulate matter has an
177 aerodynamic equivalent diameter of <2.5 microns (referred to as PM_{2.5}), having a lifespan
178 of up to 2 weeks and contributing to atmospheric haze and low visibility . NH₃ is transported
179 by wind and deposited not far away from the source where it can promote plant growth in
180 areas where N was previously a limiting factor . An example of this phenomenon is the
181 eutrophication in catchments, including algal blooms that impair water quality and can harm

182 ecosystems and people . Dry N deposition is a continuous process where the gases and
183 aerosols from the atmosphere are transferred to the surface by atmospheric turbulence and
184 diffusion, whereas wet deposition is the intermittent removal of chemical species from the
185 atmosphere by precipitation. Airborne NH_3 concentrations reflect and approach a dynamic
186 equilibrium between deposition and re-emission, an ongoing exchange between the surface
187 and the atmosphere. Changes in plant species composition have been observed in natural
188 ecosystems where high N deposition occurs . There is a loss of biodiversity because N-
189 intolerant plants become depleted and N-tolerant plants become dominant . An example of
190 this effect is the direct damage to sensitive vegetation such as lichens and bryophytes that
191 occur even at very low NH_3 concentrations of around $1 \mu\text{g NH}_3 \text{ m}^{-3}$. Moreover, the
192 deposition of N onto soils with a low buffering capacity can result in soil acidification and
193 consequently basic cation depletion . Harm to human beings is caused by the fine particles
194 that are inhaled deeply into the lungs and impair respiratory function and cardiovascular
195 health . In addition, high concentrations (25 ppm) of atmospheric NH_3 , as it is an irritant
196 compound, have a negative effect on animal production in confined buildings and farm
197 workers should avoid prolonged exposure .

198

199 **4. Detection and quantification of ammonia**

200 We review here the methods currently available to measure NH_3 and discuss some of the
201 previous and significant developments in the field. The instant value of NH_3 concentration in
202 the air, a measure that the currently available sensors usually provide, is only useful to meet
203 the requirements of regulations in different activities. For some farming activities, regulators
204 require instantaneous measurements of NH_3 concentrations, but these measurements have
205 to be used in modelling platforms with other data to estimate wider emissions and impacts of
206 airborne ammonia. To be able to manage emissions from agriculture there is a need for
207 sensors to identify and quantify different sources, including point, area and volume sources,
208 and to identify major contributors. Monitoring programmes should provide data on NH_3
209 concentrations across a range of different scales and environments linking sources and

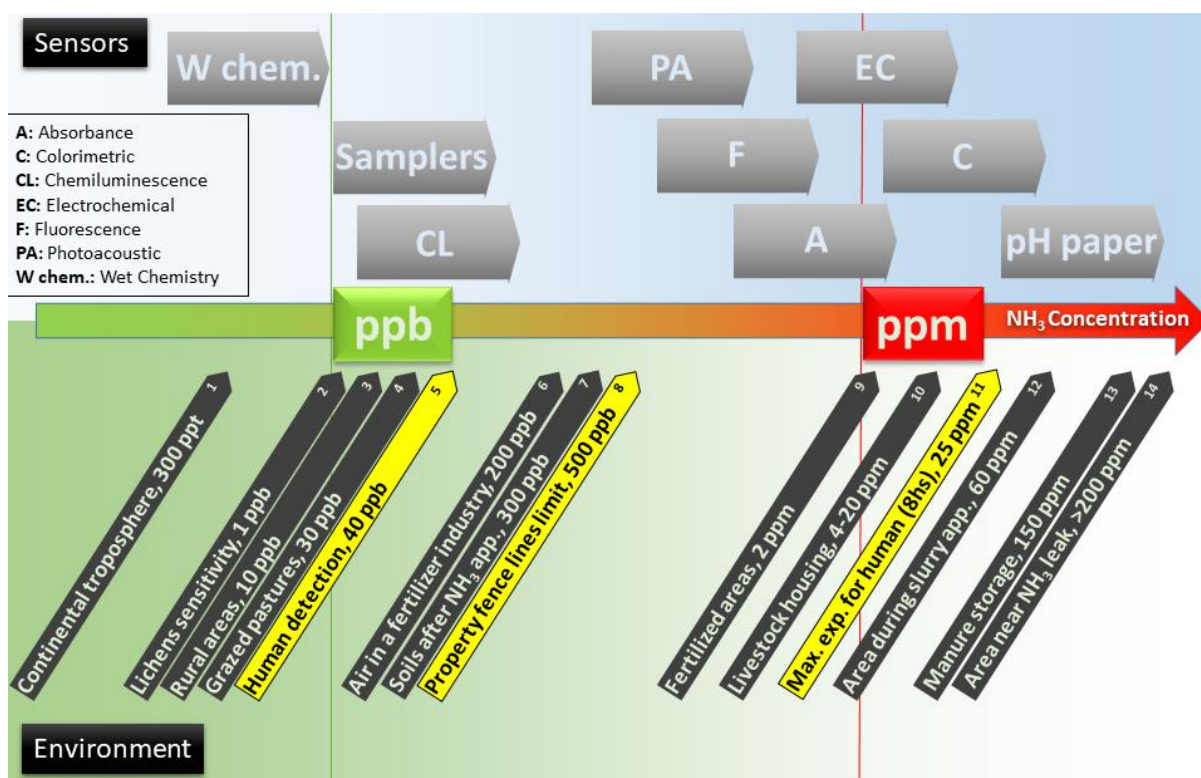
210 receptors. At present a major challenge is to detect the low concentrations occurring in the
211 troposphere (0.3–6 ppb) . Moreover, when the aim is to provide meso-scale and regional
212 patterns, in traditional monitoring schemes, low-frequency (weekly, monthly) sampling is
213 usually selected, which presents the challenge of delivering data over long periods. Most of
214 the available sensors require a qualified operator to set up, calibrate, maintain and de-
215 commission the measurements, which creates issues with staffing and costs when
216 monitoring large areas or when there is a need for high frequency sampling. One of the
217 biggest challenges is to design monitoring that can be used successfully by non-specialists
218 e.g. farmers. A range of monitoring techniques is required that covers different levels of
219 complexity and detail e.g. cheap and easy-to-deploy methods that non-specialists can use
220 for basic reconnaissance and scoping of sources, and more sophisticated methods can be
221 used to identify individual source contributions and performance in complex or specialised
222 studies. Quantifying NH₃ gas emissions needs devices that measure concentrations but also
223 modelling methods to scale concentrations and to calculate emission rates . Also, important
224 critical information is needed to achieve accurate estimations of emission rates, like
225 information on livestock housing facilities, feeds, temperature, wind speed, ventilation rates,
226 among others. Several modelling approaches have been used to estimate NH₃ emission
227 rates, and in order to obtain robust estimates they must take account of variables like wind
228 speed, humidity and air temperature at different heights – which may be measured or
229 derived from numerical weather prediction models. There are particular challenges when
230 measuring NH₃ at low concentrations, because the polarity and geometry of the molecule
231 make it highly soluble in water so it tends to adsorb onto surfaces such as air sampling
232 tubes, which can result in significant errors when concentrations are low .

233

234 For review purposes, the range of sensors available can be classified by different aspects,
235 such as cost, temporal resolution, and level of automation. In this review, sensors were
236 classified by the analytical technique used to detect NH₃ molecules. The sensors covered
237 are only those considered suitable for agricultural applications. These sensors need to be

238 portable, and be selective for NH_3 so that measurements are not compromised by
239 interference from other ambient chemicals. They also need to have a wide range of
240 operation to measure concentrations found in farming and sensitive ecosystem situations,
241 including the low concentrations that occur in some environments. Some sensors are
242 suitable for measurements in livestock housing and manure storage situations, whereas
243 others are suitable for measuring lower concentrations of NH_3 from fields (Figure 2 and
244 Table 1). As shown in Table 2, only high-cost equipment (e.g. chemiluminescent or
245 photoacoustic, cost $> \$25,000$) is suitable for directly measuring low concentrations of NH_3
246 ($< 0.1 \text{ ppm}$). Lower-cost sensors (electrochemical types, cost $\$500 - \2000) are generally
247 designed for higher-concentration situations e.g. for NH_3 leak detection ($> 1 \text{ ppm}$). An
248 alternative to high-cost sensors is the use of passive and active samplers that can
249 accumulate NH_3 over long periods (usually between 1 week and 1 month), which enables
250 them to detect low time-averaged concentrations. Moreover, an interesting new device can
251 provide information about the location of a source without needing a power supply. This is a
252 directional passive air sampler which resolves concentrations into $12 \times 30^\circ$ sectors, and
253 combines a static carousel containing passive samplers with a rotatable inlet that aligns with
254 the wind. Passive samplers tend to be cheaper and easier to deploy than active (powered)
255 samplers, but they provide lower temporal resolution and still need laboratory analysis to
256 obtain an average value of NH_3 concentration.

257



258

259 **Figure 2.** Reported NH₃ concentrations in different environments and sensor techniques for
 260 different concentration; see Table 1 for details. The yellow arrows indicate established limits
 261 for NH₃ in air. 1) , 2) , 3), 4) , 5) , 6) , 7) , 8) , 9) , 10) , 11) , 12) , 13) , 14) .

262 **Table 1. Concentrations and limits for airborne ammonia**

263 Typical concentrations are shown for different source and receptor situations, ranging from remote background locations to near-source
 264 storage and leakage situations

| Source/Environment | Reported concentrations* | | | | Reference |
|---------------------------------------|--------------------------|-----|------------------------|-----------------------------------|-----------|
| Continental Troposphere | 0.1-1.5 | ppb | 0.13-1.9 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Lichens sensitivity | 1 | ppb | 1.3 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Rural areas | 10 | ppb | 13 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Urban areas | 10.9-15.3 | ppb | $1.4-2.0 \times 10^1$ | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Industrial areas | 20 | ppb | 2.6×10^1 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Grazed pastures | 40 | ppb | 5.2×10^1 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Odour threshold for human detection | 40 | ppb | 5.2×10^1 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Near-road | 41.3 | ppb | 5.4×10^1 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Air in fertiliser industry | 30-380 | ppb | $0.39-4.9 \times 10^2$ | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Soils after anhydrous ammonia applied | 400 | ppb | 5.2×10^2 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Property fence lines limit | 500 | ppb | 6.5×10^2 | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |
| Dairy wastewater lagoons | 0.4-1.6 | ppm | $0.52-2.1 \times 10^3$ | $\mu\text{g NH}_3 \text{ m}^{-3}$ | |

| | | | | |
|---|---------|-----|--------------------------|------------------------------------|
| Areas after manure or synthetic fertilise application | 0.01-5 | ppm | 0.01-6.5x10 ³ | µg NH ₃ m ⁻³ |
| Beef cattle housing | 4 | ppm | 5.2x10 ³ | µg NH ₃ m ⁻³ |
| Dairy Housing | 0.8-10 | ppm | 0.1-1.3x10 ⁴ | µg NH ₃ m ⁻³ |
| Hog housing | 5-28 | ppm | 0.7-3.6x10 ⁴ | µg NH ₃ m ⁻³ |
| Poultry houses | 20 | ppm | 2.6x10 ⁴ | µg NH ₃ m ⁻³ |
| Maximum exposure for humans (8hs) | 25 | ppm | 3.3x10 ⁴ | µg NH ₃ m ⁻³ |
| Area during slurry application | 60 | ppm | 7.8x10 ⁴ | µg NH ₃ m ⁻³ |
| Manure storage | 150 | ppm | 2.0x10 ⁵ | µg NH ₃ m ⁻³ |
| Area near an ammonia leak | 100-600 | ppm | 1.3-7.8x10 ⁵ | µg NH ₃ m ⁻³ |

265 *Transformation rate of NH₃ concentrations: 1 µg m⁻³ ≅ 0.774 ppb.

266 **Table 2.** Reported ammonia sensors and their measured concentrations. The table is separated in commonly reported concentration for each
 267 class and outstanding sensors.

| Technique or method based | Usual concentration | | Sample rate | Concentration of selected sensors | | Sample rate | Use | Reference |
|---------------------------|---------------------|--------------------|-------------------|-----------------------------------|-----------------|-------------|--|-----------|
| | LOD | Range | | LOD | Range | | | |
| Wet chemistry | 0.1 ppb | 0.1 ppb – 1000 ppm | > min. | 0.58 ppt | 5.5-30 ppt | - | NH ₃ concentrations below ppb levels and reference methods. | |
| Chemiluminescence | 0.2 1 ppm | 0.25-100 ppm | > min. | 60 ppb | 0.31-59.6 ppm | - | Field instrumentation for low ppb NH ₃ concentrations | |
| Passive Collectors | 0.2 ppb | 0.2 -100 ppb | > hours – months. | - | >0.2 ppb | - | NH ₃ diffusion tubes are useful for providing long-term estimates. | |
| Photoacoustic | 0.3 ppm | 0.3-10 ppm | > sec. | 0.1 ppb | 0.1 ppb - 3 ppm | 5 L/min | Laboratory instrumentation for NH ₃ concentrations below ppm levels | |
| Fluorescence | 0.5 | 0.5-50 | > min. | 30 | 30-380 | 15 | Laboratory instrumentation for NH ₃ | |

| | | | | | | | |
|-----------------|------------|---------------------|--------|------------|-----------------|---------|--|
| | ppm | ppm | | ppb | ppb | L/min | concentrations of low ppm levels |
| Absorbance | 0.5 ppm | 0.1-10 ppm | > sec. | 3 ppb | 3 ppb-10 ppm | ~6 /min | Field sensors for NH ₃ concentrations of low ppm levels |
| Electrochemical | 1 ppm | 1-50 ppm | > min. | 0.5 ppm | 0.5-5 ppm | 6 /min | Field sensors for NH ₃ concentrations of low ppm levels |
| Colorimetric | 10 ppm | 10-300 ppm | > min. | 10 ppm | 10-200 ppm | 4 /min | Field sensors for NH ₃ concentrations of low ppm levels |
| pH papers | 50 ppm | 50 - 1000 ppm | > min. | 50 ppm | 50-300 ppm | 30 /min | Field sensors for NH ₃ concentrations above 50 ppm levels |

268

LOD:

Limit

of

detection.

269 **4.1. Types of Sensors**

270 Based on the main system used to detect ammonia, there are two broad types of sensors:
271 electrochemical and spectrometric. There are also two main strategies for obtaining an
272 analytical signal from the NH_3 molecule: direct measurement of NH_3 itself, or indirect
273 measurement of a compound that derives from a reaction with NH_3 . Most methods described
274 here rely on collecting gaseous NH_3 in an acidic medium and then measuring the
275 concentration. There is a need to distinguish between gaseous and particulate ammonia.
276 The volume of air passed through a sampler is recorded and the NH_3 concentration in the air
277 is calculated.

278

279 **4.1.1. Electrochemical sensors**

280 The principle of electrochemical sensors involves detecting the change in an electrical signal
281 (resistance, potential, current) caused by adsorption of NH_3 into different materials.
282 Voltametric or potentiometric determinations were developed typically to measure NH_3 in
283 aqueous samples, for which the detection limits (~1-100 ppm) are much higher than for air
284 samples. Although amperometric NH_3 sensors offer high sensitivity, they are characterised
285 by low selectivity because the NH_3 signal cannot be readily distinguished from interfering
286 signals due to other compounds that are present. Because of this interference, these types
287 of sensors are not suitable for agricultural applications and so are not discussed further.
288 Generally, those studies that present new developments in electrochemical sensing are
289 conducted by material scientists, which could explain why their application in agriculture is
290 rarely discussed. In addition, the concentrations investigated in these studies (1-10,000
291 ppm) tend to be higher than the concentrations applicable to agricultural NH_3 situations
292 (0.01-100 ppm). Nevertheless, the chemical alarm systems used to signal potentially unsafe
293 levels of NH_3 in housing facilities do not require very sensitive sensors, so that small and
294 low-power electronic devices are feasible in these situations. Overall, semiconductor and
295 metal-oxide gas sensors seem best-suited for these chemical alarm applications.

296

297 Several new materials have been developed which can change their electrical conductivity
298 as a function of the NH_3 concentration in air. Chemi-resistive materials have been used to
299 detect a number of oxidizing or reducing gases such as NH_3 , based on the charge transfer
300 that can occur between the sensing element and the redox gas. Typically these detectors
301 incorporate semiconductors that can be categorized as metal oxide, conductive polymers, or
302 combinations of metal nanoparticles and conductive polymers that result in promising
303 composite materials. Metal oxides-based NH_3 sensors have shown good performance
304 compared to carbon materials, and are competitive in analytical performance in relation to
305 conductive polymers. The main disadvantage of metal-oxide-based technologies is the
306 requirement for activation energy to convert the low conductivity of the materials into
307 semiconductors. In metal-oxide-based sensors, the activation energy is usually supplied by
308 using temperatures above 200°C , while conductive polymers are activated through a
309 “doping” synthesis that generates delocalized charges in the sensing element.

310

311 In recent decades, polyaniline-based materials have emerged as the preferred choice
312 among polymer-based sensor materials, because of their cost-effectiveness, ease-of-
313 preparation, and superior sensing performance. Most up-to-date developments have
314 improved stability and repeatability by combining polyaniline with other NH_3 -sensing
315 materials, including combinations of polyaniline nanocomposites with metal oxides,
316 graphene, carbon nanotubes, and other carbon nanomaterials. These developments allow
317 polymer-based sensing materials to readily transduce the sorption of NH_3 into an electrical
318 signal at room temperature. Nevertheless, the mechanistic understanding of the mix of this
319 polymer with carbon nanomaterials or metal oxides remains elusive, representing a
320 challenge for the next generation of these sensors. The disadvantages of the conductive
321 material-based sensors are that they require regular maintenance and lack sensitivity and
322 selectivity. For example, conducting polymers need regular regeneration to prevent loss of
323 sensitivity. Most of these developments are not suitable for the agricultural environment
324 where other redox gases are present (N_2O , NO_2 , H_2S and CH_4). Moreover, most of these

325 sensors provide only first order data, reading only one signal value per sample, so no
326 multivariate calibration could be applied to resolve interferences . In addition, there is a need
327 to use non-linear regression models, which adds an extra layer of complexity to these
328 electrochemical determinations. A few minutes per sample is needed to achieve
329 measurements in the order of several ppm which is a high concentration for most
330 agricultural purposes, except for alarm systems in livestock housing facilities . More complex
331 methodologies could be used to address the problem of sensitivity. For example, Timmer et
332 al. (2004) used miniaturization and pre-concentration by membranes and an acid trap to
333 reach the required concentration levels (ppb). Another approach that addresses selectivity is
334 the use of indirect measurements of electrolyte conductivity detection . However, these
335 methods seem to be unsuitable for long term sensing in dusty environments such as
336 agricultural facilities, because their membranes and filters get clogged easily.

337

338 4.1.2. **Spectrometry based-sensors**

339 Optical devices based on spectrometry can meet stringent requirements for sensing, such as
340 strong resistance to electromagnetic noise, high stability, low power consumption, and
341 compatibility with explosive environments.

342

343 4.1.2.1. **Absorbance-based and colorimetric sensors**

344 The most common absorbance-based and colorimetric sensors involve direct measurements
345 using long-path instruments and NH_3 -derived compounds, and indirect measurements of
346 changes in pH. The methods have a broad range of applications, and sensor selection
347 depends mainly on the NH_3 concentrations to be measured. The various methods cover a
348 calibration range between 1 and 18,000 ppm of NH_3 , and one study reports a limit of
349 detection of 1 ppb_v NH_3 . Continuous monitoring over large areas or long paths can require
350 low detection limits, and absorbance-based sensors seem to be the best option for these
351 situations (See Table 2). Direct measurements can be done with open-path sensors at
352 infrared (IR) or ultra-violet (UV) wavelengths. The methods can be used to integrate NH_3

353 concentrations over several metres of air path up to 120 meters . Another method involving a
354 long optical path is cavity-ringdown spectroscopy which has been used successfully to
355 identify NH₃ emission sources, and when used with real-time data it can explain the
356 correlation between concentrations of NH₃ and other precursor gases for PM_{2.5} in urban air .
357 The main disadvantages of the methods are that the initial costs for equipment and set up
358 are high, that qualified personnel are needed to deploy and maintain the sensors, and that
359 interferences from dust, water vapour and other gases (e.g. NO_x, CO₂, O₂, etc.) commonly
360 occur in agricultural environments . Invalid data from long-path instruments due to dust or
361 laser reflector alignment issues could be avoided using the latest chemometrics approaches
362 that can predict values with uncalibrated interferences . Moreover, miniaturized systems
363 could solve the problem of needing large sample volumes for absorption spectroscopy, or
364 they could enhance the absorption signal by using specific and very sensitive colour
365 reactions that are already available .

366
367 Chemicals for deriving NH₃ concentrations can be held on a variety of surfaces. The most
368 commonly used methods for deriving NH₃ are the Berthelot, Nessler and pyridine-pyrazole
369 reactions, or their modifications . There are two commonly used arrangements that use
370 colour changes to infer a sample gas concentration. One arrangement uses gas tube
371 samplers that are coated inside with colour-change reagent, and are attached to a hand
372 pump that sucks a pre-defined volume of air per stroke . Tubes are available with different
373 measurement ranges e.g. a range of 0.25-30 ppm is suitable for NH₃ in livestock buildings .
374 However, the sensitivity of the tubes is generally too low (>2 ppm) for measuring outdoor
375 NH₃ concentrations. The second arrangement uses a chemcassette system where a coil of
376 paper embedded with chemicals is coupled to a measuring device . The paper acts as both
377 a sampling medium, and as an analysis facility for detecting and measuring nanogram
378 amounts of the target gas. These devices use a small length of paper tape per sample,
379 allowing scheduled automatic measurements. On exposure to the target gas, the tape
380 changes colour in direct proportion to the sample gas concentration, and a photo-optical

381 system measures the colour intensity change from which the sampled gas concentration is
382 determined .

383

384 Other thin-films samplers are reported in the literature but they are still not widely used. An
385 example is the change in optical absorption of a thin dye-doped nanostructure polypyrrole
386 film . Another sensor uses polyaniline deposited on the surface of a polyethylene tube . Also,
387 there are non-specific sensors available like pH papers that measure indirectly the presence
388 of NH_3 in air . This method provides inexpensive *in situ* records for estimating NH_3
389 concentrations by matching the colour change against a calibrated chart. Other sensors use
390 patterns of different dyes to obtain values of NH_3 and to discriminate different interferences
391 like SO_2 , Cl_2 , CO_2 or volatile amines . Chemometrics analysis is used for multivariate
392 calibration when several dyes are used at the same time . Because these indirect methods
393 have low sensitivity and precision, they could be suitable for preliminary and rapid analysis in
394 high concentration environments like slurry lagoons and housing facilities.

395

396 **4.1.2.2. Fluorescence-based sensors**

397 Fluorescence is not frequently used to measure air quality in agricultural situations because
398 few relevant compounds fluoresce, compared to those that absorb radiation . However, there
399 is a growing interest in replacing conventional organic dyes with stable semiconductor or
400 carbon-based photo-luminescent nanomaterials for fluorescence-based sensing.
401 Fluorescence sensors offer relatively high sensitivity (~50 ppb), easier design strategies,
402 lower cost, and higher portability. Photo-luminescent carbon dots (CDs) are better than
403 semiconductor nanomaterials in terms of their high biocompatibility, low toxicity and lower
404 cost . To date, fluorescent CDs have been successfully exploited in a wide range of
405 applications and a recent study uses fluorescent determination in solid and liquid samples
406 from agriculture, based on simultaneous extraction and fluorimetric labelling of NH_3 , followed
407 by fluorescence measurement. The fluorimetric labelling reagent is the o-phthalaldehyde
408 (OPA), and the reaction is based on the deprotonation of a pH indicator .

409

410 As the NH_3 molecule is not fluorescent by itself, quantifications are done using derivatization
411 reactions like that used for the sensor in section 4.1.2.1. The signal recorded is the emission
412 of photons after irradiation of the derivatization reagent. A trapping solution or new porous
413 matrices like sol-gel or latexes have been developed to obtain a fluorescent signal from the
414 non-fluorescent NH_3 molecule . Thin films involve the reversible absorption and desorption of
415 NH_3 over periods of about one minute, that change the fluorescent signal of the polymer .
416 Future developments could use such fluorescence sensitivity to measure NH_3 , and current
417 applications of fluorescence-based sensors often measure NH_3 concentrations down to
418 about 5-50 ppm.

419

420 Another fluorescent sensor uses an array of seven chemical materials with fluorescent
421 properties as the sensing elements . These kinds of materials can identify interactions
422 between molecules such as p-p molecular complex action, bond formation, acid-base
423 interactions, physical adsorption, and Van der Waal forces. The sensor elements interact
424 with specific gases that can change their fluorescent and photochemical properties, so that
425 the change in their fluorescence can be detected. This approach has been successfully used
426 in detecting NH_3 once is dissolved in water, but could also be a good starting point to obtain
427 rapid and inexpensive sensors for NH_3 in air. Also, chemometrics could be used to calibrate
428 or to discriminate contributions from other gases present in media, as it has been done with
429 colour dye arrays .

430

431 **4.1.2.3. Photoacoustic based-sensors**

432 This optical technique involves the absorption of modulated laser radiation with a specific
433 wavelength of ~1530 nm for the NH_3 molecule. The deactivation of the excited molecule via
434 collisions is converted into a modulation frequency that induces acoustic waves that can be
435 monitored with a low-noise microphone. The instrument layout consists of a laser source, a
436 resonant photoacoustic (PA) cell, a microphone to monitor the sound waves and an

437 electronic control module for laser modulation and signal processing . The PA spectroscopic
438 gas detection technique has numerous advantages, making it a very promising candidate for
439 NH₃ detection in agriculture . PA provides continuous, selective and automatic
440 measurements with outstanding long-term stability . Carbon dioxide (CO₂) and water vapour
441 are the major potential interferences, but these can be addressed by using an isotopic ¹³CO₂
442 laser with a different wavelength to suppress the signals . Although PA is a very sensitive
443 technique, it still lacks the robustness needed for use in agricultural environments because
444 the sensors have a significant cross-sensitivity to nitrous oxide (N₂O) and methane (CH₄) .
445 Moreover, electronic developments are needed to reduce the cost of the equipment (>
446 \$10,000) in order to allow large-scale use of PA cells in commercial sensors. Most
447 developments using this technique measure in the range of 0.1 ppm to 100 ppm , so it could
448 be used for monitoring NH₃ in poultry houses, dairy wastewater lagoons and at property
449 fence lines where the regulations mandate a maximum concentration of 0.5 ppm .

450

451 4.2. Use of monitoring to support emission estimates

452 4.2.1. Emission factors

453 Several approaches have been developed with different complexity levels to estimate NH₃
454 emissions over large areas. The simplest method is collecting field data through surveys and
455 multiplying each identified source by an emission factor (EF) . For example, official estimates
456 in several countries have been done using livestock numbers and amount of N fertilisers
457 sold per year, multiplying these values by an average EF for each type of livestock and
458 fertiliser . To achieve more accurate results, further developments added more variables to
459 the models because the NH₃ emission rate is affected by temperature, wind speed, air
460 humidity, soil type, slurry pH, among others . Specific EF have been developed for each
461 individual emission source with information such as manure management system, housing
462 facilities and season . The official NH₃ emission estimates in China include sources like
463 livestock excreta, fertiliser application, type of agricultural soil, nitrogen-fixing plants, crop
464 residue compost, biomass burning, urine from rural populations, chemical industry, waste

465 disposal and traffic . The EFs are disaggregated both spatially and temporally according to
466 the climatic conditions and local practices across the country. The next step increasing
467 complexity and accuracy is inferring NH₃ emissions combining survey data with atmospheric
468 dispersion models. This type of approach has demonstrated that is possible to estimate, with
469 reasonable low bias, NH₃ emissions from different sources across large areas with high
470 resolution .

471

472 **4.2.2. Emission inventories**

473 Collecting data through farm surveys make the information independent from (subjective)
474 expert judgment. The use of farm surveys to collect field data is an affordable method to
475 populate NH₃ inventories that are critical to evaluate policy changes in the agricultural sector
476 and to verify compliance to international agreements . However, these inventories suffer
477 from a number of inadequacies: i) poor spatial resolution, few measurements for large areas,
478 leading generally to overestimations for non-agricultural zones and underestimations for
479 emissions from farms; ii) poor temporal resolution, long term values underestimating the
480 fertiliser application season; and iii) EFs are not specific for each source, average default
481 values are used and several factors are left out of calculations .

482

483 **4.2.3. Monitoring networks and alternative approaches**

484 Higher spatial and temporal resolution and more reliable data are obtained adding
485 measurements collected by sensors that are used to feed dispersion models. Large-area
486 measurements are performed with commercial passive samplers , calibrated monthly
487 against NH₃ active sampling devices . An example using a combination of the dispersion
488 model Fine Resolution NH₃ Exchange (FRAME) and a network of passive diffusion samplers
489 is the NH₃ emission monitoring in the UK . FRAME describes the main atmospheric
490 processes (emission, diffusion, chemistry and deposition) taking place in a column of air
491 moving along straight-line trajectories following specified wind directions, taking into account
492 also the transboundary chemical composition imported from other areas of Europe . Adding

493 more variables to the calculations like the size and shape of the source, the locations of
494 targets, the dynamics and magnitude of each source, the meteorological conditions, the
495 background concentration and a set of passive diffusion sensors which integrate over a few
496 hours to weekly periods helped improving the accuracy of the estimates of NH_3 emissions
497 from multiple small fields located near each other . Several passive and active samplers
498 have been placed across countries to establish NH_3 monitoring networks including: the UK
499 (NAMN, National Ammonia Monitoring Network) , the Netherlands (MAN, Measuring
500 Ammonia in Nature) , Switzerland , the United States (AMoN, Ammonia Monitoring Network)
501 and Ireland . The main aim of the networks is to monitor national trends, to assess regional
502 variability and to validate model calculations.

503 Traditional monitoring networks can be expensive, thus, a solution to poor temporal and
504 spatial resolution can be crowdsourced information involving citizens in monitoring . Citizen
505 science projects have become important data sources to scientists, and an attempt to
506 estimate NH_3 concentration from butterflies abundance has been published in a recent study
507 . Others biomonitors can be used to provide valuable spatial information, like biodiversity
508 measures such as lichen surveys .

509

510 **4.2.4. Communication technologies and monitoring**

511 The national inventories of NH_3 emissions can be improved with further developments in
512 low-cost portable devices based on the techniques mentioned in the previous Section 4.1.
513 Nowadays, there is a class of wireless network technologies that enable low power
514 consumption and wide area coverage (LPWANs - Low Power Wide Area Networks) such as
515 Sigfox or LoRa technologies that fit the need of large-area deployments of NH_3 sensors.
516 Usually, these technologies have low-rate sporadic communications under unlicensed
517 frequency bands, obtaining long distance links, normally 5 km in urban or over 25 km in rural
518 or suburban areas . The use of these communication technologies together with sensitive
519 sensors could be next upgrade to national inventories and NH_3 monitoring giving a better
520 temporal resolution.

521

522 **5. Future NH₃ sensing requirements**

523 There is great potential for NH₃ sensing technology to contribute to both improving
524 inventories and to the design of emission mitigation strategies. The next generation of
525 sensors has to specifically monitor sources to avoid the harmful effects of NH₃ and to help
526 improve the efficiency in N use in the agroindustry. The deployment of appropriate sensors
527 can support data collection to be used for:

- 528 i) Improving the formulation of livestock diets. Urea excretion and subsequent NH₃
529 emissions increase with increased dietary crude protein concentration.
- 530 ii) Developing urea hydrolysis inhibitors, and investigating the effects of other mitigation
531 strategies for NH₃ emissions like soil pH control or irrigation after fertilising.
- 532 iii) Improving fertilising practices to maximize the N fertiliser value and improving
533 manure management. The magnitude of NH₃ losses depends on both weather and
534 management practices so, improving the knowledge of the factors affecting NH₃
535 emissions will help to develop better practices.
- 536 iv) Prompt decision making using real-time data, which is important for corrective actions
537 such as switching on automatic water sprays or ventilation fans when a maximum
538 threshold is reached in livestock housing.
- 539 v) Monitoring and detecting emission sources. NH₃ emitted could impair soil, air and
540 water quality by contributing to over-fertilisation, acidity and formation of particulate
541 matter (PM). Monitoring networks can generate long-term emission trends, which are
542 critical for environmental policies . Regulations could be designed for minimum
543 distances between precursor sources. Data from NH₃ monitoring networks are
544 transforming the way that we understand and model the natural environment.

545

546

547 Ammonia emissions are indeed an important problem for society, where the dominant
548 source is agriculture, creating not only an environmental problem but also nutrient losses

549 that affect the efficiency of the agroindustry. Developments in NH₃ monitoring can contribute
550 in several ways to mitigate these emissions by improving inventories to set international
551 targets and by forecasting future trajectories of the contribution from the agroindustry to
552 improve farm efficiency. Future monitoring systems built by simple and easy-to-deploy
553 sensors that sample and accumulate ammonia signals over representative periods (e.g. 1-4
554 weeks) can be used to scope farm ammonia performance in sensitive environments. These
555 systems do not need to be highly accurate but they do need to resolve individual farms or
556 significant sources e.g. passive directional samplers located up- and down- wind of intensive
557 livestock buildings. These systems should be deployable by non-specialists, who can send
558 samples to central laboratories for analysis and interpretation.

559

560 The current recommendations in the livestock industry are to avoid alkaline environments for
561 ammonium (pH >9.25), maintaining low temperature and avoiding air exchange in the
562 superficial layer of manure (NH₃ vapour equilibrium). Nevertheless, every farm has its own
563 characteristics in terms of climate, soil condition, type of manure, fertiliser applied, farm
564 facilities and equipment, etc. So, weather data acquisition is essential for interpreting air-
565 quality data, and can be either measured or extracted from numerical weather prediction
566 models. Tailored advice on manure and fertiliser management, and on livestock diets will
567 have an important impact on nutrient balances in order to avoid unnecessary costs. There is
568 also a need for activity data to be collected and reported alongside air quality data, so the
569 data can be interpreted and attributed to specific activities.

570

571 Currently and despite advances in monitoring devices, direct measurements at each
572 farmyard and plot are impractical due to the cost and labour involved. Additional research is
573 needed to extend cost-effective monitoring to large areas. An important contribution could be
574 the use of wireless technology to connect sensors to make large deployments without
575 human intervention during long periods of time. In addition, the air produced in agriculture
576 environments is a mixture of a large number of gases plus relatively high moisture content.

577 Some of the currently available sensors are prone to have severe interferences since they
578 are sensitive to water vapour and other gases than NH_3 . Therefore there is a need for new
579 measurement methods customized for agriculture environments, where interferences are
580 correctly calibrated. Future sensors should provide multivariate signals about the target
581 compounds and interferences, for the application of statistical modelling.

582

583 **6. Conclusions**

584 Agricultural emissions of NH_3 are primarily associated with livestock production where the
585 losses are notably high and there is a pressing need for new developments so that farmers
586 monitor and control NH_3 emissions from fertiliser use. New user-friendly sensors are needed
587 with enough sensitivity for monitoring low concentration (ppb). For this purpose,
588 spectroscopic sensors can provide appropriately selective and sensitive measurements
589 because the spectra from these sensors usually include information that allows interferences
590 to be removed or reduced. Also, spectroscopic sensors can be automated and miniaturized
591 to create low-cost sensors that offer real-time data for day-to-day decisions on ammonia
592 control. The key improvements required in electrochemical sensors are better sensitivity and
593 the development of sensing materials that work at room temperature. Photoacoustic sensors
594 require new advances in electronics to lower their cost. It can be concluded that there is an
595 immediate need of sensors that are more sensitive, selective, low-cost and user-friendly.
596 Sensors must be capable of dealing with the high interferences arising in agricultural
597 environments. Above all, there is an urgent need for new sensors to support the optimal use
598 of N in agriculture so that nutrient pollution in the troposphere is minimised.

599

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605

606 **Conflict of interest**

607 None.

608

609 **Disclaimer**

610 The views expressed are those of the authors and not necessarily those of the UK
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612

613 **References**

614

615 Ates, M., 2013. A review study of (bio) sensor systems based on conducting polymers. J
616 Materials Science and Engineering 33(4): 1853-1859.

617 ATSDS, 2004. Agency for Toxic Substances and Disease Registry . Public health statement
618 - Ammonia CAS 7664-41-7: 1-7.

619 Ayres, J., 2009. Long-term exposure to air pollution: effect on mortality. Didcot: Committee
620 on the Medical Effects of Air Pollutants: 1-187.

621 Azcarate, S., A. de Araújo Gomes, A. M. de la Peña and H. C. Goicoechea, 2018. Modeling
622 second-order data for classification issues: Data characteristics, algorithms, processing
623 procedures and applications. TrAC Trends in Analytical Chemistry(107): 151-168.

624 Bai, H. and G. Shi, 2007. Gas sensors based on conducting polymers. Sensors 7(3): 267-
625 307.

626 Baskar, C., N. Nesakumar, J. B. B. Rayappan and M. Doraipandian, 2017. A low power
627 ammonia sensor node embedded with a light weight non-linear analytics. *Sensors Actuators*
628 *A: Physical* 263: 357-362.

629 Battye, R., W. Battye, C. Overcash and S. Fudge, 1994. Development and selection of
630 ammonia emission factors. EPA, US. U. EPA. US, Atmospheric Research and Exposure
631 Assessments Laboratory.

632 Behera, S. N. and M. Sharma, 2010. Investigating the potential role of ammonia in ion
633 chemistry of fine particulate matter formation for an urban environment. *Science of the Total*
634 *Environment* 408(17): 3569-3575.

635 Bell, M., C. Flechard, Y. Fauvel, C. Häni, J. Sintermann, M. Jocher, H. Menzi, A. Hensen
636 and A. Neftel, 2017. Ammonia emissions from a grazed field estimated by miniDOAS
637 measurements and inverse dispersion modelling. *Atmospheric Measurement Techniques*
638 10(5): 1875-1892.

639 Bhopate, D., K. Kim, P. Mahajan, A. Gore, S. Patil and S. Majhi, 2017. Fluorescent
640 chemosensor for quantitation of multiple atmospheric gases. *Journal of Nanomed*
641 *Nanotechnology* 8(2): 1-9.

642 Bicudo, J. R., C. L. Tengman, L. D. Jacobson and J. E. Sullivan, 2000. Odor, hydrogen
643 sulfide and ammonia emissions from swine farms in Minnesota. *Proceedings of the Water*
644 *Environment Federation* 2000(3): 589-608.

645 Bittman, S., M. Dedina, C. Howard, O. Oenema and M. Sutton, 2014. Options for ammonia
646 mitigation: Guidance from the UNECE Task Force on Reactive Nitrogen, NERC/Centre for
647 Ecology & Hydrology.

648 Bittman, S. and R. Mikkelsen, 2009. Ammonia emissions from agricultural operations:
649 livestock. *Better Crops* 93(1): 28-31.

650 Bor, M., J. E. Vidler and U. Roedig, 2016. LoRa for the Internet of Things. Proceedings of
651 the 2016 International Conference on Embedded Wireless Systems and Networks (EWSN
652 '16), TU Graz, Austria.

653 Bor, M. C., U. Roedig, T. Voigt and J. M. Alonso, 2016. Do LoRa low-power wide-area
654 networks scale? Proceedings of the 19th ACM International Conference on Modeling,
655 Analysis and Simulation of Wireless and Mobile Systems, ACM.

656 Bouwman, A., D. Lee, W. Asman, F. Dentener, K. Van Der Hoek and J. Olivier, 1997. A
657 global high-resolution emission inventory for ammonia. *Global Biogeochemical Cycles* 11(4):
658 561-587.

659 Brigden, K. and R. Stringer, 2000. Ammonia and urea production: Incidents of ammonia
660 release from the Profertil urea and ammonia facility, Bahia Blanca, Argentina 2000.
661 Greenpeace reports. S. p. A. Tour. Exeter UK, Greenpeace Research Laboratories,
662 Department of Biological Sciences, University of Exeter, Exeter, UK: 1-13.

663 Brink, C. and H. Van Grinsven, 2011. Costs and benefits of nitrogen in the environment. *The*
664 *European Nitrogen Assessment: Sources, Effects and Policy Perspectives*. M. Sutton, C. M.
665 Howard, J. W. Erisman et al.: 513-540.

666 Cape, J., L. Van der Eerden, L. Sheppard, I. Leith and M. Sutton, 2009. Evidence for
667 changing the critical level for ammonia. *Environmental Pollution* 157(3): 1033-1037.

668 Carozzi, M., R. M. Ferrara, M. Acutis and R. G., 2012. Dynamic of ammonia emission from
669 urea spreading in Po Valley (Italy): relationship with nitrogen compounds in the soil.
670 Proceeding of 17th International Nitrogen Workshop. Wexford, 26-29 June 2012.

671 Changwen, D., W. Jiao, Z. Zijun, S. Yazhen and Z. Jianmin, 2015. In situ measurement of
672 ammonia concentration in soil headspace using Fourier transform mid-infrared
673 photoacoustic spectroscopy. *Journal of Pedosphere* 25(4): 605-612.

674 Chen, X., L. Lin, P. Li, Y. Dai and X. Wang, 2004. Fluorescent response of sol-gel derived
675 ormosils for optical ammonia sensing film. *Analytica Chimica Acta* 506(1): 9-15.

676 Chen, Y., Q. Zhang and D. Petkau, 2001. Evaluation of different techniques for liquid
677 manure application on grassland. *Applied Engineering in Agriculture* 17(4): 489.

678 Cho, Y. B., S. H. Jeong, H. Chun and Y. S. Kim, 2018. Selective colorimetric detection of
679 dissolved ammonia in water via modified Berthelot's reaction on porous paper. *Sensors*
680 *Actuators B: Chemical* 256: 167-175.

681 Darwish, A. M., S. Wilson, A. Balckwell, K. Taylor, S. S. Sarkisov, D. N. Patel and B. Koplitz,
682 2015. Ammonia sensor based on polymer-inorganic nano-composite thin film upconversion
683 light emitter prepared by double-beam pulsed laser deposition. *American Journal of*
684 *Materials Science* 5(3A): 8-15.

685 Das, P., J.-H. Sa, K.-H. Kim and E.-C. Jeon, 2009. Effect of fertilizer application on ammonia
686 emission and concentration levels of ammonium, nitrate, and nitrite ions in a rice field.
687 *Environmental monitoring and assessment* 154(1-4): 275-282.

688 Dasari, B., W. Taube, P. Agarwal, M. Rajput, A. Kumar and J. Akhtar, 2015. Room
689 Temperature Single Walled Carbon Nanotubes (SWCNT) Chemiresistive Ammonia Gas
690 Sensor *Sensors and Transducers* 190(7): 24-30.

691 Daughton, C., J. Cantor, M. Jones and R. Sakaji, 1982. Chapter III - Ammonia determination
692 Quantitation of oil shale wastewater quality. *A manual of analytical methods*. C. Daughton.
693 Berkeley, USA, University of California.

694 DEFRA, 2018. Code of Good Agricultural Practice (COGAP) for Reducing Ammonia
695 Emissions. London TW9 4DU, Department for Environment Food & Rural Affairs, UK
696 Government.

697 Denmead, O., J. Freney and J. Simpson, 1983. Dynamics of ammonia volatilization during
698 furrow irrigation of maize. *Soil Science Society of America Journal* 47(3)

699 Denmead, O., J. Simpson and J. Freney, 1974. Ammonia flux into the atmosphere from a
700 grazed pasture. *Science* 185(4151): 609-610.

701 Dewey, C. E., B. Cox and J. Leyenaar, 2000. Measuring ammonia concentrations in the barn
702 using the Draeger (TM) and pHydron (TM) tests. Journal of Swine Health and Production
703 8(3): 127-131.

704 Djambazov, G. and K. Pericleous, 2015. Modelled atmospheric contribution to nitrogen
705 eutrophication in the English Channel and the southern North Sea. Atmospheric
706 Environment 102: 191-199.

707 Dore, A., D. Carslaw, C. Braban, M. Cain, C. Chemel, C. Conolly, R. Derwent, S. Griffiths, J.
708 Hall and G. Hayman, 2015. Evaluation of the performance of different atmospheric chemical
709 transport models and inter-comparison of nitrogen and sulphur deposition estimates for the
710 UK. Atmospheric Environment 119: 131-143.

711 Doyle, B., T. Cummins, C. Augustenborg and J. Aherne, 2014. Ambient Atmospheric
712 Ammonia in Ireland, 2013-2014. Environmental Protection Agency of Ireland, report(193).

713 Dumitras, D., D. Dutu, C. Matei, A. Magureanu, M. Petrus and C. Popa, 2007. Laser
714 photoacoustic spectroscopy: principles, instrumentation, and characterization. Journal of
715 Optoelectronics Advanced Materials 9(12): 3655.

716 EAA, 2014. European Environment Agency. Ammonia (NH₃) emissions. European
717 Environment Agency Retrieved 02/01/2019, 2019, from [http://www.eea.europa.eu/data-and-](http://www.eea.europa.eu/data-and-maps/indicators/eea-32-ammonia-nh3-emissions-1/assessment-4)
718 [maps/indicators/eea-32-ammonia-nh3-emissions-1/assessment-4](http://www.eea.europa.eu/data-and-maps/indicators/eea-32-ammonia-nh3-emissions-1/assessment-4).

719 EAA, 2016. National Emission Ceilings Directive. European Environment Agency. Denmark,
720 European Environment Agency.

721 EPA, 2016. U.S. Environmental Protection Agency. Toxicological Review of Ammonia
722 (EPA/635/R-16/163Fa). Integrated Risk Information System. Washington, DC.

723 EUROSTAT, 2016. Agriculture, forestry and fishery statistics. Belgium, European Union,
724 2016.

725 Fan, H., Z. Peng, H. Yang and K. Zhou, 2016. A new cataluminescence-based gas sensor
726 for simultaneously discriminating benzene and ammonia. *Analytical Methods* 8(6): 1257-
727 1264.

728 Feng, L., C. J. Musto, J. W. Kemling, S. H. Lim, W. Zhong and K. S. Suslick, 2010.
729 Colorimetric sensor array for determination and identification of toxic industrial chemicals.
730 *Analytical Chemistry* 82(22): 9433-9440.

731 Fenn, L. and L. Hossner, 1985. Ammonia volatilization from ammonium or ammonium-
732 forming nitrogen fertilizers. *Advances in Soil Science* : 123-169.

733 Ferrara, R. M., M. Carozzi, P. Di Tommasi, D. D. Nelson, G. Fratini, T. Bertolini, V. Magliulo,
734 M. Acutis and G. Rana, 2016. Dynamics of ammonia volatilisation measured by eddy
735 covariance during slurry spreading in north Italy. *Agriculture, Ecosystems Environment* 219:
736 1-13.

737 Fitzpatrick, R. B., 2004. Haz-Map: information on hazardous chemicals and occupational
738 diseases. *Medical Reference Services Quarterly* 23(2): 49-56.

739 Fournier, N., V. A. Pais, M. A. Sutton, K. J. Weston, U. Dragosits, S. Y. Tang and J. Aherne,
740 2002. Parallelisation and application of a multi-layer atmospheric transport model to quantify
741 dispersion and deposition of ammonia over the British Isles. *Environmental Pollution* 116(1):
742 95-107.

743 Ganiga, M. and J. Cyriac, 2016. FRET based ammonia sensor using carbon dots. *Sensors*
744 *and Actuators B: Chemical* 225: 522-528.

745 García, M. S., R. Timmis, N. Van Dijk, J. Whyatt, I. Leith, S. Leeson, C. Braban, L.
746 Sheppard, M. Sutton and Y. Tang, 2017. Directional passive ambient air monitoring of
747 ammonia for fugitive source attribution; a field trial with wind tunnel characteristics.
748 *Atmospheric Environment* 167: 576-585.

749 Goebes, M. D., R. Strader and C. Davidson, 2003. An ammonia emission inventory for
750 fertilizer application in the United States. *Atmospheric Environment* 37(18): 2539-2550.

751 Guimard, N. K., N. Gomez and C. E. Schmidt, 2007. Conducting polymers in biomedical
752 engineering. *Progress in Polymer Science* 32(8-9): 876-921.

753 Guthrie, S., S. Giles, F. Dunkerley, H. Tabaqchali, A. Harshfield, B. Ioppolo and C. Manville,
754 2018. The impact of ammonia emissions from agriculture on biodiversity. *JOURNAL?*

755 Harper, L., V. Catchpole, R. Davis and K. Weir, 1983. Ammonia Volatilization: Soil, Plant,
756 and Microclimate Effects on Diurnal and Seasonal Fluctuations 1. *Agronomy Journal* 75(2):
757 212-218.

758 Harper, L. A., R. R. Sharpe and T. B. Parkin, 2000. Gaseous nitrogen emissions from
759 anaerobic swine lagoons: Ammonia, nitrous oxide, and dinitrogen gas. *Journal of*
760 *Environmental Quality* 29(4): 1356-1365.

761 Horswill, P., O. O'Sullivan, G. K. Phoenix, J. A. Lee and J. R. Leake, 2008. Base cation
762 depletion, eutrophication and acidification of species-rich grasslands in response to long-
763 term simulated nitrogen deposition. *Environmental Pollution* 155(2): 336-349.

764 Hu, J., L. Zhang and Y. Lv, 2018. Recent advances in cataluminescence gas sensor:
765 Materials and methodologies. *Applied Spectroscopy Reviews (VOL?)*: 1-19.

766 Huang, X., Y. Song, M. Li, J. Li, Q. Huo, X. Cai, T. Zhu, M. Hu and H. Zhang, 2012. A
767 high-resolution ammonia emission inventory in China. *Global Biogeochemical Cycles* 26(1).

768 Huszár, H., A. Pogány, Z. Bozóki, Á. Mohácsi, L. Horváth and G. Szabó, 2008. Ammonia
769 monitoring at ppb level using photoacoustic spectroscopy for environmental application.
770 *Sensors Actuators B: Chemical* 134(2): 1027-1033.

771 Ibanez, J. G., M. E. Rincón, S. Gutierrez-Granados, M. h. Chahma, O. A. Jaramillo-Quintero
772 and B. A. Frontana-Urbe, 2018. Conducting polymers in the fields of energy, environmental
773 remediation, and chemical–chiral sensors. *Chemical reviews* 118(9): 4731-4816.

774 Jalalvand, A. R., M. Roushani, H. C. Goicoechea, D. N. Rutledge and H.-W. Gu, 2018.
775 MATLAB in electrochemistry: A review. *Talanta*. INCOMPLETE

776 Jin, Z., Y. Su and Y. Duan, 2001. Development of a polyaniline-based optical ammonia
777 sensor. *Sensors and Actuators B: Chemical* 72(1): 75-79.

778 Kelleghan, D. B., E. T. Hayes, M. Everard and T. P. Curran, 2019. Mapping ammonia risk on
779 sensitive habitats in Ireland. *Science of the Total Environment* 649: 1580-1589.

780 Khan, E. A., 2016. Dap-1 ammonia tank explosion: safety and security concerns in chemical
781 process plant in Bangladesh. *International Journal of Petrochemical Science & Engineering*
782 1(1): 10-11.

783 Khan, M. A., F. Qazi, Z. Hussain, M. U. Idrees, S. Soomro and S. Soomro, 2017. Recent
784 Trends in Electrochemical Detection of NH₃, H₂S and NO_x Gases. *International Journal of*
785 *Electrochemical Science* 12(3): 1711-1733.

786 Koerkamp, P. G., J. Metz, G. Uenk, V. Phillips, M. Holden, R. Sneath, J. Short, R. White, J.
787 Hartung and J. Seedorf, 1998. Concentrations and emissions of ammonia in livestock
788 buildings in Northern Europe. *Journal of Agricultural Engineering Research* 70(1): 79-95.

789 Lakowicz, J. R., 1999. Instrumentation for fluorescence spectroscopy. *Principles of*
790 *fluorescence spectroscopy*, Springer: 25-61.

791 Lau, K. T., S. Edwards and D. Diamond, 2004. Solid-state ammonia sensor based on
792 Berthelot's reaction. *Sensors and Actuators B: Chemical* 98(1): 12-17.

793 Lavrsen Kure, J., J. Krabben, S. Vilms Pedersen, M. Carozzi and S. Sommer, 2018. An
794 Assessment of Low-Cost Techniques to Measure Ammonia Emission from Multi-Plots: A
795 Case Study with Urea Fertilization. *Agronomy Journal* 8(11): 245.

796 Le Maout, P., J.-L. Wojkiewicz, N. Redon, C. Lahuec, F. Seguin, L. Dupont, S. Mikhaylov, Y.
797 Noskov, N. Ogurtsov and A. Pud, 2018. Polyaniline nanocomposites based sensor array for
798 breath ammonia analysis. Portable e-nose approach to non-invasive diagnosis of chronic
799 kidney disease. *Sensors and Actuators B: Chemical* 274: 616-626.

800 Lei, J., C. Hou, D. Huo, Y. Li, X. Luo, M. Yang, H. Fa, M. Bao, J. Li and B. Deng, 2016.
801 Detection of ammonia based on a novel fluorescent artificial nose and pattern recognition.
802 Atmospheric Pollution Research 7(3): 431-437.

803 Li, H., C. Zhang and H. Xin, 2015. Performance of an infrared Photoacoustic single gas
804 analyzer in measuring ammonia from poultry houses. Applied Engineering in Agriculture
805 31(3): 471-477.

806 Li, X., X. Li, Z. Li, J. Wang and J. Zhang, 2017. WS2 nanoflakes based selective ammonia
807 sensors at room temperature. Sensors Actuators B: Chemical 240: 273-277.

808 Lolkema, D., H. Noordijk, A. Stolk, R. Hoogerbrugge, M. van Zanten and W. van Pul, 2015.
809 The measuring ammonia in nature (MAN) network in the Netherlands. Biogeosciences
810 12(16): 5133-5142.

811 Loubet, B., M. Carozzi, P. Voylokov, J.-P. Cohan, R. Trochard and S. Générumont, 2018.
812 Evaluation of a new inference method for estimating ammonia volatilisation from multiple
813 agronomic plots. Biogeosciences 15(11): 3439.

814 Maciak, E. and T. Pustelny, 2013. An optical ammonia (NH₃) gas sensing by means of
815 Pd/CuPc interferometric nanostructures based on white light interferometry. Sensors
816 Actuators B: Chemical 189: 230-239.

817 Manap, H., G. Dooly, S. O'Keeffe and E. Lewis, 2011. Cross-sensitivity evaluation for
818 ammonia sensing using absorption spectroscopy in the UV region. Sensors Actuators B:
819 Chemical 154(2): 226-231.

820 McGinn, S., H. Janzen, T. Coates, K. Beauchemin and T. Flesch, 2016. Ammonia emission
821 from a beef cattle feedlot and its local dry deposition and re-emission. Journal of
822 Environmental Quality 45(4): 1178-1185.

823 Meng, Z.-Y., X.-B. Xu, T. Wang, X.-Y. Zhang, X.-L. Yu, S.-F. Wang, W.-L. Lin, Y.-Z. Chen,
824 Y.-A. Jiang and X.-Q. An, 2010. Ambient sulfur dioxide, nitrogen dioxide, and ammonia at

825 ten background and rural sites in China during 2007–2008. *Atmospheric Environment* 44(21-
826 22): 2625-2631.

827 Mikkelsen, R., 2009. Ammonia emissions from agricultural operations: fertilizer. *Better Crops*
828 93(4): 9-11.

829 Miles, D., 2008. Vertical stratification of ammonia in a broiler house. *Journal of Applied*
830 *Poultry Research* 17(3): 348-353.

831 Mills, A., L. Wild and Q. Chang, 1995. Plastic colorimetric film sensors for gaseous
832 ammonia. *Microchimica Acta* 121(1-4): 225-236.

833 Misselbrook, T., S. Gilhespy, L. Cardenas, J. Willimas and U. Dragosits, 2015. Inventory of
834 Ammonia Emissions from UK, Agriculture. 2014. UK., DEFRA. Department for Environment,
835 Food and Rural Affairs.

836 Moum, S. G., W. Seltzer and T. M. Goldhaft, 1969. A simple method of determining
837 concentrations of ammonia in animal quarters. *Poultry Science* 48(1): 347-348.

838 Mount, G. H., B. Rumburg, J. Havig, B. Lamb, H. Westberg, D. Yonge, K. Johnson and R.
839 Kincaid, 2002. Measurement of atmospheric ammonia at a dairy using differential optical
840 absorption spectroscopy in the mid-ultraviolet. *Atmospheric Environment* 36(11): 1799-1810.

841 Mukhtar, S., A. Rose, S. Capareda, C. Boriack, R. Lacey, B. Shaw and C. Parnell, 2003.
842 Assessment of ammonia adsorption onto Teflon and LDPE tubing used in pollutant stream
843 conveyance.

844 Naseem, S. and A. J. King, 2018. Ammonia production in poultry houses can affect health of
845 humans, birds, and the environment—techniques for its reduction during poultry production.
846 *Environmental Science Pollution Research*: 1-25.

847 Ni, J.-Q. and A. J. Heber, 1998. Sampling and measurement of ammonia concentration at
848 animal facilities—A review. 2001 ASAE Annual Meeting, American Society of Agricultural and
849 Biological Engineers.

850 Ni, J.-Q. and A. J. Heber, 2008. Sampling and measurement of ammonia at animal facilities.
851 *Advances in Agronomy* 98: 201-269.

852 NRC, 2003. National Research Council. Air emissions from animal feeding operations:
853 Current knowledge, future needs. National Academies Press.

854 Oenema, O., H. Witzke, Z. Klimont, J. Lesschen and G. Velthof, 2009. Integrated
855 assessment of promising measures to decrease nitrogen losses from agriculture in EU-27.
856 *Agriculture, Ecosystems Environment* 133(3-4): 280-288.

857 Pan, B., S. K. Lam, A. Mosier, Y. Luo and D. Chen, 2016. Ammonia volatilization from
858 synthetic fertilizers and its mitigation strategies: a global synthesis. *Agriculture, Ecosystems*
859 *Environment* 232: 283-289.

860 Pavlovic, R. T., U. Nopmongcol, Y. Kimura and D. T. Allen, 2006. Ammonia emissions,
861 concentrations and implications for particulate matter formation in Houston, TX. *Atmospheric*
862 *Environment* 40: 538-551.

863 Phan, N.-T., K.-H. Kim, Z.-H. Shon, E.-C. Jeon, K. Jung and N.-J. Kim, 2013. Analysis of
864 ammonia variation in the urban atmosphere. *Atmospheric Environment* 65: 177-185.

865 Puchalski, M. A., M. E. Sather, J. T. Walker, C. M. Lehmann, D. A. Gay, J. Mathew and W.
866 P. Robarge, 2011. Passive ammonia monitoring in the United States: Comparing three
867 different sampling devices. *Journal of Environmental Monitoring* 13(11): 3156-3167.

868 Pushkarsky, M., M. Webber and C. Patel, 2003. Ultra-sensitive ambient ammonia detection
869 using CO₂-laser-based photoacoustic spectroscopy. *Applied Physics B* 77(4): 381-385.

870 Reche, C., M. Viana, M. Pandolfi, A. Alastuey, T. Moreno, F. Amato, A. Ripoll and X. Querol,
871 2012. Urban NH₃ levels and sources in a Mediterranean environment. *Atmospheric*
872 *Environment* 57: 153-164.

873 Reidy, B., B. Rhim and H. Menzi, 2008. A new Swiss inventory of ammonia emissions from
874 agriculture based on a survey on farm and manure management and farm-specific model
875 calculations. *Atmospheric Environment* 42(14): 3266-3276.

876 Rosenstock, T. S., E. Diaz-Pines, P. Zuazo, G. Jordan, M. Predotova, P. Mutuo, S.
877 Abwanda, M. Thiong'o, A. Buerkert and M. C. Rufino, 2013. Accuracy and precision of
878 photoacoustic spectroscopy not guaranteed. *Global Change Biology* 19(12): 3565-3567.

879 Rostami, M., S. Monaco, D. Sacco, C. Grignani and E. Dinuccio, 2015. Comparison of
880 ammonia emissions from animal wastes and chemical fertilizers after application in the soil.
881 *International Journal of Recycling of Organic Waste in Agriculture* 4(2): 127-134.

882 RoTAP., 2012. Review of Transboundary Air Pollution (RoTAP): Acidification,
883 Eutrophication, Ground Level Ozone and Heavy Metals in the UK: Summary for Policy
884 Makers, Centre for Ecology & Hydrology.

885 Sather, M. E., J. Mathew, N. Nguyen, J. Lay, G. Golod, R. Vet, J. Cotie, T. Hertel, E. Aaboe
886 and R. Callison, 2008. Baseline ambient gaseous ammonia concentrations in the Four
887 Corners area and eastern Oklahoma, USA. *Journal of Environmental Monitoring* 10(11):
888 1319-1325.

889 Schilt, S., L. Thévenaz, M. Niklès, L. Emmenegger and C. Hüglin, 2004. Ammonia
890 monitoring at trace level using photoacoustic spectroscopy in industrial and environmental
891 applications. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 60(14):
892 3259-3268.

893 Schroeck, A. M., V. Gaube, E. Haas and W. Winiwarter, 2019. Estimating nitrogen flows of
894 agricultural soils at a landscape level—A modelling study of the Upper Enns Valley, a long-
895 term socio-ecological research region in Austria. *Science of The Total Environment* 665:
896 275-289.

897 Shankar, P. and J. B. B. Rayappan, 2015. Gas sensing mechanism of metal oxides: The role
898 of ambient atmosphere, type of semiconductor and gases-A review. *Sci. Lett. J* 4: 126.

899 Sheikh, I., S. Nissa, B. Zaffer, K. Bulbul, A. Akand, H. Ahmed, D. Hasin, I. Hussain and S.
900 Hussain, 2018. Ammonia production in the poultry houses and its harmful effects.
901 International Journal of Veterinary Sciences and Animal Husbandry 3(4): 30-33.

902 Sheppard, L. J., I. D. Leith, T. Mizunuma, J. Neil Cape, A. Crossley, S. Leeson, M. A. Sutton,
903 N. van Dijk and D. Fowler, 2011. Dry deposition of ammonia gas drives species change
904 faster than wet deposition of ammonium ions: evidence from a long-term field manipulation.
905 Global Change Biology 17(12): 3589-3607.

906 Sigrist, M. W., 2015. Mid-infrared laser-spectroscopic sensing of chemical species. Journal
907 of Advanced Research 6(3): 529-533.

908 Silva, S. O., R. Magalhães, M. Marques and O. Frazão, 2016. New advances in fiber cavity
909 ring-down technology. Optics Laser Technology 78: 115-119.

910 Singles, R., M. Sutton and K. Weston, 1998. A multi-layer model to describe the atmospheric
911 transport and deposition of ammonia in Great Britain. Atmospheric Environment 32(3): 393-
912 399.

913 Sintermann, J., C. Ammann, U. Kuhn, C. Spirig, R. Hirschberger, A. Gärtner and A. Neftel,
914 2011. Determination of field scale ammonia emissions for common slurry spreading practice
915 with two independent methods. Atmospheric Measurement Techniques 4(9): 1821-1840.

916 Sommer, S. G., J. E. Olesen and B. T. Christensen, 1991. Effects of temperature, wind
917 speed and air humidity on ammonia volatilization from surface applied cattle slurry. The
918 Journal of Agricultural Science 117(1): 91-100.

919 Šraj, L. O. C., M. I. G. Almeida, S. E. Swearer, S. D. Kolev and I. D. McKelvie, 2014.
920 Analytical challenges and advantages of using flow-based methodologies for ammonia
921 determination in estuarine and marine waters. TrAC Trends in Analytical Chemistry 59: 83-
922 92.

923 Strömberg, N. and S. Hulth, 2001. An ammonium selective fluorosensor based on the
924 principles of coextraction. Analytica Chimica Acta 443(2): 215-225.

925 Sun, K., L. Tao, D. J. Miller, M. A. Zondlo, K. B. Shonkwiler, C. Nash and J. M. Ham, 2015.
926 Open-path eddy covariance measurements of ammonia fluxes from a beef cattle feedlot.
927 *Agricultural and Forest Meteorology* 213: 193-202.

928 Sutton, M., Y. Tang, B. Miners and D. Fowler, 2001. A new diffusion denuder system for
929 long-term, regional monitoring of atmospheric ammonia and ammonium. *Water, Air and Soil*
930 *Pollution: Focus* 1(5-6): 145-156.

931 Tang, Y., J. Cape and M. Sutton, 2001. Development and types of passive samplers for
932 monitoring atmospheric NO₂ and NH₃ concentrations. *The Scientific World Journal* 1: 513-
933 529.

934 Tang, Y. S., U. Dragosits, N. van Dijk, L. Love, I. Simmons and M. A. Sutton, 2009.
935 Assessment of ammonia and ammonium trends and relationship to critical levels in the UK
936 National Ammonia Monitoring Network (NAMN). *Atmospheric Ammonia*, Springer: 187-194.

937 Tang, Z., J. Yang, J. Yu and B. Cui, 2010. A colorimetric sensor for qualitative discrimination
938 and quantitative detection of volatile amines. *Sensors* 10(7): 6463-6476.

939 Tanguy, N. R., M. Thompson and N. Yan, 2018. A review on advances in application of
940 polyaniline for ammonia detection. *Sensors and Actuators B: Chemical* 257: 1044-1064.

941 Tavoli, F. and N. Alizadeh, 2013. Optical ammonia gas sensor based on nanostructure dye-
942 doped polypyrrole. *Sensors and Actuators B: Chemical* 176: 761-767.

943 The Royal Society, 2018. The impact of ammonia emissions from agriculture on biodiversity.
944 RAND Europe. S. Guthrie, S. Giles, F. Dunkerley et al. Cambridge, UK.

945 Thöni, L., P. Brang, S. Braun, E. Seitler and B. Rihm, 2004. Ammonia monitoring in
946 Switzerland with passive samplers: patterns, determinants and comparison with modelled
947 concentrations. *Environmental Monitoring and Assessment* 98(1-3): 93-107.

948 Timmer, B., W. Olthuis and A. Van Den Berg, 2005. Ammonia sensors and their
949 applications—a review. *Sensors Actuators B: Chemical* 107(2): 666-677.

950 Toda, K., J. Li and P. K. Dasgupta, 2006. Measurement of ammonia in human breath with a
951 liquid-film conductivity sensor. *Analytical chemistry* 78(20): 7284-7291.

952 UK_Government, 2019. The Environmental Permitting (England and Wales) Regulations.
953 from <http://www.legislation.gov.uk/>.

954 United Nations, E. a. S. C., 2014. Convention on Long-range Transboundary Air Pollution:
955 Guidance document on preventing and abating ammonia emissions from agricultural
956 sources.

957 USHHS, 2004. U.S. Department of Health and Human Services. Toxicological Profile For
958 Ammonia. Agency for Toxic Substances and Disease Registry. P. H. Service. US, U.S.
959 DEPARTMENT OF HEALTH AND HUMAN SERVICES.

960 Vaddella, V., P. Ndegwa, H. Joo and J. J. J. o. e. q. Ullman, 2010. Impact of separating dairy
961 cattle excretions on ammonia emissions. 39(5): 1807-1812.

962 Valente, I. M., H. M. Oliveira, C. D. Vaz, R. M. Ramos, A. J. Fonseca, A. R. Cabrita and J. A.
963 Rodrigues, 2017. Determination of ammonia nitrogen in solid and liquid high-complex
964 matrices using one-step gas-diffusion microextraction and fluorimetric detection. *Talanta*
965 167: 747-753.

966 Van Damme, M., L. Clarisse, S. Whitburn, J. Hadji-Lazaro, D. Hurtmans, C. Clerbaux and
967 P.-F. Coheur, 2018. Industrial and agricultural ammonia point sources exposed. *Nature*
968 564(7734): 99.

969 Van der Heyden, C., E. Brusselman, E. Volcke and P. Demeyer, 2016. Continuous
970 measurements of ammonia, nitrous oxide and methane from air scrubbers at pig housing
971 facilities. *Journal of Environmental Management* 181: 163-171.

972 Vantieghem, P., D. Maes, A. Kaiser and T. Merckx, 2017. Quality of citizen science data and
973 its consequences for the conservation of skipper butterflies (Hesperiidae) in Flanders
974 (northern Belgium). *Journal of Insect Conservation* 21(3): 451-463.

975 Velthof, G., C. Van Bruggen, C. Groenestein, B. De Haan, M. Hoogeveen and J. Huijsmans,
976 2012. A model for inventory of ammonia emissions from agriculture in the Netherlands.
977 *Atmospheric Environment* 46: 248-255.

978 Vieno, M., M. Heal, S. Hallsworth, D. Famulari, R. Doherty, A. Dore, Y. Tang, C. Braban, D.
979 Leaver and M. Sutton, 2014. The role of long-range transport and domestic emissions in
980 determining atmospheric secondary inorganic particle concentrations across the UK.
981 *Atmospheric Chemistry Physics* 14(16): 8435-8447.

982 Waldrip, H., N. Cole and R. Todd, 2015. Nitrogen sustainability and beef cattle feedyards: II.
983 Ammonia emissions. *The Professional Animal Scientist* 31(5): 395-411.

984 Wang, S., J. Nan, C. Shi, Q. Fu, S. Gao, D. Wang, H. Cui, A. Saiz-Lopez and B. Zhou, 2015.
985 Atmospheric ammonia and its impacts on regional air quality over the megacity of Shanghai,
986 China. *Scientific Reports* 5: 15842.

987 Wathes, C. M., T. G. Demmers and H. Xin, 2003. Ammonia concentrations and emissions in
988 livestock production facilities: guidelines and limits in the USA and UK. 2003 ASAE Annual
989 Meeting, American Society of Agricultural and Biological Engineers.

990 Webber, M. E., T. MacDonald, M. B. Pushkarsky, C. K. N. Patel, Y. Zhao, N. Marcillac and
991 F. M. Mitloehner, 2005. Agricultural ammonia sensor using diode lasers and photoacoustic
992 spectroscopy. *Measurement Science Technology* 16(8): 1547.

993 Weeser, B., J. S. Kroese, S. Jacobs, N. Njue, Z. Kemboi, A. Ran, M. Rufino and L. Breuer,
994 2018. Citizen science pioneers in Kenya—A crowdsourced approach for hydrological
995 monitoring. *Science of The Total Environment* 631: 1590-1599.

996 Wilson, L., P. Bacon, J. Bull, U. Dragosits, T. Blackall, T. Dunn, K. Hamer, M. Sutton and S.
997 Wanless, 2004. Modelling the spatial distribution of ammonia emissions from seabirds in the
998 UK. *Environmental Pollution* 131(2): 173-185.

999 Wolseley, P. A., P. W. James, M. R. Theobald and M. A. Sutton, 2006. Detecting changes in
1000 epiphytic lichen communities at sites affected by atmospheric ammonia from agricultural
1001 sources. *The Lichenologist* 38(2): 161-176.

1002 Zhang, D., Z. Wu, P. Li, X. Zong, G. Dong and Y. Zhang, 2018. Facile fabrication of
1003 polyaniline/multi-walled carbon nanotubes/molybdenum disulfide ternary nanocomposite and
1004 its high-performance ammonia-sensing at room temperature. *Sensors and Actuators B:
1005 Chemical* 258: 895-905.

1006 Zhang, Y. and L.-T. Lim, 2018. Colorimetric array indicator for NH₃ and CO₂ detection.
1007 *Sensors and Actuators B: Chemical* 255: 3216-3226.

1008 Zhao, L., R. Manuzon, M. Darr, H. Keener, A. J. Heber and J.-Q. Ni, 2009. Ammonia
1009 emissions from a commercial poultry manure composting facility. *Livestock Environment VIII,*
1010 *31 August–4 September 2008, Iguassu Falls, Brazil, American Society of Agricultural and
1011 Biological Engineers.*

1012

1013