



Title	Ammonia emissions from agriculture and their contribution to fine particulate matter: A review of implications for human health
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1 **Title:** Ammonia emissions from agriculture and their contribution to fine particulate matter: A
2 review of implications for human health

3

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20 **Abbreviations**

21	NH ₃	Ammonia
22	PM _{2.5}	Fine Particulate Matter
23	COPD	Chronic Obstructive Pulmonary Disorder
24	EDGAR	Emissions Database for Global Atmospheric Research
25	CAA	Clean Air Act
26	US	United States
27	O ₂	Oxygen
28	H ₂ O	Water
29	CO ₂	Carbon Dioxide
30	N ₂	Nitrogen Gas
31	N ₂ O	Nitrogen Dioxide
32	CERCLA	Comprehensive Environmental Response, Compensation and Liability Act
33	BAT	Best Available Techniques
34	IPPC	Integrated Pollution Prevention and Control
35	IED	Industrial Emissions Directive
36	K	Kelvin (Degrees)
37	NH ₄ ⁺	Ammonium
38	NH ₄ NO ₃	Ammonium Nitrate
39	(NH ₄) ₂ SO ₄	Ammonium Sulphate

40	NH ₄ CL	Ammonium Chloride
41	NEC	National Emissions Ceiling Directive
42	EPA	Environmental Protection Agency
43	WHO	World Health Organisation
44	ALRI	Acute Lower Respiratory Illness
45	CEV	Cerebrovascular Disease
46	IHD	Ischemic Heart Disease
47	LC	Lung Cancer
48	O ₃	Ozone

49 **Abstract**

50 Atmospheric ammonia (NH₃) released from agriculture is contributing significantly to
51 acidification and atmospheric NH₃ may have on human health is much less readily available.
52 The potential direct impact of NH₃ on the health of the general public is under-represented in
53 scientific literature, though there have been several studies which indicate that NH₃ has a direct
54 effect on the respiratory health of those who handle livestock. These health impacts can include
55 a reduced lung function, irritation to the throat and eyes, and increased coughing and phlegm
56 expulsion. More recent studies have indicated that agricultural NH₃ may directly influence the
57 early on-set of asthma in young children. In addition to the potential direct impact of ammonia,
58 it is also a substantial contributor to the fine particulate matter (PM_{2.5}) fraction (namely the US
59 and Europe); where it accounts for the formation of 30% and 50% of all PM_{2.5} respectively.
60 PM_{2.5} has the ability to penetrate deep into the lungs and cause long term illnesses such as
61 Chronic Obstructive Pulmonary Disease (COPD) and lung cancer. Hence, PM_{2.5} causes
62 economic losses which equate to billions of dollars (US) to the global economy annually. Both
63 premature deaths associated with the health impacts from PM_{2.5} and economic losses could be
64 mitigated with a reduction in NH₃ emissions resulting from agriculture. As agriculture
65 contributes to more than 81% of all global NH₃ emissions, it is imperative that food production
66 does not come at a cost to the world's ability to breathe; where reductions in NH₃ emissions
67 can be easier to achieve than other associated pollutants.

68 **Keywords:** NH₃, PM_{2.5}, Particulate Matter, Asthma, Respiratory Health, Animal Production.

69 **1. Introduction**

70 Ammonia (NH₃) is a reduced form of nitrogen which is released to the atmosphere (Behera et
71 al., 2013; Doyle et al., 2017; Sutton et al., 2008), with agriculture contributing over 81% of its
72 total global emissions (Van Damme et al., 2021). Figure 1 displays ammonia emissions across
73 the world from various sources, highlighting the high spatial variation of NH₃ globally
74 (EDGAR, 2015). Areas which are orange to red in colour (such as can be seen across Europe,
75 the Eastern United States, China, and India) emit high levels of NH₃ (greater than 16 tonnes of
76 ammonia per year per grid cell).

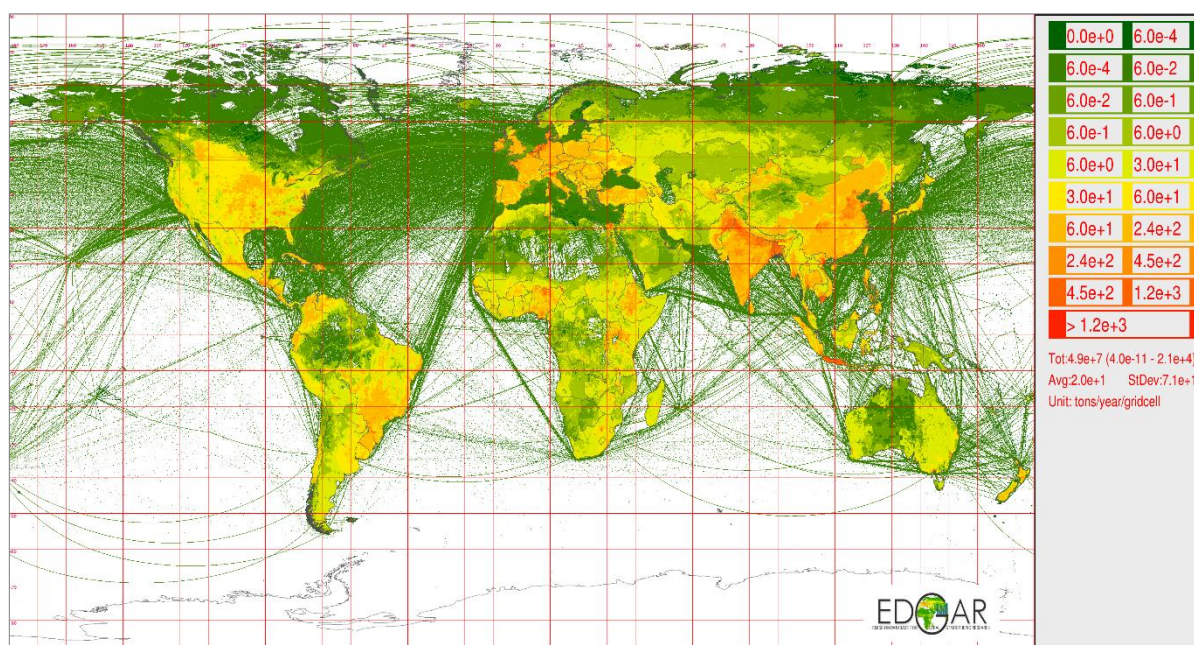


Figure 1. Spatial distribution of global ammonia emissions from various sources (EDGAR 2015). Emissions are represented as tonnes of NH₃ emitted per year (per grid cell). Areas with higher emission values are represented with an orange to red colouring e.g. Europe, Eastern US and India.

77 Environmental impacts of atmospheric NH₃ are well documented in scientific literature,
78 resulting primarily from the acidification and eutrophication of ecosystems, typically resulting
79 in species community changes (García-Gómez et al., 2014; Sutton et al., 2009). Currently, the
80 known impacts which NH₃ may have on biodiversity (particularly in sensitive habitats) are a
81 key component in the development of various national and regional policies (e.g. EPA, 2021;

82 Luesink and Michels, 2018; Shropshire Council, 2018) to better protect the environment from
83 pollution, and to comply with the Habitats Directive (92/43/EEC) (European Economic
84 Committee, 1992). In comparison to these well documented environmental impacts, and the
85 resulting policies created from such, there has been relatively little work on the potential impact
86 agricultural NH₃ may have on human health.

87 An increase in large-scale intensive agricultural operations such as concentrated animal feeding
88 operations (Schultz et al., 2019) has contributed to increasing NH₃ emissions, and the potential
89 effect of this pollutant on human health has been raised as a concern (Ge et al., 2020; Naseem
90 & King, 2018). NH₃ emissions from agriculture are responsible for 30% of all PM_{2.5} in the US
91 and Ganga Basin of India, 50% in Europe, and between 15 – 23% in China (Bauer et al., 2016;
92 Behera et al., 2010; Erisman & Schaap, 2004; Han et al., 2020). PM_{2.5} exposure can cause
93 illnesses such as chronic obstructive pulmonary disorder (COPD), lung cancer, and has even
94 been documented as a leading risk factor to premature mortality (Apte et al., 2018; Lelieveld
95 et al., 2015; Yu et al., 2000). Many precursors to PM_{2.5} are well regulated, such as nitrogen
96 oxide (NO_x) and sulphur dioxide (SO₂), though studies have shown that NH₃ is the precursor
97 which has the biggest impact on the formation of this pollutant (Brunekreef et al., 2015;
98 Megaritis et al., 2013; Thakrar et al., 2020). Therefore, it is imperative that there is an
99 understanding of how agricultural NH₃ is emitted to the atmosphere. Without understanding
100 the beginning of the chain from NH₃ sources to health impacts of PM_{2.5}, optimum reductions
101 in health impacts from this pollutant will not be reached. Knowing the extent at which
102 agriculture contributes to both NH₃ emissions and the generation of PM_{2.5} can allow for a more
103 thorough examination of the potential impacts to human health which agricultural NH₃ may
104 cause, and how these health impacts may be reduced by controlling emissions from the source.
105 This review aims to provide an understanding of both the root cause of NH₃ emissions (i.e.,
106 agriculture) and the public and socio-economic (i.e., public health) impacts this can have due

107 to PM_{2.5} formation. By focusing on these multiple links of the “NH₃ – PM_{2.5} – health impacts”
108 chain, readers may better understand the full cycle, and how source reduction of NH₃ can lessen
109 health impacts.

110 **2. Methodology**

111 The aim of this systematic literature review is to provide the reader with a clear understanding
112 of the processes involved in the release of NH₃ from agriculture, the contribution of NH₃ to
113 PM_{2.5} formation, and the resulting impacts to human health. In order for sources to be included
114 in this review, they had to meet certain criteria such as:

- 115 1. Is the article peer reviewed, or is the information from a reliable source such as
116 government or health organisations?
- 117 2. Does the article/information fall in line with the purpose of this review article?
- 118 3. Is there sufficient data provided in quantitative analysis to be representative/statistically
119 accurate?
- 120 4. Is the demographic used for qualitative analysis appropriate and representative?
- 121 5. How current or up to date is the source? If the source is not current (i.e., greater than
122 10 years old), is it important for the aim of the review or does it provide a foundational
123 concept?

124 The information provided within this review was sourced using multiple databases. Google
125 Scholar and Science Direct were used initially to gain an understanding of both the extent and
126 availability of peer reviewed research articles pertaining to this review. Descriptive search
127 terms such as *ammonia*, *particulate matter*, *PM_{2.5}*, *health impacts*, *human health*, *asthma*,
128 *COPD*, *agriculture*, *atmospheric ammonia*, *deposition and transformation*, *toxicology*, *social*
129 *economic costs*, and *abatement* were used in various searches. These keywords were combined
130 with “AND” and “OR” commands as appropriate to search a wide range of articles across the
131 search engines mentioned, as well as (but not limited to) PubMed Central (PMC), My Science
132 Work, ORCID, and internet searches using Google. Any sources which were obtained as a
133 result of an internet search were written by professionals and published in journals, government

134 agencies or health organisations. As well as using search engines, references of particular
135 interest in some articles used in the review were also studied. From these searches, 596 papers
136 were identified to meet the search criteria outlined previously. In order for a source to be
137 included in the final review, the study or information was required to include both agriculture
138 and NH₃ emissions, interactions with PM_{2.5} and subsequent health impacts, and/or data
139 pertaining to such. Many of the studies which were screened were not used in the review
140 although they met the initial search criteria, as their primary investigation did not provide
141 clarity to the subject at hand (e.g., papers which primarily focused on emission data from one
142 livestock facility at one location). In total, 135 of these sources were investigated and utilised
143 for this review. There were no instances of duplicate papers found during the inclusion process.

144

145 **3. Sources of Ammonia in Agriculture**

146 Agriculture is considered as the dominant source of atmospheric ammonia, contributing to over
147 81% of global NH₃ emissions (Van Damme et al., 2021). The primary sources of NH₃
148 emissions from agriculture include livestock and animal production, manure handling and
149 storage, livestock housing and the application of manure/slurry and artificial fertilizers to land
150 (Behera et al., 2013; Mikkelsen et al., 2011; Sutton et al., 2013) (Figure 2).

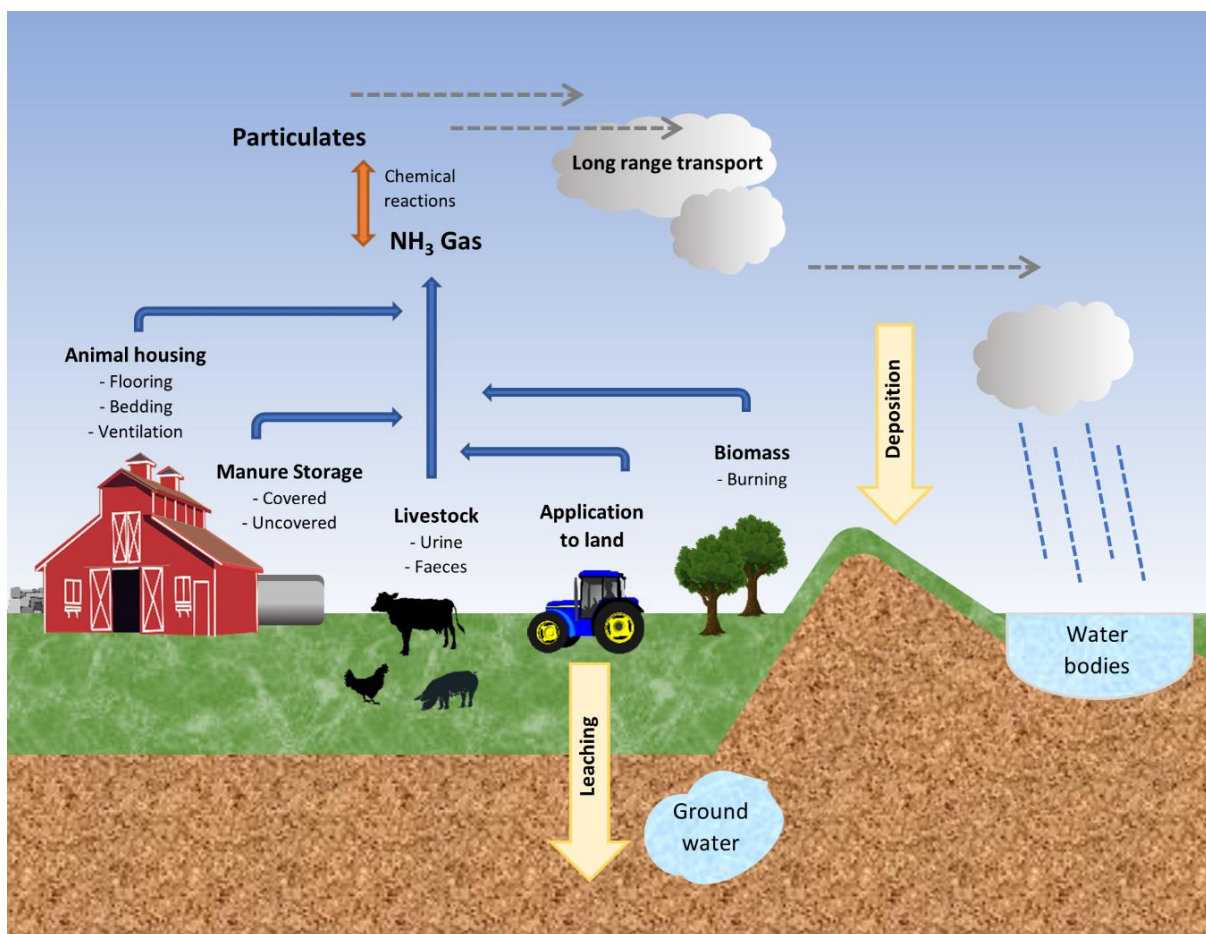


Figure 2. Potential sources of NH₃ from agriculture and routes of transport

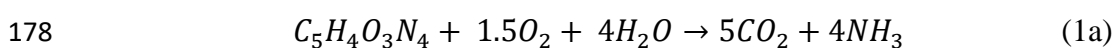
151 NH₃ is regulated to some extent under EU law where the National Emissions Ceilings (NEC)
152 Directive (2016/2284) (European Union, 2016) requires reporting of NH₃ emissions and
153 monitoring its concentration on sensitive habitats, the Habitats Directive (92/43/EEC)
154 (European Economic Committee, 1992) which requires projects to limit contributions to

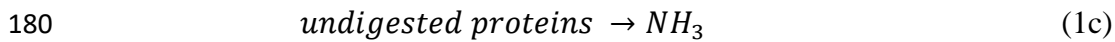
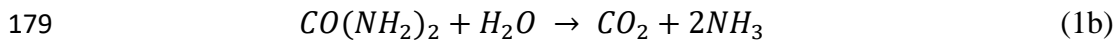
155 sensitive habitats, and the Industrial Emissions Directive (2010/75/EU) (European Union,
156 2010) which limits emissions from certain facilities.

157 The source contribution of each agricultural activity varies greatly based on national, regional
158 and local agricultural practices (Balasubramanian et al., 2020; Singles et al., 1998). Emission
159 factors are used to estimate the emission of pollutants, and are used by regulatory bodies to
160 assess emissions from various pollution sources (Faulkner & Shaw, 2008). Emission factors
161 for NH₃, which are widely used to estimate emissions from agricultural sources, are not capable
162 of accurately reflecting the temporal and spatial variations in emissions (Faulkner & Shaw,
163 2008). This is not only because of differences in agricultural practices, but also as a result of
164 variations in environmental, meteorological, and climatic conditions (Balasubramanian et al.,
165 2020; Sutton et al., 2013; Zhu et al., 2015). Von Bobruzki et al., (2010), Behera et al., (2013),
166 and Sutton et al., (2013) all provide excellent synopses highlighting the difficulties that are
167 faced with the measurement and modelling of NH₃ emissions (and concentrations) in relation
168 to agricultural sources.

169 **3.1. Livestock**

170 Excreta from livestock, including uric acid, urea and faeces, can be decomposed or volatilized
171 to form NH₃ (Behera et al., 2013). There are several microbiological processes involved in the
172 degradation of these compounds to NH₃. Uric acid in the presence of oxygen (O₂) and water
173 (H₂O) is converted to carbon dioxide (CO₂) and NH₃ by the enzyme uricase (equation 1a). Urea
174 is degraded to CO₂ and NH₃ by the enzyme urease, which is produced by various microbes
175 often present in manure (equation 1b). Undigested proteins present in manure are converted to
176 NH₃ by both the enzymes uricase and urease, as well as the action of bacterial metabolism
177 (equation 1c).





181 **Equation 1.** The process involved in ammonia production from uric acid (a), urea (b), and undigested proteins
182 (c) (Behera et al., 2013).

183 Most of the urine that is excreted onto the floor of animal housing is mineralised to ammonium
184 (NH_4^+) or transformed to NH_3 . However, this urine can also combine with faeces in animal
185 houses such as those with slat-based or solid floor systems to produce slurry (Mendes et al.,
186 2017). The resulting slurry can release gaseous emissions by bacterial degradation and
187 enzymatic reactions, resulting in the release of nitrogen gas (N_2), nitrogen dioxide (N_2O) and
188 NH_3 . Many different factors can influence gas formation and volatilization, in turn impacting
189 both the emission of NH_3 from animal houses and the resulting surrounding atmospheric
190 concentrations of this gas. These factors are mainly related to animals (e.g. genetics, diet,
191 number and weight, animal activity, and behaviour), wastes (e.g., handling, treatment, pH,
192 temperature, and surface area), environment (e.g., indoor and outdoor temperature, ventilation
193 flow, and air velocity over the manure surface) and other site-specific factors, such as the
194 presence and type of bedding materials (Blanes-Vidal et al., 2008). Cattle contribute to around
195 51% of all European NH_3 emissions, and over half of the total 80% of NH_3 emissions released
196 from agriculture in the US (CLRTAP, 2020; Liu et al., 2017). In 2014, cattle contributed to
197 approximately 44% of total manure nitrogen production globally (Zhang et al., 2017).
198 Approximately 15% of all livestock NH_3 emissions can be attributed to pig production globally
199 (Philippe et al., 2011), while poultry accounts for around 13% of global NH_3 emissions (Crippa
200 et al., 2016; Jiang et al., 2020).

201 Housing practices for livestock vary internationally, and NH_3 emissions from such are heavily
202 dependent on the indoor and outdoor temperatures, ventilation rates, and exposure of excreta
203 to the air (e.g. Blanes-Vidal et al., 2008; Blunden and Westerman, 2008; Granier et al., 1996;

204 Philippe et al., 2011). Describing housing systems broadly, cattle housing for example can
205 utilise slatted floors with slurry collection beneath (e.g., Europe and China), solid scraped
206 floors, and/or tie stalls (e.g., US) (Robbins et al., 2019; Wang et al., 2018). Cattle can also be
207 housed in naturally ventilated buildings, either year round or over the winter period with a
208 grazing period in summer (McIlroy et al., 2019). Housing in pig production typically uses both
209 concrete and slatted floors (with associated slurry storage tanks below). Poultry production
210 units can have a number of housing styles, including high-rise cage-based systems with manure
211 stored beneath cages, manure belt systems, cage free housing, or free range (Naseem & King,
212 2018).

213 Changes to housing, including ventilation rates, bedding, flooring, temperature, and manure
214 storage can all aid in mitigating emissions from this source. For livestock buildings, air
215 treatment systems (e.g. biofilters, bio-scrubbers, activated carbon filters) are considered as Best
216 Available Techniques (BAT) by the EU (Santonja et al., 2017). The incorporation of acid
217 scrubbers and bio-trickling filters in housing units can be highly effective at removing NH₃,
218 with efficiencies in the range of 91% to 99% and about 70%, respectively (Melse & Ogink,
219 2005). However, because of economic (high running costs), environmental (high energy
220 consumption, chemical use, discharge water), and technical reasons (unstable performance of
221 bio-scrubbers) these techniques are only considered suitable for intensive pig and poultry
222 livestock installations under the IPPC Directive (2008/1/EC) (now the IED (2010/75/EC))
223 (European Union, 2010; Melse et al., 2009).

224 The common slurry based, slatted system used in many cattle production facilities can benefit
225 from the introduction of straw bedding, reducing NH₃ emissions by up to 30% (Webb et al.,
226 2005). This is due to the absorption of urine into the straw bed, thus reducing the interaction of
227 NH₃ with surrounding air. However, the addition of straw can make slurry removal more
228 difficult. Implementing abatement techniques such as decreasing manure surface area,

229 introducing rapid removal of urine, and decreasing air velocity and temperature above manure
230 pits could aid in reducing NH₃ emissions from cattle houses (Hellsten, 2017). Reducing the
231 area of slatted floor, particularly in pig housing, can reduce the area in which the animals
232 defecate. In most cases, housed pigs will choose to defecate on slatted floors (cooler areas) as
233 opposed to solid floors (warmer areas where pigs choose to lay) (Aarnink et al., 1997; Ye et
234 al., 2009). These studies found that decreasing the area of slatted floor within a pig house can
235 reduce daily NH₃ emissions by 0.7g of NH₃ per fattening pig. Designing new (or re-designing
236 old) houses to have partially slatted or bedded floors could reduce NH₃ emissions by up to 20%
237 for a cost of €0 - €3 per kilogram of NH₃-N reduced (Hellsten, 2017).

238 **3.2. External slurry storage**

239 Animal wastes can be stored as slurry which combines the liquid (urine) and solid (faeces)
240 fractions, or separated into its individual liquid and solid components (Grant & Boehm, 2020).
241 Once slurry is removed from animal houses, it can be stored in a number of ways including,
242 for example, silos (concrete, steel or wooden tanks), and lagoons (Webb et al., 2005). The
243 surface area of these storage facilities plays a vital role in the level of NH₃ emitted from the
244 manure. Lagoons tend to have a higher surface area to volume ratio than silos, which entails a
245 greater emission potential when compared to tank-style storage systems (Webb et al., 2005).
246 Though lagoons also have a higher emission rate per unit area, where Kupper et al. (2020)
247 states that the baseline emissions for slurry storage are:

- 248 • 0.12 g m⁻² h⁻¹ (cattle slurry) and 0.15 g m⁻² h⁻¹ (pig slurry) stored in lagoons
- 249 • 0.08 g m⁻² h⁻¹ (cattle slurry) and 0.24 g m⁻² h⁻¹ (pig slurry) stored in tanks

250 The most effective means of reducing NH₃ emissions from storage areas is through fitting
251 covers and canopies, which can reduce emissions by up to 80% (Webb et al., 2005). Such a
252 reduction strategy was assessed for China by Xu et al. (2017), showing a country-wide

253 reduction capacity between 19 – 37% when compared to the “business as usual” emission
254 levels. The method of covered manure storage is most effective for circular tanks which are
255 made of a non-porous material such as steel (Webb et al., 2005). The use of biological
256 mechanisms can also assist in reducing NH₃ emissions when covering slurries. Blanes-Vidal
257 et al. (2009) assessed the use of aged straws for slurry cover (which contains a higher
258 microbiota than fresh straw), showing that this method had a 99% efficiency in reducing NH₃
259 emissions. This is compared to a 47% efficiency in reduction using new straw covers. These
260 authors also state that 50% of this reduction efficiency corresponds to the straw cover acting
261 as a biofilter, whereas the remaining 50% is associated with physical and chemical effects.
262 Hellsten (2017) stated that there are minimal measures deemed applicable to reduce NH₃
263 emissions from solid livestock waste systems. Covering the solid waste, applying some form
264 of litter, and keeping the waste dry are currently the only feasible options for manure
265 management in solid waste.

266 **3.3. Land application**

267 Ammonia emissions during and after land application of animal wastes (both as liquid slurries
268 and as solid manure) and synthetic nitrogenous fertilizers, contributes to a significant
269 proportion of the total NH₃ emissions from agriculture (Misselbrook et al., 2005). A review of
270 324 studies by Ma et al. (2021) estimated that emissions of NH₃ from manure application were
271 3.79 Tg N/year, and for synthetic fertilizer application were 12.32 Tg N/year. Ma et al. (2021)
272 also describes that China, India, and the US produce 60% of total NH₃ emissions associated
273 with this synthetic fertilizer application.

274 One of the most important factors in NH₃ emissions from land application is the method of
275 how manure is spread on the fields (Bourdin et al., 2014; Edwards, 2020; Hyde et al., 2003).
276 Currently, broad spreading (e.g., splash plate) application is the cheapest method of land-
277 spreading but is associated with the highest levels of NH₃ emissions compared to any other

278 approach (Edwards, 2020). NH_3 volatilization from nitrogen fertilizer or slurry/manure
279 application also depends strongly on localized environmental and meteorological parameters,
280 such as daily maximum and minimum temperature, radiation, precipitation, relative humidity,
281 and wind velocity at 10 m height, as well as soil moisture content, dry matter content of the
282 slurry/manure, and the leaf area of the crop/product (e.g. Anderson et al., 2003; Bash et al.,
283 2013; Fu et al., 2015; Häni et al., 2016; Misselbrook et al., 2005).

284 By applying alternative techniques of manure spreading such as shallow injection, band
285 application, or direct incorporation, an emissions reduction potential of approximately 95%
286 could be reached (Hellsten, 2017). For mineral fertilizer application, switching from urea to
287 other fertilizer types (such as calcium ammonium nitrate), or the use of urease inhibitor (e.g.
288 protected urea), could also substantially reduce NH_3 emissions (Forrestal et al., 2016; Lichiheb
289 et al., 2019).

290 **3.4. Meteorological and seasonal variations**

291 Alongside temperature, humidity, and other meteorological conditions, Jiang et al. (2020)
292 states that rain is one of the primary driving forces when it comes to the application of manure
293 or fertilizer to land, with semi-dry tropic regions presenting an emission rate five times greater
294 than that of colder, wet climates. In these humid or tropical areas, the emission of NH_3 from
295 controlled poultry houses is three times greater than that of dry, cold locations. In many
296 European countries, application of manure and fertilizer to land coincides with the dry seasons,
297 thus resulting in higher emissions from this source (land application) during summer periods
298 (Backes et al., 2016). Seasonal patterns in atmospheric NH_3 variations were noted by Doyle et
299 al. (2017), referencing several studies which all experienced similar patterns dependent on both
300 weather, climate, and agricultural activities (Tang et al., 2004; Zbieranowski & Aherne, 2012).
301 Temperature and climate also have an impact on biomass burning, which although is not the
302 most prominent source of agricultural NH_3 in the environment, still plays an important role in

303 emissions (Bray et al., 2021). Biomass burning in the US accounts for approximately 10% of
304 NH₃ emissions (with agriculture accounting for approximately 82% of this) and usually occurs
305 mostly in the summer and autumn (Bray et al., 2021). Across the globe, there is a high
306 variability of both the frequency and intensity of these fires (Oenema et al., 2012).

307 The volatilization of NH₃ following land application has been shown to be significantly
308 correlated to meteorological and soil factors such as air and soil temperature, humidity, solar
309 radiation, rainfall and wind speed at the time of, and after, application (e.g. Braschkat et al.,
310 1997; Générmont and Cellier, 1997; Menzi et al., 1998; Misselbrook et al., 2005; Moal et al.,
311 1995; Søggaard et al., 2002; Sommer and Hutchings, 2001). In Blanes-Vidal et al. (2012)
312 seasonal NH₃ concentrations measured at five rural locations during spring and summer
313 seasons were $85 \pm 25\%$ and $20 \pm 26\%$ higher than annual averages at each region (mean \pm
314 standard deviation), while NH₃ concentrations during autumn and winter were lower than
315 annual averages ($-47 \pm 2\%$ and $-58 \pm 10\%$, respectively). Similar patterns were found by
316 Skjøth et al. (2011) which determined the annual course of NH₃ emissions for a site in
317 Germany. A year of NH₃ monitoring carried out in Ireland identified a Pearson correlation of
318 0.9 with ambient temperature, with peak concentrations observed in June, showing trends
319 coinciding with seasonally variable agricultural practices (Kelleghan et al., 2021). As well as
320 seasonal variation, diurnal variation in emissions also occur as a result of indoor temperature
321 and animal activity in-house (Kelleghan et al., 2020; Schauburger et al., 2018; Sun et al., 2008).
322 As this variation in NH₃ emissions and concentrations occurs both seasonally and diurnally, it
323 highlights the need for intensive long term monitoring of both.

324 Ammonia emissions have also been shown to be correlated to changes in climate e.g. global
325 emissions increase of 42% for a warming of 5°C (Sutton et al., 2013). These long-term trends
326 and projections, known as climate signals, have epidemiological relevance for human health.
327 This means that expected global warming is counteracting the efforts to reduce NH₃ emissions.

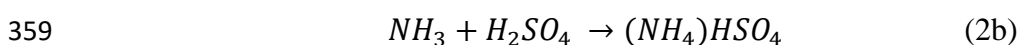
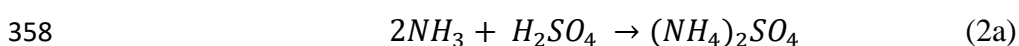
328 For the 2050s, NH₃ emissions from confined livestock buildings, predominantly used for
329 fattening pigs and poultry, are expected to increase by about 15 - 20% (relative to 2007) due to
330 the increase of temperature (Simpson et al., 2014). The sensitivity to global warming for the
331 investigated agricultural emission categories were assessed by Skjøth & Geels (2013), showing
332 the highest sensitivity for livestock buildings and manure storage, manure handling, application
333 of fertilizer, and grazing animals. Skjøth & Geels (2013) expect an emission increase up to
334 40% due to the global warming signal. Under the assumption of a constant linear trend of
335 anthropogenic warming until 2050, NH₃ emissions from livestock buildings will increase by
336 about 11% between 1981 and 2050 from livestock buildings. For the last four decades, a mean
337 trend of a 1.6% increase per decade was determined for confined livestock buildings
338 (Schauberger et al., 2018). For the storage of manure, Aarnink & Elzing found that a 1 Kelvin
339 (K) increase in storage temperature would result in a 10% increase in the emission rate of NH₃
340 (Aarnink & Elzing, 1998). This emphasises the urgent need to reduce emissions as much as
341 possible, as increasing emissions in the future due to climate change will exacerbate any
342 associated environmental and human health impacts.

343 **4. The Fate of Ammonia and Potential Health Implications**

344 **4.1. The transformation of NH₃ to PM_{2.5}**

345 Once NH₃ is released from agricultural sources, it can then travel in a gaseous form through
346 the atmosphere for short or long distances (Philippe et al., 2011). When NH₃ interacts with a
347 surface, it adheres and is removed from the atmosphere (i.e. dry deposition to plant surfaces),
348 or following chemical transformation, it is deposited remotely through rainfall (wet deposition)
349 (Asman et al., 1998). The rate of both dry and wet deposition is dependent on a number of
350 factors including meteorological conditions, the physical and chemical properties of the
351 pollutant and surrounding surface conditions, and concentrations of other atmospheric
352 pollutants (Doyle et al., 2017).

353 Chemical reactions in the atmosphere involving NH₃ contribute significantly to the generation
354 of PM_{2.5} (Giannakis et al., 2019). Gaseous NH₃ reacts with aerosols containing sulphuric and
355 nitric acids to create particulates such as ammonium nitrate (NH₄NO₃), ammonium sulphate
356 ((NH₄)₂SO₄), and ammonium chloride (NH₄CL) (Gong et al., 2013). Bauer et al. (2016)
357 explains how phase partitioning between inorganic aerosols is driven primarily by NH₃.



361 **Equation 2.** The conversion of NH₃ to ammonium sulphate and ammonium nitrate (Mensink & Deutsch, 2008)

362 Gaseous NH₃ is highly reactive in the atmosphere and is deposited quickly, with a deposition
363 velocity reaching 14 cm/second (Phillips et al., 2004). High concentrations of NH₃ deposited
364 locally can have serious effects on foliage and vegetation (Doyle et al., 2017; Krupa, 2003). In
365 comparison, resulting NH₄⁺ is deposited at a much slower rate, usually through wet deposition,

366 at a rate of approximately 0.2 cm/second (Asman et al., 1998; Phillips et al., 2004). This
367 transformation of gaseous NH₃ to NH₄⁺ aerosols can allow for this pollutant to move much
368 further from the point source, and be deposited through wet deposition (Phillips et al., 2004).

369 Despite the significant role that NH₃ has in the formation of PM_{2.5}, it has been relatively lagging
370 in terms of regulation when in comparison to other precursors such as SO₂ and NO_x (Megaritis
371 et al., 2013). In fact, NH₃ can increase the rate of reactions in the atmosphere of both SO₂ and
372 NO_x, resulting in an even larger concentration of particulates (Brunekreef et al., 2015). In
373 Europe for example, the NEC Directive required that in 2020, emissions of SO₂ and NO_x were
374 to be reduced by 59% and 42% respectively, but NH₃ emissions were to be reduced by only
375 6% in comparison (Brunekreef et al., 2015). In the US, while the Clean Air Act (CAA) grants
376 federal authorities the permission to regulate NH₃, and the Comprehensive Environmental
377 Response, Compensation and Liability Act (CERCLA) requires some reporting of this
378 pollutant, there are no definitive regulations in terms of emission thresholds for NH₃ (USDA
379 Agricultural Air Quality Task Force, 2014). China, although considered as a global ammonia
380 hotspot, does not regulate NH₃ in any way (Plautz, 2018).

381 Kanter et al. (2020) provides an excellent overview on how environmental sinks (such as water
382 and climate) are often put to the front of decision making in terms of policies regarding nitrogen
383 emissions. From their examination of 2726 policies across 186 countries, only 28 of 1390 sink-
384 focused policies addressed the impacts of nitrogen across multiple-sinks. This can have
385 inadvertent impacts on other sectors, and cause pollution swapping to occur. For example, to
386 reduce nitrate run-off into waterways, the US increased the use of manure lagoons which are a
387 large source of NH₃ emissions (Aillery et al., 2005). Several studies have suggested that one
388 of the most efficient forms of PM_{2.5} reduction is to reduce agricultural NH₃ emissions,
389 particularly in areas where agriculture is a key contributor to reduced air quality (e.g. Backes
390 et al., 2016; Megaritis et al., 2013; Zhang et al., 2020). By reducing the formation of PM_{2.5}, the

391 associated health impacts of this pollutant would also be reduced. Therefore, an integrated
392 approach for regulating precursors of PM_{2.5}, particularly with a focus on NH₃ and health, would
393 be highly beneficial to reduce the overall creation of secondary PM_{2.5}.

394 4.2. How NH₃ and PM_{2.5} impact human health

395 NH₃ is an irritant, which can have severe effects on human health when over exposure occurs
396 (Sundblad et al., 2004). In an agricultural setting, exposure to extremely high levels of NH₃ is
397 uncommon, often occurring only during farming accidents (National Research Council (US)
398 Committee, 2008). However, exposure to lower concentrations over longer periods may still
399 have a negative impact on human health. The most frequently reported health complaints from
400 NH₃ exposure include eye, nose, and throat irritation, headache, nausea, diarrhea, hoarseness,
401 sore throat, cough, chest tightness, nasal congestion, palpitations, shortness of breath, stress,
402 drowsiness, and alterations in mood (Schiffman and Williams, 2005; Wing and Wolf, 2000)
403 (Figure 3).

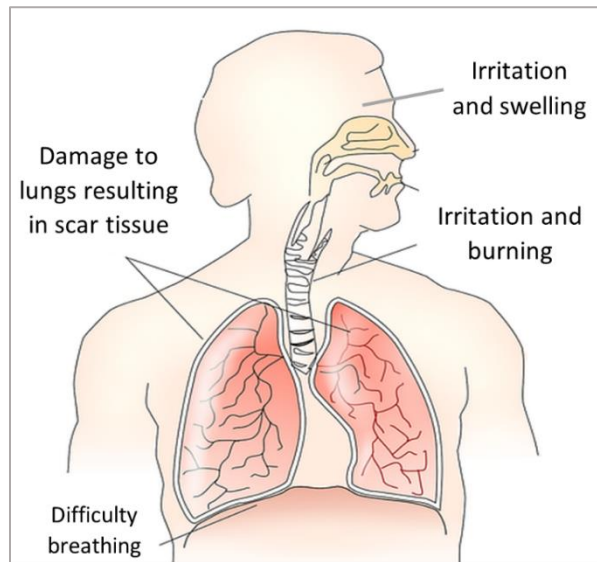


Figure 3. The direct effects of NH₃ on the respiratory tract, showing some of the most commonly reported symptoms

404 Irritation from exposure to ammonia can begin at concentrations as low as 5 ppm (eye
405 discomfort) but are more noticeable at concentrations around 30 ppm, with irritation beginning

406 to occur in the upper respiratory tract (National Research Council (US) Committee, 2008;
407 Sundblad et al., 2004). Acute or accidental exposures to high concentrations (usually above 50
408 ppm) of NH₃ gases can result in tracheal burns as well as damage to bronchial tissues, but
409 exposures such as these are quite rare thanks to occupational exposure limits and health and
410 safety protocols in many work places (National Research Council (US) Committee, 2008).
411 Although exposure to high levels of NH₃ are rare, some agricultural occupations (such as
412 intensive livestock production housing) can result in longer exposure times to levels of NH₃
413 that would not occur in other occupations (Naseem & King, 2018). It is therefore important to
414 ensure exposure within such facilities (and other similar occupations) is within regulated limits.
415 The 2018 Code of Practice for the Chemical Agents Regulations (Health and Safety Authority,
416 2018) based on the European standards states that the occupational exposure limit value for
417 NH₃ should be no more than 20 ppm (14 mg/m³) over an eight hour interval and no more than
418 50 ppm (36 mg/m³) over a 15 minute interval, while the US exposure limits for NH₃ are 25
419 ppm (18 mg/m³) for eight-hour intervals (CDC, 2019).

420 Although direct exposure to agricultural NH₃ can be problematic for human health, when
421 assessed as a precursor to PM_{2.5}, it has a much greater potential impact to human health. The
422 World Health Organisation (2013) describes PM_{2.5} as particles which are small enough to
423 penetrate the thoracic region of the respiratory system once inhaled. Long-term exposure to
424 PM_{2.5} is considered a higher risk to human health than exposure to PM₁₀ (Cohen et al., 2017;
425 Erisman & Schaap, 2004). It is estimated that over 90% of people from across the world are
426 exposed to PM_{2.5} concentrations that exceed the WHO guidelines of 10 µg/m³ (Health Effects
427 Institute, 2019). Exposure to PM_{2.5} can have adverse effects on lung development in children
428 and can cause respiratory irritation in people with pre-existing conditions such as asthma and
429 heart disease (World Health Organisation, 2013). In 2015, 2017, and 2019, exposure to PM_{2.5}
430 pollution posed a significant risk to premature mortality and declining health as stated in the

431 corresponding Global Burden of Disease (GBD) studies (Bu et al., 2021; Collaborators GBD
432 2019 Risk Factors, 2020; Lelieveld et al., 2015). The 2017 GBD study found that exposure to
433 PM_{2.5} was responsible for 4.58 million deaths globally (Bu et al., 2021). In 2015, outdoor
434 PM_{2.5} pollution was identified as the fifth greatest risk factor for health, with ambient
435 PM_{2.5} exposure responsible for 4.2 million deaths worldwide. Increasing PM_{2.5} concentrations
436 can cause serious implications to human health, such as worsening asthma symptoms (18%
437 increase in occurrence with an increase of 10 µg/m³) and an increased risk of cardiopulmonary
438 mortality (a rise of 6 – 13% per 10 µg/m³ PM_{2.5} under long-term exposure) (WHO, 2013; Yu
439 et al., 2000). Reduced life expectancy is also associated with increasing PM_{2.5} levels, as is
440 highlighted in a study conducted by Apte et al. (2018). These authors state that the probability
441 of death above the age of 60 increases with exposure to PM_{2.5} depending on risk factors and
442 disease rates. For example, in low-income and high mortality rate countries such as areas of
443 South Asia, the probability of living to the age of 85 from 60 years old would increase by 20%
444 if the risk of exposure to PM_{2.5} was lowered. In high income areas where exposure to PM_{2.5} is
445 primarily low, the survival rate for this 25-year age range is approximately 50%. In Europe,
446 modelled PM_{2.5} concentrations were found to peak in central Europe with annual mean
447 concentrations ranging between 16 – 18 µg/m³ (Apte et al., 2018). Although this range is below
448 the threshold limit laid out by European standards of 25 µg/m³, it is in exceedance of the WHO
449 guideline of 10 µg/m³. Apte et al. (2018) stated that in 2016, 95% of the population across the
450 globe resided in areas where PM_{2.5} exceeded these WHO guidelines. In the US, the reported
451 national average for PM_{2.5} concentrations in 2019 was approximately 8.5 µg/m³, which is
452 below the US standard of 12 µg/m³ for primary PM_{2.5} and 15 µg/m³ for secondary PM_{2.5} (US
453 EPA, 2020).

454 **5. PM_{2.5}, Agriculture and Health**

455 **5.1. Health impacts of PM_{2.5} as a result of ammonia emissions**

456 There is currently a chain that exists between NH₃ emissions, the formation of PM_{2.5}, and the
457 subsequent impacts that this PM_{2.5} has on human health. Increasing PM_{2.5} emissions are likely
458 to cause an elevated risk of premature mortality, aggravation of pre-existing conditions, and an
459 overall decline in health among at risk populations (e.g. Lelieveld et al., 2015; Spencer and
460 Van Heyst, 2018; World Health Organisation, 2013). To break this chain (or at least to
461 significantly reduce associated impacts), reducing potential emissions of NH₃ primarily from
462 agriculture has been highlighted as a key approach (Gu et al., 2021). Reducing NH₃ emissions
463 from agriculture would aid in reducing PM_{2.5} generation, leading to better air quality and
464 reduced human health impacts.

465 Lelieveld et al. (2015) estimated that in 2010, 3.15 million premature deaths globally were a
466 result of exposure to PM_{2.5}, with 650,000 of these directly resulting from agricultural
467 emissions. Health impacts which were analysed in this study were chronic obstructive
468 pulmonary disease (COPD), acute lower respiratory illness (ALRI), cerebrovascular disease
469 (CEV), ischaemic heart disease (IHD) and lung cancer (LC). The study provided information
470 relating to the top 15 countries in 2010 where premature mortality is linked to outdoor air
471 pollution, specifically PM_{2.5} and ozone (O₃). It is important to note that although O₃ is included
472 in the statistics within this study, only 142,000 of the deaths mentioned were attributable to O₃
473 exposure and were linked only to COPD. In five of these 15 countries, air pollution resulting
474 from agriculture was the leading contributor to premature deaths, and the second leading cause
475 in several others (Lelieveld et al., 2015). For the people who died prematurely as a result of
476 PM_{2.5} exposure, the following percentage of these deaths were attributed to outdoor air
477 pollution (PM_{2.5} and O₃ exposure) resulting from agriculture: Ukraine (52%), Germany (45%),
478 Russia (43%), Japan (38%), and Turkey (29%) (Figure 4). The second leading cause of

479 premature mortality linked to PM_{2.5} (and O₃) in these countries were power generation (Russia,
480 Turkey & Ukraine), land traffic (Germany) and industry (Japan).

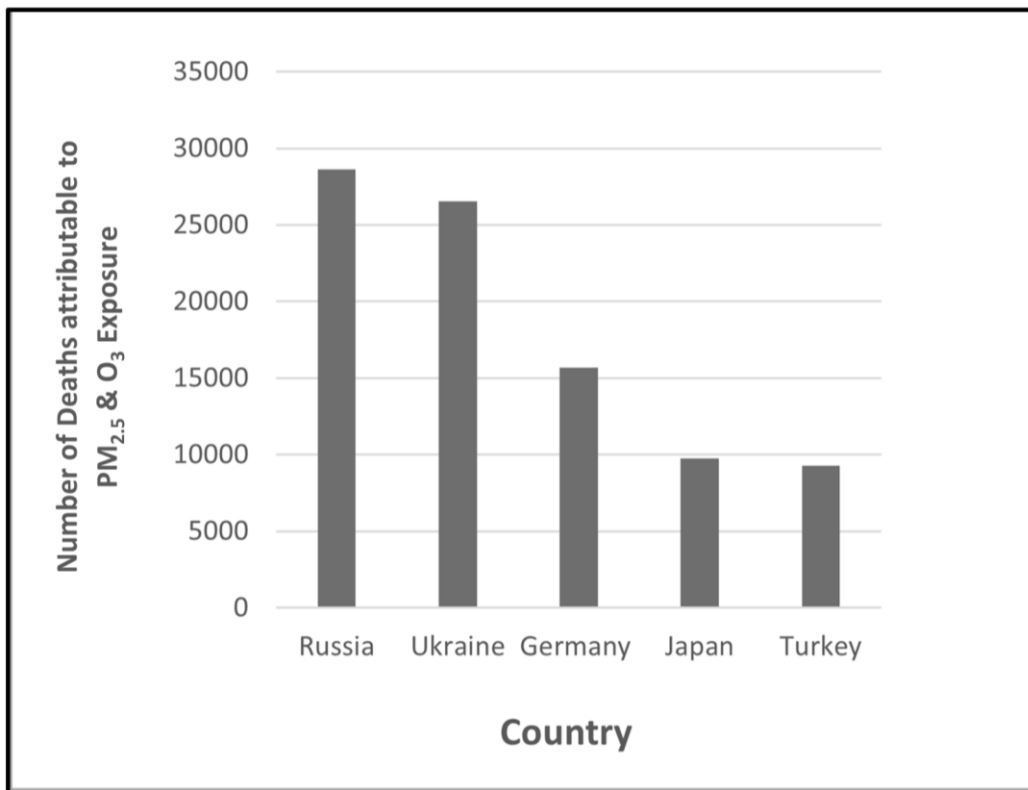


Figure 4. Number of premature deaths (age > 5 or < 30) as a result of exposure to PM_{2.5} and O₃ from agriculture in 2010 (data obtained from Lelieveld et al., 2015)

481 A similar study conducted by Malley et al. (2021) assessed premature mortality as a result of
482 exposure to PM_{2.5} attributable to agricultural emissions using information from the 2019 GBD.
483 Adults over 30 years of age were examined for COPD, IHD, ischaemic stroke (IS), LC, and
484 Type 2 diabetes. Children less than 5 years of age were assessed for lower respiratory
485 infections. The authors estimate that in 2018, 44 million tonnes of NH₃ were emitted globally,
486 mostly due to manure management and application. This study found that the health burden
487 (i.e., premature deaths) associated with PM_{2.5} as a result of agriculture was 537 thousand deaths,
488 18% less than that of Lelieveld et al. (2015). Of these deaths, 358,000 (9% of all PM_{2.5}
489 associated mortalities) were a direct result from the contribution of NH₃ emissions to PM_{2.5}

490 formation. The percentage of premature deaths as a result of exposure to PM_{2.5} from NH₃
491 emissions was highest in North America (90%), East and Southeast Asia (88%), and Western
492 Europe (79%) (Malley et al., 2021).

493 Balasubramanian et al. (2021) assessed various literature on PM_{2.5} from the global food cycle.
494 This study found that contributors of particular importance included livestock production, land-
495 use changes, manure management and agricultural waste burning. From this analysis,
496 Balasubramanian et al. (2021) estimated that 890,000 annual deaths attributable to PM_{2.5} (a
497 total of 23% of deaths resulting from PM_{2.5} exposure based on the 2015 GBD study) are a result
498 of the global food system. Their work highlighted that these deaths could be higher due to
499 underestimated NH₃ emissions, and the overall scarcity of emission data from all stages of the
500 global food system. Thakrar et al. (2020) studied anthropogenic emissions of primary and
501 secondary PM_{2.5} in the US, and the activities and processes which contribute to their release
502 and formation. The authors used models (used for fine scale pollution damage mapping) to
503 assess the deaths attributable to these activities and processes. Thakrar et al. (2020) linked a
504 total of 100,000 (maximum: 107,000, minimum: 88,000) deaths attributable to primary PM_{2.5}
505 and secondary PM_{2.5} precursors in the US (SO₂, NO_x and NH₃). Of five major sectors
506 associated with emissions, they ranked agriculture as the third highest regarding associated
507 deaths (19,000), above residential emissions (12,700) and the electricity sector (10,200). This
508 work estimated livestock rearing and fertilizer applications as the activities which contributed
509 the most to these agricultural deaths (8,400 and 3,700 respectively). Approximately a fifth of
510 the total 100,000 deaths predicted were associated directly with the precursor NH₃ (17,400),
511 higher than that of SO₂ (10,900) and only slightly less than NO_x (19,100). As mentioned
512 previously, NO_x and SO₂ are tightly regulated in terms of air quality in many countries, whereas
513 the targets for the reduction of NH₃ are much less in comparison. One of the recommendations
514 from Thakrar et al. (2020) is to tighten regulations on emissions (particularly NH₃) from

515 agriculture, thus adding to the suggestion that reducing NH₃ emissions will subsequently
516 reduce health impacts from PM_{2.5}. Similarly, Balasubramanian et al. (2021) suggest that air
517 pollution policies and legislation should include NH₃ given how imperative it is in the
518 formation of PM_{2.5}.

519 Erisman & Schaap (2004) state that approximately 50% of all PM_{2.5} in Europe arises from the
520 production of secondary inorganic aerosols from NH₃. In agricultural dominated regions such
521 as North Carolina in the US, the limitation of PM_{2.5} formation could be explained by acid-gas
522 (sulphate and nitrate) availability relative to NH₃ (Walker et al., 2006). Behera & Sharma
523 (2010) examined the formation of PM_{2.5} from NH₃ in the Ganga Basin region of India. This
524 study concluded that the majority of chemical components of the PM_{2.5} examined were
525 (NH₄)₂SO₄, NH₄NO₃ and NH₄Cl. The authors stated that 30% of the PM_{2.5} fraction mass was
526 attributed to inorganic secondary aerosol formation from NH₃ reactions, with a particular
527 emphasis on the contribution NH₃ has during the winter months. Similarly, Bauer et al. (2016)
528 examined the input which agriculture has on the formation of PM_{2.5} using the NASA Goddard
529 Institute for Space Studies Earth System ModelE2 (Bauer et al., 2016). The results of this
530 examination found that ammonium and nitrate were the primary contributors to the formation
531 of PM_{2.5} from agricultural pollution in the Eastern US, China, and Europe. This study found
532 that in the US (particularly in the East) there was an annual PM_{2.5} concentration above 14
533 µg/m³, with agricultural pollution being responsible for 30% of all fine particulate formation.
534 In contrast, PM_{2.5} as a result of agriculture was determined to be responsible for 55% of all
535 human-activity induced pollution in Europe (Bauer et al., 2016). Pinder and Adams (2007)
536 state that in the US, reducing NH₃ by 23% (January) and 21% (July) can result in a 2 µg/m³
537 reduction in PM_{2.5}. This reduction in PM_{2.5} would see 70% of Eastern US areas which currently
538 exceed 15 µg/m³ achieve annual averages below this value.

539 Climate signals can have epidemiological relevance for human health, as mentioned
540 previously. In particular, the climate signal for NH₃ emissions was shown by Geels et al. (2015)
541 through a simulation for the 2080s. This simulation found that, in regions with high NH₃
542 emissions such as Germany, Poland, Netherlands and Belgium, PM_{2.5} concentrations will be
543 affected, which will result in a Europe-wide increase of chronic mortality by 4%. Pozzer et al.
544 (2017) showed that a 50% reduction of agricultural emissions could prevent the mortality
545 attributable to air pollution by about 250,000 people per year worldwide, amounting to
546 reductions of 30%, 19%, 8%, and 3% over North America, Europe, East Asia, and South Asia,
547 respectively. Megaritis et al. (2013) utilised a chemical transport model (CTM) to assess the
548 potential of reducing PM_{2.5} in response to 50% reductions in precursors (SO₂, NO_x and NH₃)
549 and seasonal temperature changes (winter and summer) across Europe. Reducing NH₃
550 emissions was found to be the most effective control method to reduce PM_{2.5} emissions in both
551 summer (5.5% PM_{2.5} reduction) and winter (4% PM_{2.5} reduction) periods. These reductions
552 were highest in Western and Central Europe, with PM_{2.5} reductions of 2.3 µg/m³ (15%, summer
553 period) and 1.5 µg/m³ (10%, winter period) respectively. Reducing NH₃ emissions was found
554 to greatly reduce the formation of ammonium nitrate and ammonium sulphate, leading to
555 potentially substantial reductions in total PM_{2.5} (Megaritis et al., 2013) Although reducing NO_x
556 and SO₂ also had an impact on PM_{2.5}, Megaritis et al. (2013) states that there is high spatial
557 variability with NO_x reductions, and SO₂ is dependent on the availability of NH₃. Not only does
558 the availability of NH₃ influence the generation of ammonium sulphate, but it has also been
559 suggested that the presence of nitrates in the PM_{2.5} fraction is more damaging to health than
560 other particle components (such as NO_x and SO₂), aiding in the need for a greater focus on the
561 regulation of NH₃ emissions as a precursor to PM_{2.5} (Brunekreef et al. 2015).

562 Rural populations living near agricultural and animal production activities are exposed to
563 different environmental factors, including NH₃ exposures (Blanes-Vidal, 2017). Various

564 authors have demonstrated an increased occurrence of physical symptoms and conditions, such
565 as coughs, wheezing, nasal irritation, shortness of breath and asthma, among these populations
566 (Blanes-Vidal et al., 2014; Radon et al., 2001; Schinasi et al., 2011). Exposures above irritation
567 limits of these various pollutants (such as NH₃, H₂S etc.) can cause a series of health symptoms
568 through direct physiological mechanisms. Typically, these symptoms occur at the time of
569 exposure and subside after a short interval (Schiffman et al., 2004). However, for sensitive
570 individuals such as children, the elderly, and asthmatic patients, exposure to this air pollution
571 may induce health symptoms that persist for longer periods of time as well as aggravate existing
572 medical conditions (Schiffman et al., 2004). Heederik et al. (2007) stated that determining
573 which health effects may arise because of particular gaseous pollution (from NH₃, H₂S or CO₂)
574 is controversial and has not been well examined. Other articles such as those by Gehring et al.
575 (2010) and Ryan & Holguin (2010) suggest that exposure to NH₃, and PM_{2.5} resulting from
576 NH₃, can trigger asthmatic reactions in children with pre-existing asthmatic conditions, but
577 more importantly could also play a role in the onset of asthma in very young children. More
578 recent studies such as Pavilonis et al. (2013) and Holst et al. (2018) aimed to assess the role
579 exposure to agricultural emissions (such as NH₃ and PM_{2.5}) may have on early onset asthma in
580 children. Pavilonis et al. (2013) showed that children living in the vicinity of animal production
581 facilities, with a larger relative environmental exposure to gases from animal wastes, had a
582 significantly increased odds of both asthma (OR=1.51, p=0.014 asthma) and medication for
583 wheeze (OR=1.38, p=0.023) (Pavilonis et al., 2013). Holst et al. (2018) examined a population
584 of children who developed asthma from their first to sixth birthday (n = 335,629) in Denmark
585 and aimed to determine if ammonia and ammonium played any part in the development of this
586 condition. These authors used national pollutant concentration information from the year 2008
587 for their study and found that the annual mean concentration was 1.99 ppb (~ 1.4 µg/m³) for
588 NH₃, 1.19 ppb (~ 0.8 µg/m³) for NH₄⁺, and 7.10 µg/m³ for PM_{2.5}. From their study, Holst et al.

589 (2018) determined that there was a clear positive exposure-response association between NH_3
590 and the early onset of asthma. This study found that children who were exposed to higher
591 concentrations of NH_3 were 1.74 times more likely to develop asthma when compared to
592 children who were exposed to the lowest level of NH_3 . In particular, the authors of this study
593 suggest that NH_4^+ contributes the most to early onset of asthma in children in this region. In
594 comparison, direct exposure to $\text{PM}_{2.5}$ was not found to be associated with the onset of asthma
595 in young children in this study.

596 In contrast, a study which was carried out in the Yakima Valley region of the United States
597 assessed the correlation between ambient NH_3 concentrations and reported asthma symptoms
598 and medication use (Loftus et al., 2015). The mean NH_3 concentrations surrounding the homes
599 of the study participants (51 school aged children) ranged from 0.2 to $238 \mu\text{g}/\text{m}^3$ over the course
600 of the 13-month study. The authors determined from this study that there was no statistical
601 significance between increased NH_3 exposure and worsened asthma health as reported by these
602 children. However, the study did indicate that there was an association between measured lung
603 function and NH_3 exposure at the participant's home. The authors of this study note several
604 potential limitations including parental bias, participants avoidance of outdoor areas on high
605 NH_3 days, and non-differential measurement error. It is important to note that studies such as
606 these are a relatively new area of focus, and there is still need for a more in-depth analysis of
607 all contributing factors to asthma as a result of exposure to agricultural emissions (including
608 but not limited to bioaerosols, microbiota, odour, and psychological symptoms). Primarily, the
609 reduction of NH_3 emissions from agriculture should be the overall focus and will inevitably aid
610 in reducing the $\text{PM}_{2.5}$ fraction which forms as a result of these emissions, which is well
611 documented in terms of health impacts. By reducing these emissions, air quality can be

612 improved, declines in human health can be avoided, health costs from these illnesses can be
613 lowered, and a better quality of life can be achieved.

614 **5.2. The economic benefit of reducing NH₃ and PM_{2.5}**

615 Policies and decisions are often made on the basis of social costs and reduction benefits, and
616 this is no different for NH₃ or PM_{2.5} (Brink et al., 2011). In this regard, social costs can be
617 considered as anything an individual or a community may have to change or limit when
618 modifying their nitrogen management techniques. Incorporating NH₃ mitigation measures into
619 policy and implementation of these techniques in agricultural practices could impose
620 potentially significant costs. For example, Zhang et al. (2020) states that a possible 38 – 67%
621 of NH₃ from agriculture could be mitigated at a cost of 6 – 11 billion US\$ (€5.2 – €9.5 billion)
622 in China. In comparison, the societal benefits are considered to all positively contribute to
623 human welfare and can assist in both protecting human health and benefiting the economy over
624 time. Where the mitigation measures for China were upwards of 6 billion US\$, the overall
625 societal benefits because of these mitigations were estimated at 18 – 42 billion US\$ (~ €15.8 -
626 €36.9 billion) (Zhang et al., 2020). In the EU, a mean annual cost of health to Member States
627 ranges between €2 – €36 (~ US\$2.50 - \$41) per kg N for NH₃ (Brink et al., 2011). Pinder and
628 Adams (2007) state that in the US, reducing NH₃ (particularly in the winter periods) is
629 significantly more cost effective for PM_{2.5} reductions than reducing NO_x and SO₂ in regions
630 across the Eastern US. Gu et al., (2021) states that in comparison to the reduction methods
631 available for nitrogen oxide emissions, the abatement of ammonia on a global level is only 10%
632 of the cost associated with NO_x reductions.

633 Reducing NH₃ emissions can lower PM_{2.5} concentrations, and contribute to a reduction in
634 premature mortality globally, subsequently aiding in both economic and health cost savings.
635 Giannadaki et al. (2018) modelled that reducing agricultural emissions (particularly NH₃) could
636 result in the prevention of over 200,000 deaths per year across 59 countries. These authors

637 examined 59 countries and applied a “value of statistical life” (the willingness to pay for
638 avoiding the risk of a premature death) to monetize potential health outcomes by reducing
639 agricultural emissions. An estimation of the costs which would be incurred to reduce NH₃
640 emissions to a level which meets national reduction commitments were also highlighted in this
641 study. Giannadaki et al. (2018) states that this reduction would cost a total of €70 million - €89
642 million (€43 - €2176 annually per farm in the EU, depending on abatement strategies
643 implemented and current farm emissions). Within the EU, this decrease in emissions could see
644 a reduction in mortality rate of 18%, resulting in an annual saving to the economy of
645 approximately €75.6 billion (~ US\$86 billion). Giannakis et al. (2019) estimates that the cost
646 to implement all four major NH₃ abatement strategies to decrease emissions (including reduced
647 nitrogen feed, manure storage, low emission fertilizer application techniques and animal
648 housing) to the required EU levels would cost approximately €4.3 billion (5.0 billion US\$) for
649 Europe alone. These authors state that implementing these abatement strategies would have the
650 greatest effect on premature mortality reduction in Germany, followed by the United Kingdom
651 and Italy with reductions of 930, 928 and 448 deaths per year, respectively.

652 In the US, agriculture is considered as one of the leading causes of gross external damage
653 (economic loss) due to premature mortality, costing the economy approximately 230 billion
654 US\$ (€197 billion) (Tschofen et al., 2019). Within the agricultural sector, livestock and animal
655 husbandry are the main contributors to gross external damage, accounting for approximately
656 50% of all economic loss due to premature death. A study of a pig facility in North Carolina
657 suggested that PM_{2.5} resulting from NH₃ leads to higher health costs than any other agricultural
658 pollutant, costing the US healthcare system anywhere between US\$10 (~ €8.50) and US\$73 (~
659 €64) per kilogram of NH₃ (Stokstad, 2014). Domingo et al. (2021) states that improving the
660 efficiency of NH₃ application to land (either by method of application or timing of application
661 period) at a cost of 0.8 to 3.2 US\$ per kg/NH₃ can see an economic benefit between 33.4 and

662 42.4 US\$ per kg/NH₃ solely due to the reduction of PM_{2.5} formation. In China, the previously
663 mentioned reduction of NH₃ by 38 – 67% was estimated to reduce premature mortalities by
664 between 90 – 240 thousand people, due to a reduction in PM_{2.5} concentrations of approximately
665 8 – 20% (Zhang et al., 2020). In economic terms, this could save over 10 – 26 billion US\$ in
666 health costs in China alone.

667 **6. Results and Discussion**

668 Agriculture is the primary contributor to global NH₃ emissions, playing a major role as a
669 precursor to secondary PM_{2.5} (Brunekreef et al., 2015; Erisman and Schaap, 2004; Thakrar et
670 al., 2020). The production of livestock, storage of associated manure, and its use as organic
671 fertilizer, alongside synthetic fertilizers, are key components in the emission of NH₃ from
672 agriculture (Environmental Protection Agency, 2018; Ma et al., 2021; Zhang et al., 2017). The
673 emission of NH₃ from agriculture varies from region to region and depends heavily on the
674 source from which it is emitted (Balasubramanian et al., 2020; Singles et al., 1998). This high
675 spatial variability of NH₃ alongside its seasonal and diurnal variations (Blanes-Vidal et al.,
676 2008; Kelleghan et al., 2020) highlights the need for representative long term monitoring of
677 both emissions and atmospheric concentrations of this pollutant. Incorporating alternative
678 approaches such as switching from the use of splash plates to trailing shoe or injection for land
679 application, or implementing manure store covers, offers ways of reducing NH₃ emissions (e.g.,
680 Hellsten, 2017; Xu et al., 2017).

681 The reduction of NH₃ emissions where possible is imperative, as current projections suggest
682 emissions are set to continue to rise with increasing temperatures due to the current climate
683 crisis (Skjøth & Geels, 2013; Sutton et al., 2013), alongside the increasing intensification of
684 agriculture to meet global food demands (Malley et al., 2021). Failure to incorporate low
685 emission techniques to reduce NH₃ will see an increase in negative impacts from this pollutant
686 across the globe due to these predicted increases. It is clear that NH₃ emissions are responsible
687 for a large portion of the PM_{2.5} fraction of air pollution (Bauer et al., 2016; Pinder & Adams,
688 2007; Thakrar et al., 2020). Although current health impacts from NH₃ as an individual
689 pollutant are more likely for those who work in or near sources (Naseem & King, 2018), the
690 contribution of NH₃ to particulate matter formation has the potential to impact human health
691 on a population level (e.g. Giannakis et al., 2019). This PM_{2.5} can have negative effects on

692 human health including the development of chronic health issues and premature death (e.g.
693 Lelieveld et al., 2015; Thakrar et al., 2020). NH_3 contributes to approximately 50% of $\text{PM}_{2.5}$ in
694 Europe, 30% in the US, and 15-23% of the $\text{PM}_{2.5}$ mass in the North China Plain (Bauer et al.,
695 2016; Erisman & Schaap, 2004; Han et al., 2020).

696 According to various studies mentioned previously, Europe can be considered as having the
697 greatest NH_3 contribution to the formation of $\text{PM}_{2.5}$ in comparison to other regions. Table 1
698 provides an overview of NH_3 emission sources in the 28 Member States in the European Union.
699 Cattle are the largest contributor to NH_3 emissions from agriculture (28%) and $\text{PM}_{2.5}$ as a result
700 of such (14%) in this region, followed by organic fertilizer use (27% NH_3 emissions, 13%
701 $\text{PM}_{2.5}$ contribution) and the use of inorganic N-fertilizers (17% NH_3 emissions, 8% $\text{PM}_{2.5}$
702 contribution) (European Environment Agency, 2021). This may explain the reasoning for the
703 EU's stricter regulations surrounding NH_3 emissions. However, even with the regulations in
704 place in Europe regarding NH_3 emissions and atmospheric concentrations, there is still a high
705 contribution percentage to $\text{PM}_{2.5}$, again highlighting the need for regulations to be implemented
706 regarding NH_3 as a precursor pollutant.

707 **Table 1.** Emissions of NH₃ (%) from various sources and their percentage contribution to PM_{2.5} in the 28 EU
 708 Member States (source data: European Environment Agency, 2021), and potential increases in these emissions
 709 based on a 5°C warming scenario discussed in Sutton et al. (2013).

Source	Contribution to NH ₃ emissions (%)	Contribution of NH ₃ emissions to PM _{2.5} fraction (%)	Potential NH ₃ emissions (%) (based on 5* increase in temp, Sutton et al. 2013)	Potential contribution of NH ₃ emissions to PM _{2.5} (%)
Cattle	28	14	40	20
Organic Fertilizer	27	13	38	19
Inorganic N- fertilizer (includes also urea application)	17	8	24	12
Pigs	11	5	15	8
Poultry	7	4	10	5
Non-agricultural	7	3	10	5
Sheep, Goats, Horses, Mules, Buffalo, Other	3	2	4	2
Other Agricultural	1	0	1	0

710 Sutton et al. (2013) states that with an increase to global temperatures of 5°C, there is a
 711 predicted increase in global NH₃ emissions of 42%. Using this 42% increase estimate, the
 712 potential increase in NH₃ emissions for these sources in Europe, and the resulting fraction of
 713 PM_{2.5} created is highlighted in Table 1. The three greatest emitters of NH₃ in Europe would see
 714 a growth in NH₃ emissions from the values mentioned above, to 40%, 38%, and 24% for cattle,
 715 organic and inorganic fertilizer use respectively. This would in turn result in a dramatic increase
 716 of the fraction of PM_{2.5} as a result of NH₃ emissions from these sectors (20%, 19% and 12%
 717 respectively), ultimately placing a greater strain on human health in these areas. Although
 718 PM_{2.5} as a result of NH₃ emissions is greatest in Europe, if current emissions are not reduced
 719 across the world, there is potential that the PM_{2.5} fraction across the globe as a result of

720 livestock production will increase. Increases such as these will have a significant impact on
721 human health and economic costs associated with such across the globe. This highlights the
722 need for rapid action to reduce NH₃ emissions to protect human health due to the current
723 climate crisis.

724 The potential contribution to PM_{2.5} production of NH₃ emissions from livestock reflect the
725 necessity of incorporating NH₃ into policies to protect human health in addition to the
726 environment. For example, in the EU, the NEC Directive focuses on reporting national NH₃
727 emissions (and deposition values) and setting emission limits, while also now requiring
728 monitoring across sensitive habitats; where the Habitats Directive encourages Member States
729 to set contribution limits to Natura 2000 sites and the Industrial Emission Directive sets
730 emission limits for pig and poultry farms of certain sizes. These directives are typically subject
731 to interpretation by each Member State with application varying from country to country. In
732 the US, NH₃ is not considered as a criteria pollutant under the CAA which would allow for the
733 regulation of this pollutant, a request that was rejected by the USEPA in 2018 (Stowell, 2018).

734 Reflecting on the data presented in Table 1, it is clear that livestock NH₃ emissions play a vital
735 role in contributing to the PM_{2.5} fraction, particularly in Europe, and should be incorporated in
736 any future discussions in the regulation of both NH₃ emissions themselves, and the control of
737 PM_{2.5}. Across the rest of the world, updating of emissions databases and monitoring of NH₃
738 should be a priority so as to understand the emissions and source contributions (such as above),
739 and in turn, provide the ability to assess source contribution to PM_{2.5} creation. As NH₃ is
740 primarily released from agriculture, there must be a balance found between the control of these
741 emissions and the ability of the industry to continue to meet the global food demand. Policy
742 makers and stakeholders must fully understand the role that NH₃ plays in terms of its emission
743 from various agricultural sectors, its reactions and transportation in the environment,
744 particularly its role in the formation of PM_{2.5} and the subsequent impact on human health. NH₃

745 is a difficult pollutant to measure due to the variability of emissions and concentrations in the
746 environment, and so regulations must begin with the implementation of long-term monitoring
747 to better understand the movement of NH₃ on a local, regional, or national scale. Understanding
748 the beginning of this chain (i.e., how NH₃ enters and moves through the environment) will
749 allow for better informed policies to be implemented to protect human health from the creation
750 of PM_{2.5} as a result of agricultural NH₃ emissions. There is an inherent need for future
751 regulations and policies surrounding NH₃ and PM_{2.5} to be reflective of both environmental
752 protection and improving human health, while ensuring neither are impacted as a result of the
753 other. By considering the issue of NH₃ abatement from a combined approach (i.e., assessing
754 impacts on agriculture, the environment, and public health), a better outcome for all sectors
755 could be reached while also reducing the socio-economic costs of NH₃ and PM_{2.5}. Through
756 understanding the source sector contributions of NH₃ emissions, a focus could be placed on
757 areas which would be high impact and low cost to implement reduction measures for NH₃,
758 subsequently reducing PM_{2.5} creation from this precursor (such as measures assessed by
759 Buckley et al., 2020; Edwards, 2020; Hellsten, 2017). This can aid in the beginnings of
760 legislation and policy discussions, and even the implementation of government grants and
761 assistance to reach set targets (e.g. LESS scheme in Ireland (Government of Ireland, 2022)),
762 providing stakeholders and policy makers with a clearer indication of where to start.

763 **7. Conclusion**

764 Numerous studies indicate that one of the most efficient ways to reduce PM_{2.5} is to reduce
765 global NH₃ emissions resulting from agriculture. This reduction in NH₃ can subsequently also
766 have a large impact in the reduction of chronic health problems, decrease premature mortality
767 across the globe and can lower monetary loss to national economies due to these premature
768 deaths. Regulations surrounding NH₃ as a precursor to PM_{2.5} formation is lacking and should

769 be incorporated into future policy and regulations across the globe to aid in bettering human
770 health from PM_{2.5} exposure.

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