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

Authors: Katie E. Wyer[a], David B. Kelleghan[b], Victoria Blanes-Vidal[c], Günther Schauberger[d] and Thomas P. Curran[a]

[a] *UCD School of Biosystems and Food Engineering, University College Dublin, Belfield, Dublin 4, Ireland.*

[b] *Teagasc, Environment, Soils and Land-Use Department, Johnstown Castle, Co. Wexford, Y35 TC97, Ireland*

[c] *Applied AI and Data Science (AID), Maersk Mc-Kinney Moller Institute, University of Southern Denmark, Odense, Denmark.*

[d] *Department of Physiology and Biophysics, University of Veterinary Medicine, Vienna, A-1210 Wien, Veterinärplatz 1, Austria.*

Corresponding Author:

Katie E. Wyer

katie.wyer@ucdconnect.ie

Abbreviations

NH_3	Ammonia
$PM_{2.5}$	Fine Particulate Matter
COPD	Chronic Obstructive Pulmonary Disorder
EDGAR	Emissions Database for Global Atmospheric Research
CAA	Clean Air Act
US	United States
O_2	Oxygen
H_2O	Water
CO_2	Carbon Dioxide
N_2	Nitrogen Gas
N_2O	Nitrogen Dioxide
CERCLA	Comprehensive Environmental Response, Compensation and Liability Act
BAT	Best Available Techniques
IPPC	Integrated Pollution Prevention and Control
IED	Industrial Emissions Directive
K	Kelvin (Degrees)
NH_4^+	Ammonium
NH_4NO_3	Ammonium Nitrate
$(NH_4)_2SO_4$	Ammonium Sulphate

NH_4CL	Ammonium Chloride
NEC	National Emissions Ceiling Directive
EPA	Environmental Protection Agency
WHO	World Health Organisation
ALRI	Acute Lower Respiratory Illness
CEV	Cerebrovascular Disease
IHD	Ischemic Heart Disease
LC	Lung Cancer
O_3	Ozone

Abstract

Atmospheric ammonia (NH_3) released from agriculture is contributing significantly to acidification and atmospheric NH_3 may have on human health is much less readily available. The potential direct impact of NH_3 on the health of the general public is under-represented in scientific literature, though there have been several studies which indicate that NH_3 has a direct effect on the respiratory health of those who handle livestock. These health impacts can include a reduced lung function, irritation to the throat and eyes, and increased coughing and phlegm expulsion. More recent studies have indicated that agricultural NH_3 may directly influence the early on-set of asthma in young children. In addition to the potential direct impact of ammonia, it is also a substantial contributor to the fine particulate matter ($PM_{2.5}$) fraction (namely the US and Europe); where it accounts for the formation of 30% and 50% of all $PM_{2.5}$ respectively. $PM_{2.5}$ has the ability to penetrate deep into the lungs and cause long term illnesses such as Chronic Obstructive Pulmonary Disease (COPD) and lung cancer. Hence, $PM_{2.5}$ causes economic losses which equate to billions of dollars (US) to the global economy annually. Both premature deaths associated with the health impacts from $PM_{2.5}$ and economic losses could be mitigated with a reduction in NH_3 emissions resulting from agriculture. As agriculture contributes to more than 81% of all global NH_3 emissions, it is imperative that food production does not come at a cost to the world's ability to breathe; where reductions in NH_3 emissions can be easier to achieve than other associated pollutants.

Keywords: NH_3, $PM_{2.5}$, Particulate Matter, Asthma, Respiratory Health, Animal Production.

1. Introduction

Ammonia (NH_3) is a reduced form of nitrogen which is released to the atmosphere (Behera et al., 2013; Doyle et al., 2017; Sutton et al., 2008), with agriculture contributing over 81% of its total global emissions (Van Damme et al., 2021). Figure 1 displays ammonia emissions across the world from various sources, highlighting the high spatial variation of NH_3 globally (EDGAR, 2015). Areas which are orange to red in colour (such as can be seen across Europe, the Eastern United States, China, and India) emit high levels of NH_3 (greater than 16 tonnes of ammonia per year per grid cell).

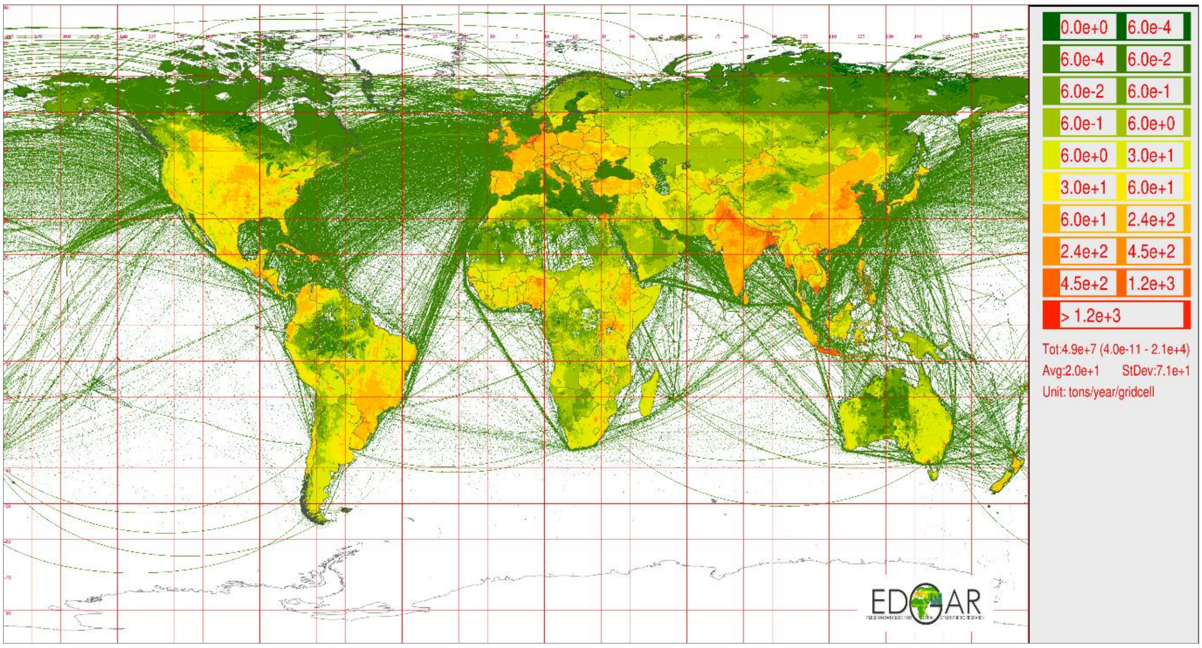

Figure 1. Spatial distribution of global ammonia emissions from various sources (EDGAR 2015). Emissions are represented as tonnes of NH_3 emitted per year (per grid cell). Areas with higher emission values are represented with an orange to red colouring e.g. Europe, Eastern US and India.

Environmental impacts of atmospheric NH_3 are well documented in scientific literature, resulting primarily from the acidification and eutrophication of ecosystems, typically resulting in species community changes (García-Gómez et al., 2014; Sutton et al., 2009). Currently, the known impacts which NH_3 may have on biodiversity (particularly in sensitive habitats) are a key component in the development of various national and regional policies (e.g. EPA, 2021;

Luesink and Michels, 2018; Shropshire Council, 2018) to better protect the environment from pollution, and to comply with the Habitats Directive (92/43/EEC) (European Economic Committee, 1992). In comparison to these well documented environmental impacts, and the resulting policies created from such, there has been relatively little work on the potential impact agricultural NH_3 may have on human health.

An increase in large-scale intensive agricultural operations such as concentrated animal feeding operations (Schultz et al., 2019) has contributed to increasing NH_3 emissions, and the potential effect of this pollutant on human health has been raised as a concern (Ge et al., 2020; Naseem & King, 2018). NH_3 emissions from agriculture are responsible for 30% of all $PM_{2.5}$ in the US and Ganga Basin of India, 50% in Europe, and between 15 – 23% in China (Bauer et al., 2016; Behera et al., 2010; Erisman & Schaap, 2004; Han et al., 2020). $PM_{2.5}$ exposure can cause illnesses such as chronic obstructive pulmonary disorder (COPD), lung cancer, and has even been documented as a leading risk factor to premature mortality (Apte et al., 2018; Lelieveld et al., 2015; Yu et al., 2000). Many precursors to $PM_{2.5}$ are well regulated, such as nitrogen oxide (NO_x) and sulphur dioxide (SO_2), though studies have shown that NH_3 is the precursor which has the biggest impact on the formation of this pollutant (Brunekreef et al., 2015; Megaritis et al., 2013; Thakrar et al., 2020). Therefore, it is imperative that there is an understanding of how agricultural NH_3 is emitted to the atmosphere. Without understanding the beginning of the chain from NH_3 sources to health impacts of $PM_{2.5}$, optimum reductions in health impacts from this pollutant will not be reached. Knowing the extent at which agriculture contributes to both NH_3 emissions and the generation of $PM_{2.5}$ can allow for a more thorough examination of the potential impacts to human health which agricultural NH_3 may cause, and how these health impacts may be reduced by controlling emissions from the source.

This review aims to provide an understanding of both the root cause of NH_3 emissions (i.e., agriculture) and the public and socio-economic (i.e., public health) impacts this can have due to $PM_{2.5}$ formation. By focusing on these multiple links of the "NH_3 – $PM_{2.5}$ – health impacts" chain, readers may better understand the full cycle, and how source reduction of NH_3 can lessen health impacts.

2. Methodology

The aim of this systematic literature review is to provide the reader with a clear understanding of the processes involved in the release of NH_3 from agriculture, the contribution of NH_3 to $PM_{2.5}$ formation, and the resulting impacts to human health. In order for sources to be included in this review, they had to meet certain criteria such as:

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

The information provided within this review was sourced using multiple databases. Google Scholar and Science Direct were used initially to gain an understanding of both the extent and availability of peer reviewed research articles pertaining to this review. Descriptive search terms such as *ammonia, particulate matter, $PM_{2.5}$, health impacts, human health, asthma, COPD, agriculture, atmospheric ammonia, deposition and transformation, toxicology, social economic costs*, and *abatement* were used in various searches. These keywords were combined with “AND” and “OR” commands as appropriate to search a wide range of articles across the search engines mentioned, as well as (but not limited to) PubMed Central (PMC), My Science Work, ORCID, and internet searches using Google. Any sources which were obtained as a result of an internet search were written by professionals and published in journals, government agencies or health organisations. As well as using search engines, references of particular interest in some articles used in the review were also studied. From these searches, 596 papers were identified to meet the search criteria outlined previously. In order for a source to be included in the final review, the study or information was required to include both agriculture and NH_3 emissions, interactions with $PM_{2.5}$ and subsequent health impacts, and/or data pertaining to such. Many of the studies which were screened were not used in the review although they met the initial search criteria, as their primary investigation did not provide clarity to the subject at hand (e.g., papers which primarily focused on emission data from one livestock facility at one location). In total, 135 of these sources were investigated and utilised for this review. There were no instances of duplicate papers found during the inclusion process.

3. Sources of Ammonia in Agriculture

Agriculture is considered as the dominant source of atmospheric ammonia, contributing to over 81% of global NH_3 emissions (Van Damme et al., 2021). The primary sources of NH_3 emissions from agriculture include livestock and animal production, manure handling and storage, livestock housing and the application of manure/slurry and artificial fertilizers to land (Behera et al., 2013; Mikkelsen et al., 2011; Sutton et al., 2013) (Figure 2).

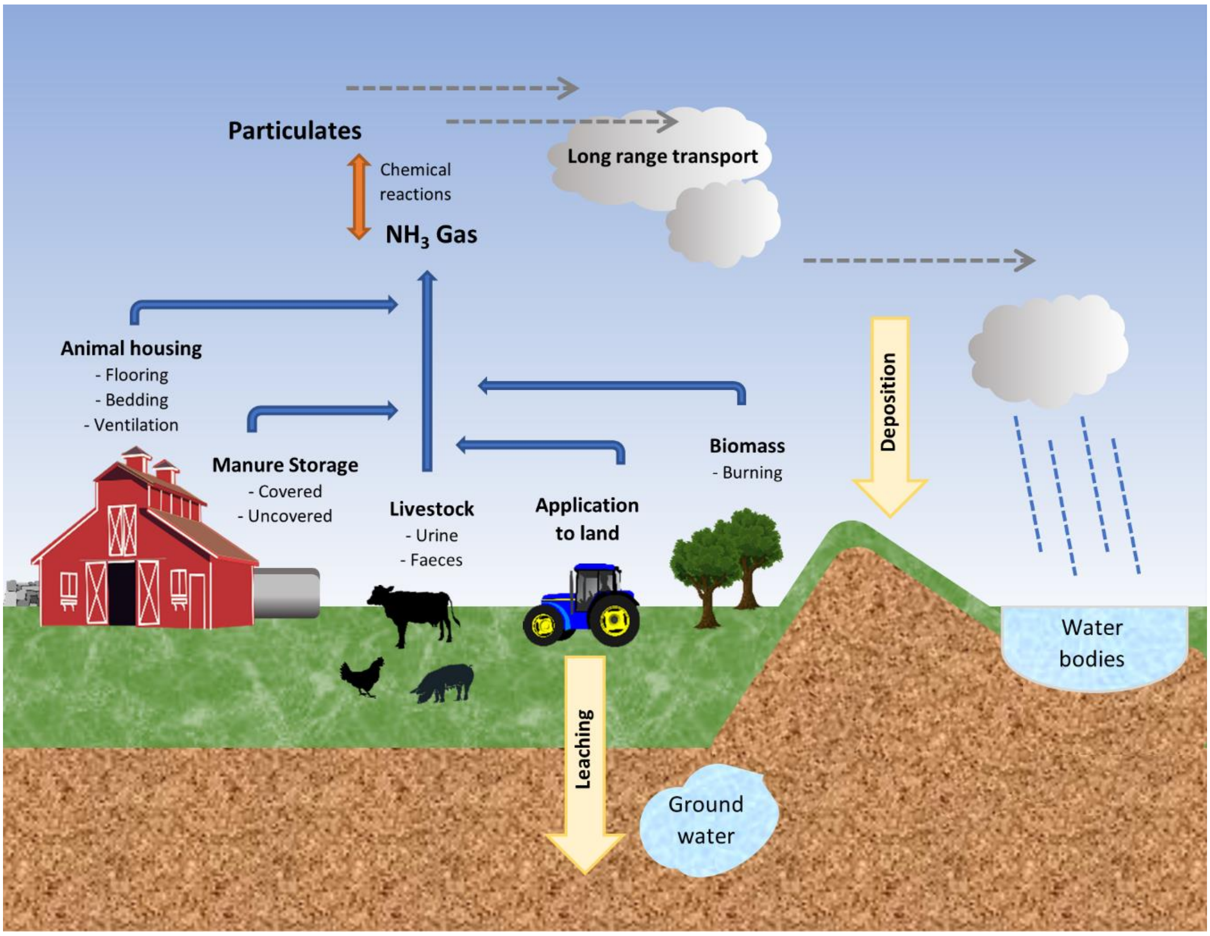

Figure 2. Potential sources of NH_3 from agriculture and routes of transport

NH_3 is regulated to some extent under EU law where the National Emissions Ceilings (NEC) Directive (2016/2284) (European Union, 2016) requires reporting of NH_3 emissions and monitoring its concentration on sensitive habitats, the Habitats Directive (92/43/EEC) (European Economic Committee, 1992) which requires projects to limit contributions to sensitive habitats, and the Industrial Emissions Directive (2010/75/EU) (European Union, 2010) which limits emissions from certain facilities.

The source contribution of each agricultural activity varies greatly based on national, regional and local agricultural practices (Balasubramanian et al., 2020; Singles et al., 1998). Emission factors are used to estimate the emission of pollutants, and are used by regulatory bodies to assess emissions from various pollution sources (Faulkner & Shaw, 2008). Emission factors for NH_3, which are widely used to estimate emissions from agricultural sources, are not capable of accurately reflecting the temporal and spatial variations in emissions (Faulkner & Shaw, 2008). This is not only because of differences in agricultural practices, but also as a result of variations in environmental, meteorological, and climatic conditions (Balasubramanian et al., 2020; Sutton et al., 2013; Zhu et al., 2015). Von Bobrutzki et al., (2010), Behera et al., (2013), and Sutton et al., (2013) all provide excellent synopses highlighting the difficulties that are faced with the measurement and modelling of NH_3 emissions (and concentrations) in relation to agricultural sources.

3.1. Livestock

Excreta from livestock, including uric acid, urea and faeces, can be decomposed or volatilized to form NH_3 (Behera et al., 2013). There are several microbiological processes involved in the degradation of these compounds to NH_3. Uric acid in the presence of oxygen (O_2) and water (H_2O) is converted to carbon dioxide (CO_2) and NH_3 by the enzyme uricase (equation 1a). Urea is degraded to CO_2 and NH_3 by the enzyme urease, which is produced by various microbes often present in manure (equation 1b). Undigested proteins present in manure are converted to NH_3 by both the enzymes uricase and urease, as well as the action of bacterial metabolism (equation 1c).

$$C_5H_4O_3N_4 + 1.5O_2 + 4H_2O \rightarrow 5CO_2 + 4NH_3 \qquad (1a)$$

$$CO(NH_2)_2 + H_2O \rightarrow CO_2 + 2NH_3 \quad (1b)$$

$$undigested\ proteins \rightarrow NH_3 \quad (1c)$$

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

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

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

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

Changes to housing, including ventilation rates, bedding, flooring, temperature, and manure storage can all aid in mitigating emissions from this source. For livestock buildings, air treatment systems (e.g. biofilters, bio-scrubbers, activated carbon filters) are considered as Best Available Techniques (BAT) by the EU (Santonja et al., 2017). The incorporation of acid scrubbers and bio-trickling filters in housing units can be highly effective at removing NH_3, with efficiencies in the range of 91% to 99% and about 70%, respectively (Melse & Ogink, 2005). However, because of economic (high running costs), environmental (high energy consumption, chemical use, discharge water), and technical reasons (unstable performance of bio-scrubbers) these techniques are only considered suitable for intensive pig and poultry livestock installations under the IPPC Directive (2008/1/EC) (now the IED (2010/75/EC)) (European Union, 2010; Melse et al., 2009).

The common slurry based, slatted system used in many cattle production facilities can benefit from the introduction of straw bedding, reducing NH_3 emissions by up to 30% (Webb et al., 2005). This is due to the absorption of urine into the straw bed, thus reducing the interaction of NH_3 with surrounding air. However, the addition of straw can make slurry removal more difficult. Implementing abatement techniques such as decreasing manure surface area, introducing rapid removal of urine, and decreasing air velocity and temperature above manure pits could aid in reducing NH_3 emissions from cattle houses (Hellsten, 2017). Reducing the area of slatted floor, particularly in pig housing, can reduce the area in which the animals defecate. In most cases, housed pigs will choose to defecate on slatted floors (cooler areas) as opposed to solid floors (warmer areas where pigs choose to lay) (Aarnink et al., 1997; Ye et al., 2009). These studies found that decreasing the area of slatted floor within a pig house can reduce daily NH_3 emissions by 0.7g of NH_3 per fattening pig. Designing new (or re-designing old) houses to have partially slatted or bedded floors could reduce NH_3 emissions by up to 20% for a cost of €0 - €3 per kilogram of NH_3-N reduced (Hellsten, 2017).

3.2. External slurry storage

Animal wastes can be stored as slurry which combines the liquid (urine) and solid (faeces) fractions, or separated into its individual liquid and solid components (Grant & Boehm, 2020). Once slurry is removed from animal houses, it can be stored in a number of ways including, for example, silos (concrete, steel or wooden tanks), and lagoons (Webb et al., 2005). The surface area of these storage facilities plays a vital role in the level of NH_3 emitted from the manure. Lagoons tend to have a higher surface area to volume ratio than silos, which entails a greater emission potential when compared to tank-style storage systems (Webb et al., 2005). Though lagoons also have a higher emission rate per unit area, where Kupper et al. (2020) states that the baseline emissions for slurry storage are:

- 0.12 g m^{-2} h^{-1} (cattle slurry) and 0.15 g m^{-2} h^{-1} (pig slurry) stored in lagoons
- 0.08 g m^{-2} h^{-1} (cattle slurry) and 0.24 g m^{-2} h^{-1} (pig slurry) stored in tanks

The most effective means of reducing NH_3 emissions from storage areas is through fitting covers and canopies, which can reduce emissions by up to 80% (Webb et al., 2005). Such a reduction strategy was assessed for China by Xu et al. (2017), showing a country-wide reduction capacity between 19 – 37% when compared to the "business as usual" emission levels. The method of covered manure storage is most effective for circular tanks which are made of a non-porous material such as steel (Webb et al., 2005). The use of biological mechanisms can also assist in reducing NH_3 emissions when covering slurries. Blanes-Vidal et al. (2009) assessed the use of aged straws for slurry cover (which contains a higher microbiota than fresh straw), showing that this method had a 99% efficiency in reducing NH_3 emissions. This is compared to a 47% efficiency in reduction using new straw covers. These authors also state that 50% of this reduction efficiency corresponds to the straw cover acting as a biofilter, whereas the remaining 50% is associated with physical and chemical effects. Hellsten (2017) stated that there are minimal measures deemed applicable to reduce NH_3 emissions from solid livestock waste systems. Covering the solid waste, applying some form of litter, and keeping the waste dry are currently the only feasible options for manure management in solid waste.

3.3. Land application

Ammonia emissions during and after land application of animal wastes (both as liquid slurries and as solid manure) and synthetic nitrogenous fertilizers, contributes to a significant proportion of the total NH_3 emissions from agriculture (Misselbrook et al., 2005). A review of 324 studies by Ma et al. (2021) estimated that emissions of NH_3 from manure application were 3.79 Tg N/year, and for synthetic fertilizer application were 12.32 Tg N/year. Ma et al. (2021) also describes that China, India, and the US produce 60% of total NH_3 emissions associated with this synthetic fertilizer application.

One of the most important factors in NH_3 emissions from land application is the method of how manure is spread on the fields (Bourdin et al., 2014; Edwards, 2020; Hyde et al., 2003). Currently, broad spreading (e.g., splash plate) application is the cheapest method of land-spreading but is associated with the highest levels of NH_3 emissions compared to any other approach (Edwards, 2020). NH_3 volatilization from nitrogen fertilizer or slurry/manure application also depends strongly on localized environmental and meteorological parameters, such as daily maximum and minimum temperature, radiation, precipitation, relative humidity, and wind velocity at 10 m height, as well as soil moisture content, dry matter content of the slurry/manure, and the leaf area of the crop/product (e.g. Anderson et al., 2003; Bash et al., 2013; Fu et al., 2015; Häni et al., 2016; Misselbrook et al., 2005).

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

3.4. Meteorological and seasonal variations

Alongside temperature, humidity, and other meteorological conditions, Jiang et al. (2020) states that rain is one of the primary driving forces when it comes to the application of manure or fertilizer to land, with semi-dry tropic regions presenting an emission rate five times greater than that of colder, wet climates. In these humid or tropical areas, the emission of NH_3 from controlled poultry houses is three times greater than that of dry, cold locations. In many European countries, application of manure and fertilizer to land coincides with the dry seasons, thus resulting in higher emissions from this source (land application) during summer periods (Backes et al., 2016). Seasonal patterns in atmospheric NH_3 variations were noted by Doyle et al. (2017), referencing several studies which all experienced similar patterns dependent on both weather, climate, and agricultural activities (Tang et al., 2004; Zbieranowski & Aherne, 2012). Temperature and climate also have an impact on biomass burning, which although is not the most prominent source of agricultural NH_3 in the environment, still plays an important role in emissions (Bray et al., 2021). Biomass burning in the US accounts for approximately 10% of NH_3 emissions (with agriculture accounting for approximately 82% of this) and usually occurs mostly in the summer and autumn (Bray et al., 2021). Across the globe, there is a high variability of both the frequency and intensity of these fires (Oenema et al., 2012).

The volatilization of NH_3 following land application has been shown to be significantly correlated to meteorological and soil factors such as air and soil temperature, humidity, solar radiation, rainfall and wind speed at the time of, and after, application (e.g. Braschkat et al., 1997; Génermont and Cellier, 1997; Menzi et al., 1998; Misselbrook et al., 2005; Moal et al., 1995; Søgaard et al., 2002; Sommer and Hutchings, 2001). In Blanes-Vidal et al. (2012) seasonal NH_3 concentrations measured at five rural locations during spring and summer seasons were 85 ± 25% and 20 ± 26% higher than annual averages at each region (mean ± standard deviation), while NH_3 concentrations during autumn and winter were lower than annual averages (−47 ± 2% and −58 ± 10%, respectively). Similar patterns were found by Skjøth et al. (2011) which determined the annual course of NH_3 emissions for a site in Germany. A year of NH_3 monitoring carried out in Ireland identified a Pearson correlation of 0.9 with ambient temperature, with peak concentrations observed in June, showing trends coinciding with seasonally variable agricultural practices (Kelleghan et al., 2021). As well as seasonal variation, diurnal variation in emissions also occur as a result of indoor temperature and animal activity in-house (Kelleghan et al., 2020; Schauberger et al., 2018; Sun et al., 2008). As this variation in NH_3 emissions and concentrations occurs both seasonally and diurnally, it highlights the need for intensive long term monitoring of both.

Ammonia emissions have also been shown to be correlated to changes in climate e.g. global emissions increase of 42% for a warming of 5°C (Sutton et al., 2013). These long-term trends and projections, known as climate signals, have epidemiological relevance for human health. This means that expected global warming is counteracting the efforts to reduce NH_3 emissions.

For the 2050s, NH_3 emissions from confined livestock buildings, predominantly used for fattening pigs and poultry, are expected to increase by about 15 - 20% (relative to 2007) due to the increase of temperature (Simpson et al., 2014). The sensitivity to global warming for the investigated agricultural emission categories were assessed by Skjøth & Geels (2013), showing the highest sensitivity for livestock buildings and manure storage, manure handling, application of fertilizer, and grazing animals. Skjøth & Geels (2013) expect an emission increase up to 40% due to the global warming signal. Under the assumption of a constant linear trend of anthropogenic warming until 2050, NH_3 emissions from livestock buildings will increase by about 11% between 1981 and 2050 from livestock buildings. For the last four decades, a mean trend of a 1.6% increase per decade was determined for confined livestock buildings (Schauberger et al., 2018). For the storage of manure, Aarnink & Elzing found that a 1 Kelvin (K) increase in storage temperature would result in a 10% increase in the emission rate of NH_3 (Aarnink & Elzing, 1998). This emphasises the urgent need to reduce emissions as much as possible, as increasing emissions in the future due to climate change will exacerbate any associated environmental and human health impacts.

4. The Fate of Ammonia and Potential Health Implications

4.1. The transformation of NH_3 to $PM_{2.5}$

Once NH_3 is released from agricultural sources, it can then travel in a gaseous form through the atmosphere for short or long distances (Philippe et al., 2011). When NH_3 interacts with a surface, it adheres and is removed from the atmosphere (i.e. dry deposition to plant surfaces), or following chemical transformation, it is deposited remotely through rainfall (wet deposition) (Asman et al., 1998). The rate of both dry and wet deposition is dependent on a number of factors including meteorological conditions, the physical and chemical properties of the pollutant and surrounding surface conditions, and concentrations of other atmospheric pollutants (Doyle et al., 2017).

Chemical reactions in the atmosphere involving NH_3 contribute significantly to the generation of $PM_{2.5}$ (Giannakis et al., 2019). Gaseous NH_3 reacts with aerosols containing sulphuric and nitric acids to create particulates such as ammonium nitrate (NH_4NO_3), ammonium sulphate ($(NH_4)_2SO_4$), and ammonium chloride (NH_4CL) (Gong et al., 2013). Bauer et al. (2016) explains how phase partitioning between inorganic aerosols is driven primarily by NH_3.

$$2NH_3 + H_2SO_4 \rightarrow (NH_4)_2SO_4 \qquad (2a)$$

$$NH_3 + H_2SO_4 \rightarrow (NH_4)HSO_4 \qquad (2b)$$

$$NH_3 + HNO_3 \leftarrow\rightarrow (NH_4)NO_3 \qquad (2c)$$

Equation 2. The conversion of NH_3 to ammonium sulphate and ammonium nitrate (Mensink & Deutsch, 2008)

Gaseous NH_3 is highly reactive in the atmosphere and is deposited quickly, with a deposition velocity reaching 14 cm/second (Phillips et al., 2004). High concentrations of NH_3 deposited locally can have serious effects on foliage and vegetation (Doyle et al., 2017; Krupa, 2003). In comparison, resulting NH_4^+ is deposited at a much slower rate, usually through wet deposition, at a rate of approximately 0.2 cm/second (Asman et al., 1998; Phillips et al., 2004). This transformation of gaseous NH_3 to NH_4^+ aerosols can allow for this pollutant to move much further from the point source, and be deposited through wet deposition (Phillips et al., 2004).

Despite the significant role that NH_3 has in the formation of $PM_{2.5}$, it has been relatively lagging in terms of regulation when in comparison to other precursors such as SO_2 and NO_x (Megaritis et al., 2013). In fact, NH_3 can increase the rate of reactions in the atmosphere of both SO_2 and NO_x, resulting in an even larger concentration of particulates (Brunekreef et al., 2015). In Europe for example, the NEC Directive required that in 2020, emissions of SO_2 and NO_x were to be reduced by 59% and 42% respectively, but NH_3 emissions were to be reduced by only 6% in comparison (Brunekreef et al., 2015). In the US, while the Clean Air Act (CAA) grants federal authorities the permission to regulate NH_3, and the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) requires some reporting of this pollutant, there are no definitive regulations in terms of emission thresholds for NH_3 (USDA Agricultural Air Quality Task Force, 2014). China, although considered as a global ammonia hotspot, does not regulate NH_3 in any way (Plautz, 2018).

Kanter et al. (2020) provides an excellent overview on how environmental sinks (such as water and climate) are often put to the front of decision making in terms of policies regarding nitrogen emissions. From their examination of 2726 policies across 186 countries, only 28 of 1390 sink-focused policies addressed the impacts of nitrogen across multiple-sinks. This can have inadvertent impacts on other sectors, and cause pollution swapping to occur. For example, to reduce nitrate run-off into waterways, the US increased the use of manure lagoons which are a large source of NH_3 emissions (Aillery et al., 2005). Several studies have suggested that one of the most efficient forms of $PM_{2.5}$ reduction is to reduce agricultural NH_3 emissions, particularly in areas where agriculture is a key contributor to reduced air quality (e.g. Backes et al., 2016; Megaritis et al., 2013; Zhang et al., 2020). By reducing the formation of $PM_{2.5}$, the associated health impacts of this pollutant would also be reduced. Therefore, an integrated approach for regulating precursors of $PM_{2.5,}$ particularly with a focus on NH_3 and health, would be highly beneficial to reduce the overall creation of secondary $PM_{2.5.}$

4.2. How NH_3 and $PM_{2.5}$ impact human health

NH_3 is an irritant, which can have severe effects on human health when over exposure occurs (Sundblad et al., 2004). In an agricultural setting, exposure to extremely high levels of NH_3 is uncommon, often occurring only during farming accidents (National Research Council (US) Committee, 2008). However, exposure to lower concentrations over longer periods may still have a negative impact on human health. The most frequently reported health complaints from NH_3 exposure include eye, nose, and throat irritation, headache, nausea, diarrhea, hoarseness, sore throat, cough, chest tightness, nasal congestion, palpitations, shortness of breath, stress, drowsiness, and alterations in mood (Schiffman and Williams, 2005; Wing and Wolf, 2000) (Figure 3).

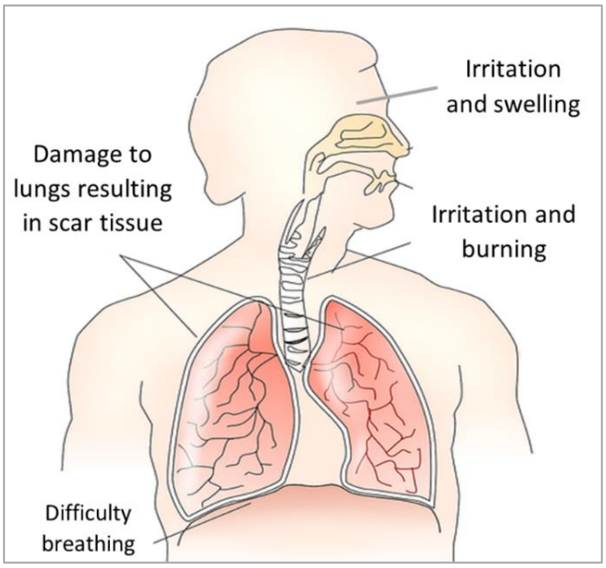

Figure 3. The direct effects of NH_3 on the respiratory tract, showing some of the most commonly reported symptoms

Irritation from exposure to ammonia can begin at concentrations as low as 5 ppm (eye discomfort) but are more noticeable at concentrations around 30 ppm, with irritation beginning to occur in the upper respiratory tract (National Research Council (US) Committee, 2008; Sundblad et al., 2004). Acute or accidental exposures to high concentrations (usually above 50 ppm) of NH_3 gases can result in tracheal burns as well as damage to bronchial tissues, but exposures such as these are quite rare thanks to occupational exposure limits and health and safety protocols in many work places (National Research Council (US) Committee, 2008). Although exposure to high levels of NH_3 are rare, some agricultural occupations (such as intensive livestock production housing) can result in longer exposure times to levels of NH_3 that would not occur in other occupations (Naseem & King, 2018). It is therefore important to ensure exposure within such facilities (and other similar occupations) is within regulated limits. The 2018 Code of Practice for the Chemical Agents Regulations (Health and Safety Authority, 2018) based on the European standards states that the occupational exposure limit value for NH_3 should be no more than 20 ppm (14 mg/m^3) over an eight hour interval and no more than 50 ppm (36 mg/m^3) over a 15 minute interval, while the US exposure limits for NH_3 are 25 ppm (18 mg/m^3) for eight-hour intervals (CDC, 2019).

Although direct exposure to agricultural NH_3 can be problematic for human health, when assessed as a precursor to $PM_{2.5}$, it has a much greater potential impact to human health. The World Health Organisation (2013) describes $PM_{2.5}$ as particles which are small enough to penetrate the thoracic region of the respiratory system once inhaled. Long-term exposure to $PM_{2.5}$ is considered a higher risk to human health than exposure to PM_{10} (Cohen et al., 2017; Erisman & Schaap, 2004). It is estimated that over 90% of people from across the world are exposed to $PM_{2.5}$ concentrations that exceed the WHO guidelines of 10 µg/m^3 (Health Effects Institute, 2019). Exposure to $PM_{2.5}$ can have adverse effects on lung development in children and can cause respiratory irritation in people with pre-existing conditions such as asthma and heart disease (World Health Organisation, 2013). In 2015, 2017, and 2019, exposure to $PM_{2.5}$ pollution posed a significant risk to premature mortality and declining health as stated in the corresponding Global Burden of Disease (GBD) studies (Bu et al., 2021; Collaborators GBD 2019 Risk Factors, 2020; Lelieveld et al., 2015). The 2017 GBD study found that exposure to $PM_{2.5}$ was responsible for 4.58 million deaths globally (Bu et al., 2021). In 2015, outdoor $PM_{2.5}$ pollution was identified as the fifth greatest risk factor for health, with ambient $PM_{2.5}$ exposure responsible for 4.2 million deaths worldwide. Increasing $PM_{2.5}$ concentrations can cause serious implications to human health, such as worsening asthma symptoms (18% increase in occurrence with an increase of 10 µg/m^3) and an increased risk of cardiopulmonary mortality (a rise of 6 – 13% per 10 µg/m^3 $PM_{2.5}$ under long-term exposure) (WHO, 2013; Yu et al., 2000). Reduced life expectancy is also associated with increasing $PM_{2.5}$ levels, as is highlighted in a study conducted by Apte et al. (2018). These authors state that the probability of death above the age of 60 increases with exposure to $PM_{2.5}$ depending on risk factors and disease rates. For example, in low-income and high mortality rate countries such as areas of South Asia, the probability of living to the age of 85 from 60 years old would increase by 20% if the risk of exposure to $PM_{2.5}$ was lowered. In high income areas where exposure to $PM_{2.5}$ is primarily low, the survival rate for this 25-year age range is approximately 50%. In Europe, modelled $PM_{2.5}$ concentrations were found to peak in central Europe with annual mean concentrations ranging between 16 – 18 µg/m^3 (Apte et al., 2018). Although this range is below the threshold limit laid out by European standards of 25 µg/m^3, it is in exceedance of the WHO guideline of 10 µg/m^3. Apte et al. (2018) stated that in 2016, 95% of the population across the globe resided in areas where $PM_{2.5}$ exceeded these WHO guidelines. In the US, the reported national average for $PM_{2.5}$ concentrations in 2019 was approximately 8.5 µg/m^3, which is below the US standard of 12 µg/m^3 for primary $PM_{2.5}$ and 15 µg/m^3 for secondary $PM_{2.5}$ (US EPA, 2020).

5. $PM_{2.5}$, Agriculture and Health

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

There is currently a chain that exists between NH_3 emissions, the formation of $PM_{2.5}$, and the subsequent impacts that this $PM_{2.5}$ has on human health. Increasing $PM_{2.5}$ emissions are likely to cause an elevated risk of premature mortality, aggravation of pre-existing conditions, and an overall decline in health among at risk populations (e.g. Lelieveld et al., 2015; Spencer and Van Heyst, 2018; World Health Organisation, 2013). To break this chain (or at least to significantly reduce associated impacts), reducing potential emissions of NH_3 primarily from agriculture has been highlighted as a key approach (Gu et al., 2021). Reducing NH_3 emissions from agriculture would aid in reducing $PM_{2.5}$ generation, leading to better air quality and reduced human health impacts.

Lelieveld et al. (2015) estimated that in 2010, 3.15 million premature deaths globally were a result of exposure to $PM_{2.5}$, with 650,000 of these directly resulting from agricultural emissions. Health impacts which were analysed in this study were chronic obstructive pulmonary disease (COPD), acute lower respiratory illness (ALRI), cerebrovascular disease (CEV), ischaemic heart disease (IHD) and lung cancer (LC). The study provided information relating to the top 15 countries in 2010 where premature mortality is linked to outdoor air pollution, specifically $PM_{2.5}$ and ozone (O_3). It is important to note that although O_3 is included in the statistics within this study, only 142,000 of the deaths mentioned were attributable to O_3 exposure and were linked only to COPD. In five of these 15 countries, air pollution resulting from agriculture was the leading contributor to premature deaths, and the second leading cause in several others (Lelieveld et al., 2015). For the people who died prematurely as a result of $PM_{2.5}$ exposure, the following percentage of these deaths were attributed to outdoor air pollution ($PM_{2.5}$ and O_3 exposure) resulting from agriculture: Ukraine (52%), Germany (45%), Russia (43%), Japan (38%), and Turkey (29%) (Figure 4). The second leading cause of premature mortality linked to $PM_{2.5}$ (and O_3) in these countries were power generation (Russia, Turkey & Ukraine), land traffic (Germany) and industry (Japan).

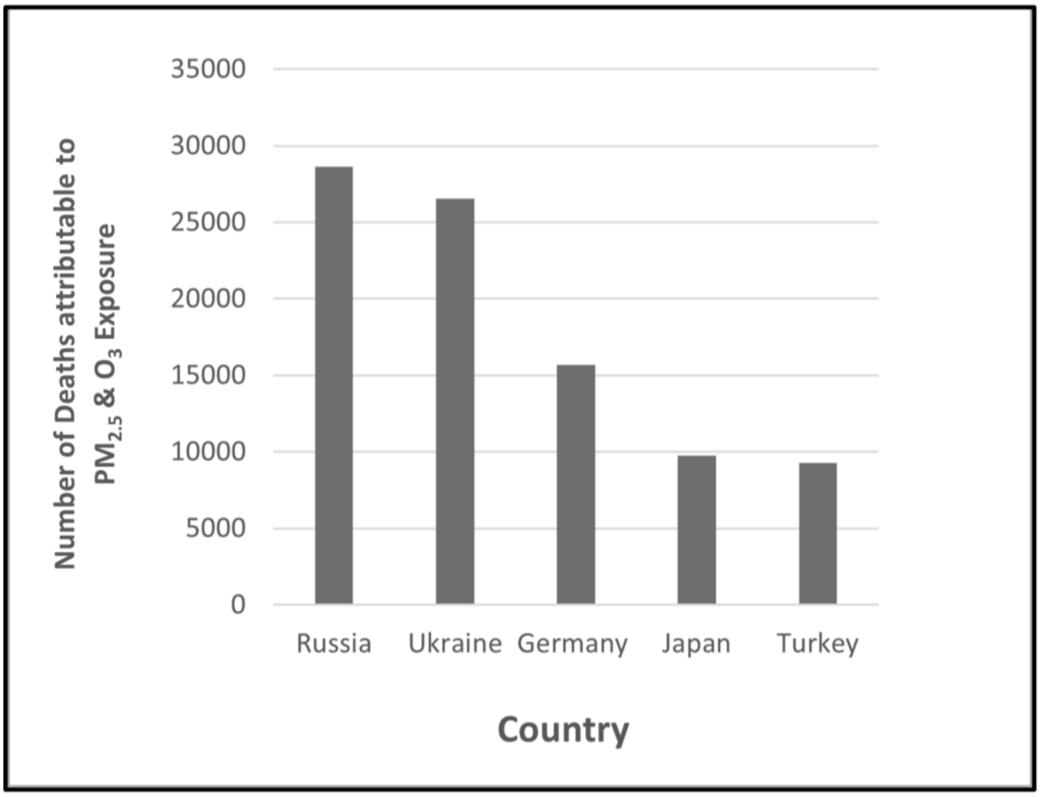

Figure 4. Number of premature deaths (age > 5 or < 30) as a result of exposure to $PM_{2.5}$ and O_3 from agriculture in 2010 (data obtained from Lelieveld et al., 2015)

A similar study conducted by Malley et al. (2021) assessed premature mortality as a result of exposure to $PM_{2.5}$ attributable to agricultural emissions using information from the 2019 GBD. Adults over 30 years of age were examined for COPD, IHD, ischaemic stroke (IS), LC, and Type 2 diabetes. Children less than 5 years of age were assessed for lower respiratory infections. The authors estimate that in 2018, 44 million tonnes of NH_3 were emitted globally, mostly due to manure management and application. This study found that the health burden (i.e., premature deaths) associated with $PM_{2.5}$ as a result of agriculture was 537 thousand deaths, 18% less than that of Lelieveld et al. (2015). Of these deaths, 358,000 (9% of all $PM_{2.5}$ associated mortalities) were a direct result from the contribution of NH_3 emissions to $PM_{2.5}$

formation. The percentage of premature deaths as a result of exposure to $PM_{2.5}$ from NH_3 emissions was highest in North America (90%), East and Southeast Asia (88%), and Western Europe (79%) (Malley et al., 2021).

Balasubramanian et al. (2021) assessed various literature on $PM_{2.5}$ from the global food cycle. This study found that contributors of particular importance included livestock production, land-use changes, manure management and agricultural waste burning. From this analysis, Balasubramanian et al. (2021) estimated that 890,000 annual deaths attributable to $PM_{2.5}$ (a total of 23% of deaths resulting from $PM_{2.5}$ exposure based on the 2015 GBD study) are a result of the global food system. Their work highlighted that these deaths could be higher due to underestimated NH_3 emissions, and the overall scarcity of emission data from all stages of the global food system. Thakrar et al. (2020) studied anthropogenic emissions of primary and secondary $PM_{2.5}$ in the US, and the activities and processes which contribute to their release and formation. The authors used models (used for fine scale pollution damage mapping) to assess the deaths attributable to these activities and processes. Thakrar et al. (2020) linked a total of 100,000 (maximum: 107,000, minimum: 88,000) deaths attributable to primary $PM_{2.5}$ and secondary $PM_{2.5}$ precursors in the US (SO_2, NO_x and NH_3). Of five major sectors associated with emissions, they ranked agriculture as the third highest regarding associated deaths (19,000), above residential emissions (12,700) and the electricity sector (10,200). This work estimated livestock rearing and fertilizer applications as the activities which contributed the most to these agricultural deaths (8,400 and 3,700 respectively). Approximately a fifth of the total 100,000 deaths predicted were associated directly with the precursor NH_3 (17,400), higher than that of SO_2 (10,900) and only slightly less than NO_x (19,100). As mentioned previously, NO_x and SO_2 are tightly regulated in terms of air quality in many countries, whereas the targets for the reduction of NH_3 are much less in comparison. One of the recommendations from Thakrar et al. (2020) is to tighten regulations on emissions (particularly NH_3) from agriculture, thus adding to the suggestion that reducing NH_3 emissions will subsequently reduce health impacts from $PM_{2.5.}$ Similarly, Balasubramanian et al. (2021) suggest that air pollution policies and legislation should include NH_3 given how imperative it is in the formation of $PM_{2.5}$.

Erisman & Schaap (2004) state that approximately 50% of all $PM_{2.5}$ in Europe arises from the production of secondary inorganic aerosols from NH_3. In agricultural dominated regions such as North Carolina in the US, the limitation of $PM_{2.5}$ formation could be explained by acid-gas (sulphate and nitrate) availability relative to NH_3 (Walker et al., 2006). Behera & Sharma (2010) examined the formation of $PM_{2.5}$ from NH_3 in the Ganga Basin region of India. This study concluded that the majority of chemical components of the $PM_{2.5}$ examined were $(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl. The authors stated that 30% of the $PM_{2.5}$ fraction mass was attributed to inorganic secondary aerosol formation from NH_3 reactions, with a particular emphasis on the contribution NH_3 has during the winter months. Similarly, Bauer et al. (2016) examined the input which agriculture has on the formation of $PM_{2.5}$ using the NASA Goddard Institute for Space Studies Earth System ModelE2 (Bauer et al., 2016). The results of this examination found that ammonium and nitrate were the primary contributors to the formation of $PM_{2.5}$ from agricultural pollution in the Eastern US, China, and Europe. This study found that in the US (particularly in the East) there was an annual $PM_{2.5}$ concentration above 14 $\mu g/m^{3}$, with agricultural pollution being responsible for 30% of all fine particulate formation. In contrast, $PM_{2.5}$ as a result of agriculture was determined to be responsible for 55% of all human-activity induced pollution in Europe (Bauer et al., 2016). Pinder and Adams (2007) state that in the US, reducing NH_3 by 23% (January) and 21% (July) can result in a 2 $\mu g/m^3$ reduction in $PM_{2.5}$. This reduction in $PM_{2.5}$ would see 70% of Eastern US areas which currently exceed 15 $\mu g/m^3$ achieve annual averages below this value.

Climate signals can have epidemiological relevance for human health, as mentioned previously. In particular, the climate signal for NH_3 emissions was shown by Geels et al. (2015) through a simulation for the 2080s. This simulation found that, in regions with high NH_3 emissions such as Germany, Poland, Netherlands and Belgium, $PM_{2.5}$ concentrations will be affected, which will result in a Europe-wide increase of chronic mortality by 4%. Pozzer et al. (2017) showed that a 50% reduction of agricultural emissions could prevent the mortality attributable to air pollution by about 250,000 people per year worldwide, amounting to reductions of 30%, 19%, 8%, and 3% over North America, Europe, East Asia, and South Asia, respectively. Megaritis et al. (2013) utilised a chemical transport model (CTM) to assess the potential of reducing $PM_{2.5}$ in response to 50% reductions in precursors (SO_2, NO_x and NH_3) and seasonal temperature changes (winter and summer) across Europe. Reducing NH_3 emissions was found to be the most effective control method to reduce $PM_{2.5}$ emissions in both summer (5.5% $PM_{2.5}$ reduction) and winter (4% $PM_{2.5}$ reduction) periods. These reductions were highest in Western and Central Europe, with $PM_{2.5}$ reductions of 2.3 µg/m^3 (15%, summer period) and 1.5 µg/m^3 (10%, winter period) respectively. Reducing NH_3 emissions was found to greatly reduce the formation of ammonium nitrate and ammonium sulphate, leading to potentially substantial reductions in total $PM_{2.5}$ (Megaritis et al., 2013) Although reducing NO_x and SO_2 also had an impact on $PM_{2.5}$, Megaritis et al. (2013) states that there is high spatial variability with NO_x reductions, and SO_2 is dependent on the availability of NH_3. Not only does the availability of NH_3 influence the generation of ammonium sulphate, but it has also been suggested that the presence of nitrates in the $PM_{2.5}$ fraction is more damaging to health than other particle components (such as NO_x and SO_2), aiding in the need for a greater focus on the regulation of NH_3 emissions as a precursor to $PM_{2.5}$ (Brunekreef et al. 2015).

Rural populations living near agricultural and animal production activities are exposed to different environmental factors, including NH_3 exposures (Blanes-Vidal, 2017). Various authors have demonstrated an increased occurrence of physical symptoms and conditions, such as coughs, wheezing, nasal irritation, shortness of breath and asthma, among these populations (Blanes-Vidal et al., 2014; Radon et al., 2001; Schinasi et al., 2011). Exposures above irritation limits of these various pollutants (such as NH_3, H_2S etc.) can cause a series of health symptoms through direct physiological mechanisms. Typically, these symptoms occur at the time of exposure and subside after a short interval (Schiffman et al., 2004). However, for sensitive individuals such as children, the elderly, and asthmatic patients, exposure to this air pollution may induce health symptoms that persist for longer periods of time as well as aggravate existing medical conditions (Schiffman et al., 2004). Heederik et al. (2007) stated that determining which health effects may arise because of particular gaseous pollution (from NH_3, H_2S or CO_2) is controversial and has not been well examined. Other articles such as those by Gehring et al. (2010) and Ryan & Holguin (2010) suggest that exposure to NH_3, and $PM_{2.5}$ resulting from NH_3, can trigger asthmatic reactions in children with pre-existing asthmatic conditions, but more importantly could also play a role in the onset of asthma in very young children. More recent studies such as Pavilonis et al. (2013) and Holst et al. (2018) aimed to assess the role exposure to agricultural emissions (such as NH_3 and $PM_{2.5}$) may have on early onset asthma in children. Pavilonis et al. (2013) showed that children living in the vicinity of animal production facilities, with a larger relative environmental exposure to gases from animal wastes, had a significantly increased odds of both asthma (OR=1.51, p=0.014 asthma) and medication for wheeze (OR=1.38, p=0.023) (Pavilonis et al., 2013). Holst et al. (2018) examined a population of children who developed asthma from their first to sixth birthday (n = 335,629) in Denmark and aimed to determine if ammonia and ammonium played any part in the development of this condition. These authors used national pollutant concentration information from the year 2008 for their study and found that the annual mean concentration was 1.99 ppb (~ 1.4 µg/m^3) for NH_3, 1.19 ppb (~ 0.8 µg/m^3) for NH_4^+, and 7.10 µg/m^3 for $PM_{2.5}$. From their study, Holst et al.

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

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

5.2. The economic benefit of reducing NH_3 and $PM_{2.5}$

Policies and decisions are often made on the basis of social costs and reduction benefits, and this is no different for NH_3 or $PM_{2.5}$ (Brink et al., 2011). In this regard, social costs can be considered as anything an individual or a community may have to change or limit when modifying their nitrogen management techniques. Incorporating NH_3 mitigation measures into policy and implementation of these techniques in agricultural practices could impose potentially significant costs. For example, Zhang et al. (2020) states that a possible 38 – 67% of NH_3 from agriculture could be mitigated at a cost of 6 – 11 billion US$ (€5.2 – €9.5 billion) in China. In comparison, the societal benefits are considered to all positively contribute to human welfare and can assist in both protecting human health and benefiting the economy over time. Where the mitigation measures for China were upwards of 6 billion US$, the overall societal benefits because of these mitigations were estimated at 18 – 42 billion US$ (~ €15.8 - €36.9 billion) (Zhang et al., 2020). In the EU, a mean annual cost of health to Member States ranges between €2 – €36 (~ US$2.50 - $41) per kg N for NH_3 (Brink et al., 2011). Pinder and Adams (2007) state that in the US, reducing NH_3 (particularly in the winter periods) is significantly more cost effective for $PM_{2.5}$ reductions than reducing NO_x and SO_2 in regions across the Eastern US. Gu et al., (2021) states that in comparison to the reduction methods available for nitrogen oxide emissions, the abatement of ammonia on a global level is only 10% of the cost associated with NO_x reductions.

Reducing NH_3 emissions can lower $PM_{2.5}$ concentrations, and contribute to a reduction in premature mortality globally, subsequently aiding in both economic and health cost savings. Giannadaki et al. (2018) modelled that reducing agricultural emissions (particularly NH_3) could result in the prevention of over 200,000 deaths per year across 59 countries. These authors examined 59 countries and applied a "value of statistical life" (the willingness to pay for avoiding the risk of a premature death) to monetize potential health outcomes by reducing agricultural emissions. An estimation of the costs which would be incurred to reduce NH_3 emissions to a level which meets national reduction commitments were also highlighted in this study. Giannadaki et al. (2018) states that this reduction would cost a total of €70 million - €89 million (€43 - €2176 annually per farm in the EU, depending on abatement strategies implemented and current farm emissions). Within the EU, this decrease in emissions could see a reduction in mortality rate of 18%, resulting in an annual saving to the economy of approximately €75.6 billion (~ US$86 billion). Giannakis et al. (2019) estimates that the cost to implement all four major NH_3 abatement strategies to decrease emissions (including reduced nitrogen feed, manure storage, low emission fertilizer application techniques and animal housing) to the required EU levels would cost approximately €4.3 billion (5.0 billion US$) for Europe alone. These authors state that implementing these abatement strategies would have the greatest effect on premature mortality reduction in Germany, followed by the United Kingdom and Italy with reductions of 930, 928 and 448 deaths per year, respectively.

In the US, agriculture is considered as one of the leading causes of gross external damage (economic loss) due to premature mortality, costing the economy approximately 230 billion US$ (€197 billion) (Tschofen et al., 2019). Within the agricultural sector, livestock and animal husbandry are the main contributors to gross external damage, accounting for approximately 50% of all economic loss due to premature death. A study of a pig facility in North Carolina suggested that $PM_{2.5}$ resulting from NH_3 leads to higher health costs than any other agricultural pollutant, costing the US healthcare system anywhere between US$10 (~ €8.50) and US$73 (~ €64) per kilogram of NH_3 (Stokstad, 2014). Domingo et al. (2021) states that improving the efficiency of NH_3 application to land (either by method of application or timing of application period) at a cost of 0.8 to 3.2 US$ per kg/NH_3 can see an economic benefit between 33.4 and

42.4 US$ per kg/NH_3 solely due to the reduction of $PM_{2.5}$ formation. In China, the previously mentioned reduction of NH_3 by 38 – 67% was estimated to reduce premature mortalities by between 90 – 240 thousand people, due to a reduction in $PM_{2.5}$ concentrations of approximately 8 – 20% (Zhang et al., 2020). In economic terms, this could save over 10 – 26 billion US$ in health costs in China alone.

6. Results and Discussion

Agriculture is the primary contributor to global NH_3 emissions, playing a major role as a precursor to secondary $PM_{2.5}$ (Brunekreef et al., 2015; Erisman and Schaap, 2004; Thakrar et al., 2020). The production of livestock, storage of associated manure, and its use as organic fertilizer, alongside synthetic fertilizers, are key components in the emission of NH_3 from agriculture (Environmental Protection Agency, 2018; Ma et al., 2021; Zhang et al., 2017). The emission of NH_3 from agriculture varies from region to region and depends heavily on the source from which it is emitted (Balasubramanian et al., 2020; Singles et al., 1998). This high spatial variability of NH_3 alongside its seasonal and diurnal variations (Blanes-Vidal et al., 2008; Kelleghan et al., 2020) highlights the need for representative long term monitoring of both emissions and atmospheric concentrations of this pollutant. Incorporating alternative approaches such as switching from the use of splash plates to trailing shoe or injection for land application, or implementing manure store covers, offers ways of reducing NH_3 emissions (e.g., Hellsten, 2017; Xu et al., 2017).

The reduction of NH_3 emissions where possible is imperative, as current projections suggest emissions are set to continue to rise with increasing temperatures due to the current climate crisis (Skjøth & Geels, 2013;Sutton et al., 2013), alongside the increasing intensification of agriculture to meet global food demands (Malley et al., 2021). Failure to incorporate low emission techniques to reduce NH_3 will see an increase in negative impacts from this pollutant across the globe due to these predicted increases. It is clear that NH_3 emissions are responsible for a large portion of the $PM_{2.5}$ fraction of air pollution (Bauer et al., 2016; Pinder & Adams, 2007; Thakrar et al., 2020). Although current health impacts from NH_3 as an individual pollutant are more likely for those who work in or near sources (Naseem & King, 2018), the contribution of NH_3 to particulate matter formation has the potential to impact human health on a population level (e.g. Giannakis et al., 2019). This $PM_{2.5}$ can have negative effects on human health including the development of chronic health issues and premature death (e.g. Lelieveld et al., 2015; Thakrar et al., 2020). NH_3 contributes to approximately 50% of $PM_{2.5}$ in Europe, 30% in the US, and 15-23% of the $PM_{2.5}$ mass in the North China Plain (Bauer et al., 2016; Erisman & Schaap, 2004; Han et al., 2020).

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

Table 1. Emissions of NH_3 (%) from various sources and their percentage contribution to $PM_{2.5}$ in the 28 EU Member States (source data: European Environment Agency, 2021), and potential increases in these emissions 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 NH3 emissions (%) (based on 5* increase in temp, Sutton et al. 2013)**	**Potential contribution of NH_3 emissions to PM2.5 (%)**
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

Sutton et al. (2013) states that with an increase to global temperatures of 5°C, there is a predicted increase in global NH_3 emissions of 42%. Using this 42% increase estimate, the potential increase in NH_3 emissions for these sources in Europe, and the resulting fraction of $PM_{2.5}$ created is highlighted in Table 1. The three greatest emitters of NH_3 in Europe would see a growth in NH_3 emissions from the values mentioned above, to 40%, 38%, and 24% for cattle, organic and inorganic fertilizer use respectively. This would in turn result in a dramatic increase of the fraction of $PM_{2.5}$ as a result of NH_3 emissions from these sectors (20%, 19% and 12% respectively), ultimately placing a greater strain on human health in these areas. Although $PM_{2.5}$ as a result of NH_3 emissions is greatest in Europe, if current emissions are not reduced across the world, there is potential that the $PM_{2.5}$ fraction across the globe as a result of livestock production will increase. Increases such as these will have a significant impact on human health and economic costs associated with such across the globe. This highlights the need for rapid action to reduce NH_3 emissions to protect human health due to the current climate crisis.

The potential contribution to $PM_{2.5}$ production of NH_3 emissions from livestock reflect the necessity of incorporating NH_3 into policies to protect human health in addition to the environment. For example, in the EU, the NEC Directive focuses on reporting national NH_3 emissions (and deposition values) and setting emission limits, while also now requiring monitoring across sensitive habitats; where the Habitats Directive encourages Member States to set contribution limits to Natura 2000 sites and the Industrial Emission Directive sets emission limits for pig and poultry farms of certain sizes. These directives are typically subject to interpretation by each Member State with application varying from country to country. In the US, NH_3 is not considered as a criteria pollutant under the CAA which would allow for the regulation of this pollutant, a request that was rejected by the USEPA in 2018 (Stowell, 2018).

Reflecting on the data presented in Table 1, it is clear that livestock NH_3 emissions play a vital role in contributing to the $PM_{2.5}$ fraction, particularly in Europe, and should be incorporated in any future discussions in the regulation of both NH_3 emissions themselves, and the control of $PM_{2.5}$. Across the rest of the world, updating of emissions databases and monitoring of NH_3 should be a priority so as to understand the emissions and source contributions (such as above), and in turn, provide the ability to assess source contribution to $PM_{2.5}$ creation. As NH_3 is primarily released from agriculture, there must be a balance found between the control of these emissions and the ability of the industry to continue to meet the global food demand. Policy makers and stakeholders must fully understand the role that NH_3 plays in terms of its emission from various agricultural sectors, its reactions and transportation in the environment, particularly its role in the formation of $PM_{2.5}$ and the subsequent impact on human health. NH_3

is a difficult pollutant to measure due to the variability of emissions and concentrations in the environment, and so regulations must begin with the implementation of long-term monitoring to better understand the movement of NH_3 on a local, regional, or national scale. Understanding the beginning of this chain (i.e., how NH_3 enters and moves through the environment) will allow for better informed policies to be implemented to protect human health from the creation of $PM_{2.5}$ as a result of agricultural NH_3 emissions. There is an inherent need for future regulations and policies surrounding NH_3 and $PM_{2.5}$ to be reflective of both environmental protection and improving human health, while ensuring neither are impacted as a result of the other. By considering the issue of NH_3 abatement from a combined approach (i.e., assessing impacts on agriculture, the environment, and public health), a better outcome for all sectors could be reached while also reducing the socio-economic costs of NH_3 and $PM_{2.5}$. Through understanding the source sector contributions of NH_3 emissions, a focus could be placed on areas which would be high impact and low cost to implement reduction measures for NH_3, subsequently reducing $PM_{2.5}$ creation from this precursor (such as measures assessed by Buckley et al., 2020; Edwards, 2020; Hellsten, 2017). This can aid in the beginnings of legislation and policy discussions, and even the implementation of government grants and assistance to reach set targets (e.g. LESS scheme in Ireland (Government of Ireland, 2022)), providing stakeholders and policy makers with a clearer indication of where to start.

7. Conclusion

Numerous studies indicate that one of the most efficient ways to reduce $PM_{2.5}$ is to reduce global NH_3 emissions resulting from agriculture. This reduction in NH_3 can subsequently also have a large impact in the reduction of chronic health problems, decrease premature mortality across the globe and can lower monetary loss to national economies due to these premature deaths. Regulations surrounding NH_3 as a precursor to $PM_{2.5}$ formation is lacking and should be incorporated into future policy and regulations across the globe to aid in bettering human health from $PM_{2.5}$ exposure.

Acknowledgments

The authors would like to acknowledge the Department of Agriculture, Food, and the Marine and the researchers in the Triple A (Abating Ammonia in Agriculture) Project, particularly the coordinator Dr. Dominika Krol in Teagasc.

Funding: This research was undertaken as part of the Triple A (Abating Ammonia in Agriculture) Project (2019R554) funded by the Irish Department of Agriculture, Food, and the Marine.

References

Aarnink, A. J. A., & Elzing, A. (1998). Dynamic model for ammonia volatilization in housing with partially slatted floors, for fattening pigs. *Livestock Production Science*, *53*(2), 153–169. https://doi.org/https://doi.org/10.1016/S0301-6226(97)00153-X

Aarnink, A. J. A., Swierstra, D., Van Den Berg, A. J., & Speelman, L. (1997). Effect of type of slatted floor and degree of fouling of solid floor on ammonia emission rates from fattening piggeries. *Journal of Agricultural and Engineering Research*, *66*(2), 93–102. https://doi.org/10.1006/jaer.1996.0121

Aillery, M., Gollehon, N., Johansson, R., Kaplan, J., Key, N., & Ribaudo, M. (2005). *A Report from the Economic Research Service Managing Manure To Improve Air and Water Quality*. www.ers.usda.gov/

Anderson, N., Strader, R., & Davidson, C. (2003). Airborne reduced nitrogen: Ammonia emissions from agriculture and other sources. *Environment International*, *29*(2–3), 277–286. https://doi.org/10.1016/S0160-4120(02)00186-1

Apte, J. S., Brauer, M., Cohen, A. J., Ezzati, M., & Arden, C. (2018). *Ambient PM 2.5 Reduces Global and Regional Life Expectancy*. https://doi.org/10.1021/acs.estlett.8b00360

Asman, W. A. H., Sutton, M. A., & Schjorring, J. K. (1998). Ammonia: emission, atmospheric transport and deposition. *New Phytologist*, *139*(1), 27–48. https://doi.org/10.1046/j.1469-8137.1998.00180.x

Backes, A. M., Aulinger, A., Bieser, J., Matthias, V., & Quante, M. (2016). Ammonia emissions in Europe, part II: How ammonia emission abatement strategies affect secondary aerosols. *Atmospheric Environment*, *126*, 153–161. https://doi.org/10.1016/j.atmosenv.2015.11.039

Balasubramanian, S., Domingo, N. G. G., Hunt, N. D., Gittlin, M., Colgan, K. K., Marshall, J. D., Robinson, A. L., Azevedo, I. M. L., Thakrar, S. K., Clark, M. A., Tessum, C. W., Adams, P. J., Pandis, S. N., & Hill, J. D. (2021). The food we eat, the air we breathe: A review of the fine particulate matter-induced air quality health impacts of the global food system. *Environmental Research Letters*, *16*(10). https://doi.org/10.1088/1748-9326/AC065F

Balasubramanian, S., McFarland, D. M., Koloutsou-Vakakis, S., Fu, K., Menon, R., Lehmann, C., & Rood, M. J. (2020). Effect of grid resolution and spatial representation of nh3 emissions from fertilizer application on predictions of nh3 and pm2.5 concentrations in the united states corn belt. *Environmental Research Communications*, *2*(2). https://doi.org/10.1088/2515-7620/ab6c01

Bash, J. O., Cooter, E. J., Dennis, R. L., Walker, J. T., & Pleim, J. E. (2013). Evaluation of a regional air-quality model with bidirectional NH3 exchange coupled to an agroecosystem model. *Biogeosciences*, *10*(3), 1635–1645. https://doi.org/10.5194/bg-10-1635-2013

Bauer, S. E., Tsigaridis, K., & Miller, R. (2016). Significant atmospheric aerosol pollution caused by world food cultivation. *Geophysical Research Letters*, *43*, 5394–5400. https://doi.org/10.1002/2016GL068354

Behera, S. N., Sharma, M., Aneja, V. P., & Balasubramanian, R. (2013). Ammonia in the atmosphere: A review on emission sources, atmospheric chemistry and deposition on terrestrial bodies. *Environmental Science and Pollution Research*, *20*(11), 8092–8131. https://doi.org/10.1007/s11356-013-2051-9

Behera, S., Sharma, M., Aneja, V. P., & Balasubramanian, R. (2010). Investigating the potential role of ammonia in ion chemistry of fine particulate matter formation for an urban environment. *Science of the Total Environment*, *408*(17), 3569–3575. https://doi.org/10.1016/j.scitotenv.2010.04.017

Behera, S., Sharma, M., Aneja, V. P., & Balasubramanian, R. (2013). Ammonia in the atmosphere: A review on emission sources, atmospheric chemistry and deposition on terrestrial bodies. *Environmental Science and Pollution Research*, *20*(11), 8092–8131. https://doi.org/10.1007/s11356-013-2051-9

Blanes-Vidal, V., Baelum, J., Schwartz, J., & Christensen, L. P. (2014). Respiratory and sensory irritation symptoms among residents exposed to low-to-moderate air pollution from biodegradable wastes. *Journal of Exposure Science and Environmental Epidemiology*, *24*, 388–397. https://doi.org/10.1038/jes.2014.20

Blanes-Vidal, V., Hansen, M. N., Pedersen, S., & Rom, H. B. (2008). Emissions of ammonia, methane and nitrous oxide from pig houses and slurry: Effects of rooting material, animal activity and ventilation flow. *Agriculture, Ecosystems and Environment*, *124*, 237–244. https://doi.org/10.1016/j.agee.2007.10.002

Blanes-Vidal, V., Hansen, M. N., & Sousa, P. (2009). Reduction of Odor and Odorant Emissions from Slurry Stores by Means of Straw Covers. *Journal of Environmental Quality*, *38*(4), 1518–1527. https://doi.org/10.2134/jeq2008.0412

Blanes-Vidal, V., Nadimi, E. S., Ellermann, T., Andersen, H. V., & Løfstrøm, P. (2012). Perceived annoyance from environmental odors and association with atmospheric ammonia levels in non-urban residential communities: A cross-sectional study. *Environmental Health: A Global Access Science Source*, *11*(1), 1–10. https://doi.org/10.1186/1476-069X-11-27

Blanes-Vidal1, V. (2017). Living in the Clean, Stress-Free and Quiet Countryside: Does the Reality Match the Dream? Residential Exposure to Environmental Health Stressors in the Danish Countryside. *Human Ecology*, *45*(4), 545–552. https://about.jstor.org/terms

Blunden, J., & Westerman, P. W. (2008). Measurement and analysis of ammonia and hydrogen sulfide emissions from a mechanically ventilated swine confinement building in North Carolina. *Atmospheric Environment*, *42*(14), 3315–3331. https://doi.org/10.1016/J.ATMOSENV.2007.06.040

Bourdin, F., Sakrabani, R., Kibblewhite, M. G., & Lanigan, G. J. (2014). Effect of slurry dry matter content, application technique and timing on emissions of ammonia and greenhouse gas from cattle slurry applied to grassland soils in Ireland. *Agriculture, Ecosystems and Environment*, *188*(2014), 122–133. https://doi.org/10.1016/j.agee.2014.02.025

Bouwman, A. F., Boumans, L. J. M., & Batjes, N. H. (2002). Estimation of global NH3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global Biogeochemical Cycles*, *16*(2), 8–14.

Braschkat, J., Mannheim, T., & Marschner, H. (1997). Estimation of ammonia losses after application of liquid cattle manure on grassland. *Zeitschrift Für Pflanzenernährung Und Bodenkunde*, *160*(2), 117–123. https://doi.org/10.1002/jpln.19971600202

Bray, C. D., Battye, W. H., Aneja, V. P., & Schlesinger, W. H. (2021). Global emissions of NH3, NOx, and N2O from biomass burning and the impact of climate change. *Journal of the Air and Waste Management Association*, *71*(1), 102–114. https://doi.org/10.1080/10962247.2020.1842822

Brink, C., Van Grinsven, H., Jacobsen, B. H., Rabl, A., Gren, I., Holland, M., Hicks, K., Brouwer, R., Dickens, R., Willems, J., Termansen, M., Velthof, G., Alkemade, R., Oorschot, M. Van, & Webb, J. (2011). Chapter 22: Costs and benefits of nitrogen in the environment. In M.A. Sutton (Ed.), *The European Nitrogen Assessment* (pp. 513–540).

Cambridge University Press.

Brunekreef, B., Harrison, R. M., Künzli, N., Querol, X., Sutton, M. A., Heederik, D. J. J., & Sigsgaard, T. (2015). Reducing the health effect of particles from agriculture. *The Lancet Respiratory Medicine*, *3*(11), 831–832. https://doi.org/10.1016/S2213-2600(15)00413-0

Bu, X., Xie, Z., Liu, J., Wei, L., Wang, X., Chen, M., & Ren, H. (2021). Global PM2.5-attributable health burden from 1990 to 2017: Estimates from the Global Burden of disease study 2017. *Environmental Research*, *197*, 111123. https://doi.org/10.1016/J.ENVRES.2021.111123

Buckley, C., Krol, D., Lanigan, G. J., Donnellan, T., Spink, J., Hanrahan, K., Boland, A., Forrestal, P., Humphreys, J., Murphy, P., NiFhlatharta, N., Richards, K., Shalloo, L., Wall, D., Waters Editors, S., & Krol, D. J. (2020). *An Analysis of the Cost of the Abatement of Ammonia Emissions in Irish Agriculture to 2030*. https://www.teagasc.ie/media/website/publications/2020/NH3-Ammonia-MACC.pdf

CDC. (2019). *CDC - NIOSH Pocket Guide to Chemical Hazards - Ammonia*. https://www.cdc.gov/niosh/npg/npgd0028.html

CLRTAP. (2020). *Assessment Report on Ammonia – 2020*. https://doi.org/10.1038/s43016-020-0113-y

Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., & Dandona, R. (2017). Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. *The Lancet*, *389*(10082), 1907–1918.

Collaborators GBD 2019 Risk Factors. (2020). Global burden of 87 risk factors in 204 countries and territories, 1990–2019: a systematic analysis for the Global Burden of Disease Study 2019. *The Lancet*, *396*(10258), 1223–1249. https://doi.org/10.1016/S0140-6736(20)30752-2

Crippa, M., Janssens-Maenhout, G., Dentener, F., Guizzardi, D., Sindelarova, K., Muntean, M., Van Dingenen, R., & Granier, C. (2016). Forty years of improvements in European air quality: Regional policy-industry interactions with global impacts. *Atmospheric Chemistry and Physics*, *16*(6), 3825–3841. https://doi.org/10.5194/ACP-16-3825-2016

Domingo, N. G. G., Balasubramanian, S., Thakrar, S. K., Clark, M. A., Adams, P. J., Marshall, J. D., Muller, N. Z., Pandis, S. N., Polasky, S., Robinson, A. L., Tessum, C. W., Tilman, D., Tschofen, P., & Hill, J. D. (2021). Air quality-related health damages of food. *Proceedings of the National Academy of Sciences of the United States of America*, *118*(20). https://doi.org/10.1073/pnas.2013637118

Doyle, B., Cummins, T., Augustenborg, C., & Aherne, J. (2017). *Ambient Atmospheric Ammonia in Ireland, 2013-2014*. www.epa.ie

EDGAR. (2015). *EDGAR - Emissions Database for Global Atmospheric Research*. Global Air Pollutant Emissions - EDGAR v5.0. https://edgar.jrc.ec.europa.eu/gallery?release=v50_AP&substance=NH3&sector=TOTALS

EDGAR. (2017). *EDGAR - Emissions Data Maps*. https://edgar.jrc.ec.europa.eu/gallery.php?release=v50_AP&substance=NH3&sector=TOTALS

Edwards, R. (2020). *Low Emission Slurry Spreading Systems help to reduce ammonia emissions Robert Edwards Sustainable Land Management Branch*. www.daera-ni.gov.uk/services/daera-online-services, Environmental Protection Agency. (2018). *Ireland's Transboundary Gas Emissions 1990-2016* (Issue March). https://www.epa.ie/pubs/reports/air/airemissions/Irelands

EPA. (2021). *Licence Application Guidance: Assessment of the impact of ammonia and nitrogen on natura 2000 sites from intensive agriculture installations*. http://www.epa.ie/terminalfour/AppropAssess/index.jsp

Erisman, J. W., & Schaap, M. (2004). The need for ammonia abatement with respect to secondary PM reductions in Europe. *Environmental Pollution, 129*(1), 159–163. https://doi.org/10.1016/j.envpol.2003.08.042

European Economic Committee. (1992). Council directive 92/43/EEC of 21 May 1992 on the conservation of natural habitats and of wild fauna and flora. In *Off. J. Eur. Communities* (p. L206/7).

European Environment Agency. (2021, August 11). *National emissions reported to the Convention on Long-range Transboundary Air Pollution (LRTAP Convention)* . https://www.eea.europa.eu/data-and-maps/data/national-emissions-reported-to-the-convention-on-long-range-transboundary-air-pollution-lrtap-convention-15

European Union. (2010). *Directive 2010/75/EU of the European Parliament and of the Council of 24 November 2010 on industrial emissions (integrated pollution prevention and control)* (pp. 17–119). https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A32010L0075

European Union. (2016). *Directive (EU) 2016/2284 of the European Parliament and of the Council of 14 December 2016 on the reduction of national emissions of certain atmospheric pollutants, amending Directive 2003/35/EC and repealing Directive 2001/81/EC* (pp. 1–31). Off. J. Eur. Union L344/1 . https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=uriserv%3AOJ.L_.2016.344.01.0001.01.ENG

Faulkner, W. B., & Shaw, B. W. (2008). Review of ammonia emission factors for United States animal agriculture. *Atmospheric Environment*, *42*(27), 6567–6574. https://doi.org/10.1016/j.atmosenv.2008.04.021

Forrestal, P. J., Harty, M., Carolan, R., Lanigan, G. J., Watson, C. J., Laughlin, R. J., Mcneill, G., Chambers, B. J., & Richards, K. G. (2016). Ammonia emissions from urea, stabilized urea and calcium ammonium nitrate: Insights into loss abatement in temperate grassland. *Soil Use and Management*, *32*(June), 92–100. https://doi.org/10.1111/sum.12232

Fu, X., Wang, S. X., Ran, L. M., Pleim, J. E., Cooter, E., Bash, J. O., Benson, V., & Hao, J. M. (2015). Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model. *Atmospheric Chemistry and Physics*, *15*(12), 6637–6649.

García-Gómez, H., Garrido, J. L., Vivanco, M. G., Lassaletta, L., Rábago, I., Àvila, A., Tsyro, S., Sánchez, G., González Ortiz, A., González-Fernández, I., & Alonso, R. (2014). Nitrogen deposition in Spain: Modeled patterns and threatened habitats within the Natura 2000 network. *Science of the Total Environment*, *485–486*(1), 450–460. https://doi.org/10.1016/j.scitotenv.2014.03.112

Ge, X., Schaap, M., Kranenburg, R., Segers, A., Reinds, G. J., Kros, H., & De Vries, W. (2020). Modeling atmospheric ammonia using agricultural emissions with improved spatial variability and temporal dynamics. *Atmos. Chem. Phys*, *20*, 16055–16087. https://doi.org/10.5194/acp-20-16055-2020

Geels, C., Andersson, C., Hänninen, O., Lansø, A. S., Schwarze, P. E., Skjøth, C. A., & Brandt, J. (2015). Future premature mortality due to O<inf>3</inf>, Secondary inorganic aerosols and primary PM in Europe — Sensitivity to changes in climate, anthropogenic emissions, population and building stock. *International Journal of Environmental Research and Public Health*, *12*(3), 2837–2869. https://doi.org/10.3390/ijerph120302837

Gehring, U., Wijga, A. H., Brauer, M., Fischer, P., De Jongste, J. C., Kerkhof, M., Oldenwening, M., Smit, H. A., & Brunekreef, B. (2010). Traffic-related air pollution and the development of asthma and allergies during the first 8 years of life. *American Journal of Respiratory and Critical Care Medicine*, *181*(6), 596–603. https://doi.org/10.1164/rccm.200906-0858OC

Génermont, S., & Cellier, P. (1997). A mechanistic model for estimating ammonia volatilization from slurry applied to bare soil. *Agricultural and Forest Meteorology*, *88*(1–4), 145–167. https://doi.org/10.1016/S0168-1923(97)00044-0

Giannadaki, D., Giannakis, E., Pozzer, A., & Lelieveld, J. (2018). Estimating health and economic benefits of reductions in air pollution from agriculture. *Science of the Total Environment*, *622–623*, 1304–1316. https://doi.org/10.1016/j.scitotenv.2017.12.064

Giannakis, E., Kushta, J., Bruggeman, A., & Lelieveld, J. (2019). Costs and benefits of agricultural ammonia emission abatement options for compliance with European air quality regulations. *Environmental Sciences Europe*, *31*(1), 93. https://doi.org/10.1186/s12302-019-0275-0

Gong, L., Lewicki, R., Griffin, R. J., Tittel, F. K., Lonsdale, C. R., Stevens, R. G., Pierce, J. R., Malloy, Q. G. J., Travis, S. A., Bobmanuel, L. M., Lefer, B. L., & Flynn, J. H. (2013). Role of atmospheric ammonia in particulate matter formation in Houston during summertime. *Atmospheric Environment*, *77*, 893–900. https://doi.org/10.1016/J.ATMOSENV.2013.04.079

Government of Ireland. (2022, February). *Low Emission Slurry Spreading (LESS) Equipment*

Scheme. https://www.gov.ie/en/service/d4b800-low-emission-slurry-spreading-less-equipment-scheme/

Granier, R., Guingand, N., & Massabie, P. (1996). Influence of hygrometry, temperature and air flow rate on the evolution of ammonia levels. *Journées de La Recherche Porcine*, *28*, 209–216.

Grant, R. H., & Boehm, M. T. (2020). Ammonia emissions from differing manure storage facilities at two midwestern free-stall dairies. *Atmosphere*, *11*(10). https://doi.org/10.3390/atmos11101108

Gu, B., Zhang, L., Dingenen, R. Van, Vieno, M., Grinsven, H. J. Van, Zhang, X., Zhang, S., Chen, Y., Wang, S., Ren, C., Rao, S., Holland, M., Winiwarter, W., Chen, D., Xu, J., & Sutton, M. A. (2021). Abating ammonia is more cost-effective than nitrogen oxides for mitigating PM2.5 air pollution. *Science*, *374*(6568), 758–762. https://doi.org/10.1126/SCIENCE.ABF8623/SUPPL_FILE/SCIENCE.ABF8623_SM.PDF

Han, X., Zhu, L., Liu, M., Song, Y., & Zhang, M. (2020). Numerical analysis of the impact of agricultural emissions on PM2.5 in China using a high-resolution ammonia emissions inventory. *Atmospheric Chemistry and Physics Discussions*, *3*, 1–31. https://doi.org/10.5194/acp-2019-1128

Häni, C., Sintermann, J., Kupper, T., Jocher, M., & Neftel, A. (2016). Ammonia emission after slurry application to grassland in Switzerland. *Atmospheric Environment*, *125*, 92–99. https://doi.org/10.1016/j.atmosenv.2015.10.069

Health and Safety Authority. (2018). *Health and Safety Authority Draft 2018 Code of Practice for the Chemical Agents Regulations 2*.

Health Effects Institute. (2019). State of Global Air 2019. *Health Effects Institute.*, 24. https://doi.org/https://www.stateofglobalair.org/sites/default/files/soga_2019_report.pdf

Heederik, D., Sigsgaard, T., Thorne, P. S., Kline, J. N., Avery, R., Bønløkke, J. H., Chrischilles, E. A., Dosman, J. A., Duchaine, C., Kirkhorn, S. R., Kulhankova, K., & Merchant, J. A. (2007). Health effects of airborne exposures from concentrated animal feeding operations. *Environmental Health Perspectives*, *115*(2), 298–302. https://doi.org/10.1289/ehp.8835

Hellsten, S. (2017). *Ammonia emissions in Sweden Inventories, projections and potential for reduction*. www.ivl.se

Hellsten, S. (2017b). *Ammonia emissions in Sweden Inventories, projections and potential for reduction*. www.ivl.se

Holst, G., Thygesen, M., Pedersen, C. B., Peel, R. G., Brandt, J., Christensen, J. H., Bønløkke, J. H., Hertel, O., & Sigsgaard, T. (2018). Ammonia, ammonium, and the risk of asthma. *Environmental Epidemiology*, *2*(3), e019. https://doi.org/10.1097/ee9.0000000000000019

Hyde, B. P., Carton, O. T., O'Toole, P., & Misselbrook, T. H. (2003). A new inventory of ammonia emissions from Irish agriculture. *Atmospheric Environment*, *37*(1), 55–62. https://doi.org/10.1016/S1352-2310(02)00692-1

Jiang, J., Stevenson, D. S., Uwizeye, A., Tempio, G., & Sutton, M. A. (2020). A climate-dependent global model of ammonia emissions from chicken farming. *Biogeosciences*, *Preprint*. https://doi.org/10.5194/bg-2020-192

Kanter, D. R., Chodos, O., Nordland, O., Rutigliano, M., & Winiwarter, W. (2020). Gaps and opportunities in nitrogen pollution policies around the world. *Nature Sustainability*, *3*(11), 956–963. https://doi.org/10.1038/s41893-020-0577-7

Kelleghan, D. B., Hayes, E. T., Everard, M., & Curran, T. P. (2020). *Assessment of the Impact of Ammonia Emissions from Intensive Agriculture Installations on Special Areas of Conservation and Special Protection Areas*. www.epa.ie

Kelleghan, D. B., Hayes, E. T., Everard, M., Keating, P., Lesniak-Podsiadlo, A., & Curran, T. P. (2021). Atmospheric ammonia and nitrogen deposition on Irish Natura 2000 sites: Implications for Irish agriculture. *Atmospheric Environment*, *261*, 118611. https://doi.org/10.1016/J.ATMOSENV.2021.118611

Krupa, S. V. (2003). Effects of atmospheric ammonia (NH3) on terrestrial vegetation: A review. *Environmental Pollution*, *124*(2), 179–221. https://doi.org/10.1016/S0269-7491(02)00434-7

Kupper, T., Häni, C., Neftel, A., Kincaid, C., Bühler, M., Amon, B., & VanderZaag, A. (2020). Ammonia and greenhouse gas emissions from slurry storage - A review. *Agriculture, Ecosystems & Environment*, *300*, 106963. https://doi.org/10.1016/J.AGEE.2020.106963

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., & Pozzer, A. (2015). The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature*, *525*(7569), 367–371. https://doi.org/10.1038/nature15371

Lichiheb, N., Myles, L., Personne, E., Heuer, M., Buban, M., Nelson, A. J., Koloutsou-Vakakis, S., Rood, M. J., Joo, E., Miller, J., & Bernacchi, C. (2019). Implementation of the effect of urease inhibitor on ammonia emissions following urea-based fertilizer application at a Zea mays field in central Illinois: A study with SURFATM-NH3 model. *Agricultural and Forest Meteorology*, *269–270*, 78–87. https://doi.org/https://doi.org/10.1016/j.agrformet.2019.02.005

Liu, Z., Liu, Y., Murphy, J. P., & Maghirang, R. (2017). Ammonia and methane emission factors from cattle operations expressed as losses of dietary nutrients or energy. *Agriculture (Switzerland)*, *7*(3), 1–12. https://doi.org/10.3390/agriculture7030016

Loftus, C., Yost, M., Sampson, P., Torres, E., Arias, G., Vasquez, V. B., Hartin, K., Armstrong, J., Tchong-French, M., Vedal, S., Bhatti, P., & Karr, C. (2015). Ambient ammonia exposures in an agricultural community and pediatric asthma morbidity. *Epidemiology*, *26*(6), 794–801. https://doi.org/10.1097/EDE.0000000000000368

Luesink, H., & Michels, R. (2018). *Ammonia regulations near nature areas in Denmark and the Netherlands compared*. https://doi.org/10.18174/445690

Ma, R., Zou, J., Han, Z., Yu, K., Wu, S., Li, Z., Liu, S., Niu, S., Horwath, W. R., & Zhu-Barker, X. (2021). Global soil-derived ammonia emissions from agricultural nitrogen fertilizer application: A refinement based on regional and crop-specific emission factors. *Global Change Biology*, *27*(4), 855–867. https://doi.org/10.1111/GCB.15437

Malley, C. S., Hicks, W. K., Kulyenstierna, J. C. I., Michalopoulou, E., Molotoks, A., Slater, J., Heaps, C. G., Ulloa, S., Veysey, J., Shindell, D. T., Henze, D. K., Nawaz, O., Anenberg, S. C., Mantlana, B., & Robinson, T. P. (2021). Integrated assessment of global climate, air pollution, and dietary, malnutrition and obesity health impacts of food production and consumption between 2014 and 2018. *Environmental Research Communications* , *3*. https://doi.org/10.1088/2515-7620/ac0af9

McIlroy, J. P., McGeough, K. L., Laughlin, R. J., & Carolan, R. (2019). Abatement of ammonia emissions from dairy cow house concrete floor surfaces through additive application. *Biosystems Engineering*, *188*, 320–330. https://doi.org/10.1016/J.BIOSYSTEMSENG.2019.10.016

Megaritis, A. G., Fountoukis, C., Charalampidis, P. E., Pilinis, C., & Pandis, S. N. (2013).

Sciences ess Atmospheric Chemistry and Physics Climate of the Past Geoscientific Instrumentation Methods and Data Systems Response of fine particulate matter concentrations to changes of emissions and temperature in Europe. *Atmos. Chem. Phys*, *13*, 3423–3443. https://doi.org/10.5194/acp-13-3423-2013

Megaritis, A. G., Fountoukis, C., Charalampidis, P. E., Pilinis, C., & Pandis, S. N. (2013). Response of fine particulate matter concentrations to changes of emissions and temperature in Europe. *Atmospheric Chemistry and Physics*, *13*(6), 3423–3443. https://doi.org/10.5194/acp-13-3423-2013

Melse, R W, & Ogink, N. W. M. (2005). Air scrubbing techniques for ammonia and odor reduction at livestock operations: Review of on-farm research in the Netherlands. *Transactions of the American Society of Agricultural Engineers*, *48*(6), 2303–2313. https://www.scopus.com/inward/record.uri?eid=2-s2.0-32244449217&partnerID=40&md5=261a95440613149b9977dcde681b184f

Melse, Roland W, Ogink, N. W. M., & Rulkens, W. H. (2009). Air Treatment Techniques for Abatement of Emissions from Intensive Livestock Production . *The Open Agriculture Journal*, *3*, 6–12.

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

Mensink, C., & Deutsch, F. (2008). On the Role of Ammonia in the Formation of PM2.5. In C. Borrego & A. I. Miranda (Eds.), *Air Pollution Modeling and Its Application XIX* (pp. 548–556). Springer Netherlands. https://doi.org/10.1007/978-1-4020-8453-9_60

Menzi, H., Katz, P. E., Fahrni, M., Neftel, A., & Frick, R. (1998). A simple empirical model based on regression analysis to estimate ammonia emissions after manure application. *Atmospheric Environment*, *32*(3), 301–307. https://doi.org/10.1016/S1352-2310(97)00239-2

Mikkelsen, M. H., Albrektsen, R., & Gyldenkærne, S. (2011). *Danish emission inventories for agriculture: Inventories 1985-2009* (Vol. 810). NATIONAL ENVIRONMENTAL RESEARCH INSTITUTE.

Misselbrook, T. H., Nicholson, F. A., & Chambers, B. J. (2005). Predicting ammonia losses following the application of livestock manure to land. *Bioresource Technology*, *96*(2), 159–168. https://doi.org/10.1016/j.biortech.2004.05.004

Mkhabela, M. S., Gordon, R., Smith, E., Madani, A., & Burton, D. (2008). Measurement of odour emissions using micrometeorological techniques following application of hog slurry to grass. *Canadian Biosystems Engineering / Le Genie Des Biosystems Au Canada*, *50*(6), 37–45. http://www.scopus.com/inward/record.url?eid=2-s2.0-35648987848&partnerID=40

Moal, J. F., Martinex, J., Guiziou, F., & Coste, C. M. (1995). Ammonia volatilization following surface-applied pig and cattle slurry in France. *The Journal of Agricultural Science*, *125*(2), 245–252.

Naseem, S., & King, A. J. (2018). Ammonia production in poultry houses can affect health of humans, birds, and the environment—techniques for its reduction during poultry production. In *Environmental Science and Pollution Research* (Vol. 25, Issue 16, pp. 15269–15293). Springer Verlag. https://doi.org/10.1007/s11356-018-2018-y

National Research Council (US) Committee. (2008). *Ammonia Acute Exposure Guideline Levels*. National Academies Press (US). https://www.ncbi.nlm.nih.gov/books/NBK207883/

Oenema, O., Velthof, G., Amann, M., Klimont, Z., & Winiwarter, W. (2012). Emissions from agriculture and their control potentials. In *TSAP Report #3, Version 1.0, DG-Environment of the European Commission* (Issue June). http://webarchive.iiasa.ac.at/Admin/PUB/Documents/XO-12-013.pdf

Pavilonis, B. T., Sanderson, W. T., & Merchant, J. A. (2013). Relative exposure to swine animal feeding operations and childhood asthma prevalence in an agricultural cohort HHS Public Access. *Environ Res*, *122*, 74–80. https://doi.org/10.1016/j.envres.2012.12.008

Philippe, F. X., Cabaraux, J. F., & Nicks, B. (2011). Ammonia emissions from pig houses: Influencing factors and mitigation techniques. *Agriculture, Ecosystems and Environment*, *141*(3–4), 245–260. https://doi.org/10.1016/j.agee.2011.03.012

Phillips, S. B., Arya, S. P., & Aneja, V. P. (2004). Ammonia flux and dry deposition velocity from near-surface concentration gradient measurements over a grass surface in North Carolina. *Atmospheric Environment*, *38*(21), 3469–3480. https://doi.org/10.1016/j.atmosenv.2004.02.054

Pinder, R. W., & Adams, P. J. (2007). Ammonia Emission Controls as a Cost-Effective Strategy for Reducing Atmospheric Particulate Matter in the Eastern United States. *Environmental Science and Technology*, *41*, 380–386. https://doi.org/10.1021/es060379a

Plautz, J. (2018). Piercing the haze. *Science, 361*(6407), 1060–1063.

Pozzer, A., Tsimpidi, A. P., Karydis, V. A., De Meij, A., & Lelieveld, J. (2017). Impact of agricultural emission reductions on fine-particulate matter and public health. *Atmospheric Chemistry and Physics*, *17*(20), 12813.

Radon, K., Danuser, B., Iversen, M., Jo È Rres Z, R., Monso, E., Opravil, U., Weber, C., Donham, K. J., & Nowak, D. (2001). Respiratory symptoms in European animal farmers. *European Respiratory Journal*, *17*, 747–754.

Robbins, J. A., Roberts, C., Weary, D. M., Franks, B., & Keyserlingk, M. A. G. von. (2019). Factors influencing public support for dairy tie stall housing in the U.S. *PLOS ONE*, *14*(5). https://doi.org/10.1371/JOURNAL.PONE.0216544

Ryan, P. H., & Holguin, F. (2010). Traffic pollution as a risk factor for developing asthma: Are the issues resolved? In *American Journal of Respiratory and Critical Care Medicine* (Vol. 181, Issue 6, pp. 530–531). https://doi.org/10.1164/rccm.200912-1850ED

Santonja, G., Georgitzikis, K., Maria Scalet, B., Montobbio, P., Roudier, S., & Delgado Sancho, L. (2017). *Best Available Techniques (BAT) Reference Document for the Intensive Rearing of Poultry or Pigs - Industrial Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control)*.

Schauberger, G., Piringer, M., Mikovits, C., Zollitsch, W., Hörtenhuber, S. J., Baumgartner, J., Niebuhr, K., Anders, I., Andre, K., Hennig-Pauka, I., & Schönhart, M. (2018). Impact of global warming on the odour and ammonia emissions of livestock buildings used for fattening pigs. *Biosystems Engineering*, *175*, 106–114. https://doi.org/10.1016/j.biosystemseng.2018.09.001

Schiffman, S. S., Walker, J. M., Dalton, P., Lorig, T. S., Raymer, J. H., Shusterman, D., & Williams, C. M. (2004). Potential health effects of odor from animal operations, wastewater treatment, and recycling of byproducts - PubMed. *Journal of Agromedicine*, *9*(2), 397–403. https://pubmed.ncbi.nlm.nih.gov/19785232/

Schiffman, S. S., & Williams, C. M. (2005). Science of odor as a potential health issue. *Journal of Environmental Quality*, *34*(1), 129–138.

https://doi.org/10.2134/jeq2005.0129a

Schinasi, L., Horton, R. A., Guidry, V. T., Wing, S., Marshall, S. W., & Morland, K. B. (2011). Air pollution, lung function, and physical symptoms in communities near concentrated swine feeding operations. *Epidemiology*, *22*(2), 208–215. https://doi.org/10.1097/EDE.0b013e3182093c8b

Schultz, A. A., Peppard, P., Gangnon, R. E., & Malecki, K. M. C. (2019). Residential proximity to concentrated animal feeding operations and allergic and respiratory disease. *Environment International*, *130*, 104911. https://doi.org/10.1016/j.envint.2019.104911

Shropshire Council. (2018). Assessing the impact of ammonia and nitrogen on designated sites and Natural Assets from new and expanding livestock units (LSUs) (Version 1). In *Interim NH3 Guidance Note* (pp. 1–27). https://shropshire.gov.uk/media/10859/interim-guidance-note-on-ammonia-emitting-developments-apr-2018.pdf

Simpson, D., Andersson, C., Christensen, J. H., Engardt, M., Geels, C., Nyiri, A., Posch, M., Soares, J., Sofiev, M., Wind, P., & Langner, J. (2014). Impacts of climate and emission changes on nitrogen deposition in Europe: A multi-model study. *Atmospheric Chemistry and Physics*, *14*(13), 6995–7017. https://doi.org/10.5194/acp-14-6995-2014

Singles, R., Sutton, M. A., & Weston, K. J. (1998). A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain. *Atmospheric Environment*, *32*(3), 393–399. https://doi.org/10.1016/S1352-2310(97)83467-X

Skjøth, C. A., & Geels, C. (2013). The effect of climate and climate change on ammonia emissions in Europe. *Atmos. Chem. Phys.*, *13*(1), 117–128. https://doi.org/10.5194/acp-13-117-2013

Søgaard, H. T., Sommer, S. G., Hutchings, N. J., Huijsmans, J. F. M., Bussink, D. W., &

Nicholson, F. (2002). Ammonia volatilization from field-applied animal slurry-the ALFAM model. *Atmospheric Environment*, *36*(20), 3309–3319. https://doi.org/10.1016/S1352-2310(02)00300-X

Sommer, S. G., Genermont, S., Cellier, P., Hutchings, N. J., Olesen, J. E., & Morvan, T. (2003). Processes controlling ammonia emission from livestock slurry in the field. *European Journal of Agronomy*, *19*(4), 465–486.

Sommer, S. G., & Hutchings, N. J. (2001a). Ammonia emission from field applied manure and its reduction—invited paper. *European Journal of Agronomy*, *15*(1), 1–15.

Sommer, S. G., & Hutchings, N. J. (2001b). Ammonia emission from field applied manure and its reduction - Invited paper. In *European Journal of Agronomy* (Vol. 15, Issue 1, pp. 1–15). Elsevier. https://doi.org/10.1016/S1161-0301(01)00112-5

Sommer, S. G., & Olesen, J. E. (1991). Effects of Dry Matter Content and Temperature on Ammonia Loss from Surface-Applied Cattle Slurry. *Journal of Environmental Quality*, *20*(3), 679–683. https://doi.org/10.2134/jeq1991.00472425002000030029x

Sommer, S. G., & Olesen, J. E. (2000). Modelling ammonia volatilization from animal slurry applied with trail hoses to cereals. *Atmospheric Environment*, *34*(15), 2361–2372. https://doi.org/10.1016/S1352-2310(99)00442-2

Spencer, J., & Van Heyst, B. (2018). A review of particulate matter emissions and impacts on human health: A focus on Canadian agricultural and rural emission sources. *Canadian Society for Bioengineering*, *60*(1). https://doi.org/10.7451/CBE.2018.60.6.9

Stokstad, E. (2014). Ammonia pollution from farming may exact hefty health costs. *Science*, *343*(6168), 238. https://doi.org/10.1126/science.343.6168.238

Stowell, R. (2018, January 19). *Ammonia Loss and Emission Reporting: Considerations for*

Cattle Operations | UNL Water. University of Nebraska-Lincoln. https://water.unl.edu/article/animal-manure-management/ammonia-loss-and-emission-reporting

Sun, G., Guo, H., Peterson, J., Predicala, B., & Laguë, C. (2008). Diurnal odor, ammonia, hydrogen sulfide, and carbon dioxide emission profiles of confined swine grower/finisher rooms. *Journal of the Air and Waste Management Association*, *58*(11), 1434–1448. https://doi.org/10.3155/1047-3289.58.11.1434

Sundblad, B. M., Larsson, B. M., Acevedo, F., Ernstgård, L., Johanson, G., Larsson, K., & Palmberg, L. (2004). Acute respiratory effects of exposure to ammonia on healthy persons. *Scandinavian Journal of Work, Environment and Health*, *30*(4), 313–321. https://doi.org/10.5271/sjweh.800

Sutton, M. A., Reis, S., & Baker, S. M. H. (2009). Atmospheric Ammonia: Detecting emission changes and environmental impacts. In *Atmospheric Ammonia: Detecting Emission Changes and Environmental Impacts*. Springer Netherlands. https://doi.org/10.1007/978-1-4020-9121-6

Sutton, M.A., Reis, S., Riddick, S. N., Dragosits, U., Nemitz, E., Theobald, M. R., Tang, Y. S., Braban, C. F., Vieno, M., Dore, A. J., Mitchell, R. F., Wanless, S., Daunt, F., Fowler, D., Blackall, T. D., Milford, C., Flechard, C. R., Loubet, B., Massad, R., … de Vries, W. (2013). Towards a climate-dependent paradigm of ammonia emission and deposition. *Philosophical Transactions of the Royal Society B: Biological Sciences*, *368*(1621). https://doi.org/10.1098/rstb.2013.0166

Sutton, Mark A., Erisman, J. W., Dentener, F., & Möller, D. (2008). Ammonia in the environment: From ancient times to the present. In *Environmental Pollution* (Vol. 156, Issue 3, pp. 583–604). https://doi.org/10.1016/j.envpol.2008.03.013

Tang, Y. S., van Dijk, N., Love, L., & et al. (2004). *Ammonia Monitoring in Northern Ireland, Final Report*.

Thakrar, S. K., Balasubramanian, S., Adams, P. J., Azevedo, I. M. L., Muller, N. Z., Pandis, S. N., Polasky, S., C. Arden Pope, I., Robinson, A. L., Apte, J. S., Tessum, C. W., Marshall, J. D., & Hill, J. D. (2020). Reducing Mortality from Air Pollution in the United States by Targeting Specific Emission Sources. *Environmental Science & Technology Letters*, *7*(9), 639–645. https://doi.org/10.1021/ACS.ESTLETT.0C00424

Theobald, M. R., Bealey, W. J., Tang, Y. S., Vallejo, A., & Sutton, M. A. (2009). A simple model for screening the local impacts of atmospheric ammonia. *Science of the Total Environment*, *407*(23), 6024–6033. https://doi.org/10.1016/j.scitotenv.2009.08.025

Tschofen, P., Azevedo, I. L., & Muller, N. Z. (2019). Fine particulate matter damages and value added in the US economy. *Proceedings of the National Academy of Sciences of the United States of America*, *116*(40), 19857–19862. https://doi.org/10.1073/pnas.1905030116

US EPA. (2020, June 8). *Particulate Matter (PM2.5) Trends | National Air Quality: Status and Trends of Key Air Pollutants | US EPA*. https://www.epa.gov/air-trends/particulate-matter-pm25-trends

USDA Agricultural Air Quality Task Force. (2014). *Ammonia Emissions : What To Know Before You Regulate* (Issue October). http://www.nrcs.usda.gov/wps/portal/nrcs/detail/national/air/taskforce/?cid=stelprdb1268645

Van Damme, M., Clarisse, L., Franco, B., Sutton, M. A., Willem Erisman, J., Wichink Kruit, R., van Zanten, M., Whitburn, S., Hadji-Lazaro, J., Hurtmans, D., Clerbaux, C., & Coheur, P.-F. (2021). Global, regional and national trends of atmospheric ammonia derived from a decadal (2008-2018) satellite record OPEN ACCESS RECEIVED Global, regional and national trends of atmospheric ammonia derived from a decadal (2008-2018) satellite record. *Environmental Research Letters*, *16*. https://doi.org/10.1088/1748-9326/abd5e0

Von Bobrutzki, K., Braban, C. F., Famulari, D., Jones, S. K., Blackall, T., Smith, T. E. L., Blom, M., Coe, H., Gallagher, M., Ghalaieny, M., McGillen, M. R., Percival, C. J., Whitehead, J. D., Ellis, R., Murphy, J., Mohacsi, A., Pogany, A., Junninen, H., Rantanen, S., … Nemitz, E. (2010). Field inter-comparison of eleven atmospheric ammonia measurement techniques. *Atmospheric Measurement Techniques*, *3*(1), 91–112. https://doi.org/10.5194/amt-3-91-2010

Walker, J. T., Robarge, W. P., Shendrikar, A., & Kimball, H. (2006). Inorganic PM2. 5 at a US agricultural site. *Environmental Pollution*, *139*(2), 258–271.

Wang, X., Ledgard, S., Luo, J., Guo, Y., Zhao, Z., Guo, L., Liu, S., Zhang, N., Duan, X., & Ma, L. (2018). Environmental impacts and resource use of milk production on the North China Plain, based on life cycle assessment. *Science of The Total Environment*, *625*, 486–495. https://doi.org/10.1016/J.SCITOTENV.2017.12.259

Webb, J., Menzi, H., Pain, B. F., Misselbrook, T. H., Dämmgen, U., Hendriks, H., & Döhler, H. (2005). Managing ammonia emissions from livestock production in Europe. *Environmental Pollution*, *135*(3 SPEC. ISS.), 399–406. https://doi.org/10.1016/j.envpol.2004.11.013

WHO. (2013). *Health effects of particulate matter*. http://www.euro.who.int/pubrequest

Wing, S., & Wolf, S. (2000). Articles Intensive Livestock Operations. *Environmental Health Perspectives*, *108*(3), 233–238.

World Health Organisation. (2013). *Health effects of particulate matter* . http://www.euro.who.int/pubrequest

Xu, P., Koloutsou-Vakakis, S., Rood, M. J., & Luan, S. (2017). Projections of NH3 emissions from manure generated by livestock production in China to 2030 under six mitigation scenarios. *Science of The Total Environment*, *607–608*, 78–86. https://doi.org/10.1016/j.scitotenv.2017.06.258

Ye, Z., Saha, C. K., Li, B., Tong, G., Wang, C., Zhu, S., & Zhang, G. (2009). Effect of environmental deflector and curtain on air exchange rate in slurry pit in a model pig house. *Biosystems Engineering*, *104*(4), 522–533. https://doi.org/10.1016/j.biosystemseng.2009.09.015

Yu, O., Sheppard, L., Lumley, T., Koenig, J. Q., & Shapiro, G. G. (2000). Effects of ambient air pollution on symptoms of asthma in seattle-area children enrolled in the CAMP study. *Environmental Health Perspectives*, *108*(12), 1209–1214. https://doi.org/10.1289/ehp.001081209

Zbieranowski, A. L., & Aherne, J. (2012). Spatial and temporal concentration of ambient atmospheric ammonia in southern Ontario, Canada. *Atmospheric Environment*, *62*, 441–450. https://doi.org/10.1016/J.ATMOSENV.2012.08.041

Zhang, B., Tian, H., Lu, C., Dangal, S. R. S., Yang, J., & Pan, S. (2017). Global manure nitrogen production and application in cropland during 1860-2014: A 5 arcmin gridded global dataset for Earth system modeling. *Earth System Science Data*, *9*(2), 667–678. https://doi.org/10.5194/ESSD-9-667-2017

Zhang, X., Gu, B., van Grinsven, H., Lam, S. K., Liang, X., Bai, M., & Chen, D. (2020). Societal benefits of halving agricultural ammonia emissions in China far exceed the abatement costs. *Nature Communications*, *11*(1), 1–10. https://doi.org/10.1038/s41467-

020-18196-z

Zhu, L., Henze, D. K., Bash, J. O., Cady-Pereira, K. E., Shephard, M. W., Luo, M., & Capps, S. L. (2015). Sources and Impacts of Atmospheric NH3: Current Understanding and Frontiers for Modeling, Measurements, and Remote Sensing in North America. *Current Pollution Reports*, *1*(2), 95–116. https://doi.org/10.1007/s40726-015-0010-4