

Emissions inventory of greenhouse gases and ammonia from livestock housing and manure management

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Abstract: An emission inventory is a database on the amount of pollutants released into the atmosphere. The anthropogenic emissions of air pollutants and greenhouse gases are detrimental to the environment and the ecosystems. Therefore, reducing emissions is crucial. One key issue is to inventory these emissions, and consequently databases on anthropogenic emissions will be available for making decisions on implementing suitable mitigation strategies. Such investigations aim at developing national emissions inventory for domestic livestock and to identify possible abatement techniques in order to reduce these emissions. Therefore, the objectives of this study are to introduce and define the emissions inventories, review the emission inventory guides, introduce the relation between the emissions inventory and livestock buildings and manure stores and the relevant emission factors and algorithms, review the tools for processing the emissions inventories (e.g. models, software), show the evaluation and improvement methods of emissions inventories, review the emissions abatement techniques, and present examples and paradigms of available national emissions inventories for several countries.

Keywords: emissions inventory, greenhouse gases, methane, nitrous oxide, ammonia, particulate matter, livestock buildings, manure, emission factors, emissions abatement techniques

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1 Introduction

Emissions from agriculture have great environmental impact and relevant political importance, and animal husbandry is a major source of atmospheric pollutants, such as methane (CH₄), nitrous oxide (N₂O), carbon dioxide (CO₂), and ammonia (NH₃). Carbon dioxide, methane and nitrous oxide are greenhouse gases (GHG) that contribute to global warming, while ammonia is an atmospheric pollutant and responsible for eutrophication and soil acidification (Haussermann et al., 2006; Reinhardt-Hanisch, 2008; Samer et al., 2011a,b). Ammonia emissions from agricultural industries are a significant source of atmospheric reactive nitrogen, which can lead to negative environmental consequences such as ecosystem change and formation of fine particulate

(Faulkner and Shaw, 2008). Ammonia may also form secondary particulate matter (PM). Particulate emissions also have an adverse impact on human health. Emissions of NH₃, Nitric oxide (NO), and non-methane volatile organic compounds (NMVOCs) arise from the excreta of agricultural livestock deposited in and around buildings and collected as liquid slurry, solid manure or litter-based farmyard manure. NMVOCs and NO are involved in the formation of ozone, which near the surface of the Earth can have an adverse effect on human health and plant growth (EMEP/EEA emission inventory guidebook, 2009; updated June 2010). Most of the N₂O emissions occurred after the deposition of manure on soil during cattle grazing, while CH₄ is mainly produced during the period when cattle manure remained in livestock buildings and in outside storage facilities (Gac et al., 2007). A detailed knowledge of the processes of NH₃ transfer from the manure and transport to the free atmosphere will contribute to development of techniques

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and housing designs that will contribute to the reduction of NH_3 emission to the atmosphere (Sommer et al., 2006). Livestock NH_3 emissions are higher in areas characterized by intensive livestock production with diet manipulation and land spreading offering the greatest potential for NH_3 abatement options (Carew, 2010). About 94% of global anthropogenic emissions of NH_3 to the atmosphere originate from the agricultural sector of which close to 64% is associated with livestock management (FAO, 2006). Excessive levels of NH_3 emissions contribute to eutrophication and acidification (Schuurkes and Mosello, 1988). Enteric fermentation and manure management account for 35% to 40% of the total anthropogenic CH_4 emissions and 80% of CH_4 release from agriculture (FAO, 2006). Livestock activities contribute with 65% of the global anthropogenic N_2O emissions and account for 75% to 80% of the emission from agriculture (FAO, 2006). Globally, CO_2 , CH_4 and N_2O , contribute 60%, 15% and 5%, respectively, to the anthropogenic greenhouse effect (Guo and Zhou, 2007). The Intergovernmental Panel on Climate Change (IPCC, 2007) CH_4 and N_2O are greenhouse gases with global warming potentials of 23 and 296 times that of CO_2 , respectively.

Zervas and Tsiplakou (2012) stated that GHG emissions are expected to cause global warming which results in extreme weather changes that could affect crop yields and productivity, food supplies and food prices. It is also expected that climate change will have an impact on animal metabolism and health, reproduction and productivity. On the other hand, the expected increased demand of animal origin products in the coming years will increase the reared animal numbers and consequently GHG emissions. They outlined the main GHGs emitted from livestock which are CO_2 , CH_4 and N_2O , coming from respiration, enteric fermentation and manure management respectively, with CH_4 and N_2O having the highest global warming potential. The guidelines of the IPCC (2006) stated that CO_2 emissions from livestock are not estimated because annual net CO_2 emissions are assumed to be zero – the CO_2 photosynthesized by plants is returned to the atmosphere as respired CO_2 . A portion of the C is returned as CH_4

and for this reason CH_4 requires separate consideration. Zervas and Tsiplakou (2012) stated that ruminant livestock has the highest contribution to these GHG emissions with small ruminants share being 12.25% of the total GHG emissions from livestock's enteric and manure CH_4 , and manure N_2O in CO_2 equivalent, producing 9.45 kg CO_2 equivalent per kg body weight with the respective values for cattle, pigs and poultry being 5.45, 3.97 and 3.25. Since the production systems significantly affect the GHG emissions, the grazing, livestock crop complex, and intensive ones account for 30.5%, 67.29% and 5.51% for total CH_4 emission (from enteric fermentation and manure management) and 24.32%, 68.11% and 7.57% for N_2O respectively. Taking into account the positive and negative impacts of small ruminant livestock production systems on the environmental aspects in general, they recommended that a number of potentially effective measures should be taken and the appropriate mitigation technologies should be applied in order to reduce effectively and essentially the GHG emissions to the atmosphere, with no adverse effects on intensification and increased productivity of small ruminants production systems. However, small ruminants are not key categories in many countries; therefore, this would be the case of countries with a high production of small ruminants in extensive systems.

Following the adoption of the United Nations Economic Commission for Europe (UNECE) Gothenburg protocol (Protocol to the 1979 convention on long range transboundary air pollution to abate acidification, eutrophication and ground-level ozone, United Nations Economic Commissions for Europe (UNECE), Geneva.), the members struggle to achieve significant reduction in national NH_3 emissions. Similarly, the United Nations Framework Convention on Climate Change (UNFCCC) adopted the Kyoto protocol (Protocol that set binding obligations on the industrialized countries to reduce their emissions of greenhouse gases). The protocol was adopted on December 11th, 1997, in Kyoto, Japan, and entered into force on February 16th, 2005. As of September 2011, 191 states have signed and ratified the protocol), the members exert efforts to reducing the GHG emissions. In order to achieve reduction of GHG and

NH₃ emissions, it is essential to inventory these emissions.

An emission inventory is a database of the amount of pollutants released into the atmosphere. The anthropogenic emissions of air pollutants and greenhouse gases are detrimental to the environment and the ecosystems. Therefore, reducing emissions is critical. One key issue is to inventory these emissions, and as consequently emissions databases will be available for making decisions on implementing suitable mitigation strategies. The objectives of such investigations are to develop a national emissions inventory for domestic livestock and to identify possible abatement techniques to reduce these emissions. The inventory can be developed using data derived from farm surveys and national statistical records in a country and through several years, as well as Tier 1 and 2 emission factors for enteric fermentation and manure management as proposed by the Intergovernmental Panel on Climate Change. The developed inventory should be compared with other countries, in terms of pollutant emissions per unit of land area or human population. The results can be then used, with data from other agricultural sectors, to develop a national inventory for the entire agricultural sector of a country (Aljaloud et al., 2011). A product based metric (emissions/kg product) can be launched, which allows analyzing profitability, food security and environmental conflicts. When an emission inventory is developed, it can be thereafter used to develop an abatement/mitigation strategy for livestock farms as well as at national level which focuses on abating the highest emission levels that are released through determined stages of manure management (from housing to field), this to adhere to the commitments imposed by Gothenburg and Kyoto Protocols.

Reidy et al. (2008a) stated that NH₃ emissions from agriculture commonly account for > 80% of the total NH₃ emissions. Accurate agricultural NH₃ emission inventories are therefore required for reporting within the framework of the Gothenburg Protocol of the UN Convention on Long-range Transboundary Air Pollution. To allow a coordinated implementation of the Protocol, different national inventories should be comparable. A core group of emission inventory experts therefore

developed a network and joint program to achieve a detailed overview of the best inventory techniques currently available and compiled and harmonized the available knowledge on emission factors (EFs) for nitrogen (N)-flow emission calculation models and initiated a new generation of emission inventories.

Existing emission inventory approaches mainly rely on expert judgment for information on farm and manure management. To detect the relatively small changes of total annual NH₃ emissions required under the Gothenburg Protocol, expert judgment is considered insufficient. Therefore, a new NH₃ emission inventory based on a detailed representative stratified survey on farm and manure management conducted on 1950 farms was presented by Reidy et al. (2008b). Emissions estimated within a National Emission Inventory should be compared to emissions estimated using Guidebook emission factors and National statistics for a specific year or period. The objective is to explore the quality of both methods and to find major differences and similarities (Dröge et al., 2010).

Chadwick et al. (2011) stated that slurry and farmyard manure are an inevitable consequence of livestock products generated from housed animals. These manures are recycled back to land for plants to use the nutrients they contain. However, since they contain inorganic N, microbial sources of C and water, they provide the essential substrates required for the microbial production of N₂O and CH₄. These greenhouse gases can be produced and emitted at each stage of the “manure management continuum”, being the livestock building, manure stores, manure treatment and manure spreading to land. The contribution that manure management makes to total national agricultural emissions of N₂O and CH₄ varies, but it can exceed 50% in countries reporting to the UNFCCC in 2009 (Chadwick et al., 2011). On farm management decisions interact with environmental controls such as temperature and water availability of key microbial processes (i.e., nitrification, denitrification, methanogenesis, CH₄ oxidation), affecting the magnitude of emissions from each stage of the manure management continuum (Chadwick et al., 2011). Reidy et al. (2009) mentioned that each country uses its own excretion model

and defines cattle groups in different ways and with different degrees of detail (in breed, feeding regimes). They added that there were fewer published results of NH_3 emissions from solid manure than for slurry and the introduction of litter leads to more complex interactions (e.g. immobilization and mineralization) and greater, and highly variable, emissions of other N gases. In addition, the state of development of the models with respect to immobilization and denitrification is quite different. They concluded that it is important to get a better overview of the existing knowledge, an identification of research gaps and a thorough re-editing of some of the models (especially DYNAMO and MAM) for processes other than NH_3 emission.

Comparing complete inventories to independent efforts in assessing emissions, e.g. atmospheric measurements combined with source apportionment, allows better understanding and quantifying the reliability of inventory data (Winiwarter et al., 2009). A number of emission factors (EFs) can be proposed for developing ammonia emissions inventories (Faulkner and Shaw, 2008). Besides the production facilities, the EFs for animal feeding operations should also be considered (Faulkner and Shaw, 2008). Seasonal distribution of NH_3 emissions should be considered when developing emissions inventory. Significant uncertainty exists in the seasonal distribution of NH_3 emissions since the predominant sources are animal husbandry and fertilizer application. Previous studies that estimated bottom-up and top-down NH_3 emissions have provided the most comprehensive information available about the seasonality of NH_3 emissions. Inverse modeling (top-down) results suggest that the prior NH_3 emission estimates should be increased in summer and decreased in winter, while results for spring and fall are questionable due to precipitation prediction biases. A final conclusion from this study is that total NH_x (NH_3 and aerosol NH_4^+) air concentration data are essential to quantitative top-down analyses of NH_3 emissions that can extend beyond what is possible using precipitation chemistry data (Gilliland et al., 2006). Fugitive emissions may affect the accuracy of emissions inventory, where fugitive emissions are emissions caused by wind

shear, material transfer processes or other mechanical forces from non-point sources, e.g. natural particles like sea salt and windblown dust (Winiwarter et al., 2009).

Merino et al. (2011) stated that quantification of greenhouse gas emissions is reported annually in the National Emission Inventory. They added that a mathematical approach should be used to assess uncertainties of estimated emission factors (EF) in order to update the annual CH_4 and N_2O emissions. Additionally, they mentioned that emissions from manure management have the largest uncertainty due to the high natural variability of manure. The more accurate animal data are available, the lower uncertainty is expected. This is the case in the intensive production systems. Webb et al. (2005) found that among the larger sources of NH_3 emissions, those from buildings housing cattle and pigs on straw were the most uncertain. Sommer et al. (2006) stated that livestock excreta and manure stored in housing, in manure stores, in beef feedlots, or cattle hardstandings are the most important sources of NH_3 in the atmosphere. Inventories have shown that animal housing, stored animal manure, and exercise areas account for about 69–80% of the total emission of NH_3 in Europe (ECETOC, 1994; Hutchings et al., 2001). Therefore, the objectives of this study are to introduce and define the emissions inventories, review the emission inventory guides, introduce the relation between the emissions inventory and livestock buildings and manure stores and the relevant emission factors and algorithms, review the tools for processing the emissions inventories (e.g. models, software), show the evaluation and improvement methods of emissions inventories, review the emissions abatement techniques, and present examples and paradigms of available national emissions inventories for several countries.

2 Definition of emissions inventory

An emission inventory is a database of the amount of pollutants released into the atmosphere. An emission inventory usually includes the total emissions for one or more specific air pollutants, originating from all source categories (agriculture, energy, industry, waste, and some other sources) in a certain geographic area and within a

specified time period, usually a specific year. Generally, an emission inventory provides the following information: (1) the types of activities that cause emissions, (2) the chemical or physical identity of the pollutants included, (3) the geographical area covered, (4) the time span over which emissions are estimated, (5) the methodology that was implemented to inventory the emission(s). The emission inventories are developed for scientific applications as well as for policy processes and decision making.

3 Emission inventory guides

There are two guides: the air pollutant emission inventory guidebook of the European Environment Agency (EEA) and the Cooperative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe (EMEP), and the guidelines of the Intergovernmental Panel on Climate Change (IPCC). Dröge et al. (2010) stated that knowledge of emissions and releases of pollutants to the environment is crucial for every environmental pollution problem. Information on emissions is therefore an absolute requirement in understanding environmental problems and monitoring progress in solving these problems. Dammgén and Webb (2006) mentioned that the reduction of emissions of air pollutants is subject of international conventions, which include reporting of emissions in accordance with guidelines or guidebooks provided. Within the Convention on the Long-range Transboundary Air Pollution (LRTAP), the Atmospheric Emission Inventory Guidebook describes the methodology. With respect to emissions from agricultural sources, in particular from animal husbandry, this guidebook has undergone major modifications: the calculation procedure making use of partial emission factors for the various sources of emissions (animal house, storage, manure application, etc.) were replaced by a mass flow concept for both nitrogen and carbon species. The necessity to update both the Guidebook and the IPCC Guidelines as complementary tools to describe agricultural emissions and mass flows is emphasized.

3.1 EMEP/EEA emission inventory guide

Dröge et al. (2010) mentioned that in the field of

long-range transboundary air pollution, countries in both Europe and North-America have agreed to decrease the impacts of air pollution below a level that is damaging human health and ecosystems by ratifying the UNECE Convention on Long-Range Transboundary Air Pollution (LRTAP). The aim of the Convention is “that Parties shall endeavor to limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution. Parties develop policies and strategies to combat the discharge of air pollutants through exchanges of information, consultation, research and monitoring”. The LRTAP Convention and its protocols include important pollutants like NO_x, SO_x, NH₃, NMVOC, CO, particulate matter, heavy metals and persistent organic pollutants. Parties to the LRTAP convention have agreed to annually report atmospheric emissions and are required to set up an emission inventory. As a minimum, parties shall use the latest version of the EMEP/EEA Air Pollutant Inventory Guidebook, but most countries have set up their own inventory, which uses country specific information to supplement the information from the Guidebook.

The EMEP/EEA air pollutant emission inventory guidebook (formerly called the EMEP CORINAIR emission inventory guidebook) provides guidance on estimating emissions from both anthropogenic and natural emission sources. It is designed to facilitate reporting of emission inventories by countries to the UNECE Convention on Long-range Transboundary Air Pollution and the EU National Emission Ceilings Directive. The EEA publishes the Guidebook, with the UNECE’s Task Force on Emission Inventories and Projections (EMEP/EEA, 2009).

3.2 IPCC guidelines for national inventories

The 2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines) provide methodologies for estimating national inventories of anthropogenic emissions by sources and removals by sinks of greenhouse gases. The 2006 IPCC Guidelines were prepared in response to an invitation by the Parties to the UNFCCC. They may assist Parties in fulfilling their commitments under the UNFCCC on reporting on inventories of anthropogenic emissions by sources and

removals by sinks of greenhouse gases not controlled by the Montreal Protocol, as agreed by the Parties. The 2006 IPCC Guidelines are in five volumes. Volume 1 describes the basic steps in inventory development and offers the general guidance in greenhouse gas emissions and removals estimates based on the authors' understanding of accumulated experiences of countries over the period since the late 1980s, when national greenhouse gas inventories started to appear in significant numbers. Volumes 2 to 5 offer the guidance for estimates in different sectors of economy, which are: Energy, Industrial Processes and Product Use, Agriculture, Forestry and Other Land Use, and Waste. Volume 4 (Chapter 10) provides guidance on methods to estimate emissions of methane from Enteric Fermentation in livestock, and methane and nitrous oxide emissions from Manure Management (IPCC, 2006).

4 Livestock buildings and emissions inventory

Sommer et al. (2006) stated that process-based models can be an accurate and cost-effective means of estimating emissions from a discrete source. This is especially relevant for predicting or monitoring the impact of emissions from buildings, outdoor yards, and manure stores of a large livestock production unit on adjacent sensitive area(s). In mechanically ventilated buildings ventilation rate often determines NH_3 emissions. While data on ventilation rate may be available for models of emission from individual buildings or farms, such data will not be available for national-scale models. Surrogates for ventilation rates may be available based on ambient temperature and wind speed and ambient data may also be used to calculate conditions within naturally ventilated buildings. However, to be accurate such meta-models would require detailed information of the number, age, and weight of animals within buildings and again, this may be available to use for individual buildings or farms but will not be available for national-scale models except via census data of total numbers of livestock, buildings, and averages/distributions of animals within those buildings. Such information is also known as activity data, which, in the context of calculating NH_3 emissions, may be defined as

data quantifying agricultural practices that have an influence on NH_3 emissions, for example, housing systems. They concluded that the limiting factor in our ability to model emissions from buildings housing livestock is knowledge of what is in those buildings and how they are managed. They added that the following information is needed to make accurate estimates of national NH_3 emissions from buildings housing livestock, hardstandings, and manure stores: (1) animal numbers, (2) the housing period for all types of cattle and for sheep, (3) the amount of time cattle spend on hardstandings and the proportion of cattle that use them, (4) the proportions of cattle, all classes, housed on slurry- or straw-based systems, (5) The proportions of cattle and pig slurry stored in aboveground tanks, lagoons, and weeping walls, and (6) the adoption of covers for slurry stores.

4.1 Emission factors for livestock barns and manure storages

Animal manure from housing is a mixture of feces and urine, bedding material (straw, wood shavings, sawdust, sphagnum, etc.), spilt feed and drinking water, and water used for washing floors. Housing systems are often adapted to the category of housed animal such as calves, dairy cows, sows, fatteners etc., where most cattle buildings are naturally ventilated (Sommer et al., 2006). The quantification of gaseous emissions from naturally ventilated buildings is sophisticated (Samer et al., 2011a,b,c,d).

Faulkner and Shaw (2008) stated that ammonia emissions from agricultural industries are a significant source of atmospheric reactive nitrogen, which can lead to negative environmental consequences such as ecosystem change and formation of fine particulate. They presented recently developed ammonia emission factors (EFs) from literature for animal feeding operations, including production facilities for beef and dairy cattle, swine, and poultry. Several studies have investigated the emission factors from livestock buildings. Samer et al. (2011a) listed that the emission factors for dairy cows (lactating Holstein Friesian) –through summer- were 63.5, 270, 23842, and 15.5 $\text{kg year}^{-1} \text{LU}^{-1}$ (LU: livestock unit and is equivalent to 500 kg animal live mass) for NH_3 , CH_4 , CO_2 , and N_2O respectively.

On the other hand, the emission factors –through winter– were 32, 169, 20431, and 17.3 kg year⁻¹ LU⁻¹ for NH₃, CH₄, CO₂, and N₂O respectively. However, the aforementioned values differ discernibly from study to another depending on the number of experiments, time length, climatic conditions...etc (Samer et al., 2011a,b,c,d; Samer et al., 2012b). Winiwarter et al. (2009) mentioned that due to particulate matter (PM) specific circumstances such as the large number of sources, very different release pathways and differences of the individual particles in terms of chemical composition or size, it is very difficult to appropriately handle measurement conditions to arrive at adequate emission factors, especially when emission points cannot be defined clearly.

Gupta et al. (2007) reported seasonal variation in CH₄ and N₂O emission rate from solid storage of bovine manure in Delhi as well as emission factors and emission inventory from manure management systems in India. Emission flux observed in the year 2002-2003 was 4.29 ± 1, 4.84 ± 2.44 and 12.92 ± 4.25 mg CH₄ kg⁻¹ dung day⁻¹, as well as 31.29 ± 4.93, 72.11 ± 16.22 and 6.39 ± 1.76 mg N₂O kg⁻¹ dung day⁻¹ in winter, summer and rainy seasons, respectively. CH₄ emission factors varied from 0.8 to 3.3 kg hd⁻¹ year⁻¹ for bovines and were lower than IPCC-1996 default values. N₂O emission factors varied from 3 to 11.7 mg hd⁻¹ year⁻¹ from solid storage of manure. Inventory estimates were found to about 698 ± 27 Gg CH₄ from all manure management systems and 2.3 ± 0.46 tons of N₂O from solid storage of manure for the year 2000.

O'Shaughnessy and Altmaier (2011) stated that the swine concentrated animal feeding operations (CAFOs) emit hydrogen sulfide (H₂S) from both housing structures and waste lagoons. An average emission flux rate of 0.57 mg m⁻² s⁻¹ was determined for swine CAFO lagoons. Using the average total animal weight (kg) of each CAFO, an average emission factor of 6.06 × 10⁻⁷ μg yr⁻¹ m⁻² kg⁻¹ was calculated. From studies that measured either building or lagoon emission flux rates, building fluxes, on a floor area basis, were considered equal to lagoon flux rates. Costa and Guarino (2009) investigated yearly emission factor of dust and greenhouse gases through continuous measurements in swine husbandry. They

found that the mean yearly emission factor of PM₁₀ was measured in 2 g d⁻¹ LU⁻¹ for the weaning room, 0.09 g d⁻¹ LU⁻¹ for the farrowing room, 2.59 g d⁻¹ LU⁻¹ for the fattening room and 1.23 g d⁻¹ LU⁻¹ for the gestation room. The highest PM₁₀ concentration and emission per LU was recorded in the fattening compartment while the lowest value was recorded in the farrowing room. The yearly emission factor for CO₂ was measured in 5,997 g d⁻¹ LU⁻¹ for the weaning room, 1,278 g d⁻¹ LU⁻¹ for the farrowing room, 13,636 g d⁻¹ LU⁻¹ for the fattening room and 8,851 g d⁻¹ LU⁻¹ for the gestation room. The yearly emission factor for CH₄ was measured in 24.57 g d⁻¹ LU⁻¹ for the weaning room, 4.68 g d⁻¹ LU⁻¹ for the farrowing room, 189.82 g d⁻¹ LU⁻¹ for the fattening room and 132.12 g d⁻¹ LU⁻¹ for the gestation room. The yearly emission factor for N₂O was measured in 3.62 g d⁻¹ LU⁻¹ for the weaning room, 0.66 g d⁻¹ LU⁻¹ for the farrowing room, 3.26 g d⁻¹ LU⁻¹ for the fattening room and 2.72 g d⁻¹ LU⁻¹ for the gestation room. It can be inferred that the emission factors of the different gases differs depending not only the barn being investigated but also on specific parts/compartments inside the barn. This should be considered when inventorying the emissions.

4.2 Algorithms for determining emissions from buildings and manure stores

Sommer et al. (2004) developed algorithms for calculating methane and nitrous oxide emissions from manure management. They found that biogenic emissions of CH₄ and N₂O from animal manure are stimulated by the degradation of volatile solids (VS) which serve as energy source and a sink for atmospheric oxygen. They presented algorithms which link carbon and nitrogen turnover in a dynamic prediction of CH₄ and N₂O emissions during handling and use of liquid manure (slurry). A sub-model for CH₄ emissions during storage relates CH₄ emissions to VS, temperature and storage time, and estimates the reduction in VS. A second sub-model estimates N₂O emissions from field-applied slurry as a function of VS, slurry N and soil water potential, but emissions are estimated using default emission factors. Anaerobic digestion of slurry and organic waste produces CH₄ at the expense of VS. Accordingly, the model predicted a 90% reduction of

CH₄ emissions from outside stores with digested slurry, and a >50% reduction of N₂O emissions after spring application of digested as opposed to untreated slurry. Their study indicates that simple algorithms to account for ambient climatic conditions may significantly improve the prediction of CH₄ and N₂O emissions from animal manure. Sommer et al. (2006) developed algorithms for determining ammonia emission from buildings housing cattle and pigs and from manure stores.

Keener and Zhao (2008) presented a modified N-balance method, which, although it does not distinguish for losses of N as N₂, NH₃ or NO_x's, it does determine accurately the upper limit on NH₃-N emissions for both forced and naturally ventilated livestock facilities. The method is based on nitrogen and ash contents and ratio of the inputs (feed, bedding, water, and animals) and outputs (products and waste) of the animal-production systems and does not require measuring total masses of manure from the system. Generalized equations for all classes of livestock operations were presented. This N-balance method is simple, low cost and accurate in predicting the upper limits on NH₃ emissions in the air from livestock facilities.

4.3 Emissions inventory based on farm survey

Reidy et al. (2008b) stated that existing emission inventory approaches mainly rely on expert judgment for information on farm and manure management. To detect the relatively small changes of total annual ammonia (NH₃) emissions required under the Gothenburg Protocol, expert judgment is considered insufficient. They presented, therefore, NH₃ emission inventory based on a detailed representative stratified survey on farm and manure management conducted on 1950 farms. The survey data was used to calculate NH₃ emissions with the emission model DYNAMO for each farm participating in the survey. This allowed the effects of the variability of farm and manure management parameters among farms on the NH₃ emissions to be fully taken into account. Weighted emission factors per animal for 24 livestock categories and 36 farm classes were used to upscale to the national inventory. The stratified sampling and the individual farm calculations allow the comparison of emissions from specific regions and altitudes and the

study of the variability among farms. This emission inventory approach permits a more detailed analysis of the regional distribution of NH₃ emissions as well as a more robust and standardized monitoring of the future development of emissions.

National inventories of N emissions to the atmosphere from cattle depend on reliable information about husbandry practices, with appropriate spatial and temporal resolution. Sheppard and Bittman (2011) quantified the prevalence of pasture and forage management practices that impact N intake and NH₃ emissions, with implications for N₂O emissions using a survey of 1400 beef cattle farmers approximately. The survey data were coupled to a mass balance model to inventory NH₃ emissions by accounting for total ammoniacal (i.e., ammonia and ammonium) N (TAN) from excretion through to land spreading. As inputs, the model requires excretion fractions, census of agriculture animal populations and detailed farm management practices from the survey. They stated that the N intake by grazing cattle is especially difficult to quantify. Early season grazing, when forages have elevated crude protein (>300 g CP/kg dry matter), indicated potential for elevated emissions in May and June. Such exceptional crude protein concentrations during spring grazing may be important for N emissions, especially NH₃. They added that beef cattle were reported by producers to spend 7%–30% of their time congregated, such as around water sources, feed or shade trees. This level of biosphere activity was attributed in the model to ~20% increases in NH₃ emissions relative to open pasture, and similar or higher effects would be expected for N₂O emissions.

4.4 Whole farm systems models

Crosson et al. (2011) stated that in order to comply with the United Nations Framework Convention on Climate Change (UNFCCC) greenhouse gas (GHG) emissions reporting requirements, the Intergovernmental Panel on Climate Change (IPCC, 2006) developed guidelines for calculating national GHG inventories in a consistent and standard framework. Although appropriate for national level accounting purposes, IPCC methodologies lack the farm level resolution and holistic

approach required for whole farm systems analysis. Thus, whole farm systems modeling is widely used for farm level analysis. Whole farm systems models are an appropriate tool to develop and measure GHG mitigation strategies for livestock farms. Therefore, developing a methodology for estimating emission at farm level reaching the national level would be an asset. Variation in farm system parameters, and the inherent uncertainties associated with emission factors, can have substantial implications for reported agricultural emissions and thus, uncertainty or sensitivity analysis in any modeling approach is needed. Although there is considerable variation among studies in relation to quality of farm data, boundaries assumed, emission factors applied and co-product allocation approach, we suggest that whole farm systems models are an appropriate tool to develop and measure GHG mitigation strategies for livestock farms.

5 Tools for processing emission inventories

5.1 Mass-Flow models

The mass-flow approach to estimating NH_3 emissions has evolved from initially estimating only NH_3 emissions (Cowell and ApSimon, 1998), to including processes such as immobilization, mineralization, nitrification and denitrification in order to properly quantify the TAN flow (Webb and Misselbrook, 2004) and to improve the accuracy of the NH_3 emission calculations. Webb and Misselbrook (2004) described a mass-flow approach to estimating NH_3 emissions from livestock production at national scale. NH_3 is emitted from a pool of ammoniacal-N (TAN) in livestock excreta. This pool is not added to during manure management, but is depleted by losses as gaseous emissions and leachate and by immobilization in litter. At each stage of manure management, a proportion of TAN will be lost, mainly as NH_3 , and the rest passed on to the next stage. This approach enables rapid and easy estimation of the consequences of abatement at one stage of manure management (upstream) on NH_3 losses at later stages of manure management (downstream). Such a model facilitates scenario analysis of abatement options and cost-curve production. Model output is most sensitive

to variation in estimates of the length of the housing period for cattle. Thus, the collation of accurate data on factors such as the length of the housing period and other 'activity' data, are as important in compiling accurate inventories of national emissions as improving the accuracy of emission factors. Priorities for research should be to accurately quantify the relationship between NH_3 emissions from livestock buildings and the proportion of the day those buildings are occupied, and to characterize and quantify the transformations of N that take place during storage of litter-based manures.

Gac et al. (2007) developed a methodology based on (1) national data concerning livestock and rearing practices and (2) a mass-flow approach which was developed to quantify NH_3 , CH_4 and N_2O emissions resulting from manure management. They performed a literature review to determine emission factors for each animal type and each management stage. Afterwards, they have developed a Microsoft Access® database containing these emission factors, the census data and manure compositions, allowing the calculation of gaseous emissions by the mass-flow approach. From this database, a national gas emissions inventory resulting from manure management was drawn up.

Reidy et al. (2009) and Reidy et al. (2008a) compared six N-flow models used to calculate national NH_3 emissions from litter-based systems and slurry-based systems, respectively, in different European countries using standard data sets. It was found that output of the models tested proved to be much more variable for solid manure than for slurry. The variability of emissions found in practice is likely to be much greater for straw-based systems than for slurry systems. They added that the differences in estimates of NH_3 emissions decreased as estimates of immobilization and other N losses increased. Since immobilization and denitrification depend also on the C:N ratio in manure, there would be advantages to include C flows in mass-flow models. This would also provide an integrated model for the estimation of emissions of methane, non-methane VOCs and carbon dioxide. Estimation of these would also enable an estimate of mass loss, calculation of the N and TAN concentrations

in litter-based manures and further validation of model outputs.

Wang et al. (2011) developed a detailed inventory framework to estimate N_2O and CH_4 emissions from UK agriculture using the IPCC approach. The inventory framework model was illustrated by combining relevant emission factors with agricultural census data for England, Wales, Scotland and Northern Ireland for the year 2000 to derive country-specific emission estimates which were summed to derive the UK total. The framework enables simple assessment to be made of the impact on national emissions of using different emission factors (EFs) (e.g. site- or local-specific compared with IPCC default factors). The framework was used to calculate the average annual emissions of nitrous oxide N_2O and CH_4 for specific livestock and crops, and amounts lost through volatilization, leaching and runoff for each country in the UK. The framework provides a simple, realistic and transparent approach to estimating national emissions and can easily be updated.

Bannink et al. (2011) stated that the protocol for the National Inventory of agricultural greenhouse gas emissions in The Netherlands includes a dynamic and mechanistic model of animal digestion and fermentation as an Intergovernmental Panel on Climate Change (IPCC) Tier 3 approach to estimate enteric CH_4 emission by dairy cows. The model predicts CH_4 emission by considering various dietary characteristics, including the types of carbohydrate, protein, fat, intrinsic degradation characteristics of feeds, as well as ruminal fractional passage rates, fluid volume and acidity, instead of assuming a fixed CH_4 energy conversion factor in the Tier 2 approach.

Pouliot et al. (2012) highlighted the similarities and differences in how emission inventories and datasets were developed and processed across North America and Europe for the Air Quality Model Evaluation International Initiative (AQMEII) project and then characterized the emissions for the two domains. They focused specifically on the creation of “model-ready” gridded emission datasets for 2006 across the two continental study domains. They discussed the practice of creating and processing the two inventories with a

focus on emission factors, spatial allocation, temporal variability, speciation of PM and VOCs, and the mechanics of distributing the data and supporting emission algorithms to the modeling community. The spatial and temporal distribution on common scales was compared for the pollutants of primary concern: NO_x , VOCs, SO_2 , $PM_{2.5}$, CO, and NH_3 . Because of differences of population distribution, emissions across North America tend to be more heterogeneous in spatial coverage than in Europe. The temporal patterns in the estimated emissions are largely the result of assumptions used to characterize human activity, with the exception of “natural” emissions, which are modulated by meteorological variability, and emissions from large electric generating units in the U.S., which have the benefit of continuous emission monitors that provide hourly resolved profiles. Emission estimates in both study domains are challenged by several important but poorly characterized emission source sectors, notably road dust, agricultural operations, biomass burning, and road transport. One important outcome of this comparison of 2006 emissions between Europe and North America is the greater understanding provided into how the emission estimates developed for the AQMEII project impact regional air quality model performance. On the other hand, the Emission Database for Global Atmospheric Research (EDGAR) provides global past and present-day anthropogenic emissions of air pollutants and greenhouse gases by country and on a $0.1^\circ \times 0.1^\circ$ grid. The current development of EDGAR is a joint project of the European Commission JRC Joint Research Centre and the Netherlands Environmental Assessment Agency.

Velthof et al. (2012) stated that NH_3 emissions from agriculture are quantified using a nitrogen (N) flow approach, in which the NH_3 emission is calculated from the N flows and NH_3 emission factors. Because of the direct dependency between NH_3 volatilization and Total Ammoniacal N (TAN; ammonium-N + N compounds readily broken down to ammonium) an approach based on TAN is preferred to calculate NH_3 emission instead of an approach based on total N. They developed a TAN-based NH_3 -inventory model, called NEMA

(National Emission Model for Ammonia). The total N excretion and the fraction of TAN in the excreted N are calculated from the feed composition and N digestibility of the components. TAN-based emission factors were derived or updated for housing systems, manure storage outside housing, manure application techniques, N fertilizer types, and grazing.

Hellsten et al. (2008) provided an updated spatial NH₃ emission inventory for the UK for the year 2000, based on an improved modeling approach and the use of updated input datasets of a model called AENEID1 (Atmospheric Emissions for National Environmental Impacts Determination) which its methodology constituted the basis for the updated model AENEID2 that distributes NH₃ emissions from a range of agricultural activities, such as grazing and housing of livestock, storage and spreading of manures, and fertilizer application, at a 1-km grid resolution over the most suitable landcover types.

5.2 Software systems for emission inventories

Samaali et al. (2007) stated that emission databases need to be processed for several purposes, e.g., policy and scientific, and so require flexible and fast handling. As a result, the creation and use of convenient interactive tools allowing easy and fast processing are of high importance. They presented software devoted to emission inventories processing and applied to the actualization of the ESCOMPTE inventory. The ESCOMPTE inventory is a yearly high-resolution (1 km²) emission inventory created for use in chemical transport models to study ozone pollution. This tool includes several functions allowing the update of emission inventories at different temporal and spatial resolutions based on the reference ESCOMPTE inventory. The actualized yearly and hourly inventories are found consistent with the hypothesis involved in emission inventory creation. Due to its flexible architecture, several other applications related to air quality analysis can be performed such as the creation of emission scenarios allowing the evaluation of abatement strategies and regulation effects on anthropogenic emissions. Also, the architecture of the software allows an easy application to other emission inventories provided that they have

similar or adaptable structure compared to the ESCOMPTE one.

Winiwarter and Schimak (2005) stated that atmospheric emission inventories are important tools for environmental decision making. The need to include transparency and reproducibility in emission calculation also fostered the development of environmental software systems for emission inventories. In general such software systems are organized as databases which contain as much emission related information as is available. For their own concept they introduced a layer of emission information as interim results, which were stored as specific tables. While this structure is needed for allowing computations at an acceptable response time, it is also used to differentiate detail levels between a user's sphere and an expert's sphere. In the user's sphere, it is expected that any user adapts the evaluation algorithms without the need to know about limitations in emission calculations. Still also the expert's sphere is openly accessible, allowing for full transparency of the calculation procedures. Considering uncertainty of input data and the impossibility to perform full validation, much of the information contained in an inventory will not contribute to improve the results in terms of emissions. Nevertheless, this information is still valued as a contribution to an expert system, which may become crucial at a different application of an inventory. Emission inventory systems generally contribute to comparability between data from diverse inputs, especially as they are able to support error-checking procedures, an important task due to the large number of data providers.

6 Evaluation and Improvement of Emissions Inventories

The 2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines) provide General Guidance and Reporting (in Volume 1) which includes: Approaches to Data Collection, Uncertainties, Methodological Choice and Identification of Key Categories, Time Series Consistency, Quality Assurance/Quality Control (QA/QC) and Verification, Precursors and Indirect Emissions, and Reporting Guidance and

Tables. The contents of this volume provide general guides to develop high quality emissions inventories (IPCC, 2006). Similarly, The EMEP/EEA air pollutant emission inventory guidebook provides guides on: Key category analysis and methodological choice, Data collection, Time series consistency, Uncertainties, Inventory management, Improvement and quality assurance and control QA/QC (EMEP/EEA, 2009). On the other hand, the Executive Body for the Convention on Long-Range Transboundary Air Pollution (2007a), reported methods and procedures for the technical review of air pollutant emission inventories.

In the early phase of the evolution of the emissions inventories, Rypdal and Flugsrud (2001) developed a sensitivity analysis as a tool for systematic reductions in GHG inventory uncertainties. They elucidated that a greenhouse gas emission inventory consists of a large number of input parameters, many of which have high uncertainties. The Kyoto protocol requires accurate emission data. It is, consequently, important to reduce the inventory uncertainty by improving the input parameters and methodologies in a cost-efficient manner. They defined a key parameter as one that has significant effect on the total emissions or trend and their uncertainty. Key parameters may be ranked according to their contribution to total emissions and trend uncertainty. Quantitative uncertainty estimates of emissions are not available in many countries. In order to evaluate key parameters in such inventories, they proposed a simplified approach based on thresholds. The simplified threshold approach gives insight into the inventory and identifies the key parameters. More sophisticated methods of sensitivity analysis assessments are, however, useful in order to seek specific improvements. The key parameters for determination of total emissions are the large and uncertain sources. Smaller emission sources may be key parameters for the trend determination if their source level is strongly increasing or decreasing.

Battye et al. (2003) mentioned that in terms of the potential change in mass of NH_3 emissions over regional domains, the uncertainty for soils and vegetation, and livestock waste represents a large amount, where these uncertainties stem from variations in measured emission

factors. All of the sources of NH_3 emissions are subject to variability and uncertainty. The soils and vegetation category represents one of the most important sources of uncertainty, whereas Warneck (1988) stated that plants will either absorb or give off ammonia depending on the concentration of an ammonium ion in the plant and the concentration of ammonia gas in the surrounding air. Accordingly, Battye et al. (2003) stated that the emissions estimates used in atmospheric models for vegetation should probably be linked to predicted concentrations of free ammonia, which would vary through the year and across the modeling domain. They added that the lack of monitoring data on ammonia gas is another important data gap. Without data on ammonia gas, there is little that can be done to evaluate the validity of the ammonia inventory. This lack of data also complicates any evaluations of modeled equilibria among NH_3 , NH_4^+ , NO_3^- and SO_4^{2-} . They added that, in terms of the mass of emissions, uncertainties for the livestock waste category are very large. In addition, livestock emissions are highest in rural areas and in the summer. Uncertainties in NH_3 emissions would have the strongest impact in winter, when the formation of NH_4NO_3 particulate is favored by colder temperatures, and in urban areas, where NH_3 may be the limiting component in the formation of particulate NH_4NO_3 . Although additional measurements would be helpful, a better characterization of the timing of livestock waste and fertilizer application would also reduce the uncertainty of seasonal emissions estimates.

Ramirez et al. (2008) stated that the main contributors to overall uncertainty of inventories are found to be related to N_2O emissions from agricultural soils, the N_2O implied emission factors of Nitric Acid Production, CH_4 from managed solid waste disposal on land, and the implied emission factor of CH_4 from manure management from cattle.

Dröge et al. (2010) mentioned that emissions estimated within the Dutch Emission Inventory were compared to emissions estimated using Guidebook emission factors and Dutch statistics for the year 2005. The objective was to explore the quality of both methods and to find major differences and similarities. The

comparison shows that for most sources, emission estimates are within uncertainty ranges for both methodologies, especially for sources where a higher Tier (more detailed) methodology is used to estimate the emissions. This is in line with the Guidelines which indicate that for key categories a more detailed methodology should be used. The comparison also shows some surprising differences, such as large differences in emission factors (especially Tier 1) and missing sources which have not been included in the Guidebook. This comparison is shown to be a useful tool to identify areas where improvements and further research are necessary.

Stabler et al. (2009) investigated NH_3 emissions plume from a beef feedlot in order to improve the Canadian NH_3 emission inventories and air quality forecasting capabilities. Lindley et al. (2000) investigated the uncertainties in the compilation of spatially resolved emission inventories. These investigations indicated that there are a number of variations between inventory results in terms of overall emission magnitudes and their spatial distribution. O'Shaughnessy and Altmaier (2011) conducted investigations to determine optimal settings applied to the plume dispersion model, AERMOD, for accurately determining the spatial distribution of hydrogen sulfide (H_2S) concentrations in the vicinity of swine concentrated animal feeding operations (CAFOs). They found that when simulating the spatial distribution of H_2S around a hypothetical large swine CAFO (1 M kg), concentrations within 0.5 km from the CAFO exceeded 25 ppb and dropped to 2 ppb within 6 km of the CAFO. These values compare to a level of 30 ppb that has been determined by the State of Iowa as a threshold level for ambient H_2S levels.

Bannink et al. (2011) stated that uncertainty of model predictions of CH_4 emission is determined mostly by errors in feed intake estimation, in the representation of the stoichiometry of production of volatile fatty acids (VFA) from fermented substrate, and in the acidity of rumen contents. Further uncertainty of predicted CH_4 emission can be due to errors in estimation of dietary composition of ingredients and in chemical compositions of dietary components. They added that prediction of

CH_4 should not solely focus on representing effects of nutrition on overall digestion and apparent feed utilization by cows, but that additional attention is needed to address effects of nutrition on intra-ruminal fermentation conditions, and their effects on formation of VFA and the rumen hydrogen balance.

7 Paradigms of national emissions inventories

Hyde et al. (2003) stated that the estimated total emissions from Irish agriculture were 89.9 and 91.8 kt $\text{NH}_3\text{-N}$ for 1991 and 2010, respectively. Cattle farming account for more than 75% of total emissions. The largest emission factors found included 46.9 g $\text{NH}_3\text{-N}$ lu^{-1} d^{-1} for cattle housing, 29.5 g $\text{NH}_3\text{-N}$ lu^{-1} d^{-1} for pig housing and 150 g $\text{NH}_3\text{-N}$ lu^{-1} d^{-1} for housed broilers (lu being equivalent to 500 kg live weight). They added that abatement strategies should be implemented to achieve the agreed national emission reduction target by 12% in 2010. They also found that strategies for reducing emissions from the land spreading of manure offer the greatest potential to achieve target levels.

Yamaji et al. (2003) estimated the CH_4 emissions from livestock in South, Southeast, and East Asia and found to be about 29.9 Tg CH_4 in 2000 using the Food and Agriculture Organization database and district-level data on regional activity and emission factors, considering regional specificities. These emissions consisted of 25.9 Tg CH_4 from enteric fermentation and 4.0 Tg CH_4 from livestock manure management systems. India had the greatest production, with 11.8 Tg CH_4 from livestock, primarily cattle and buffaloes. China was also a high-emission country, producing about 10.4 Tg CH_4 . They found that the total CH_4 emissions from livestock increased by an average of 2% per annum from 1965 to 2000 in Asia.

Gac et al. (2007) developed a national gas emissions inventory resulting from manure management and the data was drawn up for the year 2003 in France. Total NH_3 emissions were estimated at 382 kt N, mainly arising from cattle (72%). Greenhouse gas emissions were estimated at 14.0 Tg $\text{CO}_2\text{-Equation}$ for N_2O and 10.2 Tg $\text{CO}_2\text{-Equation}$ for CH_4 . Most of the N_2O emissions occurred after the deposition of manure on soil during

cattle grazing, while the CH₄ was mainly produced during the period when cattle manure remained in livestock buildings and in outside storage facilities.

Zhou et al. (2007) presented a systematic estimation of CH₄ and N₂O emissions during 1949–2003, based on the local measurement and IPCC guidelines, in order to investigate the GHG emissions from enteric fermentation and manure management of livestock and poultry industry in China. As far as GHG emissions are concerned among livestock, swine is found to hold major position followed by goat and sheep, while among poultry chicken has the major place and is followed by duck and geese. CH₄ emissions from enteric fermentation are estimated to have increased from 3.04 Tg in 1949 to 10.13 Tg in 2003, an average annual growth rate of 2.2%, and CH₄ emissions from manure management have increased from 0.16 Tg in 1949 to 1.06 Tg in 2003, an annual growth rate of 3.5%, while N₂O emissions from manure management have increased from 47.76 to 241.2 Gg in 2003, with an annual growth rate of 3.0%. The total GHG emissions have increased from 82.01 Tg CO₂ Equivalent in 1949 to 309.76 Tg CO₂ Equivalent in 2003, an annual growth rate of 2.4%. The estimation of livestock CH₄ and N₂O emissions in China from 1949 to 2003 is shown to be consistent with a linear growth model, and the reduction of GHG emissions is thus considered an urgent and arduous task for the Chinese livestock industry.

Aljaloud et al. (2011) developed a national emissions inventory for Saudi Arabia using data derived from farm surveys and national statistical records from 2003 to 2007, as well as Tier 1 and 2 emission factors for enteric fermentation and manure management as proposed by the Intergovernmental Panel on Climate Change. They reported that the national CH₄ emission inventory in Saudi Arabia, developed using Tier 1 default factors, increased from 87,069 t in 2003 to 100,971 t in 2007, with a CO₂ equivalent of 2,176,735 and 2,524,287 t, respectively. The contribution to the inventory varied considerably among provinces, ranging from 0.1% (Northern) to 40.6% (Riyadh) in 2007 when horse data were not included. Among animal species, dairy cattle and sheep were the biggest CH₄ producers (37.7% and

35.5%, respectively), followed by camels (13.4%), goats (11.4%), chickens (1.6%) and horses (0.4%). National CH₄ emissions consisted of 0.87 from enteric fermentation and 0.13 from manure storage. Tier 2 CH₄ emission factors were developed for dairy cattle and sheep, with each species divided into two groups (i.e., specialized high genetic merit and traditional low genetic merit), and each group of cattle and sheep were further divided into subgroups according to their physiological states. These factors for enteric fermentation and manure management were all different from Tier 1 default factors, and resulted in a decrease of 2088 t in CH₄ emission inventories (CO₂ equivalent of 52,200 t) from cattle and sheep when estimated using Tier 2, rather than Tier 1, factors. In comparison with other countries, the national CH₄ emission inventory from domestic animals in Saudi Arabia is relatively small in terms of CH₄ emissions per unit of land area or human population. These results can be used, with data from other agricultural sectors, to develop a national greenhouse gas inventory for the entire agricultural sector of Saudi Arabia.

Velthof et al. (2012) stated that the NEMA results show that the total NH₃ emission from agriculture in the Netherlands in 2009 was 88.8 Gg NH₃-N, of which 50% from housing, 37% from manure application, 9% from mineral N fertilizer, 3% from outside manure storage, and 1% from grazing. Cattle farming are the dominant source of NH₃ in the Netherlands (about 50% of the total NH₃ emission). The NH₃ emission expressed as percentage of the excreted N was 22% of the excreted N for poultry, 20% for pigs, 15% for cattle, and 12% for other livestock, which is mainly related to differences in emissions from housing systems. The calculated NH₃ emission was most sensitive to changes in the fraction of TAN in the excreted manure and to the emission factor of manure application. From 2011, NEMA has been used as official methodology to calculate the national NH₃ emission from agriculture in The Netherlands.

8 Emissions abatement techniques

Around 75% of ammonia (NH₃) emissions come from livestock production (Reinhardt-Hanisch, 2008).

Emissions occur at all stages of manure management: from buildings housing livestock; during manure storage; following manure application to land; and from urine deposited by livestock on pastures during grazing. Ammoniacal nitrogen (total ammoniacal-nitrogen, TAN) in livestock excreta is the main source of NH_3 . At each stage of manure management TAN may be lost, mainly as NH_3 , and the remainder passed to the next stage. Hence, measures to reduce NH_3 emissions at the various stages of manure management are interdependent, and the accumulative reduction achieved by combinations of measures is not simply additive. This TAN-flow concept enables rapid and easy estimation of the consequences of NH_3 abatement at one stage of manure management (upstream) on NH_3 emissions at later stages (downstream), and gives unbiased assessment of the most cost-effective measures. Rapid incorporation of manures into arable land is one of the most cost-effective measures to reduce NH_3 emissions, while covering manure stores and applying slurry by band spreader or injection are more cost-effective than measures to reduce emissions from buildings. These measures are likely to rank highly in most European countries (Webb et al., 2005). The implementation of urease inhibitors is effective in reducing ammonia emissions from cattle and pig slurry (Reinhardt-Hanisch, 2008). She added that the basic investigations on urease inhibitors afforded an important contribution to the expansion of knowledge in this area, and will lead on the other hand to develop new techniques in order to reduce the NH_3 emissions from livestock housing. Rahman et al. (2011) evaluated the effectiveness of the Digest3+3© microbial additive for reducing odor and pollutant gas emission from a swine gestation-farrowing operation, where the additive was used to treat the deep pits to be compared with other untreated pits. However, they found no significant differences in terms of odor, NH_3 , and H_2S concentrations and emissions between treated and untreated units. Overall, the microbial treatment had very little effect in reducing odor, ammonia, and hydrogen sulfide emission. Wheeler et al. (2011a) stated that amendments can be practical and cost-effective for reducing NH_3 and GHG emissions from dairy manure. They found that six

amendment products that acted as microbial digest, oxidizing agent, masking agent or adsorbent significantly can reduce NH_3 by more than 10%. They added that microbial digest/enzymes with nitrogen substrate appeared effective in reducing CH_4 fluxes. For both CH_4 and CO_2 fluxes, aging the manure slurry for 30 d can significantly reduce gas production.

Carew (2010) stated that diet manipulation to improve animal N utilization efficiency and land spreading techniques to inject or incorporate manure into the soil are the most effective measures to reduce livestock NH_3 emissions compared to housing and manure storage techniques. While biofilters, air scrubbers and urine/feces separation techniques offer potential opportunities to lower emissions, these options are expensive and impractical to implement on commercial farms. Further research is needed to understand the factors limiting livestock producers adopting mitigating strategies to reduce emissions since a whole-farm system approach can provide a modeling framework to evaluate the feasibility and cost-effectiveness of abatement measures. Sommer et al. (2004) stated that daily flushing of slurry from cattle houses would reduce total annual $\text{CH}_4 + \text{N}_2\text{O}$ emissions by 35% (CO_2 Equation), and that cooling of pig slurry in-house would reduce total annual $\text{CH}_4 + \text{N}_2\text{O}$ emissions by 21% (CO_2 Equation). Giltrap et al. (2010) investigated the effect of NI dicyandiamide (DCD) on transformations of N to nitrate (NO_3^-) and subsequent reduction to N_2O in a grazed pasture system receiving cow urine, where the DCD was able to decrease the nitrification rate. Based on this study, an issue can be raised: is the DCD able to be used as an inhibitor of N_2O emissions from floor inside a livestock building?

Liquid manure storage facilities are sources of gaseous emissions of NH_3 and greenhouse gases especially CH_4 and N_2O . Additives can reduce gaseous emissions from swine waste lagoons and pits. The additives have the potential to reduce methane emissions from anaerobic swine lagoons (Shah and Kolar, 2012). Different materials for covering liquid manure storage facilities have been investigated and are in use for mitigating odor and ammonia emissions (Sommer et al.,

1993; Williams, 2003). These materials abate also methane and nitrous oxide emissions (Berg et al., 2006). Manipulating the balance between ammonia and ammonium by lowering the pH-value of slurry is another measure to reduce emissions (Stevens et al., 1989; Oenema and Velthof, 1993; Hendriks and Vrieling, 1997; Kroodsma and Ogink, 1997; Martinez et al., 1997; Beck and Burton, 1998; Pedersen, 2003).

Ammonia and methane emissions can be controlled by pH-value. Manipulating the pH-value of slurry has an effect on the balance between ammonia and ammonium. The pH-values of untreated slurries range between seven up to eight usually. Lowering the pH reduces the gaseous emission. From former investigations it is known that a slurry pH around 5.5 can reduce ammonia emission by 80% to 90% (Al-Kanani et al., 1992; Berg et al., 2006; Husted et al., 1991; Li et al., 2006; Pain et al., 1990; Stevens et al., 1989). A slurry pH below 4.5 nearly avoids ammonia emission (Hartung and Phillips, 1994). The pH-value influences the activities of microorganisms. Higher methane production occurs, when the pH-value is between 6 and 7 (Lay et al., 1997). A slurry pH below 6 is necessary to reduce methane emission and below 5 impede methane formation (Berg et al., 2006). Whereas the use of inorganic acids has several disadvantages, using organic acids is a promising possibility to reduce not only ammonia but also methane and nitrous oxide emissions (Berg and Hoernig, 1997; Berg and Pazsiczki, 2003; Berg, 2003). Previous studies have evaluated additives for reducing gaseous emissions from manure in 5 laboratory using glass jars or plexiglass tanks and a multi-gas monitor (Wheeler et al., 2011b; Reinhardt-Hanisch, 2008; Berg et al., 2006). Zhang et al. (2008) confirmed the generally positive impacts of anaerobic and aerobic treatment on the reductions of methane and volatile organic compounds (VOCs). However, the effects of anaerobic and aerobic treatment varied over the time of storage, especially for VOCs. They indicated that to achieve significant reductions in VOC emission the storage time of anaerobic digester or aerobic reactor effluent should be limited to no more than 84 days.

On the other hand, adjusting the housing system and

the management has the potential to decrease the emissions. For instance, Dekker et al. (2011) stated that using organic laying hen husbandry in aviary systems instead of single-tiered systems has the potential to reduce emissions of NH_3 , N_2O , and CH_4 ; further reductions might be realized by changes in litter management. Regarding manure management, Haeussermann et al. (2006) stated that methane emissions can be significantly reduced by complete slurry removal between the fattening periods and subsequent cleaning of the slurry pits in pig housing. Additionally, the release of methane from indoor slurry storage can be influenced by availability of oxygen and volatile solids, pH value, substrate temperature, retention time, and presence of inhibiting compounds. These factors should be further investigated to develop emissions abatement techniques. Special considerations should be given to avoid increasing specific gas emissions while abating another one. For instance, Brink et al. (2001) found that abating agricultural emissions of NH_3 may cause releases of N_2O from this sector up to 15% higher than in the case of no NH_3 control. Ngwabie et al. (2011) stated that increased knowledge of the factors that affect emissions from livestock barns may lead to a better understanding of daily (between different days) and diurnal (within a specific day) variations in emissions, an improvement of mitigation methods and a refinement of emission models. Animal activity, animal weight, indoor air temperature and relative humidity have influence on carbon dioxide, methane, and ammonia emissions. Emission variations emphasized the need for measurements during different times within the day and during the growing period in order to obtain reliable data for assessing abatement techniques.

Sommer and Hutchings (2001) reviewed ammonia emission from field applied manure and its reduction. They found that the use of trail hoses, pre- or post-application cultivation, and reduction in slurry viscosity, choice of application rate and timing and slurry injection are considered as reduction techniques. The most effective methods of reducing ammonia emissions were concluded to be incorporation of the animal slurry and farmyard manure or slurry injection. Incorporation

should be as close to the application as possible, especially after slurry application, as loss rates are high in the first hours after application. Injection is a very efficient reduction technique, provided the slurry is applied at rates that can be contained in the furrows made by the injector tine.

Melse et al. (2009) stated that the EU emission norms for PM have come into effect recently, may be limited for continuation and/or expansion of intensive livestock operations in the near future, alongside existing ammonia and odor emission standards. Therefore, the implementation of dust abatement techniques is crucial. Mostafa (2008) listed several means for reduction of dust in/from livestock houses, such as: (1) dust suppression with spraying oil and/or water, (2) ionization, (3) cleaning the air by oxidization using chemical compounds called “oxidants”, (4) use of windbreaks, (5) implementation of de-dusters, scrubbers and filters (dry and wet).

The EMEP/EEA air pollutant emission inventory guidebook provides greenhouse emission abatement measures for animal husbandry and manure management in form of Best Available Technique (BAT) for each case, type of manure, land use, limits of applicability, emission reduction (%), availability for different farms (EMEP/EEA, 2009). On the other hand, the Executive Body for the Convention on Long-Range Transboundary Air Pollution (2007b) provided a guidance document on control techniques for preventing and abating emissions of ammonia. Moreover, the European Commission (2003) provided an Integrated Pollution Prevention and Control (IPPC) reporting on the BAT for intensive rearing of poultry and pigs.

9 Discussion

Based on a literature review of several studies, Crosson et al. (2011) criticized the IPCC methodologies which provide a set of generalized guidelines for compiling and reporting national inventories and, as such, provide a transparent and consistent framework for comparing national GHG emissions at various times. However, limitations with respect to this national and sectoral approach undermine the usefulness of the methodology for modeling at the farm level. In this

respect, whole farm modeling is widely employed for farm level GHG emissions modeling. Despite the increased flexibility and sensitivity of these models to capture farm level activity, concerns and variation relating to data sources, boundaries (i.e., which emission sources are included), emission factors and allocation approaches exist, and they limit direct comparison among studies. Thus, it is critical that assumptions made in this regard are clearly outlined. Furthermore, uncertainty analysis has a key role to play given the range of GHG emission factors reported and the inherent uncertainty involved in agricultural processes. Nevertheless, in terms of developing and assessing mitigation policies to reduce GHG emissions from livestock systems, whole farm approaches provide the most robust and comprehensive approach to developing and implementing effective strategies.

On the other hand, the livestock housing system and design greatly affect the livestock's microclimate (especially temperature and air velocity near animals) which has a direct influence on gas emissions from livestock barns. Several studies investigated the effect of barn design on animals' microenvironment, where the barn design and accordingly the management of the different components (including cooling or heating systems) inside the barn have a great effect on the indoor bioenvironment. The most important –in this context– bioenvironmental components are the temperature and the air velocity which when increase or decrease the emissions rates are directly affected (Samer, 2004; Hatem et al., 2004a,b; Hatem et al., 2006; Samer, 2008; Samer et al., 2008a,c; Ngwabie et al., 2010; Samer, 2010a,b,c; Samer, 2011a,c; Pereira et al., 2012; Samer, 2012a; Samer et al., 2012a,b; Samer et al., 2013a; Samer 2013), where the highest average NH₃ emission coincided with higher environmental temperatures (Adviento-Borbe et al., 2010). Ammonia emissions are about twice higher at manure temperatures of about 25°C than emissions at about 15°C (Ngwabie et al., 2010). Decreasing velocity, turbulence intensity and liquid temperature are shown to reduce the ammonia emission rates. The emission rates are more sensitive to the change of velocity at a low velocity compared to change of velocity at a higher

velocity range, which corresponds with the conclusion that the boundary layer thickness of velocity increases sharply when velocity is changed from 0.2 m s^{-1} to 0.1 m s^{-1} . In addition, the emission rates are more sensitive to the change of temperature at a higher temperature than at a lower liquid temperature range (Rong et al., 2009). Pereira et al. (2012) stated that increasing temperature from 5 to 35°C significantly increases potential NH_3 , CO_2 and CH_4 emissions from excreta of the housed cattle, but it does not significantly influence N_2O emissions. Also, the diet supplied to lactating cows leads to significantly higher NH_3 , N_2O and CO_2 emissions relative to heifers and dry cows. On the other hand, seasonal variations of gaseous emissions, regional conditions (i.e. climatic conditions: hot or cold climates) have direct impact on the gaseous emissions flow rates which increase in summer (Gilliland et al., 2006; Haeussermann et al., 2006; Samer, 2011d). Further, the design of the implemented ventilation system, which is mainly natural ventilation system for several livestock types, as well as the ventilation rate or the air exchange rate have direct effect of the amount of emissions flow rates of the different gases (Samer et al., 2011a,b,c,d,e,f,g; Samer et al., 2012c). The flux of emissions of harmful gases from naturally ventilated buildings is dependent on wind velocity (both speed and direction) and turbulence levels inside and over the outside of the building, thus emission mass flow is highly variable and difficult to estimate (Ngwabie et al., 2009; Van Buggenhout et al., 2009; Samer et al., 2013b). Morsing et al. (2008) hypothesized that the effects on gas emissions are as a consequence of changing airflow patterns and different types of flow in the boundary layer between the slurry and ventilation air. Odors and gases emitted from animal houses are strongly related to airflows. Haeussermann et al. (2006) found a clear influence of the indoor temperature – and therefore of the ventilation strategy – on the level of the methane emission rate for mean daily temperatures above 25°C . On the other hand, the factors-of-influence (FOI) that strongly influence the dispersion of NH_3 are: NH_3 -mass-flow, internal and external temperatures, mean and turbulent wind components in horizontal and vertical directions, atmospheric stability, and exhaust air height

where the continuous measurement of NH_3 remains a challenging and costly enterprise, in terms of capital investment, running costs or both (Von Bobrutski et al., 2010; Von Bobrutski et al., 2011). Additionally, the floor type plays an important role where according to Pereira et al. (2011) the gaseous emissions are significantly greater from the solid floor relative to the slatted floor at all temperatures (5 , 15 and 25°C). Low emission values can only be achieved by reducing the emission source surfaces, by decreasing temperature and air velocity near the source, and minimizing air volumetric flow rates throughout the livestock buildings (Bjorneberg et al., 2009; Blanes-Vidal et al., 2007; Gay et al., 2003). Nevertheless, the type of manure management and the design of the handling system (Samer et al., 2008d; Samer, 2011b; Samer, 2012b) will have direct effect on the emissions level. For instance, Moset et al. (2012) found that there was relevant transformation of the more degradable organic matter (OM) into soluble OM during the first week of aged fattening pig slurry storage. This transformation is more pronounced in the slurry with a higher initial OM concentration in raw slurry (RS) than in separated slurry (SS), indicating a higher hydrolytic, acidogenic and acetogenic activity, as well as higher rate of urea mineralization and nitrogen denitrification rate at the beginning of the storage period in RS than in SS. Special considerations for the emission rate should be given if a biogas plant has been implemented in the livestock farm, where such implementation influences considerably the emissions rate of the different gases as the manure is handled differently from aerobic stores (Samer, 2010d; Samer, 2012b). For instance, the type of manure management in caged-layer poultry has a great impact on the level of emissions, where belt/composting has less than half the total emissions of conventional deep-pit caged-layer systems (Keener and Zhao, 2008). Another issue should be considered which is the farmstead layout especially regarding the orientation of different buildings in respect to the prevailing wind as well as the distance between the adjacent buildings (Samer et al., 2008b), where such layout parameters will affect the immission and transmission of the gases and

eventually affect the accuracy of the emission factors. The abovementioned statements confirm the need to relate the emissions inventories to whole farm systems and treating each farm as a special case where farm survey would be an effective method, which agree with Crosson et al. (2011). Sommer et al. (2006) stated that a detailed knowledge of the processes of NH₃ transfer from the manure and transport to the free atmosphere will contribute to development of techniques and housing designs that will contribute to the reduction of NH₃ emission to the atmosphere.

10 Conclusions

Current emissions inventories lack of detailed information on emissions from livestock buildings. They focus on emissions from animal husbandry (mainly enteric fermentation) as well as manure storages and field application of manure. However, livestock barns encompass large contaminated areas that form an important emitting source of pollutants. Besides, manure can be stored inside the livestock barns for short periods and this is also a considerable source of emissions. One key issue is to integrate the emission factors, mass flow models and algorithms that consider emissions from livestock barns with those of animal husbandry and manure management (handling, stores, and field application) to allow developing comprehensive emissions inventories.

On the other hand, this study focuses on emissions and emissions inventories from animal husbandry, manure management and livestock buildings. Nevertheless, these emissions should be integrated with those from soils in order to form agricultural emissions inventory and further integrated with the other main sources of emissions (waste, energy, and industry) in order to make a regional/territorial emissions inventory for a specific region/province. Subsequently, the different regional emissions inventories are then compiled together to make the national inventory.

According to the issues raised in this study, it can be further concluded that:

1) It is crucial to consider the emissions from the livestock buildings in the emissions inventories, where

the enteric fermentation and the manure stores are not the only sources of emissions from animal husbandry. One key issue is to consider the manure management system inside the building (slatted floor, manure scrapper, litter-based system...), floor type, building design, and ventilation system where these parameters have discernible influence on the emissions flow rates from livestock building. Therefore, each farm should be treated as special case, where some farms may have applied emissions abatement techniques and other farms may have not, and therefore farm survey is necessary to be integrated into the emissions inventorying procedures. Furthermore, the emission rates depends not only the barn design and the housing system but also on specific parts/compartments inside the barn (e.g. the different rooms inside a swine barn). This should be considered when inventorying the emissions.

2) The contribution to a national emissions inventory varies discernibly among the different states, provinces or governorates within the same country depending on the geographical distribution of the livestock farms, where the geographical area which encompasses the largest number of livestock would be the most contributing area to the emissions inventory. However, the management practices (especially manure handling) are a detrimental factor which if effective in mitigating the emissions, they minimize the contribution of this specific area to the national inventory. The other factor is the climatic conditions (temperature, wind velocity). On the other hand, the composition of animals' feed ration should be considered.

3) Focusing on "whole farm system approach" would be an asset. Every farm should yearly submit data and info to the agricultural and environmental authorities. It is practical to use the submitted data from each farm (e.g., number, age, weight, species of the animals as well as the implemented manure handling systems, manure field application methods, use of chemical fertilizers etc.) to develop an emission inventory for each farm and then to develop an agricultural emissions inventory to a certain geographical area based on the emissions inventories of the farms located in this area, and then the higher level onwards reaching the country level. The developed

agricultural emissions inventory should be integrated to other inventories in the same area (e.g., inventories on Energy, Industrial Processes and Product Use, Forestry, Waste, and other emissions sources) to develop the national emissions inventory of a certain pollutant.

4) The calculation procedure making use of partial emission factors for the various sources of emissions (animal house, storage, manure application, etc.) is being replaced by a mass flow concept.

5) Developing emission algorithms and then environmental software for making emissions inventories would be an asset.

6) Developing processing tools for emissions inventories, especially software systems, would be an asset.

7) Emissions of the harmful gases (NH₃, CH₄ and N₂O) should be reported annually in the national emissions inventory which should be updated annually.

8) A national emissions inventory should be compared with other countries, in terms of pollutant emissions per unit of land area or human population.

9) Inventorying the emissions of the different gases

allows developing emissions mitigation strategies.

10) The uncertainty of emissions inventories represents a challenge. Therefore, continuous evaluation and improvement of emissions inventories is essential.

11) It is insufficient to inventory the emissions, rather investigating the plume, dispersion, spatial distribution and fate of emissions is necessary to improve the emissions inventories. Afterwards, developing air quality forecasting tools would be an asset.

12) Developing emissions inventory of particulate matter (PM) will be essential in the next decade.

13) Inventorying volatile organic compounds (VOCs), odors, nitric oxide (NO), hydrogen sulfide (H₂S), and non-methane volatile organic compounds (NMVOCs) might be the state-of-the-art in the near future.

14) Understanding of the processes of CH₄, N₂O and NH₃ mass transfer from the manure and transport to the free atmosphere will contribute to development of emissions abatement techniques and housing designs, and will contribute to the reduction of gaseous emissions to the atmosphere.

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